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Influence of pollutants on activity of aerosol cloud condensation nuclei (CCN) during pollution and post-rain periods in Guangzhou, southern China

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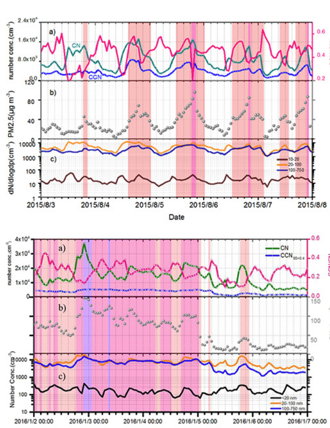
HIGHLIGHTS

- Under different levels of pollutions, particle matter ($PM_{2.5}$) and number (CN) and CCN almost showed an opposite trend to aerosol activity (CCN/CN).
- Aerosol activity (CCN/CN) did not continue to rise with increasing soluble components and $PM_{2.5}$ levels.
- Different dominated heterogeneous reactions contributed to the changes of aerosol particles both in size and compositions directly modify the aerosol activity (CCN/CN).
- The hygroscopicity of particle was eventually modified by different PM formation mechanisms both in summer and winter.

GRAPHICAL ABSTRACT



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ABSTRACT

Atmospheric pollutions have an important impact on aerosol, condensation nuclei (CN) and cloud condensation nuclei (CCN) loadings near the ground through disturbing particle size, number, chemical composition and reactions, mixing state, hygroscopicity, and so on. Aerosols and CCN were measured in urban Guangzhou during pollution and post-rain periods to examine effects of particulate pollutants on aerosol CCN activity and compare their mechanisms between summer and winter. In contrast with different levels of pollutions, particle matter ($PM_{2.5}$) and number (CN) and CCN almost showed an opposite trend to aerosol activity (CCN/CN). In summer, new particle formation (NPF) events triggered by photochemical reactions (e.g. O_3) always occurred in no-pollution daytime, and increased significantly CN and CCN as a dominant contributor to secondary aerosols.

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Under pollution conditions, the gas-to-particle transition driven by photochemical reactions guided the formation and aging processes of particles in daytime, especially in changing soluble species, whereas atmospheric oxidation and heterogeneous reactions dominated at night. In winter, stagnant weather conditions, high pollutant levels and relatively high RH were in favor of particle growing and aging through enhancing secondary particle formation and heterogeneous reactions. The wet scavenging of precipitation reduced greatly CCN amount by scouring pre-existing particles in winter, and during post-rain period the photochemical reactions did not promote the burst of secondary particle formation in the absence of ozone, compared with summer. The results may provide insights into the relationship between aerosol moisture absorption and pollution that may be useful for improving air quality.

1. Introduction

Aerosol is an important component of the atmosphere that can affect the global climate system through direct and indirect effects, and impact air quality by reducing atmospheric visibility and forming pollution in the boundary layer (IPCC, 2013). Many studies have emphasized the significant role of cloud condensation nuclei (CCN) in cloud and precipitation (PR) formation, and have also stressed their sources, compositions, and evolutionary and scavenging processes (Yum et al., 2005; Rose et al., 2008; Andreae and Rosenfeld, 2008; Seinfeld and Pandis, 2006; Rosenfeld et al., 2008; Khain, 2009; Liu et al., 2011; Yuan et al., 2017).

In general, aerosols are emitted from primary sources and produced by secondary formation through transformation of precursor gases to solid particles (Fu and Chen, 2017). The physical and chemical properties of particles are the main factors to determine aerosol activation ability that vary widely in the atmosphere, such as particle size, chemical composition, and mixing state (Yum et al., 2005; Spracklen et al., 2008; Sihto et al., 2011; Matsui et al., 2011; Wex et al., 2010). Dusek et al. (2006) proposed that aerosol cloud-nucleating ability relies on particle size more so than on chemical composition. Kuwata et al. (2008) argued that considerable variation of CCN concentrations at lower supersaturation (SS) levels may be induced by changes in chemical composition. Wang et al. (2010) analyzed the importance of detailed information on the chemical composition and mixing state of aerosols for determining particle activation properties. In addition, the response of aerosol CCN activity to different particle mixing states implies a potential contribution of anthropogenic pollutants to CCN (Lance et al., 2013; Che et al., 2016). In particular, highly diverse organic and inorganic components complicate the micro-scale properties of aerosols, such as their structural functionality, hygroscopicity, solubility and volatility (Svensmark et al., 2017; Liu et al., 2014; Arndt et al., 2017). As particles age, ambient meteorological conditions can enhance their growth and eventually change their hygroscopicity (Crosbie et al., 2015; Mallet et al., 2017). For example, high relative humidity (RH) and low air convection velocity facilitate secondary aerosol formation and thus modify particle chemistry, especially under conditions of abundant gaseous pollutants (e.g. SO_2, NO_2 and NH_3) (Wu et al., 2015; Ding et al., 2013; Kristensen et al., 2016; Wang et al., 2016). Moreover, condensed vapors encountered during the particle growth process have an effect on particle surfactant properties, affecting volume during cloud-droplet formation and changing chemical composition to influence CCN properties (Ma et al., 2016). The Raoult and Kelvin effects are well-documented explanations of the possibility of activated aerosols (Rogers and Yau, 1989).

To date, field measurements have illustrated the important relationships among aerosol, CCN and pollution in areas such as Linan (Che et al., 2016), Beijing (Gunthe et al., 2011), Hong Kong (Meng et al., 2014), Seoul (Kim et al., 2017) and Jeju Island (Kuwata et al., 2008), Amazonia (Gunthe et al., 2009) and São Paulo (Almeida et al., 2014). The mutable properties of CCN can be modified by new particle formation (NPF), particle aging and growth, and pollutants encountered in various atmospheric environments (Kalkavouras et al., 2017; Kuang et al., 2009; Kalivitis et al., 2015; Gunthe et al., 2011; Dusek et al., 2006). Yue et al. (2010) found that the NPF events dominated by sulfate and organic material (OM) formation are important contributors to CCN budget. Wiedensohler et al. (2009) reported that growing nucleation-mode particles account for up to 80% of the total CCN concentration in Beijing, in contrast with the typical dominant accumulation mode. Anthropogenic emissions can elicit changes in particle chemical composition and size distribution, and at the same time increasing particle amounts aggravate the pollution situation (Kuwata and Kondo, 2008; Wang et al., 2010; Rose et al., 2010; Kerminen et al., 2012; Leng et al., 2014). Generally, pollution results in high loading of aerosol, which impacts CCN and thus cloud cover and PR over polluted areas (Zhao et al., 2006). Measurements collected on Tai mountain demonstrated the direct effect of regional pollution on the chemical species in cloud and fog droplets, and their altitudinal differences according to long-distance transport and local air masses (Wang et al., 2012). Rosenfeld et al. (2008) described an inverse relationship between air pollution and orographic PR, indicating that a greater abundance of submicron particles suppresses the PR-forming process by acting as cloud-drop condensation nuclei.

Guangzhou, a highly populated megacity located in the core of the Pearl River Delta (PRD) region in southern China, has experienced rapid economic development and industrialization for decades. This city is currently suffering from poor air quality due to abundant anthropogenic particles and gas pollutants (Peng et al., 2014; Lai et al., 2016; Tao et al., 2017; Zheng et al., 2012). Despite improved air quality in recent years, extreme pollution or hazy days still occur during all seasons (http://www.gdep.gov.cn/). Very few studies have reported the properties of CCN influenced by pollution in southern China.

In this study, we investigated aerosols and CCN in Guangzhou during pollution and after-rain periods both in summer and winter, for the purpose of understanding how pollutants age and affect aerosol CCN activities under different atmospheric conditions, and summarizing their possible parallel transfer mechanisms. Recent studies about Chinese pollutions had revealed and come out the conclusion that the reconsideration should be taken for changes of pollution situation. Viewed solely in terms of pollutants rather than weather, our results provide insights into the relationship between aerosols and pollution that may be useful for controlling pollution.

2. Instruments and data

2.1. Observation station

Field measurements were performed in July and August 2015, and in January 2016 in Guangzhou, China. The observation station was set on the roof of a building (about 50 m above ground level) at the South China Institute of Environmental Science (SCIES), Ministry of Environmental Protection (23.07°N, 113.21°E). Detailed information about this site was provided in our previous study (Duan et al., 2017).

2.2. Measurements

A CCN counter (CCN-100; DMT, USA) equipped with a continuous 500 cm^3/min flow stream and thermal gradient was employed to