Evaluation of a new cloud droplet activation parameterization with in situ data from CRYSTAL-FACE and CSTRIPE

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[1] The accuracy of the 2003 prognostic, physically based aerosol activation parameterization of A. Nenes and J. H. Seinfeld (NS) with modifications introduced by C. Fountoukis and A. Nenes in 2005 (modified NS) is evaluated against extensive microphysical data sets collected on board the Center for Interdisciplinary Remotely Piloted Aircraft Studies (CIRPAS) Twin Otter aircraft for cumuliform and stratiform clouds of marine and continental origin. The cumuliform cloud data were collected during NASA's Cirrus Regional Study of Tropical Anvils and Cirrus Layers–Florida Area Cirrus Experiment (CRYSTAL-FACE, Key West, Florida, July 2002), while the stratiform cloud data were gathered during Coastal Stratocumulus Imposed Perturbation Experiment (CSTRIPE, Monterey, California, July 2003). In situ data sets of aerosol size distribution, chemical composition, and updraft velocities are used as input for the NS parameterization, and the evaluation is carried out by comparing predicted cloud droplet number concentrations (CDNC) with observations. This is the first known study in which a prognostic cloud droplet activation parameterization has been evaluated against a wide range of observations. On average, predicted droplet concentration in adiabatic regions is within ~20% of observations at the base of cumuliform clouds and ~30% of observations at different altitudes throughout the stratiform clouds, all within experimental uncertainty. Furthermore, CDNC is well parameterized using either a single mean updraft velocity $\overline{w}$ or by weighting droplet nucleation rates with a Gaussian probability density function of w. This study suggests that for nonprecipitating warm clouds of variable microphysics, aerosol composition, and size distribution the modified NS parameterization can accurately predict cloud droplet activation and can be successfully implemented for describing the aerosol activation process in global climate models.

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1. Introduction

[2] The radiative effects of atmospheric aerosols arise from scattering and absorption of solar and thermal infrared radiation (the "direct effect") and modulating cloud reflective properties and lifetime (the "indirect effect") [*Twomey*, 1977; *Albrecht*, 1989; *Ackerman et al.*, 2000; *Rosenfeld*, 2000; *Intergovernmental Panel on Climate Change* (*IPCC*), 2001]. A major source of the uncertainty in assessments of the indirect effect originates from complex aerosol-cloud interactions; therefore comprehensive assessment of the aerosol indirect effect (AIE) requires significant improvements in representation of aerosol-cloud interaction processes in global climate models (GCM) [*IPCC*, 2001].

[3] Formation ("activation") and subsequent growth of cloud droplets is a complex process controlled by dynamical (i.e., temperature, pressure, air parcel updraft velocity [e.g., *Pruppacher and Klett*, 1997]) and physicochemical properties of the precursor aerosol (i.e., size distribution and chemical composition [*Köhler*, 1936]). The latter can include presence of soluble gases [*Kulmala et al.*, 1993], partially soluble solutes [*Shulman et al.*, 1996], surface tension depression [*Shulman et al.*, 1996; *Facchini et al.*, 1999], accommodation coefficient changes from the presence of organic surfactants [*Feingold and Chuang*, 2002; *Nenes et al.*, 2002], and even by mass transfer limitations [e.g., *Chuang et al.*, 1997; *Nenes et al.*, 2001]. Although numerical models with a detailed treatment of cloud droplet activation have existed for many years [e.g., *Jensen and Charlson*, 1984; *Flossmann et al.*, 1985; *Pruppacher and Klett*, 1997; *Seinfeld and Pandis*, 1998; *Nenes et al.*, 2001], the computational burden associated with such simulations

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largely prohibits their use in global models; therefore treatments of aerosol-cloud interactions must rely on parameterizations, the development and validation of which represents a major challenge in GCM development [*IPCC*, 2001].

[4] Previous researchers have undertaken a variety of diagnostic investigations to derive the empirical relationships between cloud droplet number concentration (CDNC) and total aerosol mass or aerosol number concentration [*Boucher and Lohmann*, 1995; *Gultepe and Isaac*, 1996]. By using sulfate as a proxy for the atmospheric aerosol, diagnostic approaches bypass the complex physics of cloud droplet formation and allow an assessment of the aerosol indirect effect in a GCM that simulates only atmospheric sulfate concentrations [*Jones et al.*, 1994; *Boucher and Lohmann*, 1995; *Jones and Slingo*, 1996; *Feichter et al.*, 1997; *Lohmann and Feichter*, 1997; *Kiehl et al.*, 2000]. The disadvantage of diagnostic approaches is that they are based on empirical correlations from observations of limited spatiotemporal coverage. In addition, they are inherently subject to uncertainty resulting from unresolved variations in the aerosol size distribution, chemical composition and atmospheric dynamics (i.e., cloud updraft velocity fields) [*Leaitch et al.*, 1996; *Feingold et al.*, 1999; *Menon et al.*, 2002; *Lance et al.*, 2004]. As a result, diagnostic parameterizations yield a wide range in the estimates of the global annual average indirect aerosol forcing, emphasizing the need for more robust, physically based modeling approaches [*Kiehl et al.*, 2000].

[5] Prognostic, physically based parameterizations of cloud droplet formation have emerged within the last decade [*Ghan et al.*, 1993, 1995, 1997; *Abdul-Razzak et al.*, 1998; *Feingold and Heymsfield*, 1992; *Chuang and Penner*, 1995; *Lohmann et al.*, 1999; *Abdul-Razzak and Ghan*, 2000, 2002; *Nenes and Seinfeld*, 2003; *Fountoukis and Nenes*, 2005; *Abdul-Razzak and Ghan*, 2004]. As the first step, the accuracy of these parameterizations is evaluated by comparing their predicted CDNC against that derived from detailed numerical parcel models for a variety of conditions [e.g., *Abdul-Razzak et al.*, 1998; *Nenes and Seinfeld*, 2003; *Abdul-Razzak and Ghan*, 2004]. Good agreement between prognostic parameterizations and parcel models is not sufficient to ensure their successful implementation in GCMs. Prognostic parameterizations require information on turbulent updraft velocity, aerosol composition, size distribution, and mixing state. This information is required for spatial scales typically unresolved by GCMs. The approach for scaling up from cloud scale to that of GCM grid cell remains a source of uncertainty in indirect forcing estimates [*Menon et al.*, 2002]. To address this problem, subgrid-scale variability in updraft velocity is often diagnosed using probability density functions (PDF), and CDNC is calculated as a weighted average over the velocity spectrum [e.g., *Ghan et al.*, 1997]. However, the relevance of these PDFs to cloud droplet activation [e.g., *Feingold and Kreidenweis*, 2000] as well as the importance of other effects currently not included in parameterizations (i.e., influence of entrainment on droplet activation process) can only be evaluated by testing parameterizations and their implementation algorithms with observational data.

[6] In this work, we evaluate the performance of the prognostic aerosol activation parameterization of *Nenes and Seinfeld* [2003] (referred to as NS hereinafter) and its recently modified version by *Fountoukis and Nenes* [2005] (modified NS). The evaluation is done against aircraft data collected in low-level cumuliform and stratiform clouds formed in air masses of marine and continental origin. Accurate parameterization of aerosol-cloud relationships in these cloud types within GCMs is crucial; by affecting the Earth's planetary albedo, stratiform clouds play a key role in the climate system, while cumuliform clouds are important in convective transport of reactive constituents and water vapor [e.g., *IPCC*, 2001]. It is also well established that the microphysics of low-level stratiform clouds are highly sensitive to changes in droplet number concentration [e.g., *Albrecht*, 1989], as their moderate geometrical thickness is such that clouds are often on the verge of forming precipitation. As a result, slight variations in CDNC can have a large impact on the lifetime of stratiform clouds and the AIE [e.g., *Menon et al.*, 2003].

[7] The cumulus cloud data used in this study were collected during NASA's Cirrus Regional Study of Tropical Anvils and Cirrus Layers–Florida Area Cirrus Experiment (CRYSTAL-FACE, on the Web at http://cloud1.arc.nasa.gov/crystalface/), and the stratiform data were gathered during the Coastal Stratocumulus Imposed Perturbation Experiment (CSTRIPE, http://www.cstripe.caltech.edu/). Both missions employed the Center for Interdisciplinary Remotely Piloted Aircraft Studies (CIRPAS) Twin Otter aircraft. Although several studies have been conducted in the past to test cloud droplet concentration parameterizations used in GCMs [*Gultepe and Isaac*, 1996; *Lin and Leaitch*, 1997], to our knowledge, this is the first study in which the performance of a prognostic GCM aerosol activation parameterization is tested using a wide range of in situ conditions and cloud types.

2. Aerosol Activation Parameterization

[8] The NS parameterization can employ a sectional (binned) or modal (lognormal) representation of the aerosol size distribution and chemical composition. By defining separate size distributions for multiple aerosol populations, the parameterization can treat the activation of complex, externally mixed aerosols. In this work, we employ the sectional formulation; the critical supersaturation (S_c) corresponding to the section boundaries is calculated using modified Köhler theory. The theory includes the presence of surfactants and slightly soluble species [*Shulman et al.*, 1996; *Laaksonen et al.*, 1998]. The computed cloud condensation nucleus (CCN) spectrum is then included within the dynamical framework of an adiabatic parcel with a prescribed updraft velocity (or cooling rate) to compute the maximum supersaturation ($S_{\max}$) achieved in ascending air parcel. The CCN that satisfy the criteria $S_c \leq S_{\max}$ activate into cloud droplets. It is important to note that, even when the parcel supersaturation exceeds the critical supersaturation of CCN, the activation process is not instantaneous; condensational growth of droplets is explicitly considered. It has been shown that such kinetic limitations, if not taken into account, can lead to significant errors in the prediction of the number of activated droplets [*Jensen and Charlson*, 1984; *Chuang et al.*, 1997; *Nenes et al.*, 2001].

[9] Evaluation of the NS parameterization against a detailed numerical cloud parcel model, showed that, overall, NS closely follows parcel model predictions for a variety of aerosol chemical composition, size distributions and updraft velocities [*Nenes and Seinfeld*, 2003]. Despite good agreement between the NS parameterization and the cloud parcel model, *Nenes and Seinfeld* [2003] pointed out a slight underprediction in CDNC by their parameterization; it was suggested that this underprediction was likely a result of the overestimation of the water vapor condensation rate. Excessive growth of cloud drops causes depletion of the water vapor in the early stage of cloud formation, ultimately leading to the reduction of $S_{\max}$ and underestimation of the activated CCN; i.e., droplet number.

[10] *Fountoukis and Nenes* [2005] attributed the excessive growth of cloud droplets in NS parameterization to an overprediction of the modified water vapor diffusivity (D_v'). Because of gas kinetic effects, D_v' depends on droplet size, and ignoring size dependence can easily lead to more than an order of magnitude overestimation of the water vapor condensation rate [e.g., *Seinfeld and Pandis*, 1998]. An explicit treatment of a size-dependent diffusivity, however, requires an iterative process that may pose a restrictive computational burden when used in a GCM. As an alternative, *Fountoukis and Nenes* [2005] suggested applying size-averaged modified water vapor diffusivity ($\overline{D_v'}$) to the growth of all activated CCN:

$$\overline{D_v'} = \frac{\int_{D_{p\min}}^{D_{p\max}} D_v' dD_p}{\int_{D_{p\min}}^{D_{p\max}} dD_p} \tag{1}$$

where the averaging is carried out over the range $D_{p\min}$ to $D_{p\max}$ in aqueous droplet diameters, and the formulation for size-dependent diffusivity D_v' is adopted from [*Seinfeld and Pandis*, 1998]

$$D_v' = \frac{D_v}{1 + \frac{2D_v}{\alpha_c D_p}\left(\frac{2\pi M_w}{RT}\right)^{1/2}} \tag{2}$$

where α_c is the water vapor mass accommodation coefficient, M_w is the molecular weight of water (kg mol^{-1}), T is the air parcel temperature (K), and R is the universal gas constant (J mol^{-1} K^{-1}).

[11] Substitution of equation (2) into equation (1) and integration yield

$$\overline{D_v'} = D_v\left(1 + \frac{C}{\frac{D_{p\min}+D_{p\max}}{2}} \ln\left(\frac{D_{p\min}+C}{D_{p\max}+C}\right)\right) \tag{3}$$

where

$$C = \frac{2D_v}{\alpha_c}\left(\frac{2\pi M_w}{RT}\right)^{1/2}.$$

Using detailed cloud parcel model simulations, *Fountoukis and Nenes* [2005] determined optimal values of D_{pmin} and D_{pmax}. For α_c = 0.042 used in this study (see below), calculated D_{pmin} = 0.6 μm and D_{pmax} = 5 μm. The average modified diffusivity ($\overline{D_v'}$), given by equation (3), is then used for cloud droplet growth rate calculations in the modified NS parameterization.

3. Observational Data Sets

[12] Data collected on board the CIRPAS Twin Otter aircraft during the two field missions are used in this evaluation study. Low-level cumuliform clouds were the primary cloud types sampled during CRYSTAL-FACE [*Conant et al.*, 2004], while marine stratiform clouds were mainly sampled during CSTRIPE. The cloud sampling technique was similar in both field missions: several flight legs were conducted below cloud base to fully characterize the aerosol distribution and thermodynamic state of the atmosphere, followed by sampling of cloud microphysics and turbulence near cloud base. After completion of cloud base sampling, the aircraft would "spiral up" and transect the cloud at several successively higher constant altitude legs. This allowed characterization of the vertical profile of cloud microphysics and droplet size distribution. A final leg was flown above the cloud system to characterize the properties of the free tropospheric and detrained aerosols. A complete list of the parameters and sampling frequencies is given by *VanReken et al.* [2003], by *Conant et al.* [2004] and at the CSTRIPE website (http://www.cstripe.caltech.edu/).

3.1. Cumuliform Clouds

[13] The NASA CRYSTAL-FACE field mission was conducted in July 2002 in the vicinity of Key West, Florida. The CIRPAS Twin Otter aircraft provided measurements of aerosol concentration and size distribution over a range of 0.003 to 10 μm; CCN concentration at 0.2% and 0.85% supersaturations; cloud droplet number concentration and size distribution over a 0.5 to 1600 μm diameter range; aerosol chemical composition (sulfate, nitrate, ammonium and organic carbon), and updraft velocity [*VanReken et al.*, 2003; *Conant et al.*, 2004].

[14] Several closure studies were conducted using the CRYSTAL-FACE data. *VanReken et al.* [2003] evaluated aerosol-CCN closure, by comparing observed CCN concentrations with those predicted from measurements of aerosol size distribution and chemical composition. Under the assumption that the aerosol was composed entirely of ammonium sulfate, it was found that observed CCN concentrations at 0.2% and 0.85% supersaturation (S) agreed on average within 20% of that predicted by Köhler theory [*VanReken et al.*, 2003]. *Conant et al.* [2004] carried out aerosol-CDNC closure by comparing measured cloud droplet number concentrations with predictions from parcel model theory using observed updraft velocities and below-cloud aerosol and atmospheric properties. Within adiabatic cloud regions, the discrepancy between model calculated and measured CDNC was typically less than the experimental uncertainty of ~20%. Such degree of closure between aerosol size distribution, CCN spectrum and cloud droplet concentrations rules out significant errors due to the measurement techniques and/or an incomplete understanding of the processes affecting aerosol activation

Table 1. Summary of Cloud Observational Data From CRYSTAL-FACE

Flight Number–Cloud Number	$\overline{w}$, m s^{-1}	σ_w, m s^{-1}	CDNC Observed, cm^{-3}	Parameterization Predicted CDNC, cm^{-3}	
				NS	Modified NS
H4-1	1.4	0.4	567	335	405
H4-2	1.0	0.3	619	344	435
H4-3	0.9	0.3	361	247	303
C4	2.2	0.8	805	749	955
C6-1	0.9	0.35	186	185	239
C6-2	1.8	0.4	243	257	321
C6-3	1.0	0.3	354	368	484
C8-1	1.5	0.3	1036	893	1043
C8-2	1.9	0.3	895	900	1021
C10-1	1.5	0.3	2225	1510	2353
C10-2	1.2	0.3	1942	1303	2097
C11-1	2.8	0.8	1443	1374	1553
C11-2	2.4	0.5	2409	2069	2553
C12-1	2.4	1.0	450	386	443
C12-2	2.2	0.9	484	420	463
C16-1	1.1	0.1	293	185	243
C16-2	1.6	0.4	253	205	266
C17-1	1.7	0.6	447	294	385
C17-2	1.6	0.4	379	256	307
C17-3	2.4	0.7	517	420	534
Mean error[a]				−160	25

[a]The mean error (or bias) was calculated as $(1/N)\sum_{i=1}^{N}(\mathrm{CDNC}_{\mathrm{observed}} - \mathrm{CDNC}_{\mathrm{parameterized}})$, where N is number of measurements.

(see review given by *VanReken et al.* [2003]). The high degree of closure and the wide range of CCN (100 cm^{-3} to 6000 cm^{-3} at $S = 0.85\%$) [*VanReken et al.*, 2003] and cloud core drop concentration (300 cm^{-3} to 2700 cm^{-3}) [*Conant et al.*, 2004] provide an ideal data set for the assessment of aerosol activation parameterizations.

[15] During CRYSTAL-FACE, a total of nine flights, which profiled 20 clouds, were selected for this study (Table 1). Observed total particle number concentrations varied between about 400 and 7000 cm^{-3}, with, on average, higher concentrations in continental than in marine air masses. On the basis of onboard measurements with an Aerodyne aerosol mass spectrometer (AMS), aerosols were composed primarily of sulfate, ammonium and organics, with roughly a 1:1 mass ratio between sulfate and organics. The exception was 18 July (flights C10 and C11 in Table 1), on which elevated concentration of organics occurred (with roughly 1:2 mass ratio between sulfate and organics). Aerosol number concentration for flights C10 and C11 was also several times higher than those for the rest of the mission. Back trajectory analyses confirm their continental origin and suggest that the air masses might have been at ground level some 48 hours prior to being sampled by the aircraft [*VanReken et al.*, 2003].

[16] Aerosol modal diameters were composed mainly of three to four modes and did not exhibit significant variation throughout the mission. Aerosol number distribution in the subaccumulation mode (mode diameter $D_p \approx 10-100$ nm) displayed two peaks, typically in a diameter range between 10–25 nm and 50–80 nm. An accumulation mode ($D_p \approx 0.1-1$ μm) between 0.1 and 0.2 μm was also present more than half of the time. The concentration of supermicron ($D_p > 1$ μm) particles was usually very small (few particles per cm^{-3}); nonetheless, supermicron particles are included in our simulations as their water uptake can be significant enough to reduce the maximum supersaturation reached in cloud updrafts and influence droplet concentration.

[17] Chemical effects, from the presence of soluble gases (i.e., HNO_3) and various organic species, may influence the cloud droplet activation process [e.g., *Nenes et al.*, 2002]. However, as noted above, *VanReken et al.* [2003] showed that assuming the aerosol was composed of pure ammonium sulfate (consistent with the observed NH_4^+:SO_4^{2-} molar ratios) provided good aerosol-CCN closure for the CRYSTAL-FACE data set. The degree of aerosol-CDNC closure achieved by *Conant et al.* [2004] further suggests that for the Twin Otter flights considered here, chemical effects on cloud droplet activation and growth were rather minor.

[18] Entrainment of cloud-free air and droplet coalescence are two main factors that can alter observed CDNC and size distribution from adiabatic values (calculated on the basis of the subcloud measurements). Therefore careful screening of observational data sets is required before a useful comparison between parameterized and observed cloud droplet concentrations can be done. The influence of cloud droplet coalescence on measured CDNC data was removed by considering clouds with no measurable precipitation. In addition, since the observed droplet spectra showed significant variation with height inside the cumulus clouds of several kilometers in vertical extent, only observations within 50–100 m of cloud base were included for the parameterization evaluation analysis.

[19] Entrainment can reduce cloud droplet number by diluting cloud parcels with cloud-free air and by evaporating some cloud droplets to sizes of unactivated haze ($D_p <$ 1 μm) or below the threshold of the CAPS probe (∼0.5 μm). Dilution can also lower CDNC when the characteristic spatial scale of the measurements (100 m) includes both cloudy and cloud-free air volumes, termed as "cloud edges" and/or "holes" [*Hudson and Yum*, 2001]. To reduce the influence of dilution, we follow the procedure of *Conant et*

al. [2004] and consider only measurements with effective droplet diameter greater than 2.4 μm and narrow standard deviation ($\sigma < 1.5$). The liquid water content (LWC) of the remaining data, calculated as an integral over the cloud droplet size distribution, was close to the adiabatic values (determined separately for each cloud on the bases of cloud base location and the subcloud measurements of pressure, potential temperature, and water vapor mixing ratio). As the objective of this study is to evaluate the performance of NS parameterization for clouds characterized by statistical variability in updraft velocity, no further screening of data was carried out to determine mean cloud droplet concentration [e.g., *Conant et al.*, 2004].

3.2. Stratiform Clouds

[20] During CSTRIPE (July 2003), the CIRPAS Twin Otter aircraft was deployed to sample marine stratocumulus clouds near the coast of Monterey, California. The measurement strategy and instrument suite used during CSTRIPE were similar to that of CRYSTAL-FACE and will not be repeated here. The main difference in instrumentation between the two missions was modifications to one of the optical sensors of cloud, aerosol and precipitations spectrometer (CAPS) probe. Calibration tests carried out using field data from three other sensors (the Passive Cavity Aerosol Spectrometer Probe (PCASP) Model 100X, the Gerber total liquid water probe, and the Forward Scattering Spectrometer Probe (FSSP)) resulted in an estimated 40% uncertainty in droplet concentration.

[21] Eight clouds were selected for evaluation of the parameterizations. Observed total particle number concentration below cloud base varied between 500 and 2000 cm^{-3}. Aerosol chemical composition did not exhibit significant variability. Compared to CRYSTAL-FACE, aerosols sampled during CSTRIPE tended to be more acidic. They contained more sulfate and organics (with roughly 1:1 mass ratio) and much less ammonia. Four modes (three in the submicron range and one in the supermicron range) were used to describe the aerosol size distribution. The chemical composition of aerosols was assumed to be ammonium bisulfate (consistent with the observed NH_4^+:SO_4^{2-} molar ratios). Despite that on several occasions the particles contained a significant fraction of organic carbon, this simplified assumption for aerosol chemical composition can be justified by the relatively low sensitivity of CDNC to chemical relative to dynamical (i.e., updraft velocity) effects in this cloud regime. According to *Rissman et al.* [2004], for aerosol organic mass fraction (less than 50%) and the variations in updraft velocities typical of CSTRIPE (see Table 2 and discussion below), aerosol composition should have a relatively minor effect on cloud droplet number. It is further shown that discrepancies between parameterized and measured CDNC without considering aerosol chemical effects lie within the experimental uncertainty.

[22] In contrast to CRYSTAL-FACE, stratus clouds analyzed during CSTRIPE were shallower (cloud thickness was typically less than 400 m) with considerably broader spatial extent. The cloud droplet spectra showed little variation in the vertical direction within cloud depth. Therefore cloud portions selected for the parameterization evaluation, after applying the same algorithm developed for the CRYSTAL-FACE, are significantly more extensive and contain data not only from flight legs conducted at the cloud base (as it was done for CRYSTAL-FACE), but at different altitudes within the cloud.

4. Cloud Updraft Velocities

[23] The sensitivity of predicted CDNC to cloud dynamical effects was evaluated using observational updraft velocities from both missions. Two different methods for CDNC prediction are used. In the first method, CDNC is calculated using a single updraft velocity averaged over the selected clouds; in the second method, cloud droplet activation is calculated with different weighting algorithms using the probability density function (PDF) of updraft velocity (w).

4.1. Prediction of CDNC for Cumuliform Clouds Using Average Updraft Velocity

[24] Observational updraft velocities (obtained using a combination of instruments, including a five-hole gust probe on the nose of the aircraft, a Pitot-static pressure tube, a Coarse/Acquisition Code–Micro-Electro-Mechanical Systems (MEMS) Integrated GPS/INS Tactical System (C-MIGITS) GPS/inertial navigation system (INS), and the NovAtel Differential GPS) for the clouds selected from CRYSTAL-FACE exhibited substantial variability [*Conant et al.*, 2004]. However, as only the data sets from the cloud base flights were included for the parameterization evaluation, statistical variability in w was reduced (see below). Our approach is to estimate CDNCs using a single updraft velocity averaged over the selected cloud sections. Uncertainties in parameterized CDNCs attributed to cloud dynamical effects were evaluated using observed variability in w. Table 1 summarizes cloud observational data used for evaluation of the parameterization.

4.2. Prediction of CDNC for Stratiform Clouds Using PDF of Updraft Velocity

[25] Compared to CRYSTAL-FACE, clouds in CSTRIPE had significantly larger spatial extent. Therefore, compared to the mean w value, cloud sections selected for the parameterization evaluation were characterized by greater statistical variability than those in CRYSTAL-FACE. As cloud droplet activation depends strongly on vertical velocity, it is useful to evaluate two methods for representing dependence of CDNC on w. In the first method, similar to that employed in CRYSTAL-FACE, predictions were made using a single updraft velocity ($\overline{w}$) averaged over the selected cloud sections. The observed variability in w, representing several individual activation events within a cloud, was than used to estimate the uncertainty in the parameterized CDNC. As an alternative to using a single fixed value for the updraft velocity, w variability can be represented by a PDF. Several distributions have been used to account for the spatial variability of w within a stratiform cloud [e.g., *Frisch et al.*, 1995]. In this work we employ a Gaussian PDF with the mean ($\overline{w}$) and standard deviation (σ_w) of updraft velocity summarized in Table 2.

[26] For flight legs conducted at cloud base, measured w corresponds to the updraft velocity governing the maximum supersaturation reached within ascending air parcels. Therefore the PDF of w derived using observed updraft

Table 2. Summary of Cloud Observational Data From CSTRIPE

Flight Number–Cloud Number	Date	$\overline{w}$, m s^{-1}	σ_w, m s^{-1}	CDNC Observed, cm^{-3}	Parameterization Predicted CDNC, cm^{-3}			
					NS	Modified NS	Modified NS With Equation (4a)	Modified NS With Equation (4b)
CS1-1[a]	18 July	0.22	0.21	389	302	390	491	412
CS1-2	18 July	0.28	0.18	481	356	435	499	435
CS1-3	18 July	0.30	0.25	452	372	454	551	467
CS1-4	18 July	0.27	0.22	422	347	434	520	441
CS1-5	18 July	0.28	0.19	456	354	435	506	438
CS1-6	18 July	0.20	0.15	389	282	379	440	377
CS1-7[a]	18 July	0.29	0.24	452	372	442	541	459
CS2-1	21 July	0.24	0.15	275	276	301	333	299
CS2-2[a]	21 July	0.15	0.12	312	190	261	296	258
CS2-3	21 July	0.14	0.11	347	179	256	286	257
CS2-4[a]	21 July	0.18	0.07	355	216	275	287	272
CS2-5	21 July	0.22	0.17	351	268	296	335	296
CS3-1[a]	22 July	0.25	0.19	365	207	328	427	345
CS3-2[a]	22 July	0.20	0.16	426	186	277	373	298
CS3-3	22 July	0.23	0.17	518	198	309	399	324
CS3-4	22 July	0.19	0.14	442	188	266	348	302
CS3-5	22 July	0.32	0.26	400	233	392	514	415
CS3-6	22 July	0.24	0.20	332	207	319	430	343
CS4-1[a]	23 July	0.32	0.27	429	349	428	533	438
CS4-2[a]	23 July	0.17	0.13	409	222	338	383	332
CS4-3[a]	23 July	0.16	0.12	268	211	326	372	323
CS4-4	23 July	0.21	0.21	304	257	367	445	378
CS4-5	23 July	0.17	0.13	317	218	326	383	328
CS4-6	23 July	0.27	0.24	339	327	402	480	412
CS5-1	24 July	0.36	0.29	453	405	447	576	492
CS5-2	24 July	0.54	0.34	365	487	530	655	577
CS5-3	24 July	0.15	0.09	325	341	319	358	314
CS5-4	24 July	0.20	0.16	513	260	366	439	374
CS5-5	24 July	0.25	0.20	384	348	403	484	416
CS5-6	24 July	0.21	0.17	349	265	375	451	376
CS6-1[a]	25 July	0.18	0.15	315	182	226	262	227
CS6-2[a]	25 July	0.15	0.15	360	172	207	254	217
CS6-3	25 July	0.22	0.12	408	187	243	263	238
CS6-4	25 July	0.31	0.21	290	218	279	308	274
CS6-5	25 July	0.22	0.18	269	196	244	281	250
CS6-6	25 July	0.26	0.18	295	205	260	290	257
CS6-7	25 July	0.30	0.20	274	214	275	303	270
CS7-1[a]	26 July	0.20	0.18	319	258	355	427	364
CS7-2[a]	26 July	0.24	0.14	314	328	385	408	376
CS7-3	26 July	0.26	0.19	433	313	401	454	396
CS7-4	26 July	0.37	0.27	541	396	461	520	456
CS7-5	26 July	0.37	0.22	421	396	461	503	450
CS7-6	26 July	0.30	0.14	300	362	428	449	411
CS7-7	26 July	0.28	0.11	514	350	411	429	401
CS8-1[a]	27 July	0.46	0.35	315	371	414	452	402
CS8-2[a]	27 July	0.29	0.20	280	307	352	388	342
CS8-3[a]	27 July	0.44	0.26	301	364	408	433	393
CS8-4[a]	27 July	0.29	0.23	452	307	352	396	346
CS8-5	27 July	0.41	0.29	560	354	400	433	388
CS8-6	27 July	0.48	0.47	370	377	489	472	418
CS8-7[a]	27 July	0.34	0.25	449	328	374	412	364
CS8-8	27 July	0.36	0.29	451	336	382	424	375
Mean error					−94	−22	36[b]	−20[b]

[a]Cloud base flight.
[b]Only cloud base flights were used.

velocities should accurately predict droplet activation. Weighting of predicted CDNC with w is suggested [e.g., *Ghan et al.*, 1997] to account for the higher mass flux associated with stronger updrafts:

$$\mathrm{CDNC} = \int_0^\infty wN_d(w)f(w)\mathrm{d}w \Big/ \int_0^\infty wf(w)\mathrm{d}w \qquad (4a)$$

where $N_d(w)$ is the cloud droplet number concentration corresponding to a given updraft velocity. Since only the upward component of the airflow determines droplet activation and affects CDNC, both $\overline{w}$ and σ_w reported in Table 2 were estimated using only positive values of vertical velocity.

[27] For data sets collected at different altitudes within a cloud, w measured at the cloud base is not available. Thus we have chosen to estimate CDNC using measured in-cloud data of w. However, in-cloud w could be sufficiently different from cloud base w (responsible for droplet activation) to bias CDNC calculated from equation (4a). For

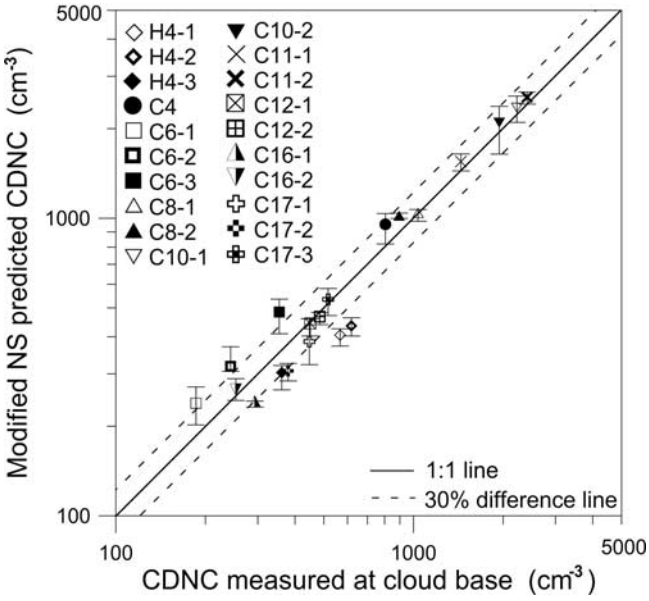

Figure 1. Comparison of cloud droplet number concentration values predicted by modified NS parameterization and measured during CRYSTAL-FACE. The error bars are calculated using 1 standard deviation of the updraft velocity reported in Table 1 (see text for details).

example, the release of latent heat may accelerate rising air parcels and shift the in-cloud PDF toward higher updraft velocities. Enhancing updrafts within the cloud does not necessarily enhance activation; droplets activated at the cloud base can deplete the moisture from vertically accelerating air and prevent in-cloud nucleation of additional droplets on interstitial aerosols [e.g., *Segal et al.*, 2003]. (The absence of a bimodal spectrum in CDNC measured at different altitudes during CSTRIPE suggests that in-cloud activation was likely not important.) For buoyancy-enhanced in-cloud updraft velocities, equation (4a) may lead to overprediction of CDNC. Therefore, for the data sets collected within a cloud, we choose an alternate PDF analysis that reduces the preferential weighting of CDNC for higher updrafts:

$$\mathrm{CDNC} = \int_0^\infty N_d(w) f(w) \mathrm{d}w \Big/ \int_0^\infty f(w) \mathrm{d}w \qquad (4b)$$

The numerical simulations of *Feingold and Kreidenweis* [2000] show that such PDF weighting is quite appropriate for cloud droplet activation in marine stratocumulus environment. The mean CDNC (averaged over all spectra) described by equation (4b) decreases with the increasing variance of the PDF (i.e., as higher w becomes more frequent) and can compensate for buoyancy-enhanced updrafts.

[28] Predictive treatment of CDNC given by equations (4a) and (4b) is particularly important for GCMs, where vertical velocities are commonly diagnosed from large-scale divergence and, because of coarse grid resolution, vertical motions are not well resolved [e.g., *Ghan et al.*, 1997]. To account for subgrid variations in aerosol activation, it is a common practice to integrate the aerosol activation over assumed Gaussian PDF of vertical velocity. Therefore, to test the suitability of NS parameterization to be incorporated in GCMs, we compare the measured CDNC with the parameterized values using different weighting algorithms given in equations (4a) and (4b).

5. Evaluation of Parameterization

[29] The performance of the NS parameterization was evaluated by comparing predicted and observed cloud droplet number concentrations. Two different versions of the parameterization with sectional and modal representation of aerosol size distribution were tested. The CDNC predicted by both versions were similar, so only the results from sectional representation of the parameterization will be discussed below. Each of the four lognormal modes used for the dry aerosol size distribution was divided into 500 size bins spaced equally in log diameter. Dry aerosol composition (NH_4^+, SO_4^{2-}) was specified on the basis of observations. The mass accommodation coefficient of water vapor (α_c) was taken to be 0.042, a value often assumed in cloud models [*Pruppacher and Klett*, 1997; *Shaw and Lamb*, 1999]. Discussion regarding the sensitivity of parameterization results to variations in α_c is given by *Fountoukis and Nenes* [2005].

[30] Figure 1 compares modified NS predicted CDNC with values measured during CRYSTAL-FACE for the conditions summarized in Table 1. This figure shows that on average, CDNC predicted by modified NS parameterization reproduces observations within experimental uncertainty (~20%). This is a robust result, suggesting that for nonprecipitating cumuliform clouds with variable microphysics and aerosol composition, the modified NS parameterization can accurately predict cloud droplet activation and growth. Good agreement between measured and parameterized CDNC (illustrated in Figure 1) is consistent with findings of *VanReken et al*. [2004] and *Conant et al.* [2004] suggesting that for the data sets collected during the CRYSTAL-FACE, a closure analysis based on a relatively simple dry aerosol chemical composition leads to accurate prediction of CDNC.

[31] Figure 2 shows modified NS parameterized versus observed CDNC for stratiform clouds. According to this figure, for the majority of data, CDNC closure was achieved within ~30%, which is less than the actual experimental uncertainty (~40%). Such agreement between parameterized and measured CDNC for the extensive data set collected during CSTRIPE suggests that the modified NS parameterization can accurately predict cloud droplet activation in stratiform clouds of various dynamics, aerosol concentration and size distribution. Compared to Figure 1, however, there is slightly larger scatter along 1:1 line. Such spread could, of course, be a result of the simplified assumption about aerosol chemical composition. However, the absence of apparent bias between measured and parameterized CDNC suggests that discrepancies can be attributed to weak updraft velocities and highly variable cloud dynamics. Table 2 shows that observed in-cloud variability in updraft velocity (σ_w) was often comparable with mean ($\overline{w}$). If species less soluble than ammonium bisulfate (i.e., organics) played a significant role in cloud droplet activation, this would cause a consistent reduction in

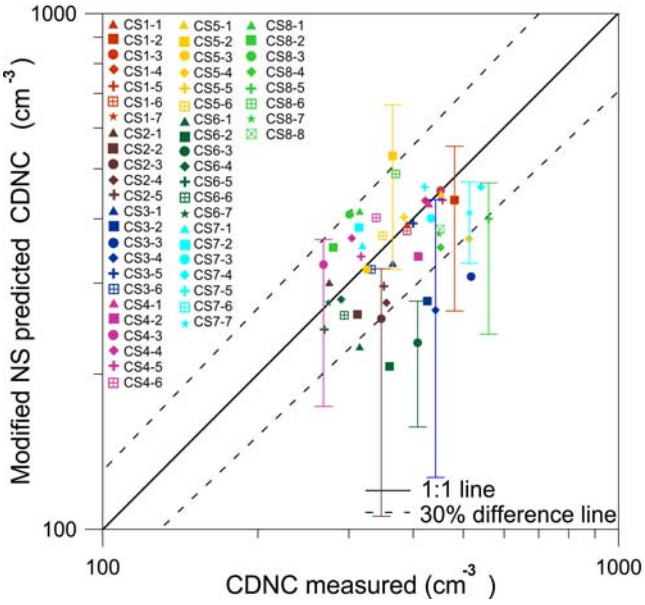

Figure 2. Comparison of cloud droplet number concentration values predicted by modified NS parameterization and measured during CSTRIPE. The error bars are calculated using 1 standard deviation of measured *w* from Table 2 and shown for the data points exhibiting the largest deviation from the observations.

the activated fraction of cloud drops. Figure 2 shows no systematic bias in CDNC closure, suggesting that the aerosol population as a whole was relatively insensitive to the presence of organic species. If we use the largest deviation from the perfect parameterization-observation line as an upper limit for the influence of aerosol chemical composition on CDNC, Figure 2 shows that the sensitivity of parameterized CDNC to the variation of *w* (shown as error bars) was, on average, 2–3 times higher compared to its sensitivity to chemical composition. This result is in agreement with *Rissman et al.* [2004], who show that for marine stratocumulus clouds with organic mass fraction and updraft velocities typically measured during CSTRIPE, the chemical effects, relative to dynamical variability, should contribute less than 40% to the observed CDNC variability.

[32] Figure 3 presents a comparison of CDNC values predicted by modified NS parameterization, using the Gaussian PDF of *w* measured during CSTRIPE. Table 2 shows that when using PDF weighting of updraft velocity, CDNC predicted for in-cloud flights was about twice accurate when equation (4b) was used. The bias toward higher predicted in-cloud CDNC values with equation (4a) is in agreement with our previous analysis that PDF weighting of cloud droplet activation with observed in-cloud updraft velocity may cause overprediction of CDNC in cumuliform clouds. Therefore, in Figure 3, CDNC was predicted using equation (4a) for the cloud base flights, while equation (4b) was used for the data collected within a cloud. Figure 3 shows an absence of systematic bias in the CDNC comparison, and suggests that with the choice of PDF weighting, the modified NS parameterization was able to achieve closure within experimental uncertainty. The lack of significant differences between Figures 2 and 3 suggests that (1) at least for the marine stratocumulus clouds considered in this study, Gaussian PDF weighting given by equations (4a) and (4b) is an appropriate formulation for predicting cloud droplet number; (2) the modified NS parameterization can be reliably used in large-scale models that account for subgrid variation in CDNC by integrating the aerosol activation over assumed Gaussian PDF of *w*.

6. Summary and Conclusions

[33] A wide range of observational data collected on board the CIRPAS Twin Otter aircraft during NASA's CRYSTAL-FACE and the CSTRIPE field campaigns have been used to evaluate the performance of the prognostic, physically based *Nenes and Seinfeld* [2003] and *Fountoukis and Nenes* [2005] cloud droplet activation parameterizations in cumuliform and stratiform cloud regimes. The modified NS parameterization performed remarkably well for a variety of aerosol chemical composition, size distribution and updraft velocities. The CDNC closures achieved for both cumuliform (~20%) and stratiform (~30%) clouds were within the estimated experimental uncertainty. The discrepancies between predicted and observed CDNC are thought to originate from cloud dynamical effects (variability in updraft velocity), rather than variation in the chemical composition of ambient aerosols. The analysis suggests that the modified NS parameterization can accurately describe droplet activation by using detailed description of the aerosol size distribution, chemical composition and updraft velocities.

[34] The extensive data set from the CSTRIPE campaign was also used to evaluate the suitability of using Gaussian probability density function (PDF) weighted vertical velocities for representing cloud droplet activation in marine stratocumulus. As updraft velocities within a cloud may

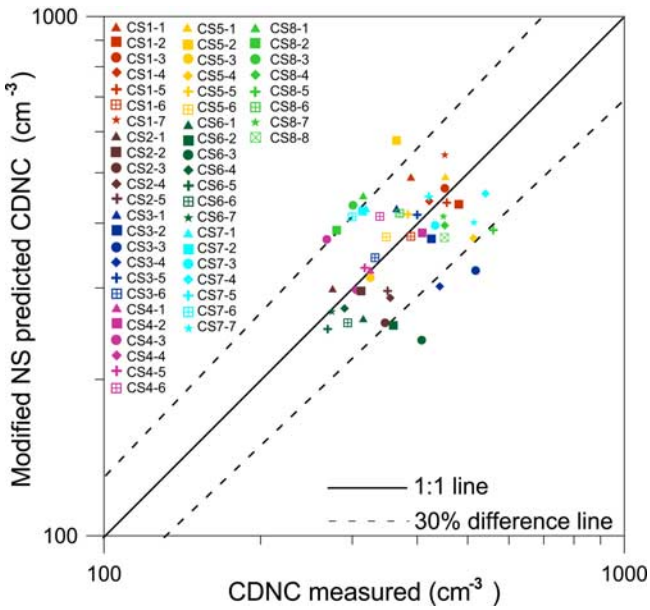

Figure 3. Comparison of cloud droplet number concentration values predicted by modified NS parameterization using Gaussian PDF of vertical velocity and measured during CSTRIPE.

differ significantly from those at the cloud base, two different formulations for CDNC prediction for in-cloud and cloud base flight legs were evaluated. The high degree of CDNC closure achieved for CSTRIPE clouds indicates that the modified NS parameterization can be reliably used in GCMs that diagnose subgrid-scale variability in w. In addition, PDF weighting given by equations (4a) and (4b) may be quite appropriate for predicting CDNC in marine stratocumulus clouds.

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