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Atmospheric Measurement Techniques Discussions

A New Method for Calculating Number Concentrations of Cloud Condensation Nulcei Based on Measurements of A Three-wavelength Humidified Nephelometer System

Jiangchuan Tao[1], Chunsheng Zhao[1], Ye Kuang[1], Gang Zhao[1], Chuanyang Shen[1], Yingli Yu[1], Yuxuan Bian[2], Wanyun Xu[2]

[1]{Department of Atmospheric and Oceanic Sciences, School of Physics, Peking University, Beijing, China}

[2]{State Key Laboratory of Severe Weather, Chinese Academy of Meteorological Sciences}

*Correspondence to: C. S. Zhao (zcs@pku.edu.cn)

Abstract

The number concentration of cloud condensation nuclei (CCN) plays a fundamental role in cloud physics. Instrumentations of direct measurements of CCN number concentration (N_{CCN}) based on chamber technology are complex and costly, thus a simple way for measuring N_{CCN} is needed. In this study, a new method for N_{CCN} calculation based on measurements of a three-wavelength humidified nephelometer system is proposed. A three-wavelength humidified nephelometer system can measure aerosol light scattering coefficient (σ_{sp}) at three wavelengths and the light scattering enhancement factor (fRH). The Angstrom exponent (Å) inferred from σ_{sp} at three wavelengths provides information on mean predominate aerosol size and hygroscopicity parameter (κ) can be calculated from the combination of fRH and Å. Given this, a look-up table that involves σ_{sp}, κ and Å is established to predict N_{CCN}. This method is validated with direct measurements of N_{CCN} using a CCN counter on the North China Plain. Results show that relative deviations between calculated N_{CCN} and measured N_{CCN} are within 30% and confirm the robustness of this method. This method enables simpler N_{CCN} measurements because the humidified nephelometer system is easily operated and stable. Compared with the method of CCN counter, another advantage of this newly proposed method is that it can obtain N_{CCN} at lower supersaturations in the ambient atmosphere.

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1. Introduction

Cloud condensation nuclei (CCN) is the aerosol particle forming cloud droplet by hygroscopic growth. CCN number concentration (N_{CCN}) plays a fundamental role in cloud micro physics and aerosol indirect radiative effect. In general, the direct measurement of N_{CCN} is achieved in a cloud chamber under super-saturated conditions (Hudson, 1989;Nenes et al., 2001;Rose et al., 2008). Due to the requirement of high accuracies of working conditions like temperatures, vapors and flow rates in cloud chambers, the direct measurement of N_{CCN} is complex and costly (Rose et al., 2008;Lathem and Nenes, 2011). Thus, developments of simplified measurements of N_{CCN} are required. In recent years, attention has been focused on measurements of aerosol optical properties (Jefferson, 2010;Ervens et al., 2007;Gasso and Hegg, 2003), which are simple and well-developed (Covert et al., 1972;Titos et al., 2016). For aerosol population free of sea salt or dust, the accumulation mode aerosol not only dominates aerosol scattering ability but also contribute most to N_{CCN}. Thus, the calculation of N_{CCN} based on measurements of aerosol optical properties is feasible, and can facilitate N_{CCN} measurement.

There are two kinds of methods to calculating N_{CCN} based on measurements of aerosol optical properties. For the first kind, N_{CCN} as well as the hygroscopicity parameter (κ) can be calculated based on measurements of a humidified nephelometer system in combination with aerosol particle number size distribution (PNSD) (Ervens et al., 2007;Chen et al., 2014). Thus additional measurements of PNSD are needed. For the second kind, N_{CCN} is calculated based on statistical relationships between N_{CCN} and aerosol optical properties, such as scattering coefficient (σ_{sp}), Angstrom Exponent (Å) and single scattering albedo (SSA) (Jefferson, 2010;Shinozuka et al., 2015). Compared with the first kind, instruments used in the second kind of methods are cheaper and easier in operation. Applications similar to the second kind are widely used in remote sensing. Earlier studies found that the aerosol volume or aerosol PNSD retrieved from remote sensing measurements can be used to calculate N_{CCN} (Gasso and Hegg, 2003;Kapustin et al., 2006). Recently, aerosol optical depth (AOD) or aerosol vertical profile is used to predict N_{CCN} directly(Ghan and Collins, 2004;Ghan et al., 2006;Andreae, 2009;Liu and Li, 2014).

In the statistical relationship between N_{CCN} and aerosol optical properties, σ_{sp} or AOD is mainly the proxy of aerosol absolute concentration, while Å or SSA can be used to reveal the variations of aerosol CCN activity. Based on Kohler theory (Köhler, 1936;Petters and Kreidenweis, 2007), aerosol CCN activity is determined by aerosol size and aerosol chemical composition which is defined as aerosol hygroscopicity. Information about aerosol size and aerosol hygroscopicity are critical to N_{CCN} prediction and their absence can lead to a deviation with factor of four (Andreae, 2009). Compared with aerosol hygroscopicity, aerosol size is more important in determining CCN activity (Dusek et al., 2006). The value of Å can provide information on mean predominate aerosol size (Brock et al., 2016;Kuang et al., 2017). As a result, N_{CCN} calculation from Å and extinction coefficient is found to be accurate to some extent (Shinozuka et al., 2015). As proxies for aerosol hygroscopicity, SSA or aerosol light scattering enhancement factor (fRH) is commonly used while not so effective. SSA is determined by the ratio between the light absorbing carbonaceous and less-absorbing components. Black carbon contributes most to the light absorbing carbonaceous and is the most important hydrophobic compositions as well. Less-absorbing components consist of inorganic salts and acids, as well as most organic compounds, which are generally hygroscopic compositions. SSA correlates positively with aerosol hygroscopicity (Rose et al., 2010) but deviates significantly due to the diversity of hygroscopicity of less-absorbing components. Thus deviations of N_{CCN} calculation based on SSA is of large errors (Jefferson, 2010). Compared with SSA, previous studies found fRH to be less effective in estimating N_{CCN}, even though fRH directly connected with aerosol hygroscopicity (Liu and Li, 2014). This may result from the significant dependence of fRH on aerosol size(Chen et al., 2014;Kreidenweis and Asa-Awuku, 2014;Kuang et al., 2017). As mentioned before, PNSD is used for better calculation of κ and N_{CCN} from fRH in previous studies (Ervens et al., 2007;Chen et al., 2014). A new method to estimate κ from fRH and Å was proposed recently (Kuang et al., 2017;Brock et al., 2016). Based on this method, fRH can be used to calculate N_{CCN} without measurements of PNSD and can be expected to improve the N_{CCN} prediction just based on measurements of aerosol optical properties.

In this study, the relationship between N_{CCN} and aerosol optical properties measured by a humidified nephelometer system is studied and a new method for N_{CCN} prediction is proposed. This new method is validated based on data observed in Gucheng campaign on the North China Plain and can be expected to improve measurements of N_{CCN} due to advantages of applying nephelometers.

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2. Methodology

2.1. Data

Data in this study are mainly measured at Gucheng (39.15N, 115.74E) during autumn in 2016 on the North China Plain (NCP). Gucheng is 100km southwest from Beijing and 40km northeast from Baoding under background pollution condition in the NCP. The observation site was surrounded by farmland and about 3km away from the Gucheng town. This campaign started on 20 October and lasted for nearly one month.

Instruments used in Gucheng campaign were located in a measurement container under temperature maintained at 25 ◦C. Ambient aerosol was sampled and dried to relative humidity (RH) lower than 30% by a inlet system consist of a PM10 inlet, a inline Nafion dryers and a RH and temperature sensor (Vaisala HMP110). Then the sample aerosol was separated by a splitter and directed into various instruments. During this campaign, aerosol scattering coefficient (σ_{sp}), aerosol optical hygroscopic growth factor (fRH), particle size-resolved activation ratio (AR) and particle number size distribution (PNSD) were obtained.

fRH as well as σ_{sp} at three wavelengths were measured by a humidified nephelometer system consisting of two nephelometers (Aurora 3000, Ecotech Inc.) and a humidifier. σ_{sp} can be described by a formula of Å: $\sigma_{sp}(\lambda)=\beta\cdot\lambda^{-Å}$, where β is the aerosol number concentration and λ is the wavelength. Thus Å can be calculated directly from σ_{sp} measured by a nephelometer. The humidifier with a Gore-Tex tube humidified the sample air up to 90% RH. A whole cycle of humidification lasted about 45minutes from 50% RH to 90% RH. Dried σ_{sp} was obtained directly from dried sample aerosol measured by one nephelometer and humidified σ_{sp} was obtained from humidified aerosol measured by another nephelometer. fRH is the ratio of the humidified σ_{sp} to the dried σ_{sp} at each RH. Detailed description of the humidified nephelometer system was illustrated in Kuang et al (2017).

The particle size-resolved AR, defined as the ratio of N_{CCN} to total particles, was measured by a system mainly consisting of a differential mobility analyzer (DMA) and a continuous-flow CCN counter (model CCN200, Droplet Measurement Technologies, USA; Roberts and Nenes (2005),;Lance et al.,(2006)). The system selected mono-disperse particles with the DMA coupled with a electrostatic classifier (model 3080; TSI, Inc., Shoreview, MN USA) and measured AR of the mono-disperse particles by a condensation particle counter (CPC model 3776; TSI, Inc.) and CCN counter. Ranges of particle size and supersaturation were 10-300nm and 0.07%-0.80%, respectively. Measurements at five supersaturations were conducted sequentially and each cycle lasted for 1 hour. Before and after the campaign, supersaturations set in this system were calibrated using ammonium sulfate (Rose et al., 2008). More information about the system are given in Deng et al. (2011) and Ma et al.(2016).

PNSD with particle diameter from 9nm to 10um was measured by a mobility particle size spectrometer (SMPS, TSI Inc., Model 3996) and an Aerodynamic Particle Sizer (APS, TSI Inc., Model 3321). SMPS consisted of a DMA, a electrostatic classifier and a CPC (model 3776; TSI, Inc., Shoreview, MN USA) and measured PNSD with diameter lower than 700nm.

In addition, PNSD and σ_{sp} from 2011 to 2014 at four campaigns (Wuqing in 2011, Xianghe in 2012 and 2013, and Wangdu in 2014) in NCP were used in this study. PNSD in these campaigns was measured by a Twin Differential Mobility Particle Sizer (TDMPS, Leibniz-Institute for Tropospheric Research (IfT), Germany) and an Aerodynamic Particle Sizer (APS, TSI Inc., Model 3321). A TSI 3563 nephelometer was used to obtain σ_{sp} at three wavelengths. Details about the four campaign can be found in Ma et al. (2011), Ma et al.(2016), Kuang et al. (2016) and Kuang et al.(2017).

2.2. Theories

Hygroscopic growth of particles at certain relative humidity can be described by κ-Köhler theory (Petters and Kreidenweis, 2007):

$$\frac{RH}{100} = \frac{g(RH)^3-1}{g(RH)^3-(1-\kappa)} \cdot \exp\left(\frac{4\sigma_{s/a}\cdot M_w}{R\cdot T\cdot D_d\cdot g\cdot \rho_w}\right) \qquad (1)$$

where g(RH) is geometric diameter growth factor, κ is the hygroscopicity parameter, S is the saturation ratio; ρ_W is the density of water; M_W is the molecular weight of water; $\sigma_{s/a}$ is the surface tension of the solution–air interface, which is assumed to be equal to the surface tension of the pure

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Discussion started: 28 July 2017

water–air interface; R is the universal gas constant; and T is the temperature.

Accounting for the impact of Å, κ_f can be derived directly from fRH (Brock et al., 2016;Kuang et al., 2017). A single-parameter parameterization scheme proposed by Brock et al. (2016) connects fRH and κ by the approximately proportional relationship between total aerosol volume and σ_{sp}:

$$f(\mathrm{RH})=1+\kappa_{sca}*\mathrm{RH}/(100-\mathrm{RH}) \quad (2)$$

where κ_{sca} is a parameter for fitting fRH curves and can determines κ_f with Å. This method was confirmed by good agreement with κ_f calculated from fRH and g(RH) (Brock et al., 2016;Kuang et al., 2017).

N_{CCN} can be calculated from size-resolved AR at a certain supersaturation (SS) and PNSD (referred to as n(logD_p)) as follows:

$$N_{CCN}=\int_{\log D_P}\mathrm{AR}(\log D_P, SS)\cdot n(\log D_P)\,\mathrm{d}\log D_P \quad (3)$$

In general, size-resolved AR curves are complicated and always replaced by a critical diameter to simplify calculation (Deng et al., 2013). The critical diameter is defined as:

$$N_{CCN}=\int_{\log D_c}^{\log D_{P,max}} n(\log D_P)\,\mathrm{d}\log D_P \quad (4)$$

where $D_{P,max}$ is the maximum diameter of the measured particle number size distribution. In other words, the integral of PNSD larger than D_c equals to the measured N_{CCN}. And a critical κ (κ_c) can be calculated by equation (1) and indicated CCN activity and hygroscopicity of particles.

3. Results

3.1. Calculation of N_{CCN} based on measurements of a Humidified Nephelometer system

Free of sea salt aerosol and dust aerosol, accumulation mode aerosol dominates both the optical scattering ability at short wavelength and the CCN activity at low supersaturation, and thus a reasonable relationship between σ_{sp} and N_{CCN} can be achieved. Figure 1 shows the size distribution of cumulative contributions of σ_{sp} at 450nm and N_{CCN} at 0.07% with various Å and κ_c, and corresponding normalized PNSDs based on data measured at the four campaigns on the North China Plain. During the four campaigns, no sea salt aerosol or dust aerosol was observed(Ma et al., 2011;Ma et al., 2016;Kuang et al., 2016;Kuang et al., 2017). For continental aerosol without sea salt or dust, Å varies from 0.5 to 1.8 and κ_c varies from 0.1 to 0.5. And as mentioned before, Å can be used as a proxy of the overall size distribution of aerosol populations, with smaller Å indicating more larger particles. In figure 1, comparisons for Å are made between 0.5 and 1.7 and for κ_c are made between 0.1 and 0.5. As larger particles contribute more to light scattering and activation, cumulative contributions of both σ_{sp} and N_{CCN} increase significantly at the diameter range of accumulation mode particles. Because more hygroscopic particles are able to activate at smaller diameters, the cumulative contribution of N_{CCN} with higher κ_c increase at smaller diameters. In general, major contributions of both σ_{sp} and N_{CCN} are made by particles from 200nm to 500nm for various Å and κ_c. This implies the feasibility of inferring N_{CCN} from aerosol optical properties.

Because smaller particles can activate at higher supersaturations while scatter less light at longer wavelengths, it's obvious that significant differences will exist between cumulative contributions of σ_{sp} and N_{CCN}. This means σ_{sp} and N_{CCN} are dominated by different particles and poor correlation between σ_{sp} and N_{CCN} can be expected. Thus the method of inferring N_{CCN} from aerosol optical properties is applicable for shorter wavelength and lower supersaturations.

Furthermore, PNSD with higher Å indicates as more Aitken mode particles and fewer accumulation mode particles. Thus large particles contribute less for both σ_{sp} and N_{CCN} when Å are higher, characterizing an increase of cumulative contribution curves at smaller diameters. In detail, differences between cumulative contribution curves with Å of 0.5 and 1.7 are about 150nm and 100nm for σ_{sp} and N_{CCN}, respectively. Changes of cumulative contributions of N_{CCN} and σ_{sp} with various Å reveal that the shape of PNSD can influence the correlation between N_{CCN} and σ_{sp}. This is confirmed by previous studies in which the Å is found to play an important role in calculating N_{CCN} from σ_{sp} (Shinozuka et al., 2015;Liu and Li, 2014).

The relationship between σ_{sp} and N_{CCN} dependent on Å and κ_c is evaluated by calculating

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Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 28 July 2017

Atmospheric Measurement Techniques
Discussions

σ_{sp} and N_{CCN} with different PNSDs classified by Å and different κ_c. In detail, ratios of N_{CCN} to σ_{sp}, referred to as AR_{sp}, are calculated to eliminate the effect of variations of particle concentrations consistent at all diameters. Results at the supersaturation of 0.07% are shown in figure 2 and AR_{sp} range from 0 to10. In general, AR_{sp} are higher for more hygroscopic particles or smaller particles. As particles become more hygroscopic, more CCN can be expected when σ_{sp} is fixed. As aerosol populations consist of more smaller CCN-active particles, the increase of σ_{sp} is weaker than that of N_{CCN}.

In detail, the sensitivity of AR_{sp} to Å also changes with Å and κ_c. When Å are higher than 1.4 and κ_c is lower than 0.2, AR_{sp} is insensitive to Å. While when Å are lower than 1 and κ_c are higher than about 0.3, AR_{sp} is more sensitive to Å than κ_c. Higher sensitivity of AR_{sp} to Å are found with higher κ_c and lower Å, which reveals that particles having more small particles and less large particles than existing particles can contribute more to N_{CCN}. This is the consequence of the sensitivity of AR_{sp} to Å resulting from the variation of small CCN-active particles, as mentioned before.

Based on the lookup-table illustrated in Figure 2, N_{CCN} at the supersaturation of 0.07% can be calculated simply from Å, κ_f and σ_{sp} which can be obtained from measurements of a humidified nephelometer system. The description of this simple method is shown in figure 3. A new look-up table needs to be made for N_{CCN} estimation at other supersaturations, which should better be less than 0.07% as mentioned in the discussion of figure 1.

One critical issue about the method is the conversion of the κ_f obtained from the humidified nephelometer system to the κ_c under super-saturated conditions. There are mainly two factors making this conversion necessary. First, closure studies of aerosol hygroscopicity found significant deviations between hygroscopicity at sub-saturated conditions and super-saturated conditions. Their difference can be expected to be about 0.1 for accumulation mode aerosol(Wu et al., 2013;Whitehead et al., 2014;Ma et al., 2016). Second, the κ_f indicates the hygroscopicity of total particles and can be quite different from aerosol hygroscopicity at a specific diameter due to variations of size distributions of particle hygroscopicity. Kuang et al. (2017) found a difference around 0.1 between

κ_f and κ inferred from g(RH) measurements for accumulation mode particles whose κ_f is no larger than 0.2. In this study, a simple conversion that κ_c is 0.2 higher than κ_f is used to calculate N_{CCN}, while for κ_f larger than 0.2, a smaller difference of 0.1 between κ_c and κ_f should be used (Kuang et al., 2017). This simplified relationship between κ_c and κ_f is applicable for two reasons. On one hand, the accurate conversion cannot be achieved without detailed information of the particle hygroscopicity, which is difficult and complicated to measure. On the other hand, a deviation of κ_c less than 0.1 generally leads to a deviation of N_{CCN} less than 20% (Ma et al., 2016), which is comparable with the deviation of CCN measurements. As a result, for a simple method of N_{CCN} calculation, this conversion is quite easy and adequate enough.

Besides aerosol size and hygroscopicity, aerosol mixing state can also affect aerosol cloud activity. When primary aerosol emissions are strong, aerosol populations are likely to be externally mixed and a realistic treatment of aerosol mixing state is critical for N_{CCN} calculation (Cubison et al., 2008;Wex et al., 2010). But for regions away from strong aerosol primary emissions, the influence of mixing state on aerosol cloud activity is small and the assumption of internal mixing state is effective for the estimation of N_{CCN} (Dusek et al., 2006;Deng et al., 2013;Ervens et al., 2010). For regions above the boundary layer where clouds form and measurements of N_{CCN} are important, this conclusion is tenable if there are no plumes(Moteki and Kondo, 2007;McMeeking et al., 2011). In the new method of this paper aerosol populations are assumed to be internally mixed. Thus this method might not be applicable for regions or air masses greatly affected by strong primary aerosol emissions. Furthermore, this new method cannot be applied for regions where sea salt or dust prevails, as mentioned before. In summary, this method can be used to calculate N_{CCN} for continental regions, especially at clouds forming heights, where aged aerosol particles dominate.

3.2. Validation based on N_{CCN} measurement

The method for calculating N_{CCN} based on measurement of the humidified nephelometer system, including the conversion of κ_c and the lookup-table, is examined using data measured in Gucheng.

Overview of data in Gucheng is shown in Figure 4. From polluted periods to clean periods, significant variations of N_{CCN} and σ_{sp} can be found but AR_{sp} of N_{CCN} to σ_{sp} stays around 5. On October 23rd and 29th, N_{CCN} and σ_{sp} are lower than 2000#/cm^3 and 500Mm^{-1}, respectively. While on

October 20th, 26th and November 3rd, N_{CCN} and σ_{sp} are higher than 2000#/cm^3 and 500Mm^{-1}, respectively. These variations of N_{CCN} and σ_{sp} are mainly due to the variation of the particle number concentration rather than the particle microphysical properties. Variations of AR_{sp} result from the variations of Å and κ_c, which indicate the variations of aerosol microphysical properties.

In general, AR_{sp} is more sensitive to variations of Å. As mentioned before, the sensitivity of AR_{sp} to Å is determined by both Å and κ_f. In detail, Å during the campaign mainly ranges from 0.5 to 15 and κ_f ranges mainly from 0.05 to 0.2, which means that κ_c ranges from 0.25 to 0.4. These values of Å and κ_f correspond a significant sensitivity of AR_{sp} to Å, as the lookup table shows in figure 2. The sensitivity of AR_{sp} to κ_c is much small and only notable during some short periods. For example, from November 5th to 7th, variations of κ_f and Å are opposite and result in nearly constant AR_{sp}. And from October 30th to November 2nd, consistent variations of Å and κ_f lead to greater variations of AR_{sp} than other periods. This weak sensitivity of AR_{sp} to κ_f may be due to the uncertainty of κ_c calculated from κ_f based on the simplified conversion.

This simplified conversion of κ_c is examined by comparing κ_f and κ_c measured in Gucheng campaign, shown in Figure 5. In general, $\Delta\kappa$ that indicates the difference between κ_f and κ_c is around 0.2 and independent from Å and κ_c. Over 80% of $\Delta\kappa$ ranges from 0.1 to 0.3 that confirms applicability of the simplified conversion of κ_c. However, a notable deviation of $\Delta\kappa$ can be found when Å is higher than 1.5. High values of Å represent existence of small particles. Compositions and mixing state of these small particles, which may be fresh emitted and experience inefficient aging processes, are diverse and likely to deviate from the simplified conversion of κ_c.

Therefore, considering the deviation of κ_c conversion and high sensitivity of AR_{sp} to κ_c when Å is higher than 1.5, the method of calculating N_{CCN} from measurements of a humidified nephelometer system may lead to significant deviation in this case which means that this method can only be adopted when Å is lower than 1.5.

Based on the lookup table of κ_c and Å, AR_{sp} is calculated and applied to calculate N_{CCN} with σ_{sp}. The calculated AR_{sp} and N_{CCN} are compared with the measured AR_{sp} and N_{CCN} shown as the green dots in Figure 6. In general, good agreements between calculations and measurements are

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2017-193
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Discussion started: 28 July 2017

achieved and relative deviations are within 30%. For the comparison of AR_{sp}, the system relative deviation is less than 10%. For the comparison of N_{CCN}, the slope and the correlation coefficient of the regression are 1.03 and 0.966, respectively.

In addition, the influence of the κ_c conversion on AR_{sp} and N_{CCN} calculation are evaluated in two ways. In the first way, $\Delta\kappa$ of the κ_c conversion is set to be 0.05 higher or lower, which means $\Delta\kappa$ of 0.25 or 0.15. The corresponding results are presented as the red dots and blue dots in Figure 6. In the second way, a constant κ_c of 0.34, which is the average of κ_c values in Gucheng campaign, is used to calculate AR_{sp} and N_{CCN}, and shown as the grey dots in Figure 6. In general, differences among calculations using various κ_c conversions are quite small. The $\Delta\kappa$ difference of 0.05 in κ_c conversion only leads to a difference of 10% for the system relative deviation. The correlation coefficient of the calculation using a constant κ_c is just a little lower than correlation coefficients of calculations using a κ_c conversion. As a result, the method of calculating N_{CCN} is insensitive to the uncertainty of the κ_c conversion.

In this study, the insensitivity of calculated N_{CCN} to κ_c conversion is partly due to the small variation of κ_f during the campaign. On one hand, the variation of κ_c can be quite large and cause non-ignorable deviations of calculated N_{CCN}. As previous studies of N_{CCN} measurement showed, the variation of κ_c is often small and a constant κ_c can be used to calculate N_{CCN} accurately (Andreae and Rosenfeld, 2008;Gunthe et al., 2009;Rose et al., 2010;Deng et al., 2013). Results in this study are similar to these previous studies. However, large variations of κ_c are also found in some other studies. In NCP, fluctuations of aerosol hygroscopicity during New Particle Formation events and soot emissions lead to significant deviations of calculated N_{CCN} from average aerosol hygroscopicity (Ma et al., 2016). On the other hand, the influence of κ_c cannot be ignored because the value of the average hygroscopicity is different in various regions during various periods. In summer of NCP, measured κ_f at sub-saturated conditions can reach up to 0.45 when inorganic compositions dominate in particles (Kuang et al., 2016). In this case, calculated N_{CCN} ignoring κ_c may be 10 times larger than measured N_{CCN}. To sum up, although the exact value of κ_c cannot be obtained from the measurement of the humidified nephelometer system, the influence of κ_c on N_{CCN} can be inferred and is found to be correct enough considering the convenience of this method. More data, especially in observations of more hygroscopic aerosol, is still needed to confirm this method.

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2017-193
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 28 July 2017

Atmospheric Measurement Techniques Discussions

4. Conclusions

N_{CCN} is a key parameter of cloud microphysics and aerosol indirect radiative effect. Direct measurements of N_{CCN} are generally conducted under super-saturated conditions in cloud chambers, and are complex and costly. The aerosols of accumulation mode contribute most to both the aerosol scattering coefficient and the aerosol CCN activity. In view of this, it is possible to predict N_{CCN} based on relationships between aerosol optical properties and the aerosol CCN activity. In this study, a new method is proposed to calculate N_{CCN} based on measurements of a humidified nephelometer system. In this method, N_{CCN} is derived from a look-up table which involves σ_{sp}, Å and κ_f, and the required three parameters can be obtained from a three-wavelength humidified nephelometer system.

Relationships between aerosol optical properties and aerosol CCN activity are investigated using datasets about aerosol PNSD measured during several campaigns in the North China Plain. The relationship between σ_{sp}, Å, κ_c and N_{CCN} is analyzed. It is found that the ratio between N_{CCN} and σ_{sp}, referred to as AR_{sp}, is determined by κ_c and Å. In light of this, it is possible to calculate N_{CCN} based only on measurements of a three-wavelength humidified nephelometer system which provides information about σ_{sp}, the hygroscopicity parameter κ and Å. However, κ derived from measurements of a humidified nephelometer system under sub-saturated conditions (termed as κ_f) differs from κ under super-saturated conditions which indicate CCN activity (termed as κ_c). As a result, the conversion from κ_f to κ_c is needed. Based on previous studies of aerosol hygroscopicity and CCN activity, a simple conversion from κ_f to κ_c with a fixed difference (referred to as $\Delta\kappa$) of 0.2 is proposed. On the basis of this simple conversion, the method of N_{CCN} prediction based only on measurements of a humidified nephelometer system is achieved under conditions without sea salt aerosol or dust aerosol.

This method is validated with measurements from a humidified nephelometer system and a CCN counter in Gucheng in 2016. During the campaign, both N_{CCN} and σ_{sp} vary with the pollution conditions. AR_{sp} is around 5 and change with Å and κ_f. The difference between κ_f and κ_c, was 0.2 ± 0.1. The agreement between the calculated N_{CCN} and the measured N_{CCN} is achieved with relative deviations less than 30%. Sensitivity of calculated N_{CCN} to conversions from κ_f to κ_c is studied by applying different kinds of conversions. Results show that calculated N_{CCN} varies little and is insensitive to the conversions, which confirms the robustness and applicability of this newly proposed method.

This study has connected aerosol optical properties with N_{CCN}, and also proposed a novel method to calculate N_{CCN} based only on measurements of a three-wavelength humidified nephelometer system. Due to the simple operation and stability of the humidified nephelometer system, this method will facilitate the real time monitoring of N_{CCN}, especially on aircrafts. In addition, measurements of the widely used CCN counter are limited to supersaturations higher than 0.07. This method is more suitable for calculating N_{CCN} at lower supersaturations, thus is more applicable for ambient measurements of clouds and fogs in the atmosphere.

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Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2017-193
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 28 July 2017

Atmospheric Measurement Techniques Discussions

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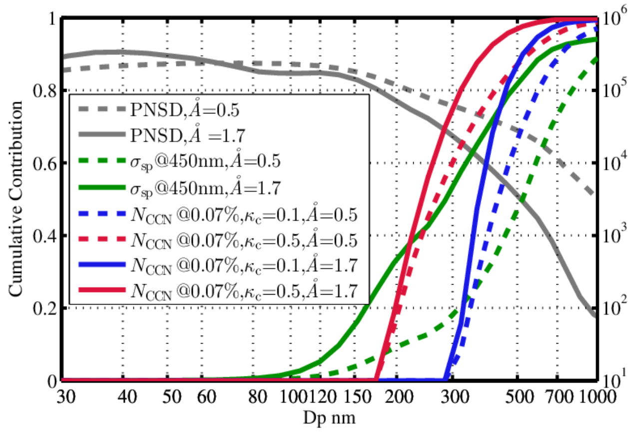

Figure 1.

Aerosol PNSD (black lines), the cumulative contribution of σ_{sp} at wavelength of 450nm (green lines) and N_{CCN} at supersaturation of 0.07% (red and blue lines) based on measurement in several campaigns in the North China Plain. Solid lines and dashed lines indicate Å of 1.7 and 0.5, respectively. Blue lines and red lines indicate κ_c of 0.1 and 0.5, respectively.

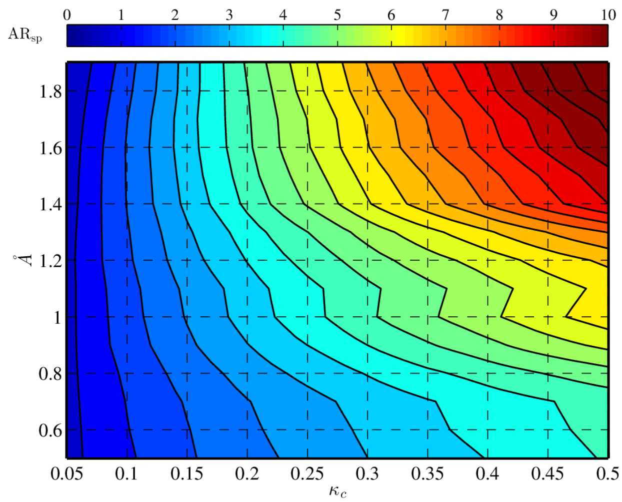

Figure 2.

Colors represent AR_{sp} (ratios between N_{CCN} and σ_{sp}) with κ_c and Å.

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Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 28 July 2017

Atmospheric Measurement Techniques
Discussions

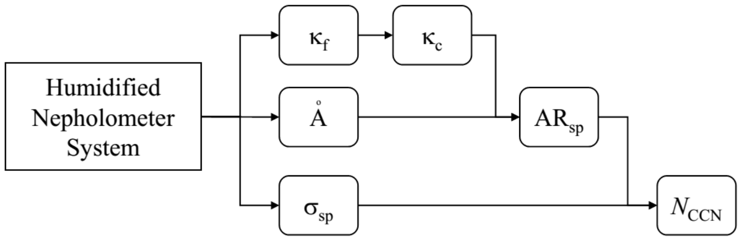

Figure 3.

The schematic chart of the N_{CCN} prediction based on measurements of a humidified nephelometer system.

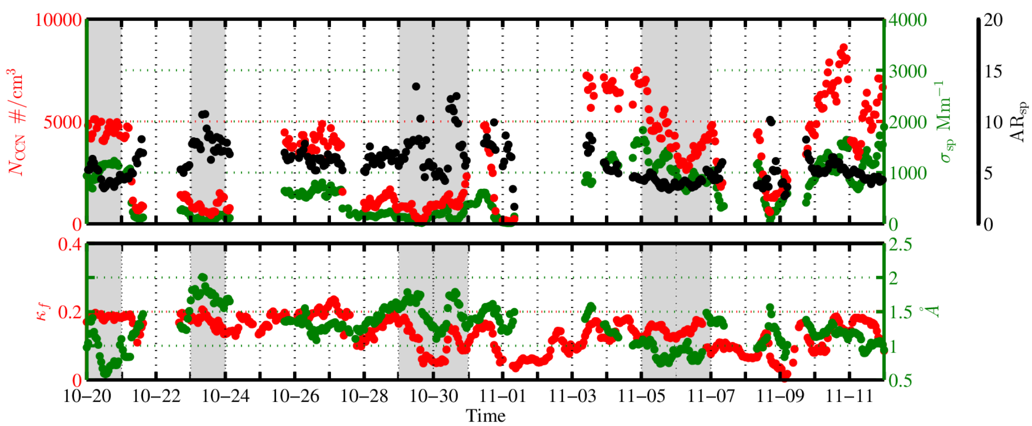

Figure 4.

Overview of measurements in Gucheng in 2016. Upper plot: time series of N_{CCN} at the supersaturation of 0.07% (red dots), σ_{sp} at the wavelength of 50nm (green dots) and their ratios (black dots), referred to as AR_{sp}. Lower plot: time series of κ_f (red dots) and Å (green dots).

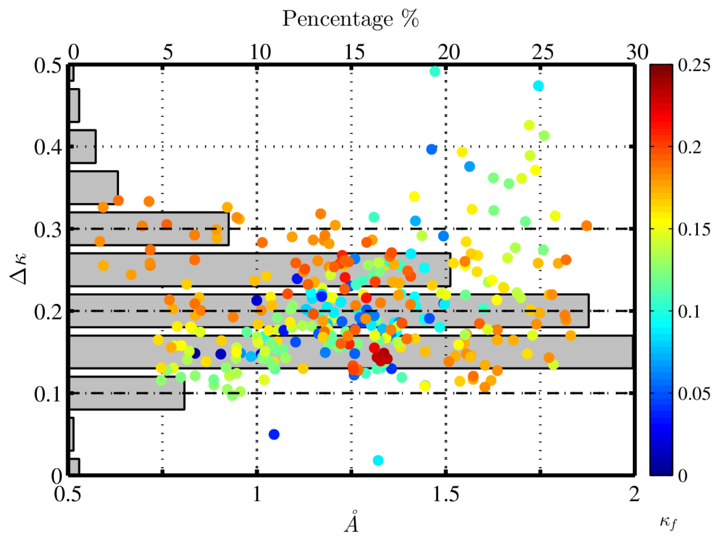

Figure 5.

Differences between κ_c and κ_f, referred to as $\Delta\kappa$, with Å (positions of dots) and κ_f (colors of dots). Bars represent percentages of $\Delta\kappa$ within different ranges.

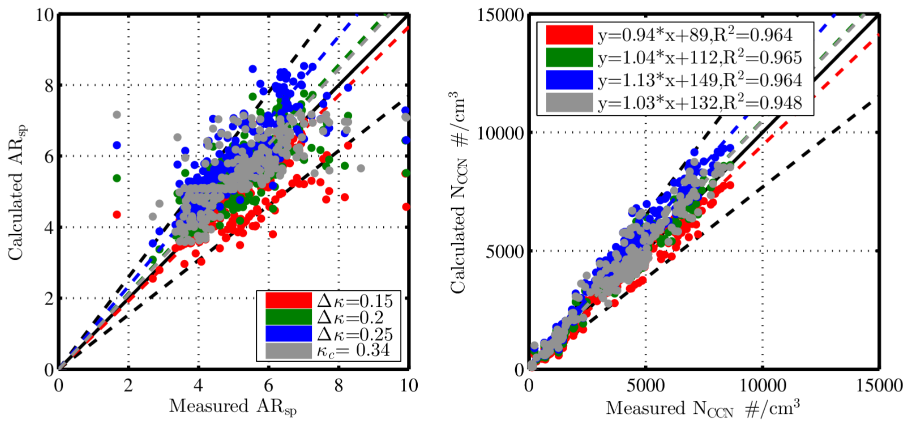

Figure 6.

Left plot: comparisons of calculated AR_{sp} and measured AR_{sp} with different conversions of κ_c from κ_f. Right plot: regressions of calculated N_{CCN} and measured N_{CCN} with different conversions of κ_c from κ_f.