An explicit cloud microphysics/LES model designed to simulate the Twomey effect

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Abstract

A three dimensional model for simulating the effect of enhanced cloud condensation nuclei (CCN) concentrations on stratocumulus clouds is presented. The model is a large-eddy simulation version of the Regional Atmospheric Modeling System (RAMS) with explicit representation of the CCN spectrum and cloud droplet spectrum. Results of two dimensional tests of the model are presented. A new droplet activation scheme based on a bi-modal size-distribution of CCN is presented. Microphysical sensitivity to a five-fold increase in initial CCN concentrations is examined. Results show that droplet concentrations increase about three-fold, effective radii decrease by 40%, and liquid water contents show less significant variations. These results are consistent with both observations and expectations. We have identified numerically-produced anomalies in the cloud microstructure at the sharp cloud-top boundaries which we believe are associated with the advection scheme. Because the focus of the research is on examining the effects of increasing CCN on cloud albedo, these numerical artifacts need to be addressed.

1. Introduction

Twomey (1974, 1977) hypothesized that increased anthropogenic sources of aerosol will result in the presence of greater numbers of cloud condensation nuclei (CCN), which will produce higher concentrations of cloud droplets and, consequently, more reflective clouds. This effect is believed to have the greatest impact on optically thin marine stratocumulus clouds. It has also been suggested (Albrecht, 1989) that enhanced CCN concentrations will suppress the rate of formation of drizzle drops. This will result in a positive feedback into the CCN-

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albedo link, since reduced drizzle in clouds with higher drop concentrations will result in larger liquid water paths and, hence, more reflective clouds. Recently Charlson et al. (1992) estimated that a 15% increase in global mean droplet concentrations in marine stratus and stratocumulus clouds results in a radiative cooling effect comparable (and opposite in sign) to current estimates of greenhouse warming.

Before this hypothesis can be placed on a sound scientific basis it is necessary to quantify the sensitivity of cloud albedo to CCN concentrations in particular cloud types. The final goal of our research is to provide quantitative estimates of this sensitivity for the special case of marine stratocumulus clouds.

Our approach is to introduce an explicit cloud microphysics scheme into the Regional Atmospheric Modeling System (RAMS) developed at Colorado State University. The RAMS is configured as a large eddy simulation (LES) model in which the major energy-containing eddies are explicitly resolved. Model simulations will produce a large cloud microphysics/macrophysics data base that will then be used by a sophisticated radiative transfer model to evaluate the impacts on cloud albedo.

The aim of this paper is two-fold: (1) to present the RAMS model in its new configuration with explicit microphysics; (2) to demonstrate the credibility of the combined explicit microphysics/LES model with the aid of an experiment showing the sensitivity of the cloud microphysical structure to enhanced CCN concentrations. To accomplish this, we have developed a new droplet activation scheme, which is described in Appendix A. In Section 2 we describe the design and implementation of the LES version of RAMS and the explicit microphysics model. In Section 3 we present results from two-dimensional runs using a sounding from a stratocumulus case observed during the First ISCCP (International Satellite Cloud Climatology Project) Regional Experiment (FIRE I). Section 4 presents a discussion of the results and outlines planned three-dimensional numerical simulations. A summary is given in Section 5.

2. Model description

2.1. RAMS as an LES model

The RAMS is a multi-purpose modeling system that has been applied to LES over inhomogeneous land surfaces (Hadfield et al., 1991, 1992; Walko et al., 1992) and to the simulation of a variety of cloud systems. In this investigation RAMS is set up as a non-hydrostatic LES model. Prognostic equations include those for the three velocity components \( (u, v, w) \), liquid water potential temperature \( (\theta_l) \); Tripoli and Cotton, 1981), perturbation Exner function \( \pi \), and total water mixing ratio \( r_w \) (the sum of vapor and liquid water mixing ratios). In all previous cloud applications a bulk microphysical scheme, which diagnoses cloud liquid water (e.g., Cotton et al., 1986), has been used. In the current explicit microphysics model, condensate is predicted using a droplet spectrum that we resolve into 25
different size bins. Prognostic equations are required for two parameters in each of these bins (see section 2.2.), and the liquid water mixing ratio $r_l$ is the sum of the mixing ratios in each bin. In RAMS, water vapor mixing ratio $r_v$ is diagnosed as the difference between $r_t$ and $r_l$. Temperature $T$ is diagnosed from the prognostic variables $\theta_1$ and $r_l$. This enables calculations of cloud supersaturation $S$:

$$S = \frac{r_v}{r_s(T)} - 1 = \frac{r_t - r_l}{r_s(T)} - 1$$

where $r_s(T)$ is the saturation mixing ratio calculated from polynomial fits to saturation vapor pressure and the diagnosed temperature $T$ (Flatau et al., 1992). In calculating $S$, both the dynamical and microphysical tendencies are accounted for. Tests of the stability of this calculation to model time step show only weak sensitivity for a doubling of the time step from 1 s to 2 s (Cotton et al., 1992).

A further modification of RAMS has been the addition of a positive definite scheme for the advection of scalar variables. The advection schemes operate in a hybrid mode with vector quantities ($u, v, w$) advected using a second-order leap-frog scheme, and scalar variables advected using a positive definite, second-order forward differencing scheme. The positive definite scheme employs the flux scheme of Tremback et al. (1987) renormalized nonlinearly as in Bott (1989).

The RAMS model includes an option for explicit feedback of bulk cloud properties on radiative heating/cooling based on the radiation parameterization developed by Chen and Cotton (1983); however, in this study the parametrization has not been implemented. Sub-grid scale diffusion is parameterized using a Smagorinsky (1963) deformation tensor modified for stability. The lower boundary condition is a surface layer parameterization based on the Louis (1979) scheme, with specified sea surface temperatures. The top boundary is a rigid lid with the option of a Rayleigh friction wave-absorbing layer in the top most grid points. Lateral boundary conditions are specified as cyclic. We have the option of spawning a sequence of finer grids which will allow us to study various processes (such as cloud top entrainment) with increasingly finer resolutions.

2.2. The explicit microphysics model

The explicit microphysics model that has been implemented in RAMS is an accurate moment-conserving scheme developed by the Tel-Aviv University Cloud Physics Group (Tzivion et al., 1987, 1989; Feingold et al., 1988). In this scheme, both the mass and the number mixing ratios are predicted for each bin. This allows the average bin mass to vary both temporally and spatially (in the mass dimension) and enables accurate transfer of mass. Tzivion et al. (1987) showed that the accuracy of this method in representing condensation and collection is comparable to that of schemes that use two to three times the number of bins, but predict on only the mass concentration in each bin (e.g., Bleck, 1970). For stratuscumulus simulations, a total of 25 bins characterize a drop spectrum in the range 1.56–504 $\mu$m (radius), with mass doubling from one bin to the next. The
25 bins are represented by both mass and number mixing ratio and therefore we require 50 prognostic equations for the drop spectrum variables. Because we are primarily concerned with non-precipitating stratocumulus clouds and clouds with only light amounts of drizzle, the specified size range should be adequate. The microphysical processes affecting the drop spectrum are droplet activation from CCN, condensation/evaporation, collision-coalescence, and sedimentation. Drop breakup is not considered, as it is expected to be negligible for these clouds.

Coupling the explicit microphysics with the dynamical model required the development of a droplet activation scheme. Initial results presented in Cotton et al. (1992) were based on Twomey's (1959) work:

$$N_r = \min[N_n, C(S \cdot 100)^k]$$

where $N_r$ is the number of drops per unit volume activated at supersaturations less than $S$, and $N_n$ describes the number concentration of CCN present in a grid volume at a given time. $C$ and $k$ are specified parameters related to the CCN size spectrum (for example, $C = 100 \text{ cm}^{-3}$, $k = 0.7$ are characteristic of a marine environment; Twomey and Wojciechowski, 1969). This type of scheme requires only the advection of the CCN number concentration and activates droplets distributed about a mean radius that is specified a priori.

In an attempt to more realistically model the dependence of the activation process on the available CCN (e.g., Kogan, 1991), we have developed an activation scheme that is based on a bi-modal size distribution of CCN. The general features of this scheme are outlined below. (A detailed description is provided in Appendix A.)

(i) The scheme explicitly resolves the size spectrum of CCN. It divides the CCN spectrum into two parts; the first describes CCN in the size range 0.01–0.1 \(\mu\text{m}\) (radius), while the second describes giant CCN with radii between 0.1 and 10 \(\mu\text{m}\). The upper and lower bounds of these spectra can easily be changed.

(ii) In each range, the CCN spectrum is described by three moments of the size distribution using a lognormal basis function. The 0th, 2nd and 3rd moments have been chosen for the purpose of these simulations; however, in general, any three moments may be used. Thus six scalar variables are used to describe the bi-modal CCN spectrum and six additional prognostic equations need to be solved. This is more efficient than a high-resolution bin description of the CCN and resolves the essential features of the activation process.

(iii) The number of newly activated droplets is calculated from the model-derived supersaturation using Kohler theory. The size spectrum of these droplets is related to the size spectrum of the dry CCN through empirical relations, derived from detailed calculations of the activation process (see Appendix A for details).

(iv) When a droplet evaporates completely, it is assumed that one particle is returned to the atmosphere (Mitra et al., 1992). In this work, the size of returned particles is assumed to conform to the size spectrum of the initial CCN and thus ignores possible processing of the CCN by the cloud. Future work will regenerate CCN in a more realistic fashion.
Using this scheme, we can initiate RAMS with a sounding of CCN and consider vertical and horizontal transport of the CCN by the explicitly represented large eddies. We can simulate their depletion by activation as well as their replenishment due to droplet evaporation. Moreover, we can simulate the interaction between rising plumes of air depleted in CCN and air streams entrained into the cloud containing environmental concentrations of CCN that are characteristic of above-boundary-layer air.

By varying the parameters of the initial CCN spectra, we hope to better represent the relationship between the spectra of newly activated droplets and that of the available CCN. Also, the inclusion of giant nuclei will allow us to study their impact on the collision-coalescence process (e.g., Johnson, 1980). Below we evaluate the impact of increasing the concentration of CCN on simulated cloud microphysical properties.

3. Results

As a preliminary test of the combined explicit microphysics model, RAMS was configured as a two-dimensional model. This is not a true LES model in that a fully three-dimensional simulation is required to properly represent turbulent eddies because vortex stretching is not permitted in two dimensions. As a result, the scale of the eddies is expected to be larger than in a three-dimensional LES simulation and entrainment processes are not properly represented. Nonetheless, these two-dimensional runs provide an opportunity to examine the non-linear coupling between the explicitly represented microphysics and the model dynamics.

The case we simulated is the well-documented FIRE I day of July 7, 1987 (e.g., Betts and Boers, 1990).

3.1. Model configuration and control case

The model domain was oriented in the east–west direction with horizontal winds set equal to the magnitude of the horizontal winds provided by the sounding (which showed little indication of directional shear thus making it amenable to two-dimensional simulations). In the horizontal, the domain was divided into 50-m intervals (80 points over the entire domain). In the vertical, the grid spacing was 50 m below 1500 m, with 10% grid stretching between consecutive levels above 1500 m, for a total of 45 points.

The model was initialized with the stratocumulus sounding of Betts and Boers (1990) with sea surface temperatures 1 K greater than those at the lowest levels and random perturbations of the surface air temperatures not exceeding 0.1 K. The time step was specified at 2 s, and this time step was also used in the microphysical routines.

A uniform CCN spectrum in the 0.01–0.1 \( \mu \)m range with a geometric mean radius \( a_g \) of 0.05 \( \mu \)m and geometric standard deviation \( \sigma_a \) of 2 was used (See Eq.
A1). The ambient CCN concentration was set to 100 cm$^{-3}$ throughout the domain. No large CCN (0.1–10 μm) were included.

The model was run for a total of 3.5 h simulated time. In general the model required about 2 h to produce well defined updrafts and downdrafts with vertical velocity maxima of $\pm 0.5$ m s$^{-1}$. Results are presented at the 3 h period. Because there was a certain amount of variability in cloud structure during the course of each run, the results presented here are not necessarily representative of the cloud structure at earlier and later times.

In the analysis of the results, we look at various horizontal averages of the different cloud properties. We define draft averages as averages taken at constant height $z$ over the extent of the draft. An updraft region is defined as a vertical column centered around the domain maximum vertical velocity; the width of the column is defined on the basis of the point where the vertical velocities decline to half their maximum values. Downdraft regions are defined similarly. In this analysis we are roughly following the data analysis methods of Schmidt and Schumann (1989) and others. We also analyze layer-averages which are averages at a given $z$ over the entire cloudy region (for cloud variables such as drop concentration and $r_l$), or over the entire domain (for CCN and thermodynamic variables). A condition imposed on this analysis is that draft regions are at least 200 m in diameter and layer-averages of cloud variables are at least 500 m in extent.

Figs. 1 and 2 show the liquid water mixing ratio $r_l$ and velocity fields, respectively, 3 h after initiation of the model. A fairly uniform cloud layer fills most of the domain and updrafts and downdrafts peak at about 1 m s$^{-1}$. Cloud top is generally constant between 725 and 775 m; however, the cloud base varies considerably between 375 and 575 m. Ill-defined cloud bases are a common phenomenon in the stratocumulus cloud decks off the coast of Southern California (e.g. Noonkester, 1984). The simulated cloud depths were coincident with observations, as were values for cloud top and estimates of cloud base. (Observational data were provided by J. Nelson in conjunction with the Third International Cloud Modeling Workshop, hereafter Nelson, 1992.)

Liquid water maxima are about 0.9 g kg$^{-1}$ and near cloud top, although local maxima often occur within the interior of the cloud (Fig. 1a). Averaged quantities, however, are more moderate, the typical layer-averaged maxima being 0.5 g kg$^{-1}$. Both the layer- and draft-averaged $r_l$ profiles show a steady, approximately linear increase with height to near the top of the cloud layer followed by a rapid decrease (Fig. 1b). Liquid water paths (LWP) are as high as 140 g m$^{-2}$ (Fig. 1c) and well correlated with regions of convective activity (Fig. 2a, c). At the edges of convective cells LWP's typically have minima between 0 and 30 g m$^{-2}$. This range of values is consistent with observations on this day by Nakajima et al. (1991) which showed LWP probabilities greatest near 75 g m$^{-2}$ and zero above 150 g m$^{-2}$.

Fig. 3a shows contours and Fig. 3b, layer and draft averages, of the droplet number concentrations $N_r$. The values of $N_r$ are of the order of 40–50 cm$^{-3}$ and are fairly constant with height, particularly within the updraft regions. Observations reported values of 50 cm$^{-3}$ for this case (Nelson, 1992). The close agree-
Fig. 1. The control run liquid water mixing ratio field \( r_t \) after 3 h: (a) contours of \( r_t \) in the \( x-z \) domain, superimposed on the wind-field. Contours are labeled in units of \( \text{g kg}^{-1} \times 100 \); (b) vertical profiles of layer-averages (solid line), updraft averages (long-dashed line) and downdraft averages (long-dash-short-dashed line). The averaging method is described in the text; (c) liquid water paths.

The \( S \) (expressed in \%) contours and average profiles (Figs. 4a, b) are well correlated with the cloud \( N_r \) and \( r_t \) fields. The peak values of \( S \) do not exceed 0.4%, and are typically of the order of 0.1%; realistic values for clouds of this type. Notice in Fig. 4a that in the vertical there is a bi-modal structure to the \( S \) field. One peak in \( S \) appears just below cloud top, and another at a level of 525–600 m.
The maximum below cloud-top could be related to cooling through evaporation above the cloud, but we believe that this is a numerical artifact related to the sharp gradient in fields at cloud top. It is thus difficult to ascertain what part, if any, of this maximum is related to well resolved physical processes. In the updraft regions the lower peak is consistent with adiabatic parcel theory which predicts a maximum in $S$ near cloud base. The existence of larger drops near cloud base (or at cloud top) would also tend to enhance $S$, since for equivalent liquid water contents, larger drops are less efficient at taking up vapor than are small droplets. The profiles of drop effective radius $r_e$ (defined by the ratio of the third to second moments of the drop spectrum) in Fig. 5b show that large drops are present near the base of the cloud in the downdraft and layer-average profiles.

Fig. 5a presents contours and average profiles of $r_e$. Values range between 12
Fig. 3. The control run drop number concentration $N_r$ after 3 h: (a) contours of $N_r$ in the $x$–$z$ domain in increments of $10 \, \text{cm}^{-3}$; (b) vertical profiles of average $N_r$ with line types as indicated in Fig. 1b.

Fig. 4. The control run supersaturation $S$ in % after 3 h: (a) contours of $S$ in the $x$–$z$ domain in increments of 0.05%. Dashed lines indicate negative values in increments of 10%; (b) vertical profiles of the average $S$ with line types as indicated in Fig. 1b.

and 18 $\mu$m throughout most of the cloud, except at cloud base, where the existence of drizzle-size drops increases $r_e$ to about 20 $\mu$m. In situ measurements (Nelson, 1992) showed effective radii in that layer in the 10–15 $\mu$m range. Although the simulated values of $r_e$ are close to the observed values, we see later that the value of $r_e$ is sensitive to the CCN spectrum used in the initial conditions.

In the updraft region, $r_e$ increases with height, in accordance with the profiles for $r_l$ and $N_r$. As shown by numerous workers (e.g., Noonkester, 1984; Bower and Choularton, 1992), a linear increase in $r_l$ and constant $N_r$ with height, imply a dependence of $r_e$ on $z^{1/3}$.

Contours of CCN concentrations $N_a$ (Fig. 6a) show the effect of activation
scavenging within the cloud layer. A number of features are noteworthy. Firstly, the average $N_a$ profiles mirror the profiles of $N_r$ as CCN are depleted by droplet activation, or, alternatively, evaporating droplets return CCN to the atmosphere (Figs. 3b, 6b). Secondly, CCN depletion is stronger in updraft regions where $S$ is higher (Fig. 6b). Thirdly, there is a relative maximum in $N_a$ just above the inversion which is probably a numerical artifact due to the sharp gradients in that region.

A series of droplet spectra is shown in Fig. 7, where once again, *draft* averages and *layer* averages have been calculated. The droplet spectra exhibit a typical
skew-symmetric form with a bimodality suggestive of collision-coalescence growth. Drop spectra of very similar form have been observed by Nicholls (1984) and Noonkester (1984). An interesting feature of these spectra is the source of droplets at about 2 μm radius, due to activation in the updraft. This peak is strong from about 375 m (not shown in Fig. 7), is still evident at 525 m (the height of maximum S—see Fig. 2) but decreases with increasing height. It is again evident

Fig. 7. The control run average droplet spectra as a function of height: (a) number size-spectra \( n(r) \) \([\text{# cm}^{-3} \text{μm}^{-1}]\) averaged over the entire cloudy layer (solid line); (b) \( n(r) \) averaged over the main updraft region (long-dashed line); and (c) \( n(r) \) averaged over the main downdraft region (long-dash-short-dashed line). See text for explanation of the conditional sampling. Each plot includes a mass distribution \( m(r) [\text{μg cm}^{-3} \text{μm}^{-1} \cdot 10^2] \), indicated by a short-dashed line.
at a height of 725 m where $S$ is fairly high probably due to numerical causes mentioned above. In the updraft, the trend for the smallest droplets to decrease in number with increasing height may be a result of their collection by larger drops, and the fact that the condensational growth out of the smallest bins is not sufficiently compensated by activation of new droplets.

In the downdraft region, droplet spectra are generally broader than those in the updraft. They comprise larger numbers of small droplets than those in the updraft region but show no evidence of activation, except at 725 m where the downdraft includes regions that are supersaturated and therefore includes spectra with a certain proportion of newly activated droplets.

In summary, we have demonstrated here that despite the limitations of two dimensions, the model simulates the marine stratocumulus fields in a reasonable
manner and that the microphysical fields are similar to those observed on the day in question.

3.2. Sensitivity to a five-fold increase in initial CCN concentrations

One of the purposes of this paper is to examine the model response to a variation in initial CCN concentrations. To this end, we repeated our control run with one difference: the initial CCN concentration was set at 500 cm$^{-3}$ (rather than 100 cm$^{-3}$). The value of $d_a$ was held at 0.05 $\mu$m but $\sigma_a$ was decreased to 1.4 (see Eq. A1). This represents a somewhat narrower CCN spectrum than in the control run but with 5 times as many particles.

Caution should be exercised when making direct comparisons between fields, since each case is evolving at a different rate. A more representative comparison would be one where spatially and temporally averaged fields are compared.
Fig. 10. The $N_i$ field. As in Fig. 3 but for $N_o = 500 \text{ cm}^{-3}$. Number concentrations are about 3 times higher than in the control run. The contour increment is 20 $\text{cm}^{-3}$.

Fig. 11. The $S$ field. As in Fig. 4 but for $N_o = 500 \text{ cm}^{-3}$. Values of $S$ are significantly lower than in the control run. The contour increment for positive $S$ is 0.05%.

Nevertheless, at the 3 h time, the cloud layers exhibit similar broad features, and a general comparison of the microphysical properties is warranted.

Results for this case are compared with the control case at 3 h after the initiation of the model. Values of $r_i$ at cloud top (Fig. 8a), and LWP’s (Fig. 8c) do not differ significantly; however, layer-averaged $r_i$ profiles (Fig. 8b) exhibit a clear two-layer structure that is not evident in the control run (Fig. 1b). The presence of a sink (and associated sharp gradient) of cloud quantities (such as liquid water and drop concentrations) associated with cloud top evaporation causes difficulties for the advective operators. We have conducted 1-dimensional tests of advection which show that oscillations of this nature are characteristic of the numerical solution upstream of an outflow (or sink) point. In simulations of the stratus
Fig. 12. The \( r_e \) field. As in Fig. 5 but for \( N_a = 500 \text{ cm}^{-3} \). The contour increment is 1 \( \mu \text{m} \).

Fig. 13. The \( N_r \) field. As in Fig. 6 but for \( N_a = 500 \text{ cm}^{-3} \). The contour increment is 20 \( \text{ cm}^{-3} \).

topped boundary layer Moeng (1986) also noted large variations in cloud top liquid water. Similar problems have been reported by others (e.g. Deardorff 1980). Thus the high values of \( r_t \) and the two layer structure are most probably numerical artifacts. It is not clear at this time why this feature is more evident in the liquid water fields of the sensitivity run, although one explanation could be that the droplets in this case are smaller (Fig. 12) and the \( r_t \) field is not eroded through sedimentation to the same extent as in the previous case.

Peak vertical velocities are about 40% less, but cover a broader region than those in the control run (Fig. 9a, c). Number concentrations are on the order of three times larger than previously (Fig. 10) regardless of the averaging method (Fig. 10b); the temporal variability in \( N_r \) is much smaller than the difference between the two cases. Supersaturation values are (as expected) much smaller
Fig. 14. Average droplet spectra. As in Fig. 7 but for the run with \( N_a = 500 \text{ cm}^{-3} \). Droplet spectra are narrower than in Fig. 7 and have fewer drizzle drops.

(Fig. 11a) than in the control, although the average profiles are qualitatively similar. The effective droplet radii are of the order of 10 \( \mu \text{m} \) (Fig. 12), i.e., approximately 40% less than in the sensitivity run, a result expected from the enhanced values of \( N_r \), together with similar \( r_i \), and consistent with the Twomey (1974) hypothesis.

Noonkester’s (1984) observations of marine stratocumulus clouds associated with marine and continental air masses bear relevance to this sensitivity study. Noonkester shows \( N_r \) increasing by about 250% over the entire cloud for a lightly polluted continental air mass case vs. a clean marine air mass case. Furthermore,
he shows smaller differences in liquid water contents. If we consider the control run as representative of the clean marine air mass, and the sensitivity run representative of the lightly polluted continental air mass, we find that the trends in liquid water contents, effective radii and number concentrations are consistent with those observed by Noonkester.

Contours of CCN (Fig. 13a) show that unlike the control run, there are still ample unactivated CCN within the cloudy region, particularly in the downdraft (Fig. 13b). In the updraft, less than half of the CCN have been activated, mainly because supersaturations have been suppressed by prior activation events and by condensational growth to existing droplets.

Droplet spectra are plotted in Fig. 14; they tend to be narrower and have fewer large drops than the spectra from the control run. Although some coalescence growth is evident, the amount of liquid water in the larger drops is small and the cloud shows no evidence of precipitation (there is close to zero liquid water below 400 m; see Fig. 8).

4. Discussion

4.1. Overview

The above simulations have supported the hypothesis that the initial CCN size spectrum is important in determining the evolution of marine stratocumulus clouds. Although, from a qualitative point of view, the sensitivity to increased CCN concentrations and to the addition of larger CCN were predictable a priori, we now have the capability of providing quantitative estimates of these effects.

A number of interesting points as well as concerns are raised by this work:

(1) When using the new activation scheme, the values of $S$ generated by the model are low ($<0.5\%$), in agreement with predicted values for marine stratocumulus clouds. Thus, a significant percentage of CCN can exist as unactivated, interstitial CCN (Hudson, 1984). This is especially true for the run with $N_o = 500 \text{ cm}^{-3}$ where we saw that about half of the CCN were not activated. For the case with $100 \text{ cm}^{-3}$, unactivated CCN constituted about 15% of the total.

(2) The highest values of $S$ often occurred near cloud top. This is believed to be a numerical artifact induced by the sharp gradients near cloud top. In a more specific study of this problem Grabowski (1989) concluded that these spurious maxima are clearly associated with "finite-difference approximations to the conservation laws for the thermodynamic variables" and are actually enhanced with increased resolution. Furthermore he concluded that cloud boundary overshoots in $S$, for the case of small cumulus in a shear free environment, exert little influence on the small scale dynamics of the cloud boundary. However, since our aim is to examine the effect of varying CCN on cloud albedo, such numerically-induced microphysical features are of concern. Work with alternate numerical operators [i.e., monotonic constraints on the advective fluxes; Grabowski and Smo-
larkiewicz (1990); Smolarkiewicz and Grabowski (1990)] is currently underway with the aim of preventing such phenomena.

(3) In all the simulations, CCN appear above the stratocumulus layer in concentrations up to 20% more than the initial ambient values. If this is a physical effect then the source of CCN could be related to droplet evaporation into the warm, dry inversion layer, which replenishes CCN to the environment. However, as discussed above, it could be a numerical artifact due to the sharp gradients at the cloud boundary.

(4) The simulation with \( N_a = 500 \text{ cm}^{-3} \) produces narrower spectra with smaller drops than does the simulation with \( N_a = 100 \text{ cm}^{-3} \) (compare Figs. 7 and 14). Nevertheless, some drizzle drops are produced when \( N_a = 500 \text{ cm}^{-3} \). If the breadth of the newly activated droplet spectrum, which is closely related to the breadth of the dry CCN spectrum (Eq. A15), is sufficiently large, then coalescence may be initiated, given long enough residence times. The relationship between the size parameters of the dry CCN spectrum \( (\sigma_a \text{ and } a_g) \) and drizzle formation is a subject that deserves further attention.

(5) The lognormal CCN spectrum Eq. (A1) produces an activity spectrum that exhibits concave downward curvature (Fig. 15). This curvature is frequently observed in measured activity spectra where \( k \) [in the \( N = C(S \cdot 100)^k \) relationship] is seen to increase with decreasing \( S \) (e.g., Hudson, 1984). Clearly, the lognormal form is preferable to the power law form in this regard. With the aid of Eqs. (A2) and (A6), the lognormal parameters that best fit observed CCN activity spectra can be calculated, and these can be used as initial conditions for the model.

(6) This study has re-focused attention on the problems one can encounter when using multimoment-based parameterizations. The model advects three moments of the CCN distribution for each size interval, and for each, the activation parameterization attempts to re-construct the lognormal parameters from the three

![Fig. 15. The power-law, best fit to the lognormal activation spectrum defined by Eq. (A6).](image-url)
moments. As reported in Clark (1973), spatial truncation error associated with the dynamics (advection and diffusion) can lead to non-physical values for the parameters of the lognormal size distribution. We have sometimes found this to be severe at the droplet activation boundaries where gradients are sharp, and the magnitude of the gradient varies with the order of the moment. In the current work, constraints were applied to ensure $a_{\text{min}} \leq a \leq a_{\text{max}}$ and $\sigma_a \geq 1$. We raise this point to re-state our concern with the results in the vicinity of boundaries of activation regions.

(7) The presentation of the results in this paper are somewhat dependent on the nature of the conditional sampling of the model data described in Section 3. Future work that compares model simulations to in situ and remote measurements will need to address the question of data averaging more carefully.

We have looked here at various spatial averages of the cloud layer but have not addressed the issue of temporal averaging of cloud layer, and the statistical differences between the simulated cases over the course of the evolution. The subject of temporal averaging is best postponed to the three-dimensional studies (see discussion next), which will more accurately simulate the cloud layer and therefore provide more meaningful comparisons.

4.2. Planned three-dimensional simulations

Our goal in constructing this model is to conduct more realistic three-dimensional LES simulations from which we can develop a parameterization scheme that is able to diagnose or predict changes in stratocumulus cloud cover, atmospheric boundary layer stability, liquid water paths, and cloud albedo, due to changes in sea surface temperatures, large-scale vertical motion and wind shear, and CCN size distributions.

These simulations will differ from our two-dimensional test in a number of respects. We will use Deardorff’s (1980) scheme as opposed to Smagorinsky’s (1963) deformation tensor when we parameterize sub grid-scale diffusion. In addition, we intend to turn on Rayleigh friction to absorb gravity waves in the vicinity of our upper boundary. We plan to increase our vertical resolution in order to better resolve cloud top entrainment processes and supersaturation fields. We have selected three cases studies for the three-dimensional simulations. One is the FIRE I case discussed here and the other two, cases observed on June 9 and June 16, 1992 during the Atlantic Stratocumulus Transition Experiment (ASTEX) experiment. The FIRE I experiment took place off the coast of Southern California; the ASTEX experiment was in the eastern Atlantic, near Porto Santo in the Madeira island chain.

At the time of this writing we can report only on a preliminary three-dimensional simulation (see Cotton et al., 1993). Many of the numerical artifacts associated with advection in the vicinity of interior outflow points are also apparent in the three dimensional run. Vertical resolution was doubled for the three dimensional run and similarly to Grabowski (1989) we saw that overshoots in $S$ were greater than previously observed. This is an important problem for LES
models with explicit microphysics since overestimates of cloud top supersaturations will enhance droplet growth, thereby impacting the cloud top radiative properties as well as the dynamical interaction between the cloud and cloud-free regions. Other features of the 3-D run were consistent with the 2-D simulations; the structure of the eddies was generally similar although with the radiation parameterization turned on the eddies were more vigorous and the cloud had a less ragged base.

5. Summary and conclusions

Two very powerful models have been combined to simulate the response of stratocumulus cloud albedo to variations in ambient CCN concentrations. The RAMS allows accurate large-eddy simulation of a horizontally homogeneous field of marine stratocumulus clouds under a variety of environmental conditions. The university of Tel-Aviv explicit microphysics model affords the opportunity to accurately simulate the evolution of droplet spectra in a cloud of horizontally and vertically varying updraft speeds and supersaturations. A new droplet activation scheme based on a bi-modal CCN spectrum has been presented. This has enabled us to investigate the effect of changing the number of CCN on cloud microphysics.

The newly configured RAMS model produces realistic simulations of a marine stratocumulus cloud layer. The two-dimensional runs show the impact of varying the environmental CCN spectrum; increases in CCN concentrations result in significantly decreased droplet sizes and increased number concentrations. Liquid water mixing ratios are not as significantly impacted. These results are consistent with what has been hypothesized (Twomey, 1974, 1977) as well as observed (e.g., Noonkester, 1984).

This study has also pointed out a number of areas of concern, namely, the advection of scalars across sharp gradients. Our results frequently exhibit numerical artifacts in these regions and future work must address this problem with alternate numerical operators. In addition, the activation parameterization which relies on a three moment description of the CCN spectrum, is compromised by these advective problems and we are currently investigating solutions to this problem using multi-bin models.

We would like to reiterate the point that our two-dimensional simulations are not meant to be definitive representations of the layer observed during FIRE I on July 7, 1987, and we expect that the results of future three-dimensional simulations could differ significantly.

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Appendix A

Description of the droplet activation scheme

The droplet activation scheme employs two CCN spectra; the first spectrum is in the range 0.01 to 0.1 μm (radius), and the second covers the range 0.1 to 10 μm. The upper and lower bounds of each region will be referred to as \( a_{\text{min}} \) and \( a_{\text{max}} \), respectively. These two regions have been chosen based on conventional classification of small and large CCN, although they can easily be adjusted if deemed necessary. In each region, the CCN spectrum is defined by three of its moments. The most relevant moments to our problem are the 0th moment (number concentration \( N_a \)), the 2nd moment (surface area \( B \)), and the 3rd moment (mass concentration \( M \)). The choice of \( N_a \) is based on the availability of measurements and on the fact that droplet concentrations will be directly dependent on \( N_a \). The ratio of \( M \) to \( B \) provides a measure of the effective radius of the particles, a parameter that is important for radiative transfer calculations. For other applications, different sets of moments can be chosen.

For the purpose of performing droplet activation calculations, a lognormal basis function has been chosen:

\[
n(a) = \frac{N_a}{(2\pi)^{1/2} \ln \sigma_a} \exp \left[ -\frac{\ln^2(a/a_g)}{2\ln^2(\sigma_a)} \right]
\]

where \( a_g \) is the geometric mean radius of the CCN particles with radius \( a \), and \( \sigma_a \) the geometric standard deviation. The lognormal form fits observed aerosol spectra well (e.g., Shettle and Fenn, 1979; Gerber, 1985), and since CCN are a subset of aerosol (particularly for internal mixing) the lognormal parameterization should also be suitable for CCN.

In the following, we outline the methodology for the activation calculations. For brevity, we deal with only one of the CCN spectra. Treatment of the second spectrum follows along identical lines. The calculations comprise three parts:

(i) Calculation of the number of activated droplets \( N_r \)

To calculate the number of droplets activated \( N_r \), we use the ambient supersaturation \( S \) calculated by the model as the critical \( S_c \) for a CCN having critical radius \( a_c \) (see, e.g., Pruppacher and Klett, 1978). This follows the Kohler theory of particle growth as a function of supersaturation. Thus,

\[
S = S_c = \left[ \frac{4A(T)^3}{27B(a_c)} \right]^{1/2}
\]

where
where $T$ is the ambient temperature; $\sigma_s$, the surface tension of the aqueous solution with the air; $\rho_s$, the density of water; $M_w$, the molecular weight of water; $M_s$, the molecular weight of the salt; $m_s$, the mass of the salt particle; $R$, the universal gas constant; $\nu$, the number of ions into which a salt molecule dissociates in water; and $\phi_s$, the osmotic coefficient for the solution.

Since $m_s = 4/3\pi\rho_s a_s^3$ ($\rho_s$ is the density of the solute), Eqs. (A2), (A3) and (A4) can be used to calculate $a$, and hence $a_c$. $N_r$ can then be written as

$$N_r = \int_{a_c}^{a_{\text{max}}} n(a) da$$  \hspace{1cm} (A5)$$

Integration of Eq. (A5) is straightforward (e.g. Feingold and Levin, 1986):

$$N_r = N_a \left[ \text{erf}(Y_{\min}) - \text{erf}(Y_{\max}) \right]$$  \hspace{1cm} (A6)$$

where $Y_{\min}$ and $Y_{\max}$ are given by Eqs. (A9) and (A10) with $n=0$ and

$$\text{erf}(z) = \frac{1}{\sqrt{(2\pi)}} \int_{-\infty}^{z} e^{-t^2/2} dt.$$  \hspace{1cm} (A7)$$

The general solution to the integral of the $n$th moment of the truncated lognormal function is given by

$$M_{\text{trunc}}^{(n)} = \int_{a_{\min}}^{a_{\max}} a^n n(a) da = M^{(n)}_{\max} \left[ \text{erf}(Y_{\max}) - \text{erf}(Y_{\min}) \right]$$  \hspace{1cm} (A8)$$

where

$$Y_{\min}^{(n)} = \left[ \frac{\ln(a_{\min}/a_g) - n\ln^2\sigma_a}{\ln\sigma_a} \right]$$  \hspace{1cm} (A9)$$

and

$$Y_{\max}^{(n)} = \left[ \frac{\ln(a_{\max}/a_g) - n\ln^2\sigma_a}{\ln\sigma_a} \right].$$  \hspace{1cm} (A10)$$

(ii) Calculation of the size parameters of the newly activated droplet spectrum

The first part of this problem is to determine the size of the dry CCN particles at 100% relative humidity (RH). Assuming equilibrium growth of CCN particles, Gerber (1985) presented a parametrization of the dependence of the lognormal parameters of a CCN spectrum as a function of RH. Here a similar approach is adopted, with the exception that non-equilibrium growth is accounted for. A detailed Lagrangian parcel model (Heymsfield and Sabin, 1989) configured with
100 bins to describe the CCN size spectrum (assumed to be composed of ammonium sulfate) is used to determine the degree to which particles achieve their equilibrium sizes. Thus, following Ivanova et al. (1977) and Kogan (1991) we determine a dry CCN radius \( a^* \) below which particles will achieve their equilibrium radii at 100% RH:

\[
a^* = 0.186 \cdot 10^{-4} w^{-0.328} \tag{A11}
\]

(with \( w \) in cm s\(^{-1} \) and \( a^* \) in cm). Note that the updraft \( w \) is a surrogate for the time available for particle growth in the subsaturated environment. Particles in strong updrafts will have less time to reach their equilibrium sizes than those in weak updrafts. At 100% RH, the size of particles greater than \( a^* \) will be determined by a parameter \( \kappa \) such that

\[
a_{100} = \kappa a \tag{A12}
\]

(see also Kogan, 1991). The parameter \( a_{100} \) is the size of a dry CCN particle at 100% RH. The parameter \( \kappa \) is given by

\[
\kappa = 2.438 w^{-0.115} a^{-0.131} \tag{A13}
\]

(with \( w \) in cm s\(^{-1} \) and \( a \) in cm). Eqs. (A11)–(A13) are valid for ammonium sulfate CCN and updrafts in the range 5–250 cm s\(^{-1} \).

We have used a broad range of input CCN spectra defined by Eq. (A1) and calculated their corresponding CCN lognormal parameters at 100% RH with the aid of (A11)–(A13). The following empirical relations were found to fit the data well:

\[
r_g = b_0 a_g^{b_0} \sigma_a^{b_2} w^{b_3} \tag{A14}
\]

\[
\sigma_r = c_0 a_g^{c_1} \sigma_a^{c_2} w^{c_3} \tag{A15}
\]

where \( r_g \) is the geometric mean radius, and \( \sigma_r \) the geometric standard deviation of the CCN at 100% RH. The coefficients in Eqs. (A14) and (A15) have been calculated using a multivariate regression to the data points and can be found in Table 1 (for the range 0.01–0.1 \( \mu m \) ) and Table 2 (for the range 0.1 \( \mu m \)–10 \( \mu m \) ). C.G.S. units are used throughout. The low standard errors of estimate (SEE) provide confidence in the method. Note that (A14) and (A15) describe the size distribution of “wet” CCN at 100% RH.

According to (A5), only some fraction of these “wet” CCN will be activated. This requires adjustment of the Eqs. (A14) and (A15) for a truncated lognormal distribution. The lower and upper bounds of truncation will be determined by \( a_c \).
Table 2  
The regression coefficients for droplet activation in the range 0.1 \( \mu m \)-10 \( \mu m \) (see Eqs. A14, A15). The coefficients are valid for 0.15 \( \mu m \) < \( a_g \) < 5 \( \mu m \); 1.1 < \( \sigma_g \) < 2.2; 5 cm s^{-1} < \( w \) < 250 cm s^{-1}.  

<table>
<thead>
<tr>
<th>( b_0 )</th>
<th>( b_1 )</th>
<th>( b_2 )</th>
<th>( b_3 )</th>
<th>SEE</th>
<th>( c_0 )</th>
<th>( c_1 )</th>
<th>( c_2 )</th>
<th>( c_3 )</th>
<th>SEE</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.182</td>
<td>0.854</td>
<td>-8.12-10^{-2}</td>
<td>-0.109</td>
<td>0.8</td>
<td>1.127</td>
<td>-4.97-10^{-2}</td>
<td>0.596</td>
<td>5.1-10^{-5}</td>
<td>7.2-10^{-2}</td>
</tr>
</tbody>
</table>

and \( a_{\text{max}} \). In each case, it must be determined with the aid of Eq. (A11) whether \( a_c \) and \( a_{\text{max}} \) are at equilibrium or non-equilibrium size. For equilibrium growth, the particle size will be given by Kohler theory; for non-equilibrium growth, the size will be determined by Eqs. (A12) and (A13).  

The lognormal parameters for the truncated CCN spectrum at 100% RH are calculated according to standard expressions. They can be found in Feingold and Levin (1986), appendix A. In the interest of brevity they will not be repeated here, but will be written only in functional form:

\[
s(r, t) = \int_0^r n(r, t_0) \frac{dr_0}{dr}, \quad (A18)
\]

where the superscript (t) denotes truncation. Parameters \( N_r \) (Eq. A6), \( r_g^{(t)} \) and \( \sigma_r^{(t)} \) together describe the parameters of the newly formed droplet spectrum.  

The next stage of this problem is to calculate the condensational growth of these newly activated droplets in the supersaturated cloud environment. This is done with the aid of the analytical solution

\[
dr dt = G[S(t) - \frac{A(T)}{r} + B(a)/r^3] \quad (A19)
\]

\( G \) determines the rate of condensational growth of a droplet, and \( A(T) \) and \( B(a) \) respectively represent the surface tension and solute correction terms to the saturation field around the droplet. \( B(a) \) is assumed to be equal to \( B(a_g) \). Because the terms in parentheses on the right hand side of Eq. (A19) are dependent on \( r \), they are calculated in the average sense; i.e., we assume that

\[
\frac{dr}{dt} = G[S(t) - \bar{y}] \quad (A20)
\]

where

\[
\bar{y} = \frac{\int_{a_g}^\infty [A(T)/r - B(a)/r^3] n(r) dr}{\int_{a_g}^\infty n(r) dr}. \quad (A21)
\]
(See also Feingold and Heymsfield, 1992.) Since \( n(r) \) is described by a log-normal distribution, Eq. (A21) is easily solved. Note that although \( G \) in Eq. (A19) is assumed to be independent of \( r \), explicit dependence on \( r \) was used in all of the calculations of growth up to 100% RH. Dependence of \( G \) on \( r \) can also be introduced in Eqs. (A19) and (A20) using a weighted average of \( G(r) \) (\( \bar{G} \)) in a similar fashion to Eq. (A21).

(iii) Updating the \( \text{ccn} \) spectrum after activation

The final step in this procedure is to update the CCN spectrum following droplet activation. \( N_a \) is reduced by an amount \( N_r \) (determined from Eq. A5) and the moments \( B \) and \( M \) are re-calculated from integrations of the truncated distribution \( n(a) \):

\[
\begin{align*}
B &= \pi \int_{a_{\text{min}}}^{a_{c}} a^2 n(a) \, da \\
M &= \frac{4}{3} \pi \rho_s \int_{a_{\text{min}}}^{a_{c}} a^3 n(a) \, da
\end{align*}
\tag{A22}
\tag{A23}
\]  

using Eq. (A8).

By explicitly resolving the size spectrum of CCN, the proposed scheme represents an improvement over the \( N=C(S \cdot 100)^k \) (Twomey, 1959) parameterization. The latter scheme cannot provide information on the size parameter of the newly activated droplet spectrum, unless allowance is made for \( k \) to change as the CCN become ingested into the cloud droplets. This would require equations for two moments of the CCN spectrum and would bring the Twomey scheme closer to the proposed scheme.

As in the proposed scheme, Kogan (1991) explicitly solved for the size spectrum of CCN using a 19-bin array. This approach is potentially very accurate. Its main drawback is its computational cost, since it requires a prognostic equation for each of the bins. By using a lognormal basis function, the present scheme reduces this overhead to only six equations (three spectral moments describing two size spectra) and treats the essential features of the activation process. Numerical tests show that at low supersaturations there is a preferential activation of larger CCN that depletes the moments to a degree that is directly proportional to the order of the moment. As the supersaturation increases, both the geometric mean radius \( a_g \) and the breadth parameter \( \sigma_a \) (defined by a ratio of \( N_a, M \) and \( B \)) become progressively smaller. The decrease in \( a_g \) as well as the narrowing of this spectrum effectively allow the upper truncation of the CCN spectrum to be accounted for.

References


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