Dear Editor,

We greatly thank the reviewers for their detailed review. Responses addressing reviewers’ comments point-by-point were uploaded (and also attached to this file). The manuscript has been revised and improved accordingly.

Best Regards

Chunsheng Zhao
Response to Referee #1:

General comment:

The authors present a study where they determined the number of cloud condensation nuclei (CCN) using a new method based on nephelometer measurements. They claim that this method is more convenient and cheaper than traditional measurements. Several studies have been published over the last 10 years that show that humidified nephelometer measurements can be used to infer CCN concentrations. They make several assumptions and use of various additional parameters. The apparent difference of the current study is the fact that no measurements of the particle number size distribution (PNSD). The manuscript contains several obscure sections and mistakes (grammar, typos). In addition, the method is poorly described and compared to previous work. Several sections are not well organized. A major revision considering my detailed comments below might help to improve the manuscript such that it may be considered for publication. In addition, the complete manuscript should be carefully proofread.

Response: Thanks for your comments. Comments are addressed point-by-point and corresponding responses are listed below. The whole manuscript is also checked.

Major comments:

1) Applicability of the new method

The caveats of the new method should be made clear in the abstract and conclusions. It is mentioned that it cannot be applied for externally mixed aerosol and particle populations with many large particles (e.g. dust, sea salt). Are there situations when delta(kappa) is too large/small that this bias will influence N(CCN)? Does the shape of the aerosol distribution play a role? Would, for example, multiple modes affect the Angstrom coefficient such that it exceeds 1.5?

Response: Thanks for the suggestion.

Hygroscopicity of both inorganic compounds and black carbon remain about the same under different saturated conditions, and the increase of hygroscopicity under supersaturated conditions are generally caused by organic compounds (Wex et al., 2009; Renbaum-Wolff et al., 2016). In ambient atmosphere, particles are consist of diverse compositions and the difference of particle hygroscopicity under different saturated conditions can be expected to be limited within a small range. However, in regions near strong sources of only one specific composition, this specific composition can completely dominate and lead to too large or too small Δκ. As a result, in regions with strong sources of a single composition, Δκ can be too large or too small and lead to significant deviations of predicted $N_{CCN}$. 
Particle number size distribution (PNSD) is important for aerosol activation and aerosol scattering. In this study, both aerosol activation and aerosol scattering are considered to be dominated by particles of accumulation mode and the shape of PNSD is taken in account to some extent by Å ranging from 0.5 to 1.5. When aerosol populations consist of large number of particles of Atiken mode which can contribute significantly to aerosol scattering, Å can exceed 1.5 and $N_{CCN}$ predicted by this new method can be overestimated.

We have added corresponding descriptions in the abstract, results and conclusions as follows:

Abstract, Line 20: “...is established to predict $N_{CCN}$. Due to the precondition for the application, this new method is not suitable for externally mixed particles, large particles (e.g. dust and sea salt) or particles near single source regions. ...”

Results, the last line of the second-to-last paragraph: “... In regions of single aerosol emissions or productions, the actual $\Delta x$ can be too large (some organic compositions, Wex et al., 2009; Renbaum-Wolff et al., 2016) or too small (inorganic compositions and black carbon) and thus is not suitable for the application of this method.”

Conclusions, the last line of second paragraph: “... under conditions without sea salt aerosol, dust aerosol, externally mixed aerosol or aerosol near single source regions.”

2) Comparison to previous studies

I suggest adding a table listing previous studies that have used optical aerosol parameters to infer $N(CCN)$. This table should include the parameters that were used (PNSD etc), air mass characteristics (aged or not), caveats of the method and comments on results/findings. This way, the necessity of measurements for various air masses will be more obvious and the applicability of the new method will be clearer. For example, the difference to the methods by Kuang et al. and Brock et al. to the current method is not fully clear.

Response: Thanks for the suggestion. We have added Table 1 and improved descriptions in the introduction. The methods proposed by Kuang et al. and Brock et al. are used to calculate hygroscopicity parameter and thus is not included in Table 1.

<table>
<thead>
<tr>
<th>Campaign</th>
<th>Air mass</th>
<th>Parameter</th>
<th>Caveats</th>
<th>Results</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>ICARTT$^1$ in the northeastern USA and Canada</td>
<td>Polluted air mass</td>
<td>fRH and PNSD</td>
<td>Calculate $N_{CCN}$ with aerosol hygroscopicity constrained by f(RH) and PNSD.</td>
<td>Predict $N_{CCN}$ at SS &gt; 0.3% with a 0.9 $R^2$.</td>
<td>Ervens et al., 2007</td>
</tr>
</tbody>
</table>
HaChi² on the North China Plain
Aged continental air mass
PNSD and fRH
Similar to Ervens et al., 2007. Calculate \( N_{\text{CCN}} \) with the hygroscopicity parameter constrained by \( f(\text{RH}) \) and PNSD.
Slopes around 1 and \( R^2 \) around 0.9.
Chen et al., 2014

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Location</th>
<th>Mass Type</th>
<th>Observation Method</th>
<th>Calculation Method</th>
<th>Additional Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>TARFOX³</td>
<td>Atlantic seaboard and ACE-2⁴</td>
<td>Polluted air mass</td>
<td>Retrieved aerosol volume from remote sensing</td>
<td>Predict ( N_{\text{CCN}} ) from aerosol volume with empirical number-to-volume concentration ratio</td>
<td>Overestimate up to 5 times</td>
</tr>
<tr>
<td>ACE-2 in northeastern Atlantic</td>
<td>Diverse air mass</td>
<td>Backscatter or extinction profile, CCN at the surface.</td>
<td>Predict ( N_{\text{CCN}} ) on most days for 0.1% SS and on 20%–40% of the days at 1% SS.</td>
<td>Ghan and Collins, 2004</td>
<td></td>
</tr>
<tr>
<td>ARM⁵</td>
<td>Climate Research Facility central site at the Southern Great Plains</td>
<td>Continental air mass</td>
<td>Backscatter (or extinction) and RH profile. fRH and CCN at surface</td>
<td>Same as Ghan and Collins, 2004.</td>
<td>Experiments CCN variance for 25%-63% of all measurements at high supersaturations</td>
</tr>
<tr>
<td>TRACE-P⁶ and ACE-Asia⁷</td>
<td>Asian outflow over the western Pacific</td>
<td>Aerosol Index (AI, the product of ambient light extinction and Å)</td>
<td>Predict ( N_{\text{CCN}} ) based on empirical relationship between AI and ( N_{\text{CCN}} )</td>
<td>AI relate well to CCN only with suitably stratified data</td>
<td></td>
</tr>
<tr>
<td>Multiple measurements</td>
<td>Diverse air mass</td>
<td>AERONET aerosol</td>
<td>Predict ( N_{\text{CCN}} ) based on empirical relationship</td>
<td>Predict ( N_{\text{CCN}} ) at SS &gt; 0.3% with a</td>
<td>Andreae, 2009</td>
</tr>
</tbody>
</table>
Estimate $N_{CCN}$ from fitting parameters for the $N_{CCN}$ activity spectra, which can be calculated based on their empirical relationships with aerosol optical properties.

Predict $N_{CCN}$ with slopes around 0.9 and $R^2$ around 0.6.

Achieve the best results by using $\sigma_{sp}$ and SSA. Weakly affect on the $\sigma_{sp}-N_{CCN}$ relationship by fRH. Deteriorate $N_{CCN}$–AOT relationship with increasing RH

<table>
<thead>
<tr>
<th>Four ARM sites</th>
<th>Polluted air mass</th>
<th>SSA, backscatter fraction and $\sigma_{sp}$</th>
<th>Predict $N_{CCN}$ with slopes around 0.9 and $R^2$ around 0.6.</th>
<th>Jefferson, 2010</th>
</tr>
</thead>
<tbody>
<tr>
<td>Multiple ARM sites around the world</td>
<td>Diverse air mass</td>
<td>RH, fRH, SSA, AOT and $\sigma_{sp}$</td>
<td>Calculate $N_{CCN}$ with $\sigma_{sp}$ (or AOT) based on their empirical relationship, whose impact RH, fRH and SSA.</td>
<td>Liu and Li, 2014</td>
</tr>
<tr>
<td>Multiple ARM sites around the world</td>
<td>Diverse air mass not dominated by dust</td>
<td>Å and extinction coefficient</td>
<td>Calculate $N_{CCN}$ with light extinction based on their empirical relationship.</td>
<td>Deviate typically within a factor of 2.0.</td>
</tr>
</tbody>
</table>

Tabel 1. Review of studies that have used aerosol optical parameters to infer $N_{CCN}$.

1 International Consortium for Atmospheric Research on Transport and Transformation.

2 Haze in China.

3 Troposphere Aerosol Radiative Forcing Experiment.

4 Second Aerosol Characterization Experiment.

5 Atmospheric Radiation Measurement.

6 Transport and Chemical Evolution over the Pacific.

7 Aerosol Characterization Experiment–Asia.
3) Clarity of method application

a) While Figure 3 is somewhat helpful, it should be extended to be the central figure of the manuscript. Labels can be added to the arrows explaining in detail what is done in each step, e.g. a reference to the respective equation would be helpful.

Response: Thanks for the suggestion. We have revised Figure 3 as follows:

\[
N_{CCN} = \sigma_{sp} \times AR_{sp}
\]

b) The comparison to measured \(N(\text{CCN})\) is useful and necessary in order to validate the new method. However, a few more details about the CCN measurements are needed. At what supersaturations were they measured (l. 116)? It is known that CCN measurements are most uncertain at low supersaturations. What supersaturation was chosen for the comparison?

Response: Thanks for the suggestion. There were five supersaturations (0.07%, 0.10%, 0.20%, 0.40% and 0.80%) and \(N_{CCN}\) at 0.07% supersaturation was chosen for the comparison, because at higher supersaturations this new method is not applicable any more. We have revised the statement in line 116 as: “Measurements at five supersaturations (0.07%, 0.10%, 0.20%, 0.40% and 0.80%) were conducted sequentially with each cycle lasted for 1 hour, and \(N_{CCN}\) at 0.07% supersaturation was used in this study.”

4) Clarity of language
At several places, the text is not clear or even wrong and should be revised. Examples include:

l. 57: Aerosol hygroscopicity is defined as the ability of an aerosol particle to take up water. Hygroscopicity is not a function of particle size.

Response: Thanks for the suggestion. We agree with the reviewer. The statement in the manuscript leads to misunderstanding and we have revised it as: “...aerosol CCN activity is determined by aerosol size and aerosol hygroscopicity. ...”

l. 68-72: It should be clarified which combination of parameters is best suited and which problems/deviations (from what?) might occur.

Response: Thanks for the suggestion. As mentioned in the response to “2) Comparison to previous studies”, we have revised line 68-72 as follows:

“...due to the diversity of hygroscopicity of less-absorbing components. Thus N_{CCN} calculation combining SSA, backscatter fraction and $\sigma_{sp}$ still lead to significant deviations, with a 0.6 $R^2$ (Jefferson, 2010). As for fRH, there was a study applied aerosol optical quantities ($\sigma_{sp}$ or aerosol optical thickness) with fRH or SSA to calculate N_{CCN} (Liu and Li, 2014). In their study, compared with the combination of SSA and aerosol optical quantities, the combination of fRH and aerosol optical quantities is found to be less effective in estimating N_{CCN}, even though fRH directly connected with aerosol hygroscopicity (Liu and Li, 2014). ...”

l. 143: ‘and can determine ‘kappa’ with A’ is unclear

Response: Thanks for the suggestion. We have revised this sentence as: “... and is found can be used to predict $\kappa_f$ in combination with $\hat{A}$ in recent studies(Brock et al., 2016; Kuang et al., 2017). This method of calculating $\kappa_f$ based on $\kappa_{sca}$ and $\hat{A}$ was confirmed by good agreement with $\kappa_f$ calculated from fRH and PNSD.”

l. 174: This text is hard to follow. At the very least, add numerical ranges for the various parameters. It would be even better to connect this discussion to a figure (either an additional one or existing one)

Response: Thanks for the suggestion. We have revised Figure 1 as follows:
Figure 1. Aerosol PNSD (black lines), the cumulative contribution of $\sigma_{sp}$ at wavelength of 450nm and 700nm (dark green lines and light green lines, respectively), the cumulative contribution of $N_{CCN}$ at supersaturation of 0.07% (dark red and dark blue lines) and the cumulative contribution of $N_{CCN}$ at supersaturation of 0.20% (light red and light blue lines) based on measurement in several campaigns in the North China Plain. Solid lines and dashed lines indicate $\Delta$ of 1.9 and 0.5, respectively. Blue lines and red lines indicate $\kappa_c$ of 0.1 and 0.5, respectively.

There is a typo error of $\Delta$ value in Figure 1. And we also revised this sentence as: “Because particles smaller than 200nm can activate at supersaturation higher than 0.07% while scatter less light at wavelengths longer than 450nm, which are shown as the light color lines in Figure 1, ...”

l. 198: ‘...which reveals that particles...' – I do not understand this fragment.

Response: Thanks for the suggestion. We have revised this sentence as: “This higher sensitivity of $AR_{sp}$ to $\Delta$ reveals that particles having mean predominante size smaller than existing particles can contribute more to $N_{CCN}$.”

l. 214: Do you mean ‘...due to size-dependent hygroscopicity’?
Response: Thanks for the suggestion. We have revised it accordingly.

l. 284 – 294: This paragraph should be rewritten as I cannot follow the line of thought. For example, you start with ‘On one hand, the variation of kappa(c) can be quite large...’ and continue later ‘On the other hand, the influence of kappa(c) cannot be ignored . . . ’ These two sentences should introduce opposing facts, but they do not.

Response: Thanks for the suggestion. We have revised this paragraph as follows:

“... First, the variation of $\kappa_c$ is not always small and can cause non-ignorable deviations of calculated $N_{CCN}$ in certain cases. As many studies of $N_{CCN}$ measurement showed, the variation of $\kappa_c$ is often small and a constant $\kappa_c$ 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 $\kappa_c$ 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). Second, the influence of $\kappa_c$ variation on $N_{CCN}$ calculation cannot be ignored because the value of the average hygroscopicity differs in various regions during various periods. In summer of NCP, measured $\kappa_f$ at sub-saturated conditions can reach up to 0.45 when inorganic compositions dominate in particles (Kuang et al., 2016). ...”

5) Structure

Essential information should be given as early as possible in the manuscript:

a) The Angstrom coefficient should be defined in the introduction or in Section 2.

Response: Thanks for the suggestion. We have defined Angstrom Exponent in Section 2 and we have revised the statement in the introduction in line 47 as: “... Angstrom Exponent ($\AA$, which is the exponent commonly used to describe the dependence of $\sigma_{sp}$ on wavelength),...”

b) Caveats of the method should be pointed out throughout the paper

Response: Thanks for the suggestion. We have added caveats in the abstract and conclusions as presented in lines 50-60 in this response.
c) It is highly confusing that in Section 2 delta(kappa) is introduced as being 0.2 and only in Section 3 a lengthy discussion of this value is given and sensitivity studies are performed. A more thorough discussion of reasons and conditions of large or small delta(kappa), respectively, should be added in the context of the applicability and accuracy of the new method. How would the results change if not a constant delta(kappa) but the exact difference for each data point in Fig 5 is used? Can we learn something from the resulting (dis)agreement as a function of A?

Response: Thanks for the suggestion.

We have added the discussion of reasons and conditions of \( \Delta \kappa \) variations in the second to last paragraph in Section 3 as follows:

“...a smaller difference of 0.1 between \( \kappa_c \) and \( \kappa_f \) should be used (Kuang et al., 2017). This simplified relationship between \( \kappa_c \) and \( \kappa_f \) is a rough estimate regardless of the complexity of differences of aerosol hygroscopicity measured by different instruments, but still used in this study for two reasons. First, the accurate conversion cannot be achieved without detailed information of the particle hygroscopicity, which is difficult and complicated to measure. Second, a deviation of \( \kappa_c \) less than 0.1 generally leads to a deviation of \( N_{\text{CCN}} \) less than 20% (Ma et al., 2016), which is comparable with the deviation of CCN measurements. As a result, for a simple method of \( N_{\text{CCN}} \) calculation, this conversion is quite easy. In addition, it is important to note that the value of the difference between \( \kappa_c \) and \( \kappa_f \) is also a rough estimate regardless of the complexity of aerosol hygroscopicity under different conditions, and the influence of \( \Delta \kappa \) deviation on \( N_{\text{CCN}} \) calculation needs to be further examined based on field observation.”

The use of exact \( \Delta \kappa \) for each data point to calculate \( N_{\text{CCN}} \) means the application of measured \( \kappa_c \) for \( N_{\text{CCN}} \) prediction based on the lookup table in Figure 2. This exclude the uncertainty of aerosol hygroscopicity in predicting \( N_{\text{CCN}} \) and highlight the impact of PNSD’s variation on \( N_{\text{CCN}} \) prediction when \( \hat{A} \) is used to estimate the influence of PNSD on the relationship between \( N_{\text{CCN}} \) and \( \sigma_{\text{sp}} \). Calculated AR_{sp} and calculated \( N_{\text{CCN}} \) with corresponding \( \hat{A} \) are shown in Figure S1. Relative deviations between calculated AR_{sp} and measured AR_{sp} are generally no higher than 30%. Compared with correlations shown in the left plot of Figure 6, whose correlation coefficients ranges from 0.5 to 0.6, the correlation between calculated AR_{sp} based on measured \( \kappa_c \) and measured AR_{sp} is better, with a correlation coefficient of 0.709. As for calculated \( N_{\text{CCN}} \) using measured \( \kappa_c \), relative deviations are mainly within 30%. Deviations of calculated AR_{sp} and calculated \( N_{\text{CCN}} \) are due to variations of PNSDs which share a same \( \hat{A} \). In addition, as for relative deviations of both calculated AR_{sp} and calculated \( N_{\text{CCN}} \), neither of them has a significant relationship with corresponding \( \hat{A} \). Besides the
uncertainty of CCN measurement, causes of calculated \( N_{CCN} \) deviations also include variations of PNSDs with a common \( A \) are almost the same for different \( A \), showing random fluctuations of PNSDs from their true values.

Figure S1. Left plot: comparisons of calculated \( A_{Rsp} \) based on measured \( \kappa_e \) and measured \( A_{Rsp} \). Right plot: regressions of calculated \( N_{CCN} \) based on measured \( \kappa_e \) and measured \( N_{CCN} \). The color of the dot are corresponding \( A \) for each data point.

6) Formatting

All parameters should be expressed in equations and should be formatted and numbered as such. For example, l. 101 and the definition of \( fRH \) (l. 106).

Response: Thanks for the suggestion. We have revised them accordingly.

7) Figures

a) The caption of Figure 2 cannot be understood without reading the text. At the very least, the parameters should be spelled out and a reference to an equation in the text should be added.

Response: Thanks for the suggestion. We have revised the caption as “Colors represent \( A_{Rsp} \) (calculated as \( A_{Rsp} = \frac{N_{CCN}}{\sigma_{sp}} \) at 450nm wavelength and 0.07% supersaturation) with different
PNSDs (classified by Å values) and different κc.

b) What are the grey bars in Figure 4?

Response: Thanks for the suggestion. The grey bars are periods when the sensitivity of ARsp to κc is notable. We have added the description in the caption of Figure 4 and in the third paragraph of Section 3.2.

c) The grey symbols in Figure 6 overlap with many other symbols. Maybe choosing open symbols would improve clarity

Response: Thanks for the suggestion. We have revised Figure 6 as follows:

Minor comments:

l. 64: Add references for the 'common use'.

Response: Thanks for the suggestion. We have added references as follows:


l. 66: This sentence needs work: 1) word missing after ‘carbonaceous’. 2) What is meant by ‘most important hydrophobic’?

Response: Thanks for the suggestion. The statement in the manuscript is confusing and we have revised this sentence as: “Black carbon dominates the absorption of solar radiation and is a main hydrophobic composition as well.”

l. 135/6: S is not included in the equation

Response: Thanks for the suggestion. It should be RH and we have revised it accordingly.

l. 164/5: Is this a result based on the literature or the current data set? If the former, add references.

Response: Thanks for the comment. It’s based on literature and we have added references as follows:


l. 191: AR(sp) can only be 0 if N(CCN) or if sigma(sp) is infinitely large. Is either of this a realistic situation?

Response: Thanks for the suggestion. σ_sp wouldn’t be infinitely large and AR_{sp} should be higher than 0. We have revised it as: “... and {AR}_{sp} is higher than 0 and lower than 10. ...”

l. 245: What ‘microphysical properties’ are you referring to here? ‘Composition’ is a chemical property
Response: Thanks for the comment. We are referring to the shape of particle size distribution and aerosol hygroscopicity, and we have revised the last two sentences in this paragraph as: “... rather than the shape of particle size distribution and aerosol hygroscopicity. Variations of ARsp result from the variations of Å and $\kappa_c$, which indicate the variations of aerosol microphysical properties and chemical compositions.”

l. 247: ‘more sensitive’ as compared to which other parameter?

Response: Thanks for the comment. It should be $\kappa_c$ and we have revised it accordingly.

l. 249: Later and in Figure 2, the range of A is up to approx. 1.5, not 15

Response: Thanks for the suggestion. We have revised it accordingly.

Technical comments

l. 2: ‘Nuclei’ misspelled

Response: Thanks for the suggestion. We have revised it.

l. 94: an inlet . . . consisting of . . . an inline . . .

Response: Thanks for the suggestion. We have revised them.

l. 109: AR has not been defined before.

Response: Thanks for the suggestion. We have revised it.

l. 128: campaigns

Response: Thanks for the suggestion. We have revised it.

l. 154: indicates

Response: Thanks for the suggestion. We have revised it.

l. 159: wavelengths

Response: Thanks for the suggestion. We have revised it.

l. 171: increases

Response: Thanks for the suggestion. We have revised it.

l. 179: remove ‘as’
Response: Thanks for the suggestion. We have revised it.
Response to Referee #2:

General comment:

This work proposed a new method to estimate number concentrations of CCN based on the humidified nephelometer measurements. The advantages of this method are more convenient and cheaper than traditional measurements, and no other measurements are needed. The manuscript fits well to the scope of AMT. Thus I recommend it to be published after the following comments listed below have been adequately addressed.

Response: Thanks for the comments. Comments are addressed point-by-point and corresponding responses are listed below.

Specific comments:

1. Lines 47-52: Please add some texts to evaluate each application. Also, I agree with another reviewer that one table should be added to summary the previous studies using aerosol optical properties to calculate NCCN.

Response: Thanks for the suggestion. We have added descriptions and a table as follows:

“... Compared with the first kind, whose $R^2$ can be about 0.9, instruments used in the second kind of methods are cheaper and easier in operation, but has a lower accuracy of $R^2$ much lower than 0.9. …”

<table>
<thead>
<tr>
<th>Campaign</th>
<th>Air mass</th>
<th>Parameter</th>
<th>Caveats</th>
<th>Results</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>ICARTT(^1) in the north eastern USA and Canada</td>
<td>Polluted air mass</td>
<td>fRH and PNSD</td>
<td>Calculate $N_{CCN}$ with aerosol hygrosopicity contrained by $f$(RH) and PNSD.</td>
<td>Predict $N_{CCN}$ at SS &gt; 0.3% with a 0.9 $R^2$.</td>
<td>Ervens et al., 2007</td>
</tr>
<tr>
<td>HaChi(^2) on the North China Plain</td>
<td>Aged continental air mass</td>
<td>PNSD and fRH</td>
<td>Calculate $N_{CCN}$ with the hygrosopicity parameter contrained by $f$(RH) and PNSD.</td>
<td>Slopes around 1 and $R^2$ around 0.9.</td>
<td>Chen et al., 2014</td>
</tr>
<tr>
<td>TARFOX(^3) Atlantic seaboard and</td>
<td>Polluted air mass</td>
<td>Retrieved aerosol volume from</td>
<td>Predict $N_{CCN}$ from aerosol volumes with empirical number-to-volume</td>
<td>Overestimate up to 5 times</td>
<td>Gasso and Hegg, 2003</td>
</tr>
<tr>
<td>Experiment</td>
<td>Air Mass</td>
<td>Measurement</td>
<td>Calculations/Results</td>
<td></td>
<td></td>
</tr>
<tr>
<td>------------</td>
<td>---------</td>
<td>-------------</td>
<td>----------------------</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>ACE-2</strong> in northeastern Atlantic</td>
<td>Diverse air mass</td>
<td>remote sensing</td>
<td>concentration ratio&lt;br&gt;Retrieves $N_{CCN}$ profile from backscatter (or extinction) vertical profile assuming their ratios are the same to the ratio at surface, which can be calculated by backscatter (or extinction) and $N_{CCN}$ measured at the surface.</td>
<td>Predict $N_{CCN}$ on most days for 0.1% SS and on 20%–40% of the days at 1% SS.</td>
<td>Ghan and Collins, 2004</td>
</tr>
<tr>
<td><strong>ARM</strong> Climate Research Facility central site at the Southern Great Plains</td>
<td>Continental air mass</td>
<td>Backscatter (or extinction) and RH profile. $fRH$ and CCN at surface</td>
<td>Explains CCN variance for 25%–63% of all measurements at high supersaturations</td>
<td>Ghan et al., 2006</td>
<td></td>
</tr>
<tr>
<td><strong>TRACE-P</strong> and <strong>ACE-Asia</strong></td>
<td>Asian outflow over the western Pacific</td>
<td>Aerosol Index (AI, the product of ambient light extinction and Å)</td>
<td>Predict $N_{CCN}$ based on empirical relationship between AI and $N_{CCN}$</td>
<td>AI relate well to CCN only with suitably stratified data</td>
<td>Kapustin et al., 2006</td>
</tr>
<tr>
<td><strong>Multiple measurements</strong></td>
<td>Diverse air mass</td>
<td>AERONET aerosol optical thickness (AOT)</td>
<td>Predict $N_{CCN}$ based on empirical relationship between AOT and $N_{CCN}$ as a power law</td>
<td>Predict $N_{CCN}$ at SS &gt; 0.3% with a 0.88 $R^2$, but have a factor-of-four range of $N_{CCN}$ at a given AOT</td>
<td>Andreae, 2009</td>
</tr>
<tr>
<td><strong>Four ARM sites</strong></td>
<td>Polluted air mass</td>
<td>SSA, backscatter fraction and $\sigma_p$</td>
<td>Estimate $N_{CCN}$ from fitting parameters for the $N_{CCN}$ activity spectra, which can be calculate based on their empirical relationships with aerosol optical properties.</td>
<td>Predict $N_{CCN}$ with slopes around 0.9 and $R^2$ around 0.6.</td>
<td>Jefferson, 2010</td>
</tr>
<tr>
<td><strong>Multiple ARM sites</strong></td>
<td>Diverse air mass</td>
<td>RH, $fRH$, SSA, AOT</td>
<td>Calculate $N_{CCN}$ with $\sigma_p$ (or AOT) based on their</td>
<td>Achieve the best results by using $\sigma_p$</td>
<td>Liu and Li, 2014</td>
</tr>
</tbody>
</table>
around the world and empirical relationship, whose impact RH, fRH and SSA. Weakly affect on the $\sigma_{sp}$–$N_{CCN}$ relationship by fRH. Deteriorate $N_{CCN}$–AOT relationship with increasing RH.

| Multiple ARM sites around the world | Diverse air mass not dominated by dust | $\bar{A}$ and extinction coefficient | Calculate $N_{CCN}$ with light extinction based on their empirical relationship. | Deviate typically within a factor of 2.0. | Shinozuka et al., 2015 |

Tabel 1. Review of studies that have used aerosol optical parameters to infer $N_{CCN}$.

1 International Consortium for Atmospheric Research on Transport and Transformation.

2 Haze in China.

3 Troposphere Aerosol Radiative Forcing Experiment.

4 Second Aerosol Characterization Experiment.

5 Atmospheric Radiation Measurement.

6 Transport and Chemical Evolution over the Pacific.

7 Aerosol Characterization Experiment–Asia.

2. Lines 172-176: I guess that the authors want to claim that the uncertainty will be smaller when performing this method for shorter wavelength and lower supersaturation. Am I correct? Concerning only one supersaturation (0.07%) was test in this study, and the relative deviation is within 30%. Therefore, I am wondering that is it possible to perform this method to higher supersaturations to check when the uncertainty will be larger than 50%.

Response: Thanks for the suggestion. Yes, the uncertainty is smaller when performing this method for shorter wavelength and lower supersaturation. We apply this method to higher supersaturations and compare calculated $AR_{sp}$ with measured $AR_{sp}$. $\Delta \kappa$ at five supersaturations are all set to be 0.2. Results are shown in Figure S2 as follows:
Figure S2. (a) to (e) Calculated AR$_{sp}$ (ratios between $N_{CCN}$ and $\sigma_{sp}$, represented as the color) based on $\sigma_{sp}$ and $N_{CCN}$ with different PNSDs classified by $\AA$ and different $\kappa_s$ at the five supersaturations. (f) Comparison between calculated AR$_{sp}$ and measured AR$_{sp}$. Colors represent supersaturations.

As the lookup table at each supersaturation shown, calculated AR$_{sp}$ is higher at higher supersaturation as a whole, which indicate more CCN with a common $\sigma_{sp}$. The same as shown in Figure 6, relative deviations of calculated AR$_{sp}$ from measured AR$_{sp}$ are generally within 30%. Calculated AR$_{sp}$ at 0.10% supersaturation are 30% higher than measured AR$_{sp}$ but still associated with measured AR$_{sp}$. For the three supersaturations higher than 0.10%, relative deviations of calculated AR$_{sp}$ from measured AR$_{sp}$ often exceed 50% and there is no significant correlation between calculated AR$_{sp}$ and measured AR$_{sp}$. Results shown in Figure S2 verify the conclusion that the uncertainty is smaller when performing this method for shorter wavelength and lower supersaturation and the this method is not applicable at supersaturations higher than 0.10%.

3. Lines 180-181: How to calculate the differences (150 nm and 100 nm)? Please explain.

Response: Thanks for the suggestion. The diameter difference of cumulative contribution between 0.5 $\AA$ and 1.7 $\AA$ is roughly estimated by the average of differences where cumulative
contributions range from 0.2 to 0.8. We have revised the statement as: “In detail, differences of cumulative contribution curves between 0.5 Å and 1.7 Å are about 150nm for $\sigma_{sp}$ and about 100nm for $N_{CCN}$. by estimating the average of differences of diameters where cumulative contributions range from 0.2 to 0.8”

4. Line 191: What are smaller CCN-active particles? Do you mean Aitken mode particles? I think the contribution of particles smaller than 100 nm to $\sigma_{sp}$ is always negligible.

Response: Thanks for the comment. Smaller CCN-active particles refers to particles smaller than the average diameter of the whole CCN-active particles but is still CCN-active. For example, particles with diameters slightly larger than $D_c$ contribute less to $\sigma_{sp}$ than particles with diameters much larger than $D_c$. We have added the corresponding description after the sentence.

5. Lines 201-203: See comment 2. It seems that you claim 0.07% is the highest supersaturation that can be applied for this method. Why? Do you have results for other supersaturations?

Response: Thanks for the suggestion. Yes, as for the five supersaturations measured in this study, 0.07% is the highest supersaturation (also the only supersaturation) that can be applied for this method. This is because $N_{CCN}$ at supersaturations higher than 0.07% are dominated by small particles more significantly than $\sigma_{sp}$ (shown in Figure 1) and the correlation between $N_{CCN}$ and $\sigma_{sp}$ become weaker. The result in Figure S2 shows that relative deviations of calculated $N_{CCN}$ at supersaturations higher than 0.07 can exceed 30% commonly.

6. Lines 206-208: Add references. Why do you think $\kappa_f$ is always lower than $\kappa_c$? Any explanations?

Response: Thanks for the suggestion. We have added reference as follows:


There are mainly two reasons why $\kappa_f$ is always lower than $\kappa_c$. First, $\kappa_f$ is calculated based on measurements under subsaturated conditions while $\kappa_c$ is calculated based on measurements under supersaturated conditions. Studies found that aerosol hygroscopicity can increase under supersaturated conditions, due to dissolution of slightly soluble substances (Wex et al., 2009), the phase separation of organic compounds (Renbaum-Wolff et al., 2016) and so on. Second, accumulation mode particles are generally most hygroscopic. $\kappa_f$ represents the average hygroscopicity of total particles and is generally lower than hygroscopicities of accumulation mode particles (Kuang et al., 2017), while $\kappa_c$ at 0.07% supersaturation indicate hygroscopicities of particles around 200nm. As a result, $\kappa_f$ is always lower than $\kappa_c$.

7. Lines 241-247 and Figure 5: How about the agreement between the retrieved and measured $\kappa_c$?

Response: Thanks for the comment. We compare the retrieved and the measured $\kappa_c$, as shown in Figure S3. For the majority of data points, relative deviations between retrieved and measured $\kappa_c$ are within about 20%. A large relative deviation much higher than 20% usually correspond to a $\AA$ higher than 1.5, which is also shown in Figure 5.
Figure S3. Comparisons between measured and retrieved $\kappa_\text{c}$ (dots) and their corresponding $\bar{\Delta}$ values (colors). The solid black line is the 1:1 line and dashed black lines indicate relative deviations of 20%.

8. Lines 248-251: The authors claim that this method can only be adopted when $\bar{\Delta}$ is lower than 1.5. Is this conclusion only based on this study or can be used in different environments?

Response: Thanks for the suggestion. This conclusion can be used in other environment, however the lookup table should be recalculated based on PNSD measured in corresponding environment.

9. I suggest the authors reorganize or recheck the text for each figure caption. More information should be included, such as gray background in Figure 2 and black & dashed lines in Figure 6.

Response: Thanks for the suggestion. There is no gray background in Figure 2 while gray backgrounds in Figure 4 are not described yet. We have revised them accordingly.

10. Technical comments:

Title: Nuclei.

Response: Thanks for the suggestion. We have revised it.

Line 36: also.

Response: Thanks for the suggestion.

Line 110: please provide DMA type.

Response: Thanks for the suggestion. It’s Model 3081 DMA and we have revised it.

Lines 111 and 120: an electrostatic classifier.

Response: Thanks for the suggestion. We have revised them.

Line 126: campaigns. Line 133: there is no S in Eq. (1), please reformulate it.

Response: Thanks for the suggestion. It should be relative humidity (RH) and we have revised it.

Line 137: explain $\kappa_f$.

Response: Thanks for the suggestion.

Line 152: indicates.
Response: Thanks for the suggestion. We have revised it.

Line 234: 0.5 to 1.5

Response: Thanks for the suggestion. We have revised it.

Lines 271-273: please add references.

Response: Thanks for the suggestion.

Line 308: changes

Response: Thanks for the suggestion. We have revised it.

There are still several grammar mistakes in the text, please carefully check.

Response: Thanks for the suggestion.
Reference:


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

Jiangchuan Tao¹, Chunsheng Zhao¹, Ye Kuang¹, Gang Zhao¹, Chuanyang Shen¹, Yingli Yu¹, Yuxuan Bian², Wanyun Xu²

[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 ($N_{\text{CCN}}$) based on chamber technology are complex and costly, thus a simple way for measuring $N_{\text{CCN}}$ is needed. In this study, a new method for $N_{\text{CCN}}$ 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 ($\sigma_{\text{sp}}$) at three wavelengths and the light scattering enhancement factor (fRH). The Angstrom exponent (Å) inferred from $\sigma_{\text{sp}}$ at three wavelengths provides information on mean predominate aerosol size and hygroscopicity parameter ($\kappa$) can be calculated from the combination of fRH and Å. Given this, a look-up table that involves $\sigma_{\text{sp}}, \kappa$ and Å is established to predict $N_{\text{CCN}}$. Due to the precondition for the application, this new method is not suitable for externally mixed particles, large particles (e.g. dust and sea salt) or particles near single source regions. This method is validated with direct measurements of $N_{\text{CCN}}$ using a CCN counter on the North China Plain. Results show that relative deviations between calculated $N_{\text{CCN}}$ and measured $N_{\text{CCN}}$ are within 30% and confirm the robustness of this method. This method enables simpler $N_{\text{CCN}}$ 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.

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 \(\kappa\) 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 \(\sigma_{sp}\), Angstrom Exponent \(\hat{\alpha}\), which is the exponent commonly used to describe the dependence of \(\sigma_{sp}\) on wavelength) and single scattering albedo (SSA) (Jefferson, 2010; Shinozuka et al., 2015). Compared with the first kind, whose \(R^2\) can be about 0.9, instruments used in the second kind of methods are cheaper and easier in operation, but has a lower accuracy of \(R^2\) much lower than 0.9. Applications similar to the second kind are widely used in remote sensing. As shown in Table 1, Earlier 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, $\sigma_{sp}$ or AOD is mainly the proxy of aerosol absolute concentration, while $\text{Å}$ or SSA can be used to reveal the variations of aerosol CCN activity, as shown in Table 1. Based on Köhler 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 $\text{Å}$ can provide information on mean predominate aerosol size (Brock et al., 2016; Kuang et al., 2017a). As a result, $N_{CCN}$ calculation from $\text{Å}$ 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 (Jefferson, 2010; Liu and Li, 2014). SSA is determined by the ratio between the light absorbing carbonaceous and less-absorbing components. Black carbon dominates the absorption of solar radiation and is a main hydrophobic composition as well. 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 $N_{CCN}$ calculation combining SSA, backscatter fraction and $\sigma_{sp}$ still lead to significant deviations, with a 0.6 $R^2$ (Jefferson, 2010). Thus deviations of $N_{CCN}$ calculation based on SSA is of large errors (Jefferson, 2010). As for fRH, there was a study applied aerosol optical quantities ($\sigma_{sp}$ or aerosol optical thickness) with fRH or SSA to calculate $N_{CCN}$ (Liu and Li, 2014). In their study, compared with the combination of SSA and aerosol optical quantities, the combination of fRH and aerosol optical quantities is found to be less effective in
estimating \( N_{CCN} \), even though \( f_{RH} \) directly connected with aerosol hygroscopicity (Liu and Li, 2014). Compared with SSA, previous studies found \( f_{RH} \) to be less effective in estimating \( N_{CCN} \), even though \( f_{RH} \) directly connected with aerosol hygroscopicity (Liu and Li, 2014). This may result from the significant dependence of \( f_{RH} \) on aerosol size (Chen et al., 2014; Kreidenweis and Asa-Awuku, 2014; Kuang et al., 2017a). As mentioned before, PNSD is used for better calculation of \( \kappa \) and \( N_{CCN} \) from \( f_{RH} \) in previous studies (Ervens et al., 2007; Chen et al., 2014). A new method to estimate \( \kappa \) from \( f_{RH} \) and \( \tilde{\kappa} \) was proposed recently (Kuang et al., 2017a; Brock et al., 2016). Based on this method, \( f_{RH} \) 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.

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 an inlet system consisting of a PM10 inlet, an 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 (\( \sigma_{sp} \)), aerosol optical hygroscopic growth factor (\( f_{RH} \)), particle size-resolved activation ratio (AR) and particle number size distribution (PNSD) were obtained.
fRH as well as $\sigma_{sp}$ at three wavelengths were measured by a humidified nephelometer system consisting of two nephelometers (Aurora 3000, Ecotech Inc.) and a humidifier. $\sigma_{sp}$ can be described by a formula of $\hat{A}$:

$$\sigma_{sp}(\lambda)=\beta\cdot\lambda\cdot\hat{A}$$  \hspace{1cm} (1)

where $\beta$ is the aerosol number concentration and $\lambda$ is the wavelength. Thus $\hat{A}$ can be calculated directly from $\sigma_{sp}$ 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 45 minutes from 50% RH to 90% RH. Dried $\sigma_{sp}$ was obtained directly from dried sample aerosol measured by one nephelometer and humidified $\sigma_{sp}$ was obtained from humidified aerosol measured by another nephelometer.

$\text{fRH/RH}$ is defined as:

$$\text{fRH/RH} = \frac{\sigma_{sp}(\text{RH})}{\sigma_{sp}}$$  \hspace{1cm} (2)

where $\sigma_{sp}(\text{RH})$ is the humidified $\sigma_{sp}$ at each RH, the ratio of the humidified $\sigma_{np}$ to the dried $\sigma_{np}$ at each RH. Detailed description of the humidified nephelometer system was illustrated in Kuang et al. (2017a).

The particle size-resolved activation ratio (AR), defined as the ratio of $N_{CCN}$ to total particles, was measured by a system mainly consisting of a differential mobility analyzer (DMA, Model 3081) 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 an 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 ($0.07\%, 0.10\%, 0.20\%, 0.40\% \text{ and } 0.80\%$) were conducted sequentially and with each cycle lasted for 1 hour, and $N_{CCN}$ at 0.07% supersaturation was used in this study. 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, an electrostatic classifier and a CPC (model 3776; TSI, Inc., Shoreview, MN USA) and measured PNSD with diameter lower than 700nm.

In addition, PNSD and $\sigma_{sp}$ 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 $\sigma_{sp}$ at three wavelengths. Details about the four campaigns can be found in Ma et al. (2011), Ma et al.(2016), Kuang et al. (2016) and Kuang et al.(2017a).

2.2. Theories

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

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

where $g(RH)$ is geometric diameter growth factor, $\kappa$ is the hygroscopicity parameter, $S_{RH}$ is the saturation ratiorelative humidity; $\rho_w$ 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 water–air interface; $R$ is the universal gas constant; and $T$ is the temperature.

Accounting for the impact of $\AA$, $\kappa_f$ can be derived directly from fRH (Brock et al., 2016; Kuang et al., 2017a). A single-parameter parameterization scheme proposed by Brock et al. (2016) connects fRH and $\kappa$ by the approximately proportional relationship between total aerosol volume and $\sigma_{sp}$:

$$f(RH) = 1 + \kappa_{sca} \cdot RH/(100-RH)$$

where $\kappa_{sca}$ is a parameter for fitting fRH curves and is found can be used to predict $\kappa_f$ in combination with $\AA$ in recent studies (Brock et al., 2016; Kuang et al., 2017a). This method of calculating $\kappa_f$ based on $\kappa_{sca}$ and $\AA$ was confirmed by good agreement with $\kappa_f$ calculated from fRH and PNSD. This method was confirmed by good agreement with $\kappa_f$.
calculated from fRH and g(RH) (Brock et al., 2016; Kuang et al., 2017a).

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

\[
N_{\text{CCN}} = \int_{\log D_p}^{\log D_{p,\text{max}}} \text{AR}(\log D_p, \text{SS}) \cdot n(\log D_p) d\log D_p \tag{3}
\]

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:

\[
N_{\text{CCN}} = \int_{\log D_c}^{\log D_{p,\text{max}}} n(\log D_p) d\log D_p \tag{4}
\]

where \( D_{p,\text{max}} \) 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 \( N_{\text{CCN}} \). And a critical \( \kappa \) (\( \kappa_c \)) can be calculated by equation (1) and indicated CCN activity and hygroscopicity of particles.

3. Results

3.1. Calculation of \( N_{\text{CCN}} \) 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 wavelengths and the CCN activity at low supersaturation, and thus a reasonable relationship between \( \sigma_{sp} \) and \( N_{\text{CCN}} \) can be achieved. Figure 1 shows the size distribution of cumulative contributions of \( \sigma_{sp} \) at 450nm and \( N_{\text{CCN}} \) at 0.07% with various \( \bar{\lambda} \) and \( \kappa_c \), 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., 2017a). For continental aerosol without sea salt or dust, \( \bar{\lambda} \) varies from 0.5 to 1.8 and \( \kappa_c \) varies from 0.1 to 0.5 (Cheng et al., 2008; Ma et al., 2011; Liu et al., 2014; Kuang et al., 2017b). And as mentioned before, \( \bar{\lambda} \) can be used as a proxy of the overall size distribution of aerosol populations, with smaller \( \bar{\lambda} \) indicating more larger particles. In figure 1, comparisons for \( \bar{\lambda} \) are made between 0.5 and 4.71.9 and for \( \kappa_c \) are made between 0.1 and 0.5. As larger particles contribute more to light scattering and activation, cumulative
contributions of both $\sigma_{sp}$ 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 $\kappa_c$ increases at smaller diameters. In general, major contributions of both $\sigma_{sp}$ and $N_{CCN}$ are made by particles from 200nm to 500nm for various $\hat{A}$ and $\kappa_c$. This implies the feasibility of inferring $N_{CCN}$ from aerosol optical properties.

Because particles smaller than 200nm can activate at supersaturations higher than 0.07% while scatter less light at wavelengths longer than 450nm, which are shown as the light color lines in Figure 1, it’s obvious that significant differences will exist between cumulative contributions of $\sigma_{sp}$ and $N_{CCN}$. This means $\sigma_{sp}$ and $N_{CCN}$ are dominated by different particles and poor correlation between $\sigma_{sp}$ 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 $\hat{A}$ indicates more Aitken mode particles and fewer accumulation mode particles. Thus large particles contribute less for both $\sigma_{sp}$ and $N_{CCN}$ when $\hat{A}$ are higher, characterizing an increase of cumulative contribution curves at smaller diameters. In detail, differences of cumulative contribution curves between 0.5 $\hat{A}$ and 1.9 $\hat{A}$ are about 150nm for $\sigma_{sp}$ and about 100nm for $N_{CCN}$, by estimating the average of differences of diameters where cumulative contributions range from 0.2 to 0.8. Differences between cumulative contribution curves with $\hat{A}$ of 0.5 and 1.7 are about 150nm and 100nm for $\sigma_{sp}$ and $N_{CCN}$, respectively. Changes of cumulative contributions of $N_{CCN}$ and $\sigma_{sp}$ with various $\hat{A}$ reveal that the shape of PNSD can influence the correlation between $N_{CCN}$ and $\sigma_{sp}$. This is confirmed by previous studies in which the $\hat{A}$ is found to play an important role in calculating $N_{CCN}$ from $\sigma_{sp}$ (Shinozuka et al., 2015; Liu and Li, 2014).

The relationship between $\sigma_{sp}$ and $N_{CCN}$ dependent on $\hat{A}$ and $\kappa_c$ is evaluated by calculating $\sigma_{sp}$ and $N_{CCN}$ with different PNSDs (classified by $\hat{A}$) and different $\kappa_c$. In detail, ratios of $N_{CCN}$ to
$\sigma_{sp}$, 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$_{sp}$-is higher than 0 and lower than 10 range from 0 to 10. In general, AR$_{sp}$ are higher for more hygroscopic particles or smaller particles. As particles become more hygroscopic, more CCN can be expected when $\sigma_{sp}$ is fixed. As aerosol populations consist of more smaller CCN-active particles, the increase of $\sigma_{sp}$ is weaker than that of $N_{CCN}$. For example, particles with diameters slightly larger than $D_c$ contribute less to $\sigma_{sp}$ than particles with diameters much larger than $D_c$.

In detail, the sensitivity of AR$_{sp}$ to $\bar{\Delta}$ also changes with $\bar{\Delta}$ and $\kappa_c$. When $\bar{\Delta}$ are higher than 1.4 and $\kappa_c$ is lower than 0.2, AR$_{sp}$ is insensitive to $\bar{\Delta}$. While when $\bar{\Delta}$ are lower than 1 and $\kappa_c$ are higher than about 0.3, AR$_{sp}$ is more sensitive to $\bar{\Delta}$ than $\kappa_c$. Higher sensitivity of AR$_{sp}$ to $\bar{\Delta}$ are found with higher $\kappa_c$ and lower $\bar{\Delta}$, which reveals that particles having mean predominate size smaller 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 $\bar{\Delta}$ 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 $\bar{\Delta}$, $\kappa_f$ and $\sigma_{sp}$ 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 $\kappa_f$ obtained from the humidified nephelometer system to the $\kappa_c$ 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 (Wex et al., 2009; Irwin et al., 2010; Good et al., 2010; Renbaum-Wolff et al. 2016). 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 $\kappa_f$ 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 size-dependent particle hygroscopicity. Kuang et al. (2017a) found a difference around 0.1 between \( \kappa_f \) and \( \kappa \) inferred from \( g(RH) \) measurements for accumulation mode particles whose \( \kappa_f \) is no larger than 0.2. In this study, a simple conversion that \( \kappa_c \) is 0.2 higher than \( \kappa_f \) is used to calculate \( N_{CCN} \), while for \( \kappa_f \) larger than 0.2, a smaller difference of 0.1 between \( \kappa_c \) and \( \kappa_f \) should be used (Kuang et al., 2017a). This simplified relationship between \( \kappa_c \) and \( \kappa_f \) is a rough estimate regardless of the complexity of differences of aerosol hygroscopicity measured by different instruments, but still used in this study 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 \( \kappa_c \) 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. In addition, it is important to note that the value of the difference between \( \kappa_c \) and \( \kappa_f \) is also a rough estimate regardless of the complexity of aerosol hygroscopicity under different conditions, and the influence of \( \Delta \kappa \) deviation on \( N_{CCN} \) calculation needs to be further examined based on field observation. In regions of single aerosol emissions or productions, the actual \( \Delta \kappa \) can be too large (some organic compositions, Wex et al., 2009; Renbaum-Wolff et al., 2016) or too small (inorganic compositions and black carbon) and thus is not suitable for the application of this method.

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 $\kappa_c$ 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 $\sigma_{sp}$ can be found but AR$_{sp}$ of $N_{CCN}$ to $\sigma_{sp}$ stays around 5. On October 23$^{rd}$ and 29$^{th}$, $N_{CCN}$ and $\sigma_{sp}$ are lower than 2000#/cm$^3$ and 500Mm$^{-1}$, respectively. While on October 20$^{th}$, 26$^{th}$ and November 3$^{rd}$, $N_{CCN}$ and $\sigma_{sp}$ are higher than 2000#/cm$^3$ and 500Mm$^{-1}$, respectively. These variations of $N_{CCN}$ and $\sigma_{sp}$ are mainly due to the variation of the particle number concentration rather than the shape of particle size distribution and aerosol hygroscopicity the particle microphysical properties. Variations of AR$_{sp}$ result from the variations of $\AA$ and $\kappa_c$, which indicate the variations of aerosol microphysical properties and chemical compositions.

In general, AR$_{sp}$ is more sensitive to variations of $\AA$ than $\kappa_c$. As mentioned before, the sensitivity of AR$_{sp}$ to $\AA$ is determined by both $\AA$ and $\kappa_f$. In detail, $\AA$ during the campaign mainly ranges from 0.5 to 1.5 and $\kappa_f$ ranges mainly from 0.05 to 0.2, which means that $\kappa_c$ ranges from 0.25 to 0.4. These values of $\AA$ and $\kappa_f$ correspond a significant sensitivity of AR$_{sp}$ to $\AA$, as the lookup table shows in figure 2. The sensitivity of AR$_{sp}$ to $\kappa_c$ is much small and only notable during some short periods (grey bars in Figure 4). For example, from November 5$^{th}$ to 7$^{th}$, variations of $\kappa_f$ and $\AA$ are opposite and result in nearly constant AR$_{sp}$. And from October 30$^{th}$ to November 2$^{nd}$, consistent variations of $\AA$ and $\kappa_f$ lead to greater variations of AR$_{sp}$ than other periods. This weak sensitivity of AR$_{sp}$ to $\kappa_f$ may be due to the uncertainty of $\kappa_c$ calculated from $\kappa_f$ based on the simplified conversion.

This simplified conversion of $\kappa_c$ is examined by comparing $\kappa_f$ and $\kappa_c$ measured in Gucheng campaign, shown in Figure 5. In general, $\Delta\kappa$ that indicates the difference between $\kappa_f$ and $\kappa_c$ is around 0.2 and independent from $\AA$ and $\kappa_c$. Over 80% of $\Delta\kappa$ ranges from 0.1 to 0.3 that confirms applicability of the simplified conversion of $\kappa_c$. However, a notable deviation of $\Delta\kappa$ can be found when $\AA$ is higher than 1.5. High values of $\AA$ 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 $\kappa_c$.

Therefore, considering the deviation of $\kappa_c$ conversion and high sensitivity of $AR_{sp}$ to $\kappa_c$ when $\hat{\Lambda}$ 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 $\hat{\Lambda}$ is lower than 1.5.

Based on the lookup table of $\kappa_c$ and $\hat{\Lambda}$, $AR_{sp}$ is calculated and applied to calculate $N_{CCN}$ with $\sigma_{sp}$. 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 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 $\kappa_c$ conversion on $AR_{sp}$ and $N_{CCN}$ calculation are evaluated in two ways. In the first way, $\Delta \kappa$ of the $\kappa_c$ 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 $\kappa_c$ of 0.34, which is the average of $\kappa_c$ 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 $\kappa_c$ conversions are quite small. The $\Delta \kappa$ difference of 0.05 in $\kappa_c$ conversion only leads to a difference of 10% for the system relative deviation. The correlation coefficient of the calculation using a constant $\kappa_c$ is just a little lower than correlation coefficients of calculations using a $\kappa_c$ conversion. As a result, the method of calculating $N_{CCN}$ is insensitive to the uncertainty of the $\kappa_c$ conversion.

In this study, the insensitivity of calculated $N_{CCN}$ to $\kappa_c$ conversion is partly due to the small variation of $\kappa_f$ during the campaign. On one hand, the variation of $\kappa_c$ can be quite large and cause non-ignorable deviations of calculated $N_{CCN}$. As previous studies of $N_{CCN}$ measurement showed, the variation of $\kappa_c$ is often small and a constant $\kappa_c$ 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 $\kappa_c$ 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 $\kappa_c$ cannot be ignored because the value of the average hygroscopicity is different in various regions during various periods. In summer of NCP, measured $\kappa_f$ 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 $\kappa_c$ may be 10 times larger than measured $N_{CCN}$. To sum up, although the exact value of $\kappa_c$ cannot be obtained from the measurement of the humidified nephelometer system, the influence of $\kappa_c$ 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.

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 $\sigma_{sp}$, Å and $\kappa_f$, 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 $\sigma_{sp}$, Å, $\kappa_c$ and $N_{CCN}$ is analyzed. It is found that the ratio between $N_{CCN}$ and $\sigma_{sp}$, referred to as $\text{AR}_{sp}$, is determined by $\kappa_c$ 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 $\sigma_{sp}$, the hygroscopicity parameter $\kappa$ and Å. However, $\kappa$ derived from measurements of a humidified nephelometer system under sub-saturated conditions (termed as $\kappa_f$)
differs from $\kappa$ under super-saturated conditions which indicate CCN activity (termed as $\kappa_c$). As a result, the conversion from $\kappa_f$ to $\kappa_c$ is needed. Based on previous studies of aerosol hygroscopicity and CCN activity, a simple conversion from $\kappa_f$ to $\kappa_c$ with a fixed difference (referred to as $\Delta \kappa$) of 0.2 is proposed. On the basis of this simple conversion, the method of $N_{\text{CCN}}$ prediction based only on measurements of a humidified nephelometer system is achieved under conditions without sea salt aerosol, dust aerosol, externally mixed aerosol or aerosol near single source regions.

This method is validated with measurements from a humidified nephelometer system and a CCN counter in Gucheng in 2016. During the campaign, both $N_{\text{CCN}}$ and $\sigma_{sp}$ vary with the pollution conditions. $AR_{sp}$ is around 5 and changes with $\tilde{A}$ and $\kappa_f$. The difference between $\kappa_f$ and $\kappa_c$, $\Delta \kappa$, was 0.2±0.1. The agreement between the calculated $N_{\text{CCN}}$ and the measured $N_{\text{CCN}}$ is achieved with relative deviations less than 30%. Sensitivity of calculated $N_{\text{CCN}}$ to conversions from $\kappa_f$ to $\kappa_c$ is studied by applying different kinds of conversions. Results show that calculated $N_{\text{CCN}}$ 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 $N_{\text{CCN}}$, and also proposed a novel method to calculate $N_{\text{CCN}}$ 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 $N_{\text{CCN}}$, 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 $N_{\text{CCN}}$ at lower supersaturations, thus is more applicable for ambient measurements of clouds and fogs in the atmosphere.

Acknowledgement

This work is supported by the National Natural Science Foundation of China (41590872, 41375134 and 41505107).

Andreae, M. O.: Correlation between cloud condensation nuclei concentration and aerosol optical thickness in remote and polluted regions, Atmospheric Chemistry and Physics, 9, 543-556, 2009.


atmospheric aerosol composition and CCN activity, Atmospheric Chemistry and Physics, 9, 7551-7575, 2009.


Kuang, Y., Zhao, C., Tao, J., Bian, Y., Ma, N., and Zhao, G.: A novel method to derive the aerosol hygroscopicity parameter based only on measurements from a humidified nephelometer system, Atmos. Chem. Phys. Discuss., 2017, 1-25, 10.5194/acp-2016-1066, 2017a.

Kuang, Y., Zhao, C. S., Tao, J. C., Bian, Y. X., Ma, N., and Zhao, G.: A novel method for deriving the aerosol hygroscopicity parameter based only on measurements from a humidified nephelometer system, Atmospheric Chemistry and Physics, 17, 6651-6662, 10.5194/acp-17-6651-2017, 2017b.


continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol
particles in theory and experiment, Atmospheric Chemistry and Physics, 8, 1153-1179, 2008.

nuclei in polluted air and biomass burning smoke near the mega-city Guangzhou, China - Part I: Size-resolved measurements and
implications for the modeling of aerosol particle hygroscopicity and CCN activity, Atmospheric Chemistry and Physics, 10, 3365-3383,
2010.

extinction of dried particles: indications of underlying aerosol processes and implications for satellite-based CCN estimates, Atmos.

hygroscopic growth on the aerosol light-scattering coefficient: A review of measurements, techniques and error sources, Atmos.

Wex, H., McFiggans, G., Henning, S., and Stratmann, F.: Influence of the external mixing state of atmospheric aerosol on derived CCN
number concentrations, Geophys. Res. Lett., 37, L10805

Whitehead, J. D., Irwin, M., Allan, J. D., Good, N., and McFiggans, G.: A meta-analysis of particle water uptake reconciliation studies,
Atmos. Chem. Phys., 14, 11833-11841, 10.5194/acp-14-11833-2014, 2014.

Herrmann, H., and Wiedensohler, A.: Relating particle hygroscopicity and CCN activity to chemical composition during the HCCT-2010
field campaign, Atmospheric Chemistry and Physics, 13, 7983-7996, 10.5194/acp-13-7983-2013, 2013.

humidities in secondary organic material produced by α-pinene ozonolysis without inorganic salts." Atmos. Chem. Phys. 16(12): 7969
7979.

Irwin, M., N. Good, et al. (2010). "Reconciliation of measurements of hygroscopic growth and critical supersaturation of
aerosol particles in central Germany." Atmos. Chem. Phys. 10(23): 11737-11752.

during the RHaMBLe discovery cruise." Atmospheric Chemistry and Physics 10(7): 3189-3203.
Figure 1.

Aerosol PNSD (black lines), the cumulative contribution of $\sigma_{sp}$ at wavelength of 450nm and 700nm (dark green lines and light green lines, respectively, green lines), and the cumulative contribution of $N_{CCN}$ at supersaturation of 0.07% (dark red and dark blue lines, red and blue lines) and the cumulative contribution of $N_{CCN}$ at supersaturation of 0.20% (light red and light blue lines) based on measurement in several campaigns in the North China Plain. Solid lines and dashed lines indicate $\lambda$ of 1.7 and 0.5, respectively. Blue lines and red lines indicate $\kappa_c$ of 0.1 and 0.5, respectively.
Figure 2.

Colors represent $\text{AR}_{sp}$ (calculated as $\text{AR}_{sp} = \frac{N_{CCN}}{\sigma_{sp}}$ at 450nm wavelength and 0.07% supersaturation) with different PNSDs (classified by $\hat{\text{A}}$ values) and different $\kappa_c$. Colors represent $\text{AR}_{sp}$ (ratios between $N_{CCN}$ and $\sigma_{sp}$) with $\kappa_c$ and $\hat{\text{A}}$. 
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), $\sigma_{sp}$ 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 A̅ (green dots). The grey bars are periods when the sensitivity of AR_{sp} to κ_c is notable.

Figure 5.

Differences between κ_c and κ_f, referred to as Δκ, with A̅ (positions of dots) and κ_f (colors of dots). Bars represent percentages of Δκ within different ranges.
Figure 6.

Left plot: comparisons of calculated $A_R$ and measured $A_R$ with different conversions of $\kappa_c$ from $\kappa_f$. Right plot: regressions of calculated $N_{CCN}$ and measured $N_{CCN}$ with different conversions of $\kappa_c$ from $\kappa_f$.

<table>
<thead>
<tr>
<th>Campaign</th>
<th>Air mass</th>
<th>Parameter</th>
<th>Caveats</th>
<th>Results</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>ICARTT$^1$ in the northeastern USA and Canada</td>
<td>Polluted air mass</td>
<td>fRH and PNSD</td>
<td>Calculate $N_{CCN}$ with aerosol hygroscopicity constrained by f(RH) and PNSD.</td>
<td>Predict $N_{CCN}$ at SS &gt; 0.3% with a 0.9 $R^2$.</td>
<td>Ervens et al., 2007</td>
</tr>
<tr>
<td>HaChi$^2$ on the North China Plain</td>
<td>Aged continental air mass</td>
<td>PNSD and fRH</td>
<td>Calculate $N_{CCN}$ with the hygroscopicity parameter constrained by f(RH) and PNSD.</td>
<td>Slopes around 1 and $R^2$ around 0.9.</td>
<td>Chen et al., 2014</td>
</tr>
<tr>
<td>TARFOX$^3$ Atlantic seaboard and ACE-2$^4$</td>
<td>Polluted aerosol volume from remote sensing</td>
<td>Retrieved aerosol volume from remote sensing</td>
<td>Predict $N_{CCN}$ from aerosol volumes with empirical number-to-volume concentration ratio</td>
<td>Overestimate up to 5 times</td>
<td>Gasso and Hegg, 2003</td>
</tr>
<tr>
<td>Location</td>
<td>Air Mass</td>
<td>Measurement</td>
<td>Relationship</td>
<td>Result</td>
<td>Reference</td>
</tr>
<tr>
<td>----------</td>
<td>----------</td>
<td>-------------</td>
<td>--------------</td>
<td>--------</td>
<td>-----------</td>
</tr>
<tr>
<td>ACE-2 in northeastern Atlantic</td>
<td>Diverse air mass</td>
<td>Backscatter or extinction profile, CCN at the surface</td>
<td>Retrieve $N_{CCN}$ profile from backscatter (or extinction) vertical profile assuming their ratios are the same to the ratio at surface, which can be calculated by backscatter (or extinction) and $N_{CCN}$ measured at the surface</td>
<td>Predict $N_{CCN}$ on most days for 0.1% SS and on 20%-40% of the days at 1% SS.</td>
<td>Ghan and Collins, 2004</td>
</tr>
<tr>
<td>ARM$^5$ Climate Research Facility central site at the Southern Great Plains</td>
<td>Continental air mass</td>
<td>Backscatter (or extinction) and RH profile, $f_{RH}$ and CCN at surface</td>
<td>Same as Ghan and Collins, 2004.</td>
<td>Explains CCN variance for 25%-63% of all measurements at high supersaturations</td>
<td>Ghan et al., 2006</td>
</tr>
<tr>
<td>TRACE-P$^6$ and ACE-Asia$^7$</td>
<td>Asian outflow over the western Pacific</td>
<td>Aerosol Index (AI), the product of ambient light extinction and Å</td>
<td>Predict $N_{CCN}$ based on empirical relationship between AI and $N_{CCN}$</td>
<td>AI relate well to CCN only with suitably stratified data</td>
<td>Kapustin et al., 2006</td>
</tr>
<tr>
<td>Multiple AERONET measurements</td>
<td>Diverse air mass</td>
<td>Aerosol optical thickness (AOT)</td>
<td>Predict $N_{CCN}$ based on empirical relationship between AOT and $N_{CCN}$ as a power law</td>
<td>Predict $N_{CCN}$ at SS &gt; 0.3% with a 0.88 $R^2$, but have a factor-of-four range of $N_{CCN}$ at a given AOT</td>
<td>Andreae, 2009</td>
</tr>
<tr>
<td>Four ARM sites</td>
<td>Polluted air mass</td>
<td>SSA, backscatter fraction and $\sigma_{sp}$</td>
<td>Estimate $N_{CCN}$ from fitting parameters for the $N_{CCN}$ activity spectra, which can be calculate based on their empirical relationships with aerosol optical properties.</td>
<td>Predict $N_{CCN}$ with slopes around 0.9 and $R^2$ around 0.6.</td>
<td>Jefferson, 2010</td>
</tr>
<tr>
<td>Multiple ARM sites around the world</td>
<td>Diverse air mass</td>
<td>RH, $f_{RH}$, SSA, AOT and $\sigma_{sp}$</td>
<td>Calculate $N_{CCN}$ with $\sigma_{sp}$ (or AOT) based on their empirical relationship, whose impact on RH, $f_{RH}$ and SSA,</td>
<td>Achieve the best results by using $\sigma_{sp}$ and SSA. Weakly affect on the $\sigma_{sp}$-$N_{CCN}$.</td>
<td>Liu and Li, 2014</td>
</tr>
</tbody>
</table>
Multiple ARM sites around the world. Diverse air mass not dominated by dust. Calculate \( N_{\text{CCN}} \) with light extinction coefficient and \( \text{extinction} \) based on their empirical relationship. Deviate typically within a factor of 2.0.

Table 1: Review of studies that have used aerosol optical parameters to infer \( N_{\text{CCN}} \).

2. Haze in China.
3. Troposphere Aerosol Radiative Forcing Experiment.
4. Second Aerosol Characterization Experiment.
5. Atmospheric Radiation Measurement.
6. Transport and Chemical Evolution over the Pacific.