CCN predictions: Is theory sufficient for assessments of the indirect effect?

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[1] This study quantitatively assesses the sensitivity of cloud droplet number (CDNC) to errors in cloud condensation nuclei (CCN) predictions that arise from application of Köhler theory. The CDNC uncertainty is assessed by forcing a droplet activation parameterization with a comprehensive dataset of CCN activity and aerosol size and chemical composition obtained during the ICARTT field campaign in August 2004. Our analysis suggests that, for a diverse range of updraft velocity, droplet growth kinetics and airmass origin, the error in predicted CDNC is (at most) half of the CCN prediction error. This means that the typical 20–50% error in ambient CCN closure studies would result in a 10–25% error in CDNC. For the first time, a quantitative link between aerosol and CDNC prediction errors is available, and can be the basis of a robust uncertainty analysis of the first aerosol indirect effect.
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1. Introduction

[2] Köhler theory, which predicts when aerosols can activate into cloud droplets, lies at the heart of all physically-based approaches used for assessments of the aerosol indirect effect. Cloud condensation nuclei (CCN) ''closure'', or, the ability to theoretically reproduce observed CCN concentrations from aerosol size distribution and chemical composition, is the ultimate test of Köhler theory. Closure for ambient aerosol has not always been successful [e.g., *Liu et al.*, 1996; *Covert et al.*, 1998; *Chuang et al.*, 2000; *Wood et al.*, 2000; *Cantrell et al.*, 2001; *Zhou et al.*, 2001; *Roberts et al.*, 2002; *VanReken et al.*, 2003; *Rissman et al.*, 2006] and has been attributed to measurement uncertainties, assumptions on aerosol mixing state and chemical composition, as well as the inability of theory to fully describe the activation of carbonaceous CCN.

[3] CDNC closure studies, in addition to estimation of CCN properties from aerosol size and composition, require observations of updraft velocity, *w*, hence are subject to larger uncertainty than CCN calculations. Nevertheless, CDNC closure has often been successful, even when CCN closure is not achieved [*Snider and Brenguier*, 2000; *Chuang et al.*, 2000; *Snider et al.*, 2003; *Conant et al.*, 2004; *Meskhidze et al.*, 2005; C. Fountoukis et al., Aerosol-cloud droplet concentration closure for clouds sampled during ICARTT 2004, manuscript in preparation, 2006, hereinafter referred to as Fountoukis et al., manuscript in preparation, 2006]. Although based upon a limited set of conditions, this finding suggests that large error in predicted CCN concentration may not necessarily yield large CDNC error. A comprehensive CCN-to-CDNC error analysis is required to assess whether this is a consistent feature of ambient clouds.

[4] This study quantitatively assesses the sensitivity of CDNC to errors in CCN concentrations, with a focus on the error arising from application of ''simple'' Köhler theory. The CDNC uncertainty is assessed for diverse cloud formation conditions by forcing a droplet activation parameterization with a comprehensive in-situ dataset of CCN and aerosol observations.

2. Methodology

[5] Our analysis employs ground-based measurements of aerosol size, composition and CCN concentrations over a climatically important range of water vapor supersaturation, *s*. Köhler theory, combined with in-situ observations of aerosol size distribution and chemical composition yields ''predicted'' CCN spectra (i.e., the aerosol number that will activate into droplets as a function of water vapor *s*). These ''observed'' and ''predicted'' CCN spectra are introduced into a droplet activation parameterization; CDNC is computed over a wide range of updraft velocities, *w* and water vapor accommodation coefficients, α. The parameterization is used instead of a cloud parcel model because CCN spectra in the former are explicitly introduced; thus errors in CCN concentration can then be explicitly related to errors in cloud droplet number. Using a cloud parcel model would be cumbersome; the minor improvements in accuracy were not sufficient to justify its usage.

2.1. CCN and Aerosol Measurements

[6] The observations dataset used in this study were obtained at the Thompson Farm (TF) site during the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) campaign (July–August 2004). TF is one of the four Atmospheric Investigation, Regional Modeling, Analysis and Prediction (AIRMAP) monitoring stations maintained by the University of New Hampshire. TF is primarily characterized by regional aerosol mixed with some local biogenic sources [e.g., *DeBell et al.* 2004]. A detailed description and analysis of the dataset is given by J. Medina et al. (Cloud Condensation Nuclei (CCN) closure on New England ambient aerosol during ICARTT 2004 field campaign: a) effects of size-

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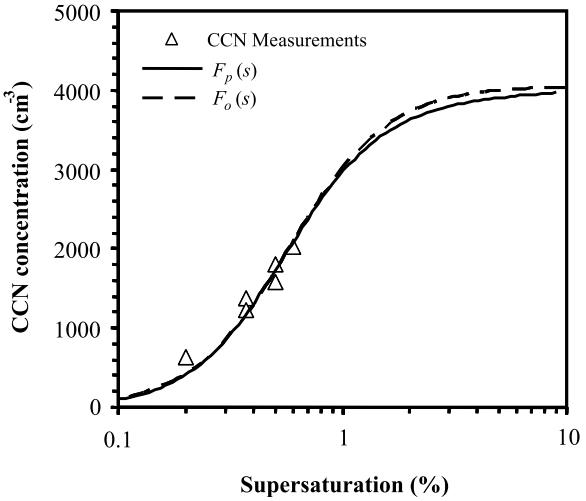

Figure 1. An example of CCN spectra obtained between 12:30 and 13:30 (LST) on August 5, 2004. The CCN observations are shown in open triangles, the dashed line represents the "observed" spectrum fit, and the solid line corresponds to "predicted" spectrum fit obtained from the observed aerosol size distribution and chemical composition.

dependent composition, manuscript in preparation, 2006, hereinafter referred to as Medina et al., manuscript in preparation, 2006). Measurements of the aerosol size distribution were obtained every two minutes with a TSI Scanning Mobility Particle Sizer (SMPS, model 3080) that included a Condensation Particle Counter (CPC, model 3010) and a long Differential Mobility Analyzer (DMA, model 3081L) [www.tsi.com]. Aerosol mobility diameter ranged between 7 and 289 nm, while the DMA operated with a sheath to aerosol flow ratio of 10:1. Simultaneously, size-resolved chemical composition was measured every 10 minutes with an Aerodyne Aerosol Mass Spectrometer (AMS) [*Jayne et al.*, 2000]. Köhler theory was then applied to obtain the "predicted" CCN spectra.

[7] CCN concentrations were measured at 0.20, 0.30, 0.37, 0.50, and 0.60% s with a Droplet Measurement Technologies, Inc. (DMT) streamwise thermal-gradient cloud condensation nuclei counter [*Roberts and Nenes*, 2005; *Lance et al.*, 2006]. CCN concentrations were measured for 6 minutes at each s, allowing a spectrum to be obtained every 30 minutes. CCN and aerosol concentrations are then hourly averaged. A total of 100 CCN spectra and aerosol size distributions, measured between August 5 and 9, are used in our analysis. The aerosol number concentration ranged between 1366–8419 cm^{-3} with an average of 3786 ± 1360 cm^{-3} and the sulfate mass fraction ranged between 0.06 and 0.54 with an average of 0.24 ± 0.09.

2.2. Cloud Droplet Formation Parameterization

[8] The *Nenes and Seinfeld* [2003] activation parameterization, together with its recent extensions by *Fountoukis and Nenes* [2005] is one of the most comprehensive, robust and flexible formulations available for global models. Its accuracy has been evaluated with detailed numerical cloud parcel model simulations [*Nenes and Seinfeld*, 2003; *Fountoukis and Nenes*, 2005] and in-situ data for cumuliform and stratiform clouds of marine and continental origin [*Meskhidze et al.*, 2005; Fountoukis et al., manuscript in preparation, 2006]. The parameterization is based on the framework of an ascending cloud parcel and utilizes the concept of "population splitting", in which droplets are classified by the proximity to their critical diameter. The latter allows the computation of parcel maximum s and CDNC from the solution of an algebraic equation. Population splitting allows for the computationally efficient and rigorous treatment of almost any aerosol size distribution function; formulations are currently available for sectional [*Nenes and Seinfeld*, 2003] and lognormal aerosol distributions [*Fountoukis and Nenes*, 2005]. The parameterization can accurately treat externally mixed aerosol with size-varying composition and complex "chemical effects" [e.g., *Nenes et al.*, 2002], such as the presence of slightly soluble species and surfactants that depress surface tension and droplet growth kinetics.

2.3. Analysis

[9] CCN observations are by nature discrete and cannot be directly used in cloud models for accurate calculation of droplet number. This issue is resolved by fitting the observations to a prescribed functional relationship; we employ the "modified power law" CCN spectrum [*Cohard et al.*, 1998, 2000],

$$F(s) = \frac{kCs^{k-1}}{(1+\eta s^2)^{\lambda}} \quad (1)$$

where $F(s)$ is the CCN concentration as a function of s (i.e., "CCN spectrum"), C is the total aerosol concentration (cm^{-3}), and k, η and λ are unitless coefficients determined from the fitting. The numerator in equation (1) is the "power law" expression of *Twomey* [1959]; *Cohard et al.* [1998, 2000] introduced the denominator so that $F(s)$ asymptotes to C at high s (∼10% for ambient aerosol).

[10] The "observed" CCN spectra, $F_o(s)$, are obtained by fitting equation (1) to two sets of constraints: *i*) the CCN measurements, and, *ii*) $F_o(s) \approx C$ when $s \geq 8\%$ (i.e., all CCN activate when $s \geq 8\%$). The "predicted" CCN spectra, $F_p(s)$, are obtained by fitting equation (1) to CCN concentrations calculated from the size distribution, chemical composition and Köhler theory. $F_o(s)$ and $F_p(s)$ are determined for each hourly average in the dataset, and, the values for k, η and λ are obtained from least squares minimization. Figure 1 presents an example of the fitting procedure. The CCN observations (open triangles) are fitted to the "observed" CCN spectrum (dashed line), while the computed CCN concentrations are fitted to the "predicted" CCN spectrum (solid line). The fitting does an excellent job of reproducing the measurements, with a residual error usually within experimental uncertainty.

[11] Figure 2 presents the intercomparison between $F_o(s)$ and $F_p(s)$ for all 100 spectra and for s between 0.1% and 1.0%. $F_o(s)$ and $F_p(s)$ on average agree to within 12% and mostly to within 50%. The error statistics and tendency for CCN overprediction is consistent with published closure studies [e.g., *Covert et al.*, 1998; *Guibert et al.*, 2003], and can arise from measurement uncertainty, simplifying assumptions regarding aerosol mixing state and composition [*Wood et al.*, 2000; *Snider and Brenguier*, 2000; Medina et al., manuscript in preparation, 2006].

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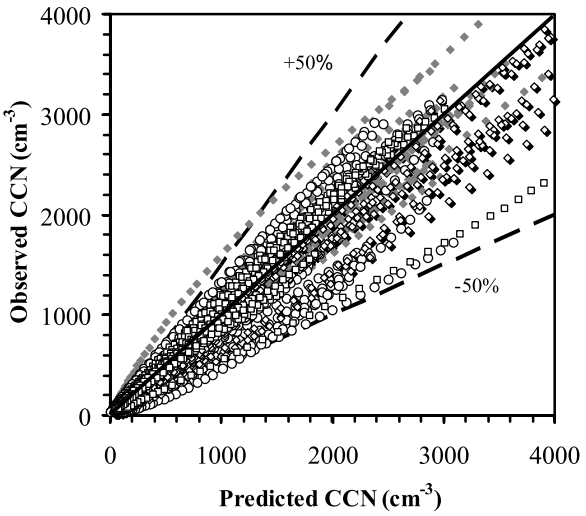

Figure 2. Observed versus predicted CCN concentrations for supersaturations ranging between 0.1 and 1%. The whole dataset used in this study is shown.

[12] CDNC is computed using the sectional version of the *Nenes and Seinfeld* [2003] parameterization with the mass transfer corrections introduced by *Fountoukis and Nenes* [2005]. $F_o(s)$ and $F_p(s)$ are discretized over 100 equally-spaced bins in s space ranging from 0.01 to 10 % s. N_d is calculated for w between 0.01 and 10 m s^{-1}, and for α between 5×10^{-3} and 1.0 [*Lance et al.*, 2004].

3. Results and Discussion

[13] Using observations as a reference, we express our simulations (for each value of w and α) in terms of a fractional CCN error, ε_{CCN}, and the fractional CDNC error ε_{CDNC},

$$\varepsilon_{CCN} = \frac{F_o(s_o) - F_p(s_o)}{F_o(s_o)}; \quad \varepsilon_{CDNC} = \frac{N_{d,o} - N_{d,p}}{N_{d,o}} \tag{2}$$

where s_o is the maximum s predicted for the "observed" CCN spectrum; $N_{d,p}$ and $N_{d,o}$ are the simulated CDNC for the "predicted" and "observed" spectra, respectively. Dividing the fractional errors yields the relative sensitivity of CDNC to CCN, Φ,

$$\Phi = \frac{\varepsilon_{CCN}}{\varepsilon_{CDNC}} = \left(\frac{N_{d,o}}{F_o(s_o)}\right)\left(\frac{F_o(s_o) - F_p(s_o)}{N_{d,o} - N_{d,p}}\right) \tag{3}$$

Φ is ideally suited for quantifying the CDNC error from application of Köhler theory. As Φ increases, CDNC becomes less sensitive to CCN concentration errors (and CCN changes as well).

[14] Figure 3 illustrates the average Φ and average parcel maximum supersaturation, s_{max}, (for all 100 spectra considered in this study), as a function of w. Calculations are shown for three values of α, reflecting currently accepted lower, average, and upper values for the parameter [*Lance et al.*, 2004]. The variability is represented by one standard deviation about the mean. Φ (as well as its variability) tends to increase at lower updrafts and higher α (Figure 3); under such conditions s_{max} is low, competition for water vapor is strong so dynamical adjustments in s_{max} partially compensate for CCN perturbations [*Ghan et al.*, 1998; *Nenes et al.*, 2001; *Rissman et al.*, 2004]. In fact, under polluted conditions and low w, the s response to increases in CCN concentrations can be strong enough to reduce CDNC, [*Ghan et al.*, 1998; *Rissman et al.*, 2004]. At higher w, Φ decreases and becomes insensitive to α because s_{max} is high hence less influenced by CCN perturbations. Perhaps the most striking feature in Figure 3 is that $\Phi \geq 2$ for almost all values of w and α considered. For updrafts above 0.2 m s^{-1}, Φ averages around 2; this means that CDNC calculations are subject to roughly half the error in CCN concentrations for a diverse set of cloud formation conditions.

[15] The preceding analysis used observations characteristic of the "polluted" Northeastern United States. It is also important to examine Φ for more pristine and "marine" environments. Lacking such observations, we reduced the concentration (i.e., the C coefficient in equation (1)) of the "observed" and "predicted" CCN spectra by a factor of 10, and then repeat the sensitivity analysis. Φ in this case is very similar to Figure 3 (not shown), only that α has a smaller impact at low w (however, still $\Phi \geq 2$). Therefore, the error in the CCN concentrations is about twice the error in the estimated CDNC, regardless of airmass origin, w and droplet growth kinetics.

4. Summary

[16] Regardless of w, droplet growth kinetics and airmass origin, we find that CDNC prediction errors are inherently less, about half (or less) of CCN prediction errors. Some profound implications arise from this finding. First, given that CCN closure is typically achieved to within 20–50%, our analysis suggests that the CDNC error resulting from application of Köhler theory currently used in global climate models is likely to be within 10–25%. This error range is corroborated with the most recent CDNC closure studies. Second, our analysis provides for the first time a quantitative link between aerosol and CDNC prediction errors. Although there is still significant uncertainty associ-

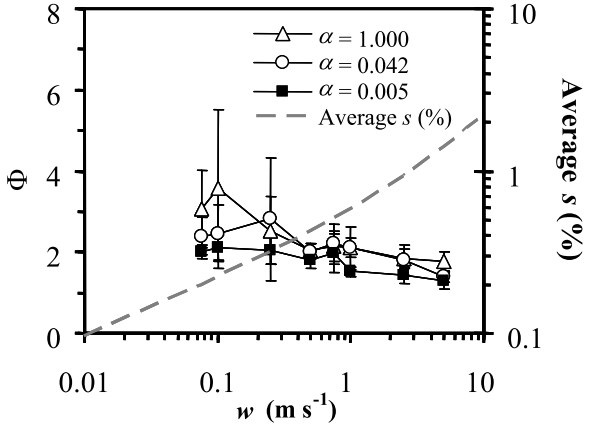

Figure 3. Average Φ and maximum supersaturation as a function of w. The variability is represented by one standard deviation about the mean. Φ is shown for three values of α. Maximum supersaturation (grey dashed line) is shown for α = 0.042.

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ated with updrafts and aerosol-cloud interactions, global aerosol models are continuously being evaluated for their predicted size distribution and chemical composition; provided that Figure 3 is applicable to a wide range of environments, one can apply our analysis to directly link uncertainty in predicted CCN to uncertainty in cloud droplet number and provides the basis for a robust uncertainty analysis of the "first" indirect effect. A first order estimation [*Seinfeld and Pandis*, 1998] suggests that a 10–25% uncertainty in global droplet concentration yields a 0.5 W m^{-2} uncertainty in indirect forcing. A comprehensive assessment would need the use of a Global Climate Model and will be the focus of a future study.

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