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1 Introduction

Over the past decade or so the evolution and equilibria of persistent decks of stratocumulus climatologically clinging to the edge of summertime subtropical highs has been an issue of increased scientific inquiry. The particular interest in the microphysical structure of these clouds stems from a variety of hypotheses which suggest that anthropogenic influences or biogenic feedbacks may alter the structure of these clouds in a manner which may be climatically significant.

Ship tracks provided early observational evidence of anthropogenic induced changes in the reflectivity or even structure of the marine boundary layer. Since these observations, most hypotheses regarding boundary layer influences on climate have been formulated by an examination of the solution space of simple models. The earliest hypothesis of this sort (and the one on the most solid footing) is due to Twomey (1974), who posited that enhanced concentrations of CCN could lead to enhanced droplet reflectivity and enhanced albedos in clouds of modest optical depths. In low lying clouds where the albedo effect dominates, the climate sensitivity to a robust perturbation in cloud albedo may be significant. Using a very simplistic dynamical model, Ackerman et al. (1993) hypothesized that in the absence of significant droplet concentrations, reduced cooling rates would be unable to generate sufficient TKE (turbulent kinetic energy) to combat the effects of large scale subsidence, eventually leading to the dramatic reduction of boundary layer depth. In such a state the boundary layer would be hypersensitive to local enhancements in ambient aerosol concentrations.

Albrecht (1989) considered the indirect influence of aerosol on cloud structure through a somewhat different pathway. He hypothesized that increases in CCN concentrations could increase the colloidal stability of clouds. Increased colloidal stability would result in reduced drizzle fluxes. In the context of his simple formulation he found that reductions in the production of drizzle would allow for more persistent cloud decks and larger fractional coverage. Recently Pincus and Baker (1994) enjoined the Albrecht hypothesis by suggesting that the effect of aerosol concentrations on cloud depth may constitute a significant feed-

back overlooked by Albrecht (1989). However, their hypothesis was based on the fact that the addition of a precipitation parameterization to a mixed layer model led to increased entrainment. The fact that entrainment is not well enough understood to be well represented in mixed layer models of the stratus topped boundary layer requires that hypotheses formulated in this manner be treated with extra suspicion.

Baker and Charlson (1990) suggested that the boundary layer may be bistable with respect to CCN production rates. Their hypothesis is that there are two stable equilibria of the ambient CCN concentrations. The first being associated with low production rates of CCN, thereby representing a balance between CCN production and precipitation scavenging. The second being associated with higher production rates, thereby representing a balance between CCN production and dry scavenging followed by sedimentation. The potential for the boundary layer to be bistable in this respect would have dramatic consequences for both types of hypotheses discussed above, as within a certain range of production rates a small change in the production of CCN could lead to a dramatic difference in boundary layer structure. The conceptual consistency of this hypothesis, despite its formulation on the basis of simplified relationships begs for further investigation.

As noted, most of these hypotheses are quite conjectural, based as they are on simple formulations of boundary layer structures and interactions between drops and aerosols. Moreover, classical formulations of susceptibility are based on looking at the change in a certain feature (say boundary layer albedo) under the perturbation of a single parameter. Unfortunately, the applicability of this type of formulation, which neglects compensating effects through an evolution of boundary layer dynamical structures has never been demonstrated. However, given the potential importance (from the perspective of the global energy balance) of the nature of the equilibrium of the cloud topped boundary layer, there is considerable interest in flushing out the validity of the hypotheses discussed above; whether it be through careful observation or detailed simulations of the range of interactions involved in a particular hypothesis.

Although proposed primarily in the context of the Twomey and Albrecht hypotheses, the objective of this research evolved more generally to address the range of cloud-aerosol interaction hypotheses. Since the hypotheses discussed above are fundamentally coupled to the dynamics of the droplet spectra their detailed consideration requires that both the droplet distribution function and its interaction with the aerosol distribution function be explicitly represented. The associated level of complexity of this approach has required significant model development, in addition to the generation of new procedures and simplified models which can be used as diagnostic tools to constrain, interpret and understand the results produced by the detailed model. In what follows we summarize our work (much of which is currently coming to fruition) in a multi-fold manner. First we consider our evaluation of the physical processes discussed above. Next we consider our progress in developing alternative modelling frameworks which constrain our results. This is followed by a consideration of the development of new and improved numerical procedures. Lastly we give an overview of our papers, presentations and collaborative work.

2 Physical Processes

2.1 The Twomey Effect

One of the primary objectives of this research has been to explore the hypothesis of Twomey. The basic approach was to couple detailed radiative calculations with detailed representations of the droplet spectra. The detailed representation of the droplet spectra was generated by the Large Eddy Simulation-Explicit Microphysics (LES-EM) model coupled to a simple mixed emissivity radiation scheme in order to drive the dynamics. Several simulations were carried out and the resultant microphysical fields were taken from the stationary regime of the turbulent simulation and used to drive a two dimensional radiative model. By comparing the radiative properties of the simulated clouds formed in environments with different CCN concentrations we were able to more accurately quantify the albedo susceptibility of stratocumulus taken to be typical of the FIRE experimental area.

2.1.1 The simulations

We performed three simulations. All of them were initialized based on the July 7, 1987 FIRE stratocumulus sounding described in Betts and Boers (1990). The simulations were designed to explore the effect of perturbed CCN concentrations on the boundary layer. First we conducted a control simulation with approximately 120 CCN cm^{-3} . The simulated boundary layer was slightly unstable with respect to the cloud top entrainment instability and as a result the cloud fraction in the stationary regime was on the order of 95 %. From the spun up control run we spun off a second simulation which was characterized by a factor of 6 enhancement in subcloud CCN concentrations. The third simulation was also spun off the control run, but in contrast to the second run, the above cloud CCN concentrations were increased by a factor of 6. As would be expected the second sensitivity run responded much more rapidly to changes in CCN concentrations.

2.1.2 The radiative calculations

In his dissertation, Dave Duda (1994) made quantitative assessments of the effects of both macrophysics and microphysics on the radiative transfer applied in three areas: remote sensing of cloud optical properties by satellites, the distribution of broadband radiative heating in stratus, and the relationship between changes in broadband cloud albedo and changes in CCN concentrations in the cloud system. This assessment was accomplished by using a newly developed multi-dimensional radiative transfer model with cloud field data produced by the RAMS/LES model.

The Spherical Harmonic Spatial Grid (SHSG) method developed by Evans (1993) was used to simulate the radiative transfer through a two dimensional cloudy atmosphere. Despite the efficiency of SHSG in computing radiances in multi-dimensional media, the computational resources presently needed for 3D radiative transfer modeling limit the size of practical model simulations, and only calculations in two dimensional clouds were completed using SHSG.

The SHSG model was extensively modified in order to compute broadband solar fluxes, and the k distribution method was used to account for gas absorption. By averaging the k distribution data over broader wavenumber and absorption coefficient ranges, the number of required computations were reduced from over 12,000 to 200, while maintaining the accuracy necessary for broadband albedo calculations (0.5%, this is roughly an order of magnitude smaller than the albedo changes produced by macrophysical or microphysical effects). Additional reduction of the number of calculations in the broadband model was limited by the problems associated with accurately accounting for the rapid variation in cloud droplet optical properties with respect to wavenumber in the bands of the broadband model. For low altitude stratus simulations, several additional computations could be ignored since the strongest k distribution weights were associated with absorption above cloud top.

Two-dimensional cross sections of the microphysical data from all three RAMS model simulations were used as input cloud property data for the broadband SHSG model. The cross sections showed that the addition of CCN above the cloud top in the first sensitivity run resulted in a 50 to 100 percent increase in the cloud droplet concentrations and a reduction of the cloud top effective radius from over 11 microns to less than 10 microns. The enhanced CCN concentrations added below cloud base increased the cloud droplet concentrations up to 110 percent compared to the control run and decreased the effective radius of the cloud top droplets to 8.5 microns. The creation of larger numbers of smaller droplets at the expense of larger droplets in the two sensitivity runs produced several changes in the cloud optical properties, and as shown below, changes in the cloud albedo.

The results of the radiative transfer calculations indicated that in unbroken marine stratus clouds the *net* horizontal transport of photons over a domain of a few kilometers was nearly zero, and the domain average broadband albedo computed in a two dimensional cross section was nearly identical to the domain average calculated from a series of independent pixel approximation (IPA) calculations of the same cross section. This matches the findings from Cahalan (1994) for monochromatic calculations in a simple cloud model, and suggests

that accurate computation of domain-averaged albedos in unbroken marine stratus can be made using IPA calculations with one dimensional radiative transfer models. The horizontal inhomogeneity does affect the cloud albedo due to the nonlinear relationship between albedo and optical depth (Cahalan, 1994) and reduces the domain average total solar cloud albedo by a relative difference of five to six percent for unbroken clouds and fifteen percent for a more inhomogeneous cross section with broken cloudiness, when compared to a perfectly homogeneous cloud with the same mean optical properties.

Given the good agreement between the domain-averaged albedos computed by the independent pixel approximation (IPA) and the multi-dimensional RTMs in this study and in Cahalan (1994), computations of the mean albedo over portions of the entire three dimensional RAMS domain were made for all three RAMS simulations using IPA calculations from a two-stream model.

Comparisons of the total solar albedos computed by this method between clouds with similar mean microphysics and different macrophysics show the relative difference in cloud albedo resulting from typical macrophysical differences in marine stratus were between three to five percent. The relative differences in cloud albedo due to microphysical changes resulting from the sixfold increase of CCN concentrations above cloud top ranged from six to nine percent. When the same increase in CCN concentration was introduced below the cloud layer, the increase in cloud albedo ranged from ten to fifteen percent, although some of the increase was due to cloud dynamical changes not associated with the effects of the additional CCN. The impact of microphysics on the cloud was greatest for small solar zenith angles.

Like the broadband albedos, local differences between the 2D and IPA computed heating rates were significant but the domain averages were very similar. The effects of PPA bias on the net flux convergence in the cloud were as large as 5 percent in the $\theta_0 = 10^\circ$ simulations, but very small at $\theta_0 = 60^\circ$. The effects of the microphysical changes on the mean net flux convergence were less than 2 percent.

Using a simplified two channel retrieval method that compared the cloud top reflectances computed from the two dimensional RAMS/LES cross sections with tabulated results from a set of plane parallel calculations, the cloud optical depth and effective radius were estimated and compared to the actual cloud properties. The results showed that the effects of cloud inhomogeneity produced local fluctuations in the reflected radiances that could be significantly different than those computed from plane parallel calculations.

The mean computed relative errors produced by these fluctuations ranged from 2 to 12 percent in the domain average R_e retrievals and 0.5 to 9 percent in the domain average τ retrievals. Horizontal inhomogeneity within a satellite pixel will also affect the retrievals due to the nonlinear relationships between the reflectance functions and the retrieved optical properties. This additional bias produced an extra 3 to 4 percent error in the effective radius retrievals and a 7 to 16 percent error in the optical depth retrievals. These error values were averaged over a range of sun/viewing geometries. In general, the errors were smallest for high sun and near nadir viewing angles, although the area-averaging effects of a satellite radiance measurement tend to make the retrievals of effective radius and optical depth fairly insensitive to the range of sun/viewing geometries used in this dissertation. Results of the radiative calculations have been presented at the 1994 AMS Conference in Nashville and at the American Geophysical Union Spring 1994 Meeting in Baltimore.

2.1.3 Repeat simulations

Because of a numerical error in the formulation of the subsidence term (which we realized only through the process of our analysis) a false divergence (in all fields) was generated which significantly cooled the entire boundary layer through the course of the simulation. This led to the development of a decoupled boundary layer associated with underlying cumulus convection. The variance in the LWP was significantly more than what was simulated in the absence of this error. Moreover the shear present in this case also complicated the analysis considerably. For these reasons the simulations are being performed anew with a corrected version of the model and a different initial sounding. However having already been through

the analysis once, the analysis of the new simulations should proceed considerably more rapidly.

2.2 Drizzle and ASTEX

Another major part of the research effort has focused on simulation of case studies from the Atlantic Stratocumulus Transition Experiment (ASTEX, 1992)¹. ASTEX provided a rich data set of in-situ and remote measurements pertaining to the aerosol-cloud-climate problem. In contrast to previous stratocumulus field campaigns (e.g., FIRE I), the ASTEX clouds tended to produce relatively large amounts of drizzle. Although drizzle rates associated with stratocumulus are small compared to precipitation from cumulus clouds (of the order of mm per day compared to mm per hour), this drizzle has a direct impact on cloud microphysical properties; precipitation sized drops frequently re-circulate within the cloud, growing as they collect smaller droplets, until their terminal velocity is too large for them to be sustained in the cloud. The drizzle process acts to deplete cloud water and droplet concentrations, with a concomitant impact on cloud optical depth and droplet effective radius. In addition, as the drizzle falls below cloud base, much of it evaporates. This redistributes heat and moisture and feeds back to boundary layer dynamics.

2.2.1 Evaluation of model against observations

Two case studies were run. The first was the June 16 1992 drizzle event observed by NOAA K_a band radar on the island of Porto Santo, while the second was a drizzle event observed on board the research vessel Malcolm Baldrige (on the same day). Model simulations of these events produced good estimates of cloud base and height, as well as the onset of drizzle.

¹FIRE Phase II: ASTEX Implementation plan, 1990. Available from the FIRE Project Office, MS 483, NASA Langley Research Center, Hampton, VA 23665-5225.

2.2.2 Numerical experiments of drizzle formation

Using the sounding from the Malcolm Baldrige, we performed a number of simulations to investigate the sensitivity of drizzle formation to cloud vertical velocities and CCN concentrations. We have shown (Feingold et al., 1995 - manuscript in preparation) that for fixed CCN concentrations, more vigorous clouds produce more drizzle because they allow drops to grow through repeated collision-coalescence cycles. In weaker clouds, droplets tend to fall out of the cloud before they have achieved significant size resulting in lower amounts of drizzle. Experiments with different CCN concentrations showed that the above factor is of comparable importance in drizzle formation to CCN concentration. In another series of experiments, we investigated the effects of the feedback of drizzle on the boundary layer dynamics. Results show that when significant amounts of drizzle reach the surface, the sub-cloud layer is stabilized and the boundary layer circulations do not penetrate through to cloud top. When only small amounts of drizzle are produced, cooling tends to be confined to the region below cloud base, resulting in destabilization and eddies which penetrate the depth of the boundary layer.

3 Alternate modelling frameworks

Using the LES-EM model we have added a new component that gives the thermodynamic state of N parcels at each timestep, where some initial distribution of parcels must be initially specified. Currently we randomly initialize the N parcels at positions below cloud base, and take $N = 500$. The ensuing trajectories are then analyzed over a period of several eddy turnover times. The information from this analysis is being used to characterize the nature of the trajectories for different boundary layer regimes in addition to the quantifying the manner in which different trajectories contribute to the simulated cloud microphysical structure.

3.1 Dynamical framework

Moreover this form of analysis is amenable to a consideration of the manner in which different dynamical frameworks generate differing cloud structures. In particular the GCSS workshop summarized below, showed that while 2-D cumulus ensemble simulations can accurately predict many features of the cloud topped boundary layer, their partitioning of the turbulent transport between the pressure terms and triple correlation terms differs dramatically from that of the 3D LES models. Moreover the 2-D simulations produce significantly greater vertical velocities variances associated with motion which is strongly organized in convective rolls. Our trajectory analysis has shown that as a consequence of this different dynamical regime associated with 2-D simulations the average cloud top residence time of parcels is about half of what it is in the 3-D simulations. Moreover the distribution of residence times is much more sharply peaked in the 2-D simulations, the cloud base velocities are much greater, and the extent of mixing (i.e. variance in conserved variables) is much less.

Because 2D models are so much more computationally efficient (depending on the grid size, one or two orders of magnitude), and because on the face of it they produce physically realistic boundary layer structures, they are appealing dynamical hosts for the consideration of a variety of microphysical processes. Consequently our results will help us better assess the applicability of 2-D models to a variety of questions. This work was presented by William Cotton as the 1995 American Meteorological Society Annual Meeting held in Dallas and is currently being written up for submission to the *J. of the Atmospheric Sciences*.

3.2 The Lagrangian parcel model

Given the ensemble of trajectories discussed above, and assuming (a verifiable assumption) that they characterize or span the dynamical behavior of the simulated clouds we may use these trajectories to drive an entraining parcel model we have developed. We then may represent the cloud in terms of the ensemble of trajectories where the microphysical evolution of the cloud is represented by the microphysical evolution along the trajectories

as represented by one of a number of microphysical models. This then defines a Lagrangian dynamical framework for our analysis. By using the identical (Eulerian) microphysical model in the parcel model as was used in the LES model we may isolate the effects of averaging (associated with the Eulerian dynamical framework) on the resultant cloud fields. We have found that vertical grid spacings of 25 m causes a slight under-prediction of the cloud base supersaturation peak, and results in the activation of too few droplets. In addition we are using this framework to understand a number of phenomena, such as cloud top supersaturation peaks, and the effect of grid scale mixing on the breadth of the droplet distribution.

Alternatively we have written a detailed microphysical model which treats the evolution of the droplet spectra in a Lagrangian manner by considering the evolution of the liquid water on an arbitrary number of aerosol classes. This approach allows for an accurate consideration of non-equilibrium effects, solute and curvature effects, gas-kinetic effects and multi-modal aerosol distributions. In this approach the physical continuity between hygroscopic aerosol and cloud drops is preserved, thereby eliminating the need for an activation/regeneration parameterization. Moreover this system can be formulated as a system of ordinary differential equations and can be solved with arbitrary accuracy. Consequently by comparing the parcel model runs with this microphysical framework to the runs with the microphysical framework used in the LES-EM model we are able to evaluate the applicability of the microphysical parameterizations and modelling assumptions used in the LES-EM model.

In addition it allows us to consider a number of physical questions of some relevance:

- (1) What is the characteristic velocity from the ensemble of trajectory which corresponds to the observed number of activated drops.
- (2) How important are non-equilibrium effects on the cloud base supersaturation structure.
- (3) How important is mixing in generating the observed cloud microphysical structure.
- (4) What is the role of subcloud variability in parcel thermodynamic properties in promoting spectral broadening.

This work was presented at

the 1995 American Meteorological Society Annual Meeting in Dallas and is currently being written up for submission to the *J. of the Atmospheric Sciences*.

4 Model Development

4.1 Advection

Advective errors tended to degrade our solutions in the vicinity of cloud top, causing high supersaturations, oscillations in the liquid water mass and drop concentration fields. This situation is commonly encountered in numerical cloud models (Grabowski, 1989). To rectify this situation we experimented with monotonic flux corrections to the higher order advection schemes (Smolarkiewicz and Grabowski, 1990). We spent a great deal of time quantifying the long time non-linear behavior of the flux correctors in 1 and 2 dimensional classical advective flows. The results were encouraging and the methodology was introduced into the model. In two dimensions we found the results were sensitive to how much diffusion was added by the flux correctors to combat the non-monotonic tendencies of the higher order schemes. The best results were associated with the peak preservers of Zalesak (1979) unfortunately this was the most computationally demanding approach as well. We proceeded with this form of flux correction, however, since the more diffusive scheme artificially degraded our inversion thereby promoting artificially high entrainment velocities, and artificially low liquid water contents. The model was run in 3-D with the new advection schemes, and most of the problems originally encountered were rectified. Cloud top supersaturations remained high, but not unreasonably so, as was the case prior to the flux corrections. This work was written up as an internal report and presented at the 1994 American Meteorological Society Annual Meeting in Nashville.

4.2 Supersaturation Calculations

An equation for the supersaturation was added. This equation models the evolution of the supersaturation over the course of a timestep and accounts for the microphysical sinks and dynamical forcings. Use of such an equation gives a much more representative

value of the activation supersaturation, and allows the for the integration of the activation condensation equations on significantly longer timesteps.

4.3 Droplet Activation

It was necessary to develop a new droplet activation scheme for use in this study. The requirements for this scheme were that it should be able to respond to cloud supersaturation and predict both the number and size of newly activated particles. Over the past three years, a number of different schemes have been tested:

4.3.1 The bimodal log normal scheme

Our first scheme was based on a bimodal log normal distribution function. The main features of this scheme are as follows:

- It explicitly resolves the size of CCN. It divides the size spectrum into two parts - small nuclei and large nuclei - with each part described by a log normal distribution function.
- Each log normal distribution is described by three of its moments. The 0th, 2nd and 3rd moments were chosen since they are most important from the point of view of radiative transfer. Thus six scalar variables are used to describe the CCN spectrum.
- The number of newly activated droplets is calculated from the model-derived supersaturation while their size is calculated from non-equilibrium growth of CCN in humid environments.

Tests of this activation scheme were conducted in the RAMS model together with the other explicit microphysical modules. A number of simulations were performed to assess the model response to a five fold increase in the CCN concentration. Results showed a three fold increase in droplet concentrations and a 40% decrease in droplet effective radius (Feingold et al., 1994a).

Our work with the activation scheme prompted a close look at a number of numerical artifacts which were evident in our simulations. The most serious was the fact that the representation of the CCN spectrum by a number of its moments had a drawback; the reconstruction of a physical size spectrum following the independent advection of each of the moments was not always possible.

4.3.2 A bin-model for activation

Based on this experience we opted for a multi-bin approach, with the CCN size spectrum divided into 6 size categories. The activation of these particles was formulated based on the activation spectra measured by an instantaneous CCN spectrometer (Hudson, 1989) during the FIRE I experiment off the coast of California (1987). This scheme meets our initial requirements, namely, that it respond to changes in the magnitude of the supersaturation and predict number and size of droplets. It has the added advantage of not making assumptions about the chemical composition of the CCN. Moreover, it alleviates the problems of reconstruction of a CCN spectrum from its moments, which we encountered with the log normal model. This activation scheme is currently available for use in LES simulations, and has been used in our studies of drizzle and the Twomey effect.

In the context of the bin model we found that the simulations were sensitive to how we distributed the regenerated CCN after a droplet evaporated. Originally we were returning CCN in accordance with the initial distribution, this led to a conversion of large CCN to small CCN. By altering the regeneration scheme so that the overall distribution of CCN was forced toward the initial distribution (a more physical scenario) we found our results to be much steadier.

4.3.3 Simple activation scheme

After considerable experimentation we felt that the bin model for CCN, despite some advantages also suffered from several disadvantages which encouraged the use of an even simpler representation of the aerosol. Because the aerosol distribution was not modelled in

detail and because it was sensitive to ad hoc assumptions in regard to regeneration in the presence of collection we felt it unwarranted to use precious memory and computer time by significantly increasing the number of aerosol categories. However using a bin model in the manner described above gives a resolution in supersaturation space proportional to the width of the bin, so that a six bin model could lead to overestimates in droplet activation on the order of 15 %.

For this reason we have been experimenting with a cumulative type activation scheme which compares the number of potentially (assuming no previous activation) activated CCN (from an assumed stationary probability distribution function of known and specified form) with the number of previously activated CCN, as represented by the number of cloud drops and some measure of previously collected and precipitated cloud drops, in order to yield the number of newly activated cloud drops at each time step. In the absence of collection or precipitation this scheme behaves perfectly and requires the addition of no new prognostic variables. In the presence of collection and precipitation one or more prognostic variables may be added to reflect the history of a parcel of air thereby accounting (in a simplified way) for precipitation and collection scavenging of aerosol.

4.4 Inclusion of solute in the bin-microphysical model

Cloud microphysical processes are well known to be dependent on the CCN forcing. By the same token, clouds also modify the CCN distribution by acting as a "laboratory" in which aqueous-phase chemistry can proceed. Various chemical reactions modify the amount of solute within droplets, so that when the cloud evaporates, the CCN distribution is different from what it was prior to cloud formation. In the absence of cloud chemistry, the process of droplet collision-coalescence also affects the CCN distribution. This process conserves solute mass but reduces the total number of drops. Because an evaporating droplet produces only one particle, coalescence will have the effect of spreading the same mass over a smaller number of particles and increasing the average CCN size. (This process is often referred to as "coalescence scavenging".)

In order to provide a framework for future studies of aqueous chemistry, and to evaluate the process of coalescence scavenging, we have extended our explicit microphysics model to keep track of solute mass in each drop bin. This requires solving an additional 25 prognostic equations (mass of solute in each of the 25 drop bins) as well as detailed book-keeping of the transfer of solute from one bin to the next due to droplet condensation/evaporation and coalescence. This task was accomplished in an efficient manner, with a relatively small increase in computation time.

4.5 Modifications to Droplet Condensation/Evaporation Calculations

The standard technique used to treat droplet condensation and evaporation has been the method of moments (Tzivion et al., 1989). The method is semi-Lagrangian and thus numerically stable, regardless of the time step. (Accuracy is affected by the time-step.) One limitation of the scheme is that it requires a power-law growth (evaporation) rate expression. When including curvature, solute and gas-kinetic effects to the rate equations, a power-law is no longer valid. Another limitation of the routine is that it is somewhat diffusive when evaporating a population of droplets. This leads to the enhanced evaporation of a portion of the droplet spectrum in the downdrafts, in addition to the maintenance of artificially high concentrations of larger drops.

We have therefore formulated an alternative condensation/evaporation scheme which is of the Eulerian type, but still fulfills our moment conservation requirements (Egan and Mahoney, 1972). This scheme requires smaller time steps, but is nevertheless efficient. It will be used in our model calculations when more accurate treatment of these processes is deemed necessary.

4.6 Aqueous Chemistry Model

As part of her M.S. thesis research, Wendy Richardson has developed an aqueous chemistry model for quantifying the chemical cloud processing. This aqueous chemistry model is appropriate for use as a stand-alone model or for incorporation into either a Eulerian, grid type model, or a lagrangian, parcel following model. This module is an extension of

a previous model (Kreidenweis, 1992). The original model had been prepared as a tool for the investigation of chemical reactions occurring in cloud drops (specifically oxidation of sulfur dioxide to sulfate) and could be used for doing bulk chemistry on one drop size only. This has been expanded to include the following:

1. Multiple bins and capacity to perform simultaneous chemistry on multiple drop sizes eliminating the need for "bulk chemistry".
2. Inclusion of internally and externally mixed aerosol as droplet base. Aerosols coded are ammonium sulfate $(NH_4)_2SO_4$, letovicite $(NH_4)_3H(SO_4)_2$, ammonium bisulfate NH_4HSO_4 , sulfuric acid H_2SO_4 , pure $NaCl$, sea salt ($NaCl$ +, includes ammonium sulfate and alkalinity), and silicon (for inclusion of an inert component).
3. Boron chemistry for inclusion of sea salt alkalinity in pH calculations (Pszenny et al., 1982).
4. CO_2 and carbonate chemistry.
5. HCl chemistry.
6. Inclusion of explicit growth equations for simultaneous growth of all droplets.
7. User friendly input and output: ppb for ambient gases, diameters for aerosol.
8. Distribution of solute masses into resultant aerosol components for aerosol regeneration.

This chemistry module has been used to investigate both the changing drop let acidity and the solute mass enhancement due to aqueous oxidation of SO_2 to SO_4 , and oxidant depletion. The effects of initial ambient concentrations of precursors, and varying conditions of drop size, liquid water content, and temperature are considered.

Model description and results were submitted to Dr. Joyce Penner (Lawrence Livermore National Laboratory, Livermore, CA) for consideration for inclusion in a global sulfur model.

The objectives of this chemical model were to compute percent depletion of SO_2 under various temperature, liquid water content, and oxidant environments, determine if and when a reagent can be considered limiting, and develop parameterizations from this information. The model used considers uptake of atmospheric oxidants (O_3 and H_2O_2) and SO_2 by cloud droplets, followed by the aqueous oxidation of SO_2 to SO_4 . Catalysts are ignored. SO_2 concentration values span the range from 1 ppt to 10 ppb. The system is considered closed, in that all vapors are depleted from their initial values. Droplet pH's are specified and held constant throughout the cloud lifetime. Values of pH=3 and pH=5 were chosen to demonstrate extremes. Cloud liquid water content, temperature, drop radius, and total number concentration are also specified and held constant. The Tables 1, 2 and 3 below show the values chosen for the model runs.

Table 1: Parameters

Cloud lifetime	Drop radius	LWC	Air density	Temperature
6 Hours	10E-4 cm	1E-6, 1E-5	2.53E+19 molecules/cm ³	289, 274 K

Table 2: Species Concentration Initial Values

ppb SO_2	10	1	.1	.01	.001	
ppb O_3	4	10	16	22	28	34
ppb H_2O_2	10	1	.1	.01	.001	

Representative results are included for high (10 ppb) and low (0.001 ppb) SO_2 initial values (at pH 3 and 5) showing the variation in the percent oxidation of SO_2 with varying initial oxidant concentrations. (See charts 1-4).

A comparison study between bulk and explicit representation of cloud drop lets was also performed. Oxidation of SO_2 to SO_4 for uniform size droplets with varying initial aerosol sizes (initial sulfate concentrations) was compared to bulk runs using the same total liquid

water, droplet number, and total initial aerosol (solute) mass. In the explicit run there was found to be a significant increase in the amount of SO_2 oxidized over that for the bulk run. The initial dependence on an initial aerosol size array decreased with both increased time and with increased droplet size.

4.7 Radiation

Previously the radiation parameterization used the top of the model as the top of the atmosphere for the purposes of radiation calculations. This resulted in too little downward longwave radiation at cloud top and too much impinging solar radiation. The radiation code was modified to include the rest of the troposphere for the purpose of radiation calculations. The above-model-top part of the troposphere does not interact dynamically with the rest of the model. Thermodynamic, and moisture profiles were interpolated from the NGM analysis files using RAMS in a meso-scale configuration.

In addition the radiation code was rewritten and its efficiency was increased by an order of magnitude with no loss in accuracy. Since the mixed emissivity type of calculations used by our model typically go as the number of levels squared a considerable increase in efficiency was achieved by defining a separate vertical grid for radiation calculations, with grid point concentrated in regions characterized by the strongest changes in thermodynamic properties.

4.8 Subgrid representation

The subgrid parameterizations received considerable attention as the solvers for the diffusion terms were rewritten and the possibility for constructing eddy diffusivities at vertical velocity points on our staggered grid was added. In addition, the surface flux routines had errors which were corrected, and the possibility of using a Deardorff type TKE closure for our subgrid terms has been added.

5 Presentations, Papers and Collaborative Work

5.1 Presentations and Papers

Work funded under this proposal was presented in two presentations at the 1994 American Meteorological Society Annual Meeting (Duda et al., 1994; Stevens et al., 1994) and in three presentations of the 1995 American Meteorological Society Annual Meeting (Stevens et al., 1995; Cotton et al., 1995; Feingold et al., 1995). Bjorn Stevens presented his work at the 1995 Spring American Geophysical Union Meeting (Stevens, 1995) and received an Outstanding Student Presentation Award for his efforts. Two invited seminars on this work were given by Bjorn Stevens. The first at the National Oceanic and Atmospheric Administration in Boulder. The second as part of the Colorado State University Applied Math Seminar. Two presentations are scheduled at the Department of Energy Science Team Meeting and the 1995 International Union of Geodesy and Geophysics General Assembly in Boulder.

In part due to his work on this project, Bjorn Stevens was the recent recipient of a National Aeronautics and Space Administration/EOS Graduate Student Fellowship in Global Change.

A list of all NIGEC supported publications is listed below in Section 8.

5.2 Collaborative Work

5.2.1 LANDSAT distributions of cloudiness

In LANDSAT observations of boundary layer clouds the probability distribution function of the liquid water path shows a qualitative change as a function of cloud fraction. In collaborative work with Bruce Wielicki, Steve Krueger and Kuanman Xu we are trying to explain LANDSAT distribution functions associated with different fractional cloud amounts, using a variety of two and three dimensional models

5.2.2 Evaluation of remote sensing techniques

We have used the model output as a surrogate for real (in-situ) cloud data to test the efficacy of new remote sensing techniques to measure cloud and drizzle microphysical properties (Frisch et al., 1995). These techniques use NOAA K_a band radar together with a microwave radiometer to measure cloud liquid water content and droplet concentration. The first 3 moments of the Doppler velocity spectrum can also be used to infer number, average size and dispersion of the drizzle drop spectrum. The simulations of this technique using model-derived data sets showed generally good agreement between remote measurements and the "real" data (Feingold et al., 1994b).

5.2.3 The GCSS working group on boundary layer clouds

William Cotton is chairman of the GEWEX Cloud Systems Study Program (GCSS) working group on Boundary Layer Clouds. Drs. Cotton and Chin-Hoh Moeng of NCAR organized and co-chaired a cloud modeling workshop in August of 1994. Bjorn Stevens participated in the Workshop. A copy of our report to GCSS is as given in Appendix A.

6 Executive Summary

An advanced cloud microphysics and large eddy simulation model of the stratus-topped marine boundary layer has been developed. Simulations of observed stratocumulus cases during FIRE I stratus and ASTEX have been performed. Preliminary simulations of the effects of enhanced CCN concentrations inserted in the marine boundary layer and above the boundary layer have been performed. Two-dimensional radiative transfer calculations with the spherical Harmonic Spatial Grid method have been performed for the enhanced CCN simulations. Calculated differences in cloud albedo due to six-fold increases in CCN concentrations above cloud top ranged from six to nine percent while the same CCN increase in the cloud layer yielded 10 to 15 percent increases in albedo. ASTEX studies have shown that drizzle production is closely related to the ability of a cloud to sustain larger drops through repeated collision-coalescence cycles.

A number of refinements to the model have been made including refined advection schemes, addition of aqueous chemistry for chemical cloud processing, refined supersaturation calculations and a bin CCN scheme.

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8 Publications Support by this Contract

8.1 Reviewed Publications

Duda, D. P., G. L. Stephens, and W. R. Cotton, 1994: Broadband solar albedo calculations in multi-dimensional marine stratus. To be submitted to *J. Atmos. Sci.*.

Feingold, G., B. Stevens, W.R. Cotton, and R.L. Walko, 1994: An explicit cloud microphysics/LES model designed to simulate the Twomey effect. *J. Atmos. Res.*, **33**, 207-233.

8.2 Conference Papers

Cotton, W.R., R.L. Walko, G. Feingold, Z. Levin, & S. Tzivion, 1992: Simulation of the Twomey effect. Preprints, 11th Conf. on Clouds & Precipitation, 17-21 Aug. 1992, Montreal, Quebec, CANADA.

Cotton, William R., Bjorn B. Stevens, Graham Feingold, and Robert L. Walko, 1992: A model for simulating the Twomey effect. Proceedings, Third International Cloud Modeling Workshop, 10-14 August 1992, Toronto, Canada, World Meteorological Organization.

Duda, D. P., G. L. Stephens, and W. R. Cotton, 1994: Impact of enhanced CCN concentrations on the radiative properties of a 3D marine stratocumulus cloud. Preprints of Eighth American Meteorological Society Conference on Atmospheric Radiation, 23-28 January, Nashville, Tennessee, American Meteorological Society, 262-264.

Stevens, B., W.R. Cotton, G. Feingold, R.L. Walko, 1994: Large eddy simulations of marine stratocumulus with explicit microphysics. Proc., 8th Conf. on Atmospheric Radiation, 23-28 January 1994, Nashville, TN, AMS.

Stevens, Bjorn, William R. Cotton, and Graham Feingold, 1995: The microphysical characteristics of convection in marine stratocumulus. Preprints, Conference on Cloud Physics, 15-20 January, 1995, Dallas, Texas.

Cotton, William R., Bjorn Stevens, and Sharon Nebuda, 1995: A question of balance — simulating microphysics and dynamics. Preprints, Conference on Cloud Physics, 15-20 January, 1995, Dallas, Texas.

8.3 Theses and Dissertations

Duda, David P., 1994: Macrophysical and microphysical influences on radiative transfer in two-dimensional marine stratus. Ph.D. dissertation, Colorado State University, Dept. of Atmospheric Science, Fort Collins, CO 80523, 202 pp. (Available as Atmos. Sci. Paper No. 565).

8.4 Other Reports

Stevens, Bjorn, 1993: A study of the theoretical behavior of ammonium sulfate aerosols in the vicinity of cloud base. Atmospheric Science Paper No. 535, Colorado State University, Dept. of Atmospheric Science, Fort Collins, CO 80523, 30 pgs.

A GCSS Working Group I-Boundary Layer Clouds 1994 Report

The major activity of the Boundary Layer Cloud working group of GCSS was the organization of a workshop in August 1994. The NCAR/GCSS Boundary Layer Cloud Workshop was held on August 16-18, 1994 in Boulder, Colorado. The purpose of this workshop was to bring together boundary layer cloud-resolving modellers, observers, and general circulation (GCM) modellers to intercompare simulations with various cloud resolving models to see how sensitive the models are to variations in numerics and physical parameterizations.

This, the first of a planned series of intercomparison workshops, was a rather simple case study. It was an idealization of the 7 July FIRE case described by Betts and Boers (1990) having a horizontally-homogeneous, nearly solid cloud deck, with no drizzle, no solar radiation, little wind shear, and weak surface heating.

A total of ten large eddy simulation (LES) groups worldwide participated in this intercomparison study. Table 1 summarizes some of the features of the models that participated

Table 3: Differences of Ten LES Codes

LES	Scientist	Advection	Time-velocity	SGS-turbulence	SGS-condensation	IR scheme
KNMI	Pier Siebesma	2nd-order FD	Leapfrog	Smagorinsky-Lilly	0 or 100%	Stephens-Welch, q_1 only
UKMO	Malcolm MacVean	2nd & 3rd FD	Leapfrog	Smagorinsky-Lilly	0 or 100%	grey-body, q_1 only
UMIST	Jacques Pasquier	2nd & 3rd FD	Leapfrog	Smagorinsky-Lilly	0 or 100%	mixed emissivity
UW	Chris Bretherton	2nd-order FD	Leapfrog	Smagorinsky-Lilly	0 or 100%	Roach-Slingo, q_1 only
MPI	Andreas Chlond	2nd & Bott FD	AB 2nd	Deardorff TKE eq.	S-D scheme	Cox-Stephens, q_1 only
NCAR	Chin-Hoh Moeng	mixed spectral-FD	AB 2nd	Deardorff TKE eq.	0 or 100%	mixed emissivity
UOK	Marat Khairoutdinov	5th-order FD in x,y	AB 3rd	Deardorff TKE eq.	0 or 100%	mixed emissivity
ARAP	Ian Sykes	2nd-order FD	Leapfrog	ARAP TEK eq.	S-D scheme	grey-body, q_1 only
WVU	Steve Lewellen	2nd-order FD	Leapfrog	ARAP TKE eq.	S-D scheme	grey-body, q_1 only
CSU	Bjorn Stevens	6th-order FD	Leapfrog	Smagorinsky-Lilly	explicit	mixed emissivity

in the study. There was only small differences among the mean wind, virtual temperature and total water fields (see Figures 1a-c), although there was quite a bit of scatter for the calculated mean cloud liquid water contents. The largest mean liquid water contents were

produced by the UMIST and UW models which is consistent with the fact that they both exhibited the highest radiative cooling rates (see Figure Fig 1e). Likewise, there is considerable scatter in cloud top heights, liquid water paths, and cloud cover (see Figures 2a-c) amongst the various models. The scatter in cloud top heights suggests large variations in average entrainment velocities among the models. I will discuss possible causes for the variation in entrainment velocities later.

The fields of buoyancy production of turbulent kinetic energy (TKE) and total kinetic energy shown in Figures 3a-b also shows considerable scatter among the models with the largest values of buoyancy production and turbulent kinetic energy near cloud top being for the UMIST model which also exhibits the largest values of cloud top cooling.

Differences among the models can be attributed to variations in the initial sounding that was actually implemented, numerics, sub-grid-scale turbulence and condensation routines, longwave radiation parameterizations, and saturation mixing ratio formulations. The cloud top cooling rates appear to be a major source of differences among the models. Moreover, the treatment of sub-grid-scale condensation also is a major difference in the models. This latter process also feeds back into the radiative cooling calculations, especially near cloud top.

A.1 Ongoing Activities

Because it was deemed that a major source of differences among the models was in their prediction of entrainment rates which depends on many factors such as cloud top radiative cooling, sub-grid-scale condensation, and liquid water prediction schemes, it was decided to establish a simple smoke-topped boundary layer case for intercomparison. Malcolm MacVean has agreed to set up this case. This case will be run entirely through electronic mail communication.

It was also proposed to consider the design of a focused experiment in which we attempt to directly measure entrainment velocities at the top of a stratus-topped marine boundary layer for the purpose of evaluating the performance of cloud-resolving models and param-

a

Horizontal Velocity Components; 3D LES

Figure 1: Vertical profile of horizontally averaged (a) vertical velocity (m/s), (b) virtual potential temperature ($^{\circ}\text{K}$), (c) total water mixing ratio (g kg^{-1}), (d) liquid water mixing ratio (g kg^{-1}), and (e) radiative heating rate (K h^{-1}).

b

Figure 1: Continued.

C

Figure 1: Continued.

d

Figure 1: Continued.

e

Figure 1: Continued.

a

Figure 2: Time evolution of (a) cloud top height (m), (b) liquid water path (g m^{-2}), and (c) cloud cover.

b

Figure 2: Continued.

C

Figure 2: Continued.

a

Figure 3: Horizontally-averaged vertical profile of (a) buoyancy production of TKE, and (b) total turbulent kinetic energy.

b

Figure 3: Continued.

eterized models in simulating entrainment. The design of the experiment is the subject of the next section.

A.2 Blueprint for Experimental Design

A.2.1 Introduction

In the Boundary Layer Cloud working group of GCSS we are fortunate to have had several recent comprehensive field campaigns such as FIRE Stratus and ASTEX which provide a wealth of information about stratocumulus clouds. These data sets are useful for furthering our understanding of the basic underlying physics of those clouds and for testing and developing models of boundary layer clouds of varying complexity.

With such a reservoir of data available to the cloudy boundary layer community one might question the need for any document defining the data requirements for the Boundary Layer Cloud Working Group. However, one can always identify gaps or weaknesses in existing field campaigns. For example, there have been few measurements of the cloudy boundary layer over land. Also, there is a dearth of information about the spectra of cloud condensation nuclei (CCN) that play such an important role in shaping cloud droplet spectra and determining if a cloud is likely to drizzle or not. Nonetheless, it is probably premature to consider the design of a major field campaign to study boundary layer clouds in the next few years.

At the NCAR/GCSS Boundary Layer Cloud Intercomparison Workshop held in Boulder, CO in August, I asked the participants what their assessment of the measurement needs are. The consensus was that a focused experiment aimed at measuring entrainment fluxes in the tops of stratocumulus clouds was needed. In fact, such an experiment is central to the theme of GCSS in which cloud-resolving models form the foundation for the construction and testing of parameterized cloudy boundary layer models for use in general circulation models (GCMs). Before synthetic data produced by cloud-resolving models will be accepted as a suitable substitute for real data to be used in developing cloud parameterization schemes, the end-user community must be convinced that the cloud-resolving

models are credible. Because the simulation of accurate entrainment velocities is crucial to predicting the evolution of stratocumulus clouds, we must demonstrate that our cloud-resolving models can predict entrainment velocities accurately. The entrainment process is so complex, however, that we do not yet know whether the dominate entraining eddies are explicitly resolved by current state of the art cloud resolving models (e.g., large eddy simulation (LES) models) or whether the unresolved eddies play a major role. Thus such an experiment is paramount to defining the credibility of LES models of the cloudy boundary layer.

What follows is a plan for measuring entrainment velocities at the top of a cloudy, marine boundary layer that is largely based on a plan proposed by Don Lenschow (personal communication).

A.2.2 Measuring Entrainment Velocity

First, it is important that the measurements be performed in a horizontally homogeneous region well away from shoreline and island effects so that reasonably steady-state cases can be obtained for comparison with model simulations. This pretty much constrains the measurement system to mobile platforms such as ships or aircraft, with aircraft being better able to obtain measurements above and in the cloud layer in relatively short times.

Measuring mean entrainment velocities is a major problem because the magnitudes are so small. For marine stratus, magnitudes are less than 0.01 m/s (Kawa and Pearson, 1989). Lenschow proposes two independent techniques for measuring entrainment velocities. The idea being that if the two techniques agree, then we can have some confidence that the measurements are correct.

The first method proposed by Lenschow is a direct measurement of a conserved tracer to obtain flux profiles through the boundary layer. The entrainment velocity is calculated from the ratio of the flux extrapolated to the top of the boundary layer to the difference in tracer concentration across the top of the boundary layer. Kawa and Pearson (1989) used this approach for ozone and total water. Lenschow notes that both these variables have

problems for this application. Both have large magnitude concentrations in the overlying atmosphere that vary both horizontally and vertically. In addition, total water is modified by drizzle and ozone has only a small sink at the surface.

Lenschow proposes that dimethylsulfide (DMS) would be an excellent tracer. It is emitted by phytoplankton in the ocean and has an atmospheric lifetime of about 2 days. Its free atmospheric concentration is usually negligible so that the concentration difference across the top of the boundary layer can be estimated accurately. A fast response DMS sampler suitable for eddy correlation analysis has not been developed yet. Lenschow is confident based on discussions with several experts (e.g., John Birks, Alan Bandy, Don Stedman, Fred Eisele, and Rich Benner) that current technology can be used for this measurement. Moreover, there is considerable enthusiasm to develop the sensor because of current interest in DMS as a source of CCN over the remote oceans.

The second approach involves diagnosing the entrainment velocity from the 'measured' mean velocity at the top of the boundary layer and the time rate of change of the boundary layer height. The entrainment velocity is the difference between the two. The change in boundary layer height can be measured in situ by repetitive penetrations of the capping inversion by aircraft or by releasing sondes. It can also be measured remotely with a lidar.

Direct measurement of the mean vertical velocity is not possible today or in the near future as its magnitude is similar to the entrainment velocity. It is, however, possible to diagnose the mean vertical velocity at cloud top by measuring the horizontal divergence at several levels in the boundary layer and the free air about the boundary layer top. Lenschow proposes that the divergence be measured with a side-looking Doppler laser system similar to forward-looking laser velocimeter flown on the NCAR Sabreliner (Keeler et al., 1987). Kristensen and Lenschow (1987) discussed the design criteria for a more complex scanning velocimeter. The advantages of this system are: (1) the measurement volume can be displaced several meters away from the aircraft; (2) Doppler shift is inherently an absolute measure of velocity; and (3) cloud droplets should not affect the accuracy.

Lenschow estimates that a combination of Inertial Navigation System (INS) and Global Positioning System (GPS) will provide an accurate measurement of airplane velocity and attitude angles with respect to the earth for these purposes.

A.2.3 Summary

In summary, we propose to carry out a focused field experiment to measure entrainment rates into the tops of marine stratocumulus clouds. The purpose for doing this is to obtain benchmark measurements of entrainment rates for testing and calibrating boundary layer cloud models. In particular, it is intended to determine the ability of LES models to properly represent entrainment rates into the tops of those clouds.

The experiment would be primarily a single aircraft experiment in which the aircraft is equipped with sensors for measuring vertical fluxes of a tracer such as DMS, a side-looking Doppler laser, a GPS, long- and short-wave upward and downward radiometers, a downward-looking aerosol backscatter lidar, and thermodynamic and cloud microphysics instrumentation. It may be desirable to have a second aircraft whose primary mission is to deploy GPS-dropwindsondes to provide a measurement of mean vertical motion in the region on a scale larger than the primary aircraft flight patterns.

The experiment would be about four to six weeks long in regions of reasonably horizontally homogeneous stratus well-removed from land or island effects. Some possible locations are summertime stratus off the California coast or off the coast of Peru where DMS production may be strong

A.3 Plans For 1995

Plans for the next intercomparison case focus on one or two of the ASTEX Lagrangian experiments. These cases provide both a clean air mass (low CCN concentrations) and a very dirty air mass case. In addition models can be used to examine simulated and observed trends for a several day period or for just a few hours in the middle of the experiment. These cases provide a wealth of data to test the model performances. Drs Chris Bretherton

and Steve Krueger have agreed to assemble this case and "lead the charge" in this second intercomparison study. Dr. Aad Van Ulden has volunteered to serve as host for the next meeting which will probably be held in the August to November 1995 time frame.

A.4 Acknowledgments

I would like to thank Don Lenschow for the work he has done in planning the details of this experiment.

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