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Consistency and applicability of parameterization schemes for the size-resolved aerosol activation ratio based on field measurements in the North China Plain

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ABSTRACT

Parameterization of the size-resolved particle activation ratio (AR) is useful for the prediction and analysis of the cloud condensation nuclei (CCN) number concentration (N_{CCN}). Critical issues for the application of AR parameterizations in models are (1) the consistency of the different parameterization schemes in terms of fitting AR curves and (2) the applicability of the estimate of N_{CCN} under different aerosol conditions. These issues are discussed in this study based on summertime measurements of the size-resolved AR in the North China Plain. By comparing parameterized AR curves, variations in the fitting parameters and application to N_{CCN} calculations, the consistency of the three existing parameterization schemes is confirmed. Based on an analysis using representative AR fitting parameters, the method commonly used to calculate N_{CCN} with a fixed AR was found to be accurate, except during periods affected by strong black carbon (BC) emissions or new particle formation (NPF) events. The N_{CCN} value was overestimated by approximately 10% when BC aerosol emissions were abundant and deviated from the 1:1 line by 15% during NPF events. The bias of the calculated N_{CCN} due to these significant emission or aerosol production events cannot be eliminated by using the appropriate representative fitting parameters. Under these circumstances, accurate prediction of N_{CCN} requires real-time aerosol hygroscopicity data based on CCN measurements. This research furthers our understanding of the relationship between aerosol cloud activation and aerosol spectra.

1. Introduction

Atmospheric aerosols can form cloud droplets under supersaturated conditions and thus influence cloud microphysics through indirect aerosol effects and chemical reactions in the atmosphere (Pruppacher and Klett, 1978; Seinfeld and Pandis, 2006; Twomey, 1974; Zhao et al., 2006). Cloud activity at specific supersaturation (SS) levels is determined by particle size and hygroscopicity (Köhler, 1936; Petters and Kreidenweis, 2007). Since both the number and hygroscopicity of particles are a function of size, the size-resolved activation ratio (AR, the ratio of cloud condensation nuclei (CCN) to total particles) and the particle number size distribution (PNSD) are needed to calculate the CCN number concentration (N_{CCN}) (Deng et al., 2013; Juranyi et al., 2010; Rose et al., 2010). Measurements of the PNSD are relatively easy to make and are commonly performed, while measurements of the size-resolved AR are difficult, often expensive, and insufficient for studies of aerosol-cloud interactions (Dusek et al., 2006; Hudson, 2007; Hudson and Da, 1996; Paramonov et al., 2015). However, size-resolved AR can be used to infer the aerosol chemical composition and mixing state because it contains information about the variation in aerosol hygroscopicity (e.g., (Good et al., 2010; Lance et al., 2013; Padro et al., 2012; Rose et al., 2011; Su et al., 2010).

Since a size-resolved AR curve usually has a simple sigmoidal shape, the size-resolved AR can be approximated by a fitting formula with three parameters. These parameters represent the proportion of CCN to total particles, the midpoint activation diameter (D_a) of the CCN and the deviation of the CCN D_a. These are the parameters needed to describe the aerosol activation ability (Gunthe et al., 2011; Padro et al., 2012; Rose et al., 2008). These AR parameters were found to be in good agreement with measurements of aerosol hygroscopicity and mixing state measured by other instruments and were applied to study the diurnal cycles of aerosol properties (Rose et al., 2011). This agreement between the AR parameters and aerosol properties measured by other instruments were also found in other studies around the world (Kawana

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et al., 2016; Mei et al., 2013; Ogawa et al., 2016). Thus, these AR parameters are effective for studying variations in aerosol properties. At present, three formulas (details of these formulas can be found in Section 2.2) are widely used in CCN measurements studies (Deng et al., 2013; Padro et al., 2012; Rose et al., 2008). To better understand the results of previous studies using different formulas, a consistent investigation of these three formulas is required. Additionally, comparisons among the three formulas will help future studies make reasonable formula choices.

A widely used practical method for determining N_{CCN} is to use the real-time PNSD measured during the same campaign and the size-resolved AR measured under similar aerosol conditions when no direct CCN measurements are available. In earlier CCN studies, the bulk AR (the ratio between N_{CCN} and the aerosol number concentration) was used as a proxy for aerosol hygroscopicity to calculate N_{CCN} with particle number concentrations (Pruppacher and Klett, 1997). However, the bulk AR is found to be quite diverse even though the overall aerosol hygroscopicity is similar in these studies (Hitzenberger et al., 2003; Burkart et al., 2011; Crosbie et al., 2015). This is because the bulk AR is a function of both aerosol hygroscopicity and PNSD and is sensitive to variation in PNSD (Dusek et al., 2006; Quinn et al., 2008; Deng et al., 2011; Morales Betancourt and Nenes, 2014). N_{CCN} can be calculated with PNSD in combination with activation parameters that represent hygroscopicity and that are independent of PNSD: critical diameter, $D_{P,cri}$ (particles larger than $D_{P,cri}$ are all activated, suitable for internally mixed particles) or the size-resolved AR (chemical compositions of particles are assumed to be externally mixed and size dependent). The variations in these activation parameters due to fluctuations in aerosol hygroscopicity are generally small, and using the representative activation parameter under some pollution conditions is effective for determining N_{CCN} from real-time PNSD measurements (e.g., Deng et al., 2013; Good et al., 2010; Irwin et al., 2011; Mochida et al., 2011; Paramonov et al., 2013; Rose et al., 2010). For continental aerosols, a fixed hygroscopicity parameter of 0.3 is usually used to calculate $D_{P,cri}$, which results in agreement between the measured N_{CCN} values and the calculated N_{CCN} values based on real-time PNSD (Andreae and Rosenfeld, 2008; Gunthe et al., 2009; Rose et al., 2010), except during periods of intense emissions or aerosol production. During new particle formation (NPF) events, secondary aerosols (SAs) can dominate the aerosol population and can lead to significant changes in aerosol hygroscopicity and the aerosol mixing state (Dusek et al., 2010; Kerminen et al., 2012; Lance et al., 2013; Levin et al., 2014; Wiedensohler et al., 2009). When the emission of BC particles is strong, the proportion of inactive particles can increase, and the effect of the aerosol mixing state on N_{CCN} is not negligible (Gunthe et al., 2011; Rose et al., 2011; Wang et al., 2010; Wex et al., 2010). In these cases, both the hygroscopicity and the mixing state of the aerosols must be specified for the precise calculation of N_{CCN} (Deng et al., 2011; Juranyi et al., 2013; Meng et al., 2014; Padro et al., 2012), and representative size-resolved AR curves are necessary for estimating N_{CCN} (Wang et al., 2013). However, in recent studies of strong emissions and production of aerosol particles in the North China Plain, significant and diverse variations of aerosol chemical composition and mixing state were found. This variability can result in large fluctuations in aerosol hygroscopicity (Cheng et al., 2009; Ma et al., 2012, 2016; Yue et al., 2011), making the use of the average size-resolved AR inappropriate for N_{CCN} calculation (Ma et al., 2016). Thus, the applicability of the average aerosol activity for N_{CCN} calculation under various aerosol conditions requires examination to improve the N_{CCN} predictions based on PNSD measurements. Based on field measurements in the North China Plain, the consistency of three parameterization schemes is tested by comparing their effectiveness at fitting AR data and their corresponding fitting parameters. Then, the applicability of calculating N_{CCN} from average parameterized AR is examined under various conditions in order to provide suggestions for improving N_{CCN} predictions based on analysis of the sensitivity of the calculated N_{CCN} to AR parameters.

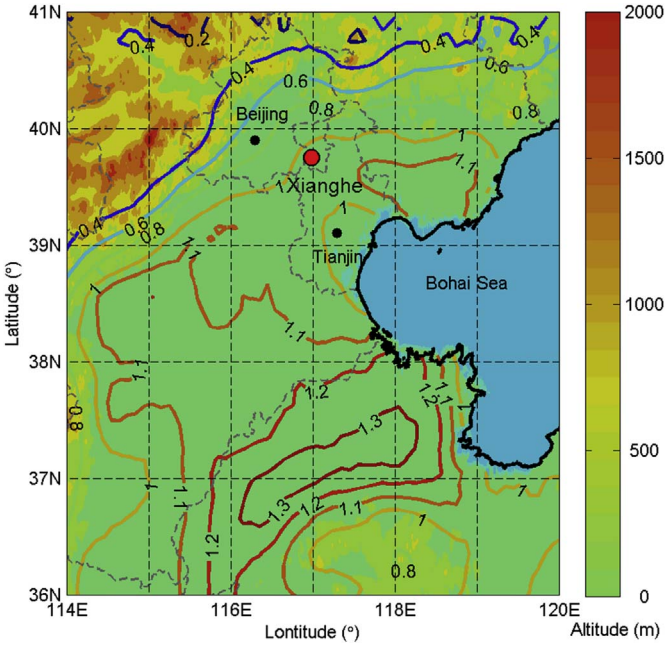

Fig. 1. A map of the North China Plain. Color represents elevation, and the contours represent the average aerosol optical depth in summer in 2012 and 2013 from MODIS (Moderate Resolution Imaging Spectroradiometer) data.

2. Methodology

2.1. Data

PNSD and aerosol size-resolved AR were measured at Xianghe station from July 22nd to August 30th, 2012, and from July 20th to August 9th, 2013. Since Xianghe is a regional station, aerosol properties observed there can represent the background aerosol characteristics of the North China Plain, as shown in Fig. 1. The North China Plain is surrounded by mountains to the west and north and by the Bohai Sea to the east. Farmland is common on the North China Plain. There are also factories, transportation networks and megacities, such as Beijing and Tianjin, that generate severe aerosol pollution conditions. More information about this station and its surrounding area can be found in Ma et al. (2016). Aerosols were sampled by a PM10 (particulate matter with aerodynamic diameter less than 10 μm) inlet, dried with a system that is capable of lowering the relative humidity to less than 30%, and measured by instruments placed within the container with temperature maintained at about 22 °C. An Aerodynamic Particle Sizer (APS Model 3320, TSI, USA) together with a Scanning Mobility Particle Sizer (SMPS, Model 3936, TSI, USA) measured PNSD between approximately 10 nm and 10 μm. Aerosol size-resolved AR curves were measured by a DMA-CCNC system.

The DMA-CCNC system consisted of an electrostatic classifier (Model 3080, TSI, USA), a condensation particle counter (CPC, Model 3772, TSI, USA), and a continuous-flow dual CCN counter (Model CCN200, Droplet Measurement Technologies, USA (Lance et al., 2006; Roberts and Nenes, 2005)). As described by Deng et al. (2013) and Ma et al. (2016), monodisperse aerosols classified by the electrostatic classifier were counted by the CPC and activated in the CCN counter. Size-resolved N_{CCN} values were measured sequentially at five supersaturations (SSs) of 0.07%, 0.10%, 0.20%, 0.40% and 0.80% at a temporal resolution of 1 h. Based on a modified algorithm (Deng et al., 2011, 2013), size-resolved aerosol AR curves can be inverted from the matched data of the CPC and CCN counter. Before and after the campaign, the sheath and sample flow rates, as well as the supersaturation of the CCN counter, were calibrated. The PNSD and the size-resolved AR were multiplied and integrated to obtain N_{CCN} at each SS: