A New Method for Distinguishing Unactivated Particles in Cloud Condensation Nuclei Measurements: Implications for Aerosol Indirect Effect Evaluation

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Abstract An ongoing challenge for cloud condensation nuclei (CCN) measurements is the inclusion of unactivated particles, which affects droplet activation parameterizations and aerosol indirect effects in models. For the first time, a reciprocal relationship between the critical diameter and critical supersaturation of activated droplets is derived from the Köhler theory to derive accurately count unactivated particles in CCN measurements. We conducted 4-day continuous observations to simultaneously measure the number concentration of CCN (NCCN) and aerosol. The results show that as supersaturation (SS) decreases from 0.186% at 25 °C, the proportion of the unactivated particles in the CCN measurements increases, reaching 88% at SS of 0.07%. Owing to the NCCN overestimation caused by NCCN-SS parameterizations with uncorrected NCCN, there is significant overestimation of aerosol indirect effects, especially under low SS conditions. After removing unactivated particles, NCCN-SS parameterizations are adjusted accordingly, which can improve simulations of aerosol-cloud-radiation interactions in models.

1. Introduction

By serving as cloud condensation nuclei (CCN), aerosols play a vital role in precipitation, radiation, and climate (Koren et al., 2014; Ramanathan et al., 2001; Rosenfeld et al., 2008; Zhang et al., 2007). Aerosol-cloud interactions (ACIs), which are currently a topic of enhanced research interest (Y. Wang, Wang et al., 2014; Y. Wang, KeunHee, 2014; Fan et al., 2018), represent the largest uncertainty in predictions of climate change (Intergovernmental Panel on Climate Change, 2013). To explore and understand the issues involving ACI, many problems need to be studied in detail, including the Twomey effect (Twomey, 1974, 1977), the Albrecht effect (Albrecht, 1989), and the dispersion effect (Liu & Daum, 2002). Essentially, one of the crucial underlying challenges is to determine the ability of aerosol particles to activate as cloud droplets, which has drawn increasing attention in recent years (Hiranuma et al., 2011; Kawana et al., 2017; Paramonov et al., 2013; Prisle et al., 2019). Hyperscopic aerosols that absorb water from the atmosphere and help initiate cloud formation are termed CCN (Lamb & Verlinde, 2011); therefore, the number concentration of CCN (NCCN) describes the ability of the aerosol population to be activated as cloud droplets. The study of ACI is in many ways an analysis of the relationship between CCN and cloud properties.

Meteorologists have long recognized the importance of CCN, and, as early as the 1960s, Twomey (1963) introduced the measurements of NCCN at a single supersaturation (SS) using a thermal-gradient diffusion cloud chamber (or static diffusion chamber). The continuous-flow parallel plate thermal diffusion chamber emerged in the 1970s (Sinnarwalla & Alofs, 1973). However, the reliability of the measurement results continues to be debated (Chuang et al., 2000; Hudson, 1983, 1989). In the 2000s, the continuous-flow streamwise thermal-gradient CCN counter from Droplet Measurement Technologies (DMT-CCNC) became commercially available with enhanced robustness and reliability (Roberts & Nenes, 2005). Based on DMT-CCNC measurements, scientists have conducted extensive field observations and laboratory experiments for CCN around the world and obtained many valuable results (e.g., Ma et al., 2016; Mochida et al., 2011; Quinn et al., 2017; Yum et al., 2007; Zhang et al., 2012).
However, unactivated particles, which are not authentic CCN at the specific SS, are included in the measurements of CCN at low SS conditions (Roberts & Nenes, 2005). The overestimation of $N_{CCN}$ caused by unactivated particles results in underestimation of droplet radii, including the effective radius and volume weighted mean radius when liquid water content is constant (Albrecht, 1989; Twomey, 1974, 1977). The underestimation of droplet radii influences cloud radiative properties and the cloud-to-rain autoconversion process, which leads to a large uncertainty in simulations of aerosol-cloud-radiation interactions in weather and climate models (Fan et al., 2012; Quaas et al., 2006). Removal of unactivated particles in CCN measurements is therefore necessary to reduce uncertainty in models. Although this problem has long been recognized, this remains an area of active research. A common approach is to consider measurements from the optical particle counter (OPC) of less than 1 $\mu$m as unactivated particles and remove them from the data (Roberts & Nenes, 2005; Rose et al., 2008). However, this method is empirical; more accurate theoretical solutions are also needed.

For the first time, we derive the reciprocal relationship between the critical diameter ($D_c$) and critical supersaturation ($SS_c$) of activated droplets to distinguish unactivated particles from activated CCN. After correcting CCN measurements, $N_{CCN}$-$SS$ parameterizations (also called as CCN activation parameterizations) are adjusted accordingly. Then, the effects of $N_{CCN}$-$SS$ parameterizations with corrected and uncorrected $N_{CCN}$ on the aerosol indirect effects are evaluated. The remainder of this paper is organized as follows. Section 2 introduces the theoretical derivation of the reciprocal relationship between $D_c$ and $SS_c$. Section 3 describes the experimental setting. Section 4 presents the experimental results and implications for the aerosol indirect effects. Conclusions are summarized in section 5.

2. Theoretical Derivation

The Köhler theory predicts the supersaturation required to activate a particle to a cloud droplet. Petters and Kreidenweis (2007) presented a method to describe the relationship between particle dry diameter ($D_d$) and CCN activity using a single hygroscopicity parameter, $\kappa$, and expanded the Köhler theory to the $\kappa$-Köhler theory:

$$S_k(D) = \frac{D^3 - D_d^3}{D^3 - (1-\kappa)D_d^3} \exp\left(\frac{A}{D}\right),$$  \hspace{1cm} (1a)

with

$$A = \frac{4\sigma_w M_w}{RT \rho_w},$$  \hspace{1cm} (1b)

where $S_k$ is the saturation ratio, $D$ is the droplet diameter, $\sigma_w$ is the surface tension of water, $M_w$ is the molecular weight of water, $R$ is the universal gas constant, $T$ is the air temperature, $\rho_w$ is the density of water, and $A$ can be considered a function of $T$. This method facilitates studying the activation process without considering the complex chemical compositions of aerosols (McFiggans et al., 2006). If we perform a first-order Taylor expansion on the right side of Eq. (1a) and ignore the higher order terms, while assuming that $D$ is much larger than $D_d$, we can derive equation (2):

$$S_k(D) = 1 + \frac{A}{D} \frac{\kappa D_d^3}{D^3}.$$  \hspace{1cm} (2)

After differentiating equation (2), $D_c$ and the critical saturation ratio ($S_c$) can be obtained as

$$D_c = \left(\frac{\kappa D_d^3}{A}\right)^{1/2},$$  \hspace{1cm} (3a)

$$S_c = 1 + \frac{2A}{3} \left(\frac{A}{\kappa D_d^3}\right)^{1/2}.$$  \hspace{1cm} (3b)
Subsequently, a reciprocal relationship between $D_c$ and $SS_c$ (equal to $(S_\circ - 1) \times 100\%$), which is a slowly varying function ($F(T)$) of $T$ (see details in the supporting information), becomes evident:

$$D_c \times SS_c = \frac{200A}{3} = F(T).$$

(4)

3. Experimental Setting

Droplets smaller than $D_c$ are haze and are considered as unactivated particles, whereas larger droplets are considered as activated droplets. Activation is the process by which unactivated particles grow through the peak of the Köhler curve and are transformed into cloud droplets. Based on the relationship between $D_c$ and $SS_c$, the CCN measurements are corrected. In the DMT-CCNC, dry aerosols undergo hygroscopic growth and condensational growth in a supersaturated column; the postgrowth spectral distribution is measured by an OPC with a measurement range of 750 to 10,000 nm. The OPC, which uses side-scattering technology, counts and sizes postgrowth droplets and particles; therefore, this method cannot distinguish between activated droplets and unactivated particles. As $F(T)$ is not sensitive to $T$, we use $T = 25$ °C as the standard with $F(T) = 139.83$ nm%. When the supersaturation is $0.1\%$ at $T = 25$ °C, $D_c$ is $1398$ nm according to equation (4). This means that only a droplet with the diameter greater than $1398$ nm is a real CCN at $0.1\%$ supersaturation. When $D_c$ is $750$ nm, $SS_c$ equals $0.186\%$ at $T = 25$ °C. Therefore, unactivated particles do not influence CCN measurements when the supersaturation is greater than $0.186\%$ at $T = 25$ °C. Conversely, when supersaturation is less than $0.186\%$, it is vital to correct the data of DMT-CCNC according to the reciprocal relationship between $D_c$ and $SS_c$ combined with the spectral distribution measured by OPC.

To evaluate the effect of unactivated particles on CCN measurements, the DMT-CCNC was first calibrated according to the manufacturer’s specification, including calibrations of supersaturation, flow, pressure, and OPC. Considering that the unactivated particles are measured by OPC when the $SS$ is less than $0.186\%$ at $25$ °C, we made changes in the supersaturation calibration. Only the data when the $SS$ is greater than $0.186\%$ were applied to the linear fitting, and the fitting result served as the $SS$ calibration line (see details in the supporting information). By this method, the influence of unactivated particles was removed in the supersaturation calibration.

After calibrating DMT-CCNC, an experiment was designed to quantify the effect of unactivated particles, growing via moisture absorption, on CCN measurements. The experimental setup is shown in Figure 1. Aerosol size distributions were measured using a Model 1000XP Wide-Range Particle Spectrometer (WPS-1000 XP, MSP Corporation, USA). The WPS-1000XP includes a neutralizer, a differential mobility analyzer, a condensation particle counter, and a laser particle spectrometer to measure the diameter and number concentration of aerosol particles in the $10$ to $10^4$ nm particle diameter range.

Experiments were performed in the laboratory of Nanjing University of Information Science and Technology (32.2°N, 118.7°E, 22 m above sea level), Nanjing, China. One end of the black carbon tube was extended out of the laboratory, and the other was connected to the instrument to measure the ambient atmosphere. The sampling site has a high aerosol background value (H. Wang, An, et al., 2014; H. Wang, Zhu, et al., 2014), which could cause a significant deviation in the counting rate of the DMT-CCNC owing to water depletion (Deng et al., 2011), making measured $N_{CCN}$ lower than actual $N_{CCN}$ (Wang et al., 2018). Therefore, the ambient atmosphere was diluted using clean air that had been passed through a filter (model number 9933-11-BQ, Parker Corporation). Two controllable air valves were used to control the intake airflow of the ambient air and the clean air; that is, the mixing ratio or the dilution ratio. The ratio of the ambient air to the clean air was $1:2$ to ensure that the total aerosol concentration entering the DMT-CCNC was below $6,000$ cm$^{-3}$ in most cases. The diluted ambient air was then dried to lower than $40\%$ humidity using a Nafion dryer. Finally, the air carrying dry aerosols was divided into two parts, entering WPS-1000XP and DMT-CCNC, respectively, to measure the dry aerosol spectrum and $N_{CCN}$. A total of $10$ different $SS$ conditions was set in DMT-CCNC, which were $0.07\%$, $0.1\%$, $0.15\%$, $0.2\%$, $0.3\%$, $0.4\%$, $0.5\%$, $0.7\%$, $0.9\%$, and $1.0\%$, respectively. Among these $SS$ conditions, only the measurement with $SS = 0.07\%$ was set for $15$ min; the others were set for $5$ min each, and so a complete $SS$ cycle was $1$ hr. Implementing this experimental procedure, we conducted $4$-day continuous observations from $30$ June to $4$ July 2019.
4. Results

4.1. Experimental Results

The time series of the 4-day measurements of WPS-1000XP and DMT-CCNC are shown in Figure 2, including the total aerosol number concentration in the range of 10 to 10^4 nm (N_{aerosol}) and N_{CCN,SS} at different SS conditions (N_{CCN,SS}). Note that the measurements shown in Figure 2 reflect the diluted air, and the actual N_{aerosol} and N_{CCN,SS} are 3 times as high as these values because of the 1:2 dilution ratio. The same applies to the following. As shown in Figure 2, N_{CCN} increases with increasing SS but is always lower than N_{aerosol}. The closed results reflect the high confidence of the measurements. In addition, to ensure the accuracy of the CCN measurements, only data with N_{aerosol} of less than 6,000 cm^{-3} are used for analysis.

Based on the relationship between D_c and SS_c (equation (4)), DMT-CCNC data are corrected at conditions of SS lower than 0.186%. As introduced in section 2, unactivated particles (diameter smaller than D_c) are removed from particle and droplet size spectra measured by OPC in DMT-CCNC; the corrected results (N_{CCN,SS}^{COR}) are indicated by the dashed lines in Figure 2. One issue that needs to be tackled is how to eliminate unactivated particles if D_c is not equal to the lower or upper diameter of each bin in the droplet size spectra. Linear interpolation is applied to solve this problem. For instance, when SS is 0.1%, D_c is 1,398 nm, which is in the second bin with a range from 1,000 to 1,500 nm; the number of unactivated particles that need to be deleted is the sum of the concentration of the first bin and 79.6% of the concentration of the second bin.

The fractions of DMT-CCNC measurements accounted for by unactivated particles are 88%, 42%, and 16% at SS of 0.07%, 0.1%, and 0.15%, respectively. As SS increases, the proportion of unactivated particles decreases. Therefore, N_{CCN} is overestimated significantly in DMT-CCNC measurements at low SS conditions. After removing unactivated particles in the CCN measurements, corrected N_{CCN} is obtained.

These corrected results are then fitted using three different equations commonly used in models. The typical N_{CCN}-SS parameterization is the Twomey’s power law equation (Twomey, 1959): N_{CCN} = C_1 \times SS^{k_1}, where C_1 corresponds to the N_{CCN} at SS = 1% and k_1 is a parameter that varies significantly. Ji and Shaw (1998) derived another parameterization: N_{CCN} = N(1-\exp(-B\times SS^{k_2})), where N is the total N_{CCN}, and parameters B and k_2 are empirical coefficients to be determined. A more general parameterization was suggested by Cohard et al. (1998): N_{CCN} = C_2 \times SS^{k_3} \times G(\mu, \frac{k_4}{2}, \frac{k_5}{2}, 1, -\gamma SS^2), where C_2 is...
proportional to $N_{CCN}$ that would be activated at an infinite SS condition, $G$ is a hypergeometric function, and $k_3$, $\mu$, and $\gamma$ are adjustable parameters depending on the aerosol properties. As shown in Figure 3, all three parameterizations with corrected $N_{CCN}$ show lower lines than those with uncorrected $N_{CCN}$ at low SS conditions. The parameterizations of Cohard et al. (1998) (blue) and Ji and Shaw (1998) (green) have similar fitting results, both of which are better than the results using the Twomey's power law equation (red).

### 4.2. Implication for Aerosol Indirect Effect Evaluation

Cloud droplet concentration ($N_c$) at cloud base is calculated using these $N_{CCN}$-SS parameterizations in many models (Fan et al., 2012; Khain, 2009; Nenes & Seinfeld, 2003). After correcting $N_{CCN}$, these $N_{CCN}$-SS parameterizations are adjusted accordingly (Figure 3). As shown in Figure 4a, compared with the parameterizations with corrected $N_{CCN}$, parameterizations with uncorrected $N_{CCN}$ significantly overestimate $N_{CCN}$, especially at low SS conditions. For instance, at SS of 0.05%, 0.1%, and 0.2%, the maximum overestimations of $N_{CCN}$ among the three parameterizations are 61%, 33%, and 12%, respectively. Therefore, it is necessary to evaluate the deviations of cloud microphysical and radiative properties caused by uncorrected $N_{CCN}$-SS parameterizations.

Cloud optical thickness ($\tau$) and single scatter albedo ($\omega_0$) are crucial to estimate the Twomey effect (Stephens, 1984; Twomey & Bohren, 1980) and can be expressed by

$$\tau \approx \frac{3}{2} W r_e^{-1},$$

$$1 - \omega_0 = 1.7 k_w r_e,$$

where $W$ is the liquid water path, $r_e$ is the effective radius of cloud droplets, and $k_w$ is the complex part of the refractive index of water. Equations (5) and (6) indicate that the Twomey effect is strongly dependent on $r_e$. Meanwhile $r_e$ is proportional to the volume weighted mean radius of cloud droplets ($r_v$) (Bower & Choularton, 1992) and can be expressed by

$$r_e = \beta \left( \frac{3q}{4\pi \rho_w N_c} \right)^{1/3} = \beta r_v,$$

where $q$ is the cloud liquid water content and $\beta$ is the scaling factor. Many studies have analyzed factors affecting $\beta$ and developed parameterizations of $\beta$ (Peng & Lohmann, 2003; Liu et al., 2008; Rostayn & Liu, 2009). Here, to focus on the effect of $N_c$ on $r_e$, $\beta$ is specified as a fixed parameter, as assumed in many climate models (Quaas et al., 2004).
According to the above equations, we first evaluate the effect of $N_{CCN}$ overestimation on the first aerosol indirect effect. As shown in Figure 4b, parameterizations with uncorrected $N_{CCN}$ significantly underestimate $\tau$ at low $SS$, which leads to overestimation of $1-\omega_0$. Both of the overestimation of $\tau$ and the underestimation of $1-\omega_0$ increase as SS decreases. For instance, at SS of 0.05%, 0.1%, and 0.2%, the maximum overestimations of $\tau$ among the three parameterizations are 17%, 10%, and 4%, respectively, and the maximum underestimations of $1-\omega_0$ are 15%, 9%, and 4%, respectively. Therefore, the overestimation by the Twomey effect is significant at low SS conditions.

In addition, parameterizations of the cloud-to-rain autoconversion process play a critical role in determining the second aerosol indirect effect (Wang et al., 2012). According to Liu et al. (2004, 2005), parameterization of the autoconversion process can be expressed by

$$P = T \times P_0,$$

where $P$ is the autoconversion rate, $P_0$ is the rate function describing the conversion rate after the onset of the autoconversion process, and $T$ is a function describing the threshold behavior of the autoconversion process. Meanwhile, $T$ can be expressed by

$$T = \frac{\int_{r_c}^{\infty} r^6 n(r) dr}{\int_{0}^{\infty} r^6 n(r) dr} \frac{\int_{r_c}^{\infty} r^3 n(r) dr}{\int_{0}^{\infty} r^3 n(r) dr},$$

where $r$ is the droplet radius, $n(r)$ is the cloud droplet size distribution, and $r_c$ is the critical radius of autoconversion process (note that $r_c$ is not related to $D_c$). The $T$ ranges from 0 to 1, with a larger $T$ indicating a
greater probability that the collision process occurs in clouds. Liu et al. (2006) derived the analytical expression of $r_c$ as follows:

$$r_c \approx 4.09 \times 10^{-4} \beta_{\text{con}}^{1/6} N_c^{1/6} q^{-1/3},$$

(10)

where $\beta_{\text{con}} = 1.15 \times 10^{23}$ is an empirical constant.

According to equations (9) and (10), we also evaluate the effect of the $N_{\text{CCN}}$ overestimation on the second aerosol indirect effect; Figure 4c shows an example. Cloud droplet size distributions were measured by a fast cloud droplet probe in seven flights in 2018 (26 January, 29 January, 30 January, 1 February, 7 February, 10 February, and 12 February) during the Aerosol and Cloud Experiments in the Eastern North Atlantic. The mean cloud droplet size distribution is fitted with a $\Gamma$ distribution: $n(r) = 0.25r^3\exp(-0.61r)$ with $R^2 = 0.89$. For this mean size distribution, $r_c$ is 11 $\mu$m and $TA$ is 0.06. As shown in Figure 4c, parameterizations with uncorrected $N_{\text{CCN}}$ overestimate $r_c$ and underestimate $TA$ at low SS, which means that precipitation initiation is delayed. At SS of 0.05%, 0.1%, and 0.2%, the maximum overestimations of $r_c$ among the three parameterizations are 8%, 5%, and 2%, respectively, and the maximum underestimations of $TA$ are 51%, 34%, and 14%, respectively. Therefore, owing to parameterizations with uncorrected $N_{\text{CCN}}$, the lifetime of clouds is overestimated and the radiation cooling effect caused by the Albrecht effect is also overestimated, especially for clouds at low SS conditions.

In summary, correction of $N_{\text{CCN}}$ and improvement of $N_{\text{CCN}}$-SS parameterizations could improve simulations of the aerosol effects on cloud microphysics and radiative properties (Fan et al., 2012; Zhao et al., 2012; Connolly et al., 2013; Wang et al., 2011; Y. Wang, KeunHee, et al., 2014). For instance, Fan et al. (2012)
showed that, compared with spectral-bin microphysics, two-moment bulk microphysics predicted much higher \( N_c \) and lower rain rates. In addition to the stronger convection predicted by bulk microphysics, overestimation of \( N_{\text{CCN}} \) caused by uncorrected \( N_{\text{CCN}}\)-SS parameterizations in bulk microphysics could be another important reason.

5. Conclusions

Aerosols serving as CCN play a vital role in the formation of clouds, thereby influencing precipitation, radiation, balance, and climate. However, unactivated particles are always present in the measurements of CCN, which is a source of uncertainty in describing cloud microphysics and cloud radiative properties. In this study, for the first time, we derived the reciprocal relationship between the \( D_c \) and \( SS_c \) of the activated droplet from the \( \kappa \)-Köhler theory; based on this relationship, the effects of unactivated particles in DMT-CCNC measurements were removed. When \( SS_c \) is greater than 0.186% at \( T \) of 25 °C, \( D_c \) is smaller than the OPC-identifiable lower limit diameter of 750 nm, which implies that there is no measurable influence of unactivated particles in the CCN measurements. Therefore, before evaluating the effect of unactivated particles on CCN measurements at low SS, we calibrated SS at SS greater than 0.186%.

After calibrations, we conducted 4-day continuous observations (30 June to 4 July 2019) in Nanjing, China, to simultaneously measure \( N_{\text{CCN}} \) and aerosol particle size distribution ranging from 10 to \( 10^4 \) nm. As SS decreases, the proportion of unactivated particles in the CCN measurements increases; at SS of 0.07%, 0.1%, and 0.15%, the proportions of the unactivated particles are 88%, 42%, and 16%, respectively. Therefore, \( N_{\text{CCN}} \) is overestimated significantly in DMT-CCNC measurements at low SS conditions. After removing unactivated particles in the CCN measurements, we obtain the corrected \( N_{\text{CCN}} \), which are then fitted using three different equations commonly used in models. The parameterizations of Cohard et al. (1998) and Ji and Shaw (1998) show similar fitting results, both of which are better than those obtained by applying Twomey’s power law equation.

Compared with \( N_{\text{CCN}}\)-SS parameterizations with corrected \( N_{\text{CCN}} \) parameterizations with uncorrected \( N_{\text{CCN}} \) significantly overestimate \( N_{\text{CCN}} \), especially at low SS conditions. Based on previous theoretical derivations, overestimation of \( N_{\text{CCN}} \) results in underestimation of the \( r_p \), 1-\( \omega_p \), and \( T_A \), which means overestimation of the cooling effect in the Twomey effect and the Albrecht effect. Meanwhile, overestimation of the corresponding radiative forcing increases as SS decreases. It is well-known that aerosol indirect effects are often overestimated in models (Fan et al., 2012; Quaas et al., 2006; Ruckstuhl et al., 2010), the correction of \( N_{\text{CCN}} \) and the improvement of \( N_{\text{CCN}}\)-SS parameterizations shed new light on this problem, especially with cloud regions under low SS conditions.

It is worth noting that the aerosols with different properties (e.g., particle size distribution, chemical composition, and mixed state) will lead to some difference in the shape of the spectrum measured by the OPC after growing in the supersaturated column, thus the fraction of DMT-CCNC measurements accounted for by the unactivated particles varies with these aerosol properties. For instance, the proportions of the unactivated particles measuring in ammonium sulfate (\( \kappa = 0.61 \) and sodium chloride (\( \kappa = 1.28 \)) lab experiments and ambient air are not identical (see details in the supporting information). Therefore, it is necessary to study the correction of CCN activation parameterization under different aerosol background regions in the future.

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