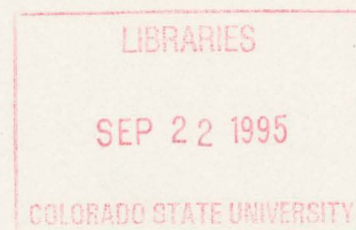


**CLOUD PROCESSING OF AEROSOL
USING A HYBRID LES/PARCEL MODEL WITH
SOLUTE-FOLLOWING MICROPHYSICS**

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ABSTRACT

CLOUD PROCESSING OF AEROSOL USING A HYBRID LES/PARCEL MODEL WITH SOLUTE-FOLLOWING MICROPHYSICS

The importance of climate forcings due to direct and indirect aerosol influences have been theorized, observed, and modeled, and are accepted on a qualitative level. Though these effects have been identified, the magnitude of their effect locally and globally has been difficult to estimate. Enhancement of the existing Regional Atmospheric Modeling System (RAMS) developed at CSU provides a way to investigate these impacts. The RAMS explicit microphysics has been expanded to include both an aerosol distribution stored in 14 size categories and separate solute mass and concentration information for each water drop size category. The initial aerosol distribution is activated according to chemical and physical principles; that is, it is not some parameterized function. The aerosol is subsequently followed as solute in each water bin, where each droplet size category independently stores a solute concentration. Because the aerosol mass is followed as solute in the drops, information on the changes in aerosol size and number due to cloud processing of an evaporating cloud is available.

Schemes for bin representation, aerosol activation, solute transfer during droplet growth, and aerosol regeneration from evaporating droplets were tested in a stand-alone box model. To determine the effectiveness of using the expanded microphysics in a full scale LES model, a series of tests using a RAMS simulation of a two-dimensional hill cap cloud were performed. A hybrid LES/parcel model was also developed. Several passive tracer trajectory environmental profiles through the hill cap cloud were determined during the full RAMS simulation. These profiles were used to drive a parcel model having the same explicit microphysics as the full RAMS simulation.

Additionally, the hybrid LES/parcel model was used to drive parcels derived from a RAMS simulation of the same cloud with bulk microphysics. Direct comparisons of

aerosol and droplet distributions in time and space were made for the full LES model and for the hybrid LES/parcel simulations. The hybrid LES/parcel model shows promise in its ability to combine very dynamically complex cloud types with complex cloud microphysics and chemistry. Cloud microphysical features, such as cloud base supersaturation maximum, were not well represented in the RAMS simulation, but were in the parcel model simulations.

Aerosol which have deliquesced and/or activated to cloud droplets serve as sites for aqueous chemical reactions which can enhance the aerosol mass. An aqueous chemistry module (Kreidenweis, 1992), appropriate for use as a stand-alone model, has been extended. The expanded version can be used to represent both externally and internally mixed aerosol. A seasalt aerosol option is included which can be used to represent marine aerosol distributions. Seasalt constituents that directly affect the chemistry are represented explicitly, along with a parameterization for seasalt alkalinity. Options for multiple droplet sizes and concurrent droplet growth have been created, for incorporation into a dynamical/ microphysical cloud model.

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1. Introduction

Human impacts on global and local climate are tremendously difficult to understand and quantify. Our species has managed to alter the chemistry of the atmosphere in the course of our energy production and other industrial pursuits both to a large degree and in a relatively short period of time (within the last 200 years). Injection of long lived greenhouse gases may have altered the radiation balance of the clean atmosphere. Gases and aerosols having shorter lifetimes may also have a significant effect on the radiation balance, both directly via light scattering and absorption and indirectly by altering the composition of and amount of cloud condensation nuclei (CCN) (Twomey, 1974 and 1977). Although greenhouse warming has garnered much publicity in recent years, the counterbalancing effect of cooling due to aerosol by both direct influences (scattering and absorption of radiation by the aerosol) and indirect influences (effect of the aerosol on cloud radiative properties and cloud lifetimes) is only more recently being recognized as equally significant. The magnitude of the global radiative forcing by aerosol is now considered by many to be of the same order as that due to greenhouse gases, but opposite in sign (Coakley and Cess, 1985; Charlson, 1992).

Particulate matter can enter the atmosphere directly, or be formed in the atmosphere by condensation of gases in gas-to-particle conversion processes. Particles can subsequently undergo further processing via gas-aerosol interactions (condensation, including deliquescence, and chemistry), aerosol-aerosol interactions (collision/coalescence), and cloud processing. Aerosol typically have a relatively short lifetime and are removed by both wet and dry deposition processes.

Cloud processing of aerosol refers to the altering of the aerosol spectrum due to the presence of cloud, and can include dynamical redistribution of aerosol, changes in aerosol due to cloud microphysics, and chemical changes (including mass enhancement) via in-cloud aqueous or gas phase chemistry. Convective cloud updrafts and downdrafts can cause a physical redistribution of aerosol and shorter lived, less well mixed trace gases. Aerosol can be removed from the atmosphere by wet

deposition. The collision and coalescence of droplets with other droplets or with interstitial aerosol in a cloud can serve to combine soluble aerosol, depleting their number, altering composition by mixing, and increasing the aerosol's average size upon the eventual evaporation of the droplets. Chemistry in cloud can cause an increase of the aerosol mass, primarily by oxidation of sulfur dioxide, SO_2 , to sulfate ion, SO_4^{2-} . On evaporation of the droplets, the mass-enhanced aerosol regenerated would subsequently activate at reduced supersaturation. In addition to the importance of aqueous chemistry in the modification of aerosol, the chemistry is also important in the reduction of atmospheric oxidants, and the acidification of cloud water and/or aerosol.

Though these effects of cloud processing on aerosol are accepted on a qualitative level, the magnitude of their influence has been difficult to estimate. The existing Regional Atmospheric Modeling System (RAMS) developed at Colorado State University (Pielke et al., 1992) provides a way to investigate these impacts, by adding aerosol to cloud simulations. The goal of this work is to develop methods for better estimates of the changes in the aerosol spectrum and its spatial distribution due to cloud chemistry, microphysics, and dynamics.

This thesis covers two distinct model types, both of which can be used to investigate cloud processing of aerosol. In one, a chemistry module has been extended for simulation of aqueous chemical processes. This aqueous chemistry model is appropriate for use as a stand-alone model or for incorporation into either an Eulerian, grid type model, or a Lagrangian, parcel following model. This module is an extension of a previous model (Kreidenweis, 1992), which assumed all drops had the same size and composition, and constant liquid water content. This has been expanded to include the following:

1. Multiple drop size.
2. Inclusion of internally and externally mixed aerosol as droplet base. Aerosols coded are ammonium sulfate $(\text{NH}_4)_2\text{SO}_4$, letovicite $(\text{NH}_4)_3\text{H}(\text{SO}_4)_2$, ammonium

bisulfate NH_4HSO_4 , sulfuric acid H_2SO_4 , pure NaCl , and silicon (for inclusion of an insoluble, relatively inert component).

3. Boron chemistry for seasalt (Pzenny, 1982)
4. CO_2 and carbonate chemistry.
5. HCl chemistry
6. Time-dependent liquid water content and a gas/liquid water balance for simultaneous growth of droplets.
7. In the case of an evaporating droplet, partitioning of solute species for aerosol regeneration.

This chemistry module has been used to investigate both the changing droplet acidity and the solute mass enhancement due to aqueous oxidation of SO_2 to SO_4^{2-} . The effects of initial ambient concentrations of precursors, and varying conditions of drop size, liquid water content, and temperature, are considered.

Another model type for the investigation of cloud aerosol processing is one in which detailed descriptions of cloud dynamics and microphysics is provided. Here, the existing RAMS explicit microphysical subroutine (Feingold et al., 1994, Stevens et al., 1995b) has been expanded to include a 14 size category, or sectional bin, aerosol distribution. The initial aerosol distribution is activated according to chemical and physical principles. Four separate methods of storage for the aerosol were considered and weighed for their consistency with activation theory. In method one, aerosol were stored as number only at some specified aerosol mass size for each size category. In methods two through four, the aerosol in each bin were represented as a total aerosol mass and total aerosol number in each bin, with different assumed distribution of aerosol in the bins.

The aerosol mass activated is incorporated into the water drop bins as solute. Each droplet size category independently stores a solute mass, and the solute can be followed through the droplet spectrum during cloud processes such as condensation, evaporation, and collision/coalescence. This solute-tracking model allows for the calculation of droplet size and solute concentration dependent chemical processes. For

example, droplet growth, which is dependent on solute effect for small drops, can be calculated for each drop size. Sulfate production, which is dependent on drop size and species present, can also be determined for each independent drop category. The explicit storage of solute also allows recovery of the mass mean size of aerosol regenerated from evaporating drops. Approximations to the regenerated aerosol size, about the known average size, were implemented and tested.

In the development of this model, a numerical scheme was required for transferring the solute through the droplet spectrum as the droplets grew via condensation of vapor. Two methods of weighting the solute transfer with the water transfer were tested. The first method weighs the solute transferred by droplet number transferred, and the second method weighs solute transfer with the water mass transferred. The expanded aerosol/solute module was tested extensively in a box model mode to determine the effects of solute weighting, and to test the use of different aerosol and water bin configurations and distribution structures.

To determine the effectiveness of using the expanded microphysics in a full scale Eulerian model, a series of tests using a RAMS simulation of a simple two-dimensional (2D) hill cap cloud were performed. First, the new microphysics was simply added to all grid calculations. These simulations will henceforth be referred to as the RAMS+ model runs. Second, in order to attempt to isolate any sources of unrealistic mixing or smoothing of distributions due to grid to grid advection of the aerosol variables, a parcel model was developed and driven by the thermodynamics experienced by a passive tracer which flowed through the hill cap cloud. This model, using the same sectional bin representation as the RAMS+ model, will be called the parcel-SB (sectional bin) model. Information on the location of the tracer allows a direct comparison of the aerosol, cloud water, and solute distributions for specific parcel locations in space and time for the two model types. Third, a different microphysical scheme was used in the parcel model. In this scheme the growth of aerosol and droplets is followed in a Lagrangian sense, eliminating computational diffusion of the solute; this will be termed the parcel-LA (Lagrangian aerosol) model. This model is used to evaluate the magnitude of computational diffusion effects on the aerosol spectrum in the first model.

2. Background

2.1 Cloud processing

Aerosols and reactive chemical species can be subject to spatial and temporal variations due to convective transport, removal effects via precipitation, and heterogeneous reactions which take place in clouds. Chatfield and Crutzen (1984) among others proposed that the convective transport of species by clouds may be important in redistributing species from lower levels of the troposphere to higher altitudes.

Experimental evidence confirms cloud impact on distributions of both aerosol and gas species (e.g., Berresheim et al., 1990). Two dimensional models have also been applied to investigate the variations in species concentrations due to the dynamical influence of clouds (Chatfield and Crutzen, 1984; 1990). There are many reactions (for instance, $\text{SO}_2 \rightarrow \text{SO}_4^{2-}$) which take place at a much greater rate in the aqueous phase.

The Regional Acid Deposition Model (RADM) was used by Walcek et al. (1990) to predict that over 65% of the sulfuric acid formed during the passage of a midlatitude storm system was formed via aqueous reactions in the cloud droplets. An increase in aerosol size due to aqueous chemistry was modeled by Easter and Hobbs (1974) and Hegg et al. (1992), among others.

Research on the cloud processing and redistribution of aerosol and atmospheric gasses includes the following topic areas:

1. The dynamical redistribution of "passive tracers": these tracers move dynamically with the air motions but do not interact in any way with the cloud water (e.g., Cotton, 1994 ; Niewiadomski, 1986; Nicholls and Weissbluth, 1988).
2. The dynamical redistribution of soluble but not reactive tracers: these might be "real" soluble gas species, but no gas or aqueous phase reactions are allowed (e.g., Cotton, 1994).

3. The redistribution of CCN via nucleation scavenging and impaction scavenging (e.g., Flossmann and Pruppacher, 1988).
4. The redistribution of reactive gases in a cloud parcel by aqueous chemistry. This box model is driven by thermodynamics implied from a constant vertical speed rising parcel to simulate vertical advection in a stratus or a cumulus cloud, or from a sinusoidal height profile simulating a wave type or hill cap cloud. Drop size dependent gas to aerosol conversion via heterogeneous chemistry is included (e.g., Ayers and Larson, 1990; Easter and Hobbs, 1974; Hegg and Hobbs, 1982; Twohy et al., 1989).
5. Eulerian type cloud model. Aqueous chemistry is included on bulk liquid water, having one or two size categories, or in multiple drops size categories (e.g., Barth et al., 1992; Flossmann, 1991).

Research in areas 1. and 2. above has shown significant differences between cloudy and clear air transport. Chatfield and Crutzen (1984) suggest that the observed global distributions of SO_2 are not what would be expected for a species with such a short atmospheric chemical lifetime if only vertical transport via eddy diffusion is assumed. They find that cloud pumping allows the rapid transport of SO_2 and its precursors up to higher levels where their lifetimes are longer. Costen, Tennille, and Levine (1988) found similar results in their study. At high altitudes, the cloud pumping caused DMS (an SO_2 precursor) to increase by 700 times the concentration of DMS computed without cloud pumping. SO_2 shows a similar trend. Even though the overall SO_2 transport via cloud pumping had been reduced by 0.47 to parameterize the effects of a sink due to cloud chemistry and precipitation, there was still a significant increase in SO_2 concentration at 10 km due to cloud pumping, a factor of 21 increase over the eddy diffusion only case.

Physical changes in the aerosol distribution by areas 3. and 4. above can alter the local and global radiation budget by direct and indirect effects. Aerosol can be removed by nucleation and impaction scavenging followed by precipitation (Flossmann, 1985; Barth

et al., 1992). Number can be decreased and aerosol size increased due to evaporation of a cloud that has undergone collision/coalescence processes. Mass can be added to aerosol by aqueous reactions in cloud drops. According to Hegg et al. (1993), unprocessed and cloud processed aerosol will have different optical properties, an example of changing direct radiation effects. Easter and Hobbs (1974) found that after a 4 minute flow through a wave cloud, mass of the aerosol was increased such that the concentration of CCN active at 0.5% supersaturation increased by 75%.

2.2 Modeling perspectives

There are three interactive perspectives of cloud modeling that may be considered in order to represent the cloud processing of aerosol and gasses. The first is the thermodynamical cloud: presented as the gross manifestations of rising and falling parcels of air that produce a cloud, alter radiation balance, and redistribute energy and mass physically in space. Second is the microphysical cloud: the homogeneous or heterogeneous nucleation of a droplet or ice particle, growth by vapor deposition and collision/coalescence mechanisms, freezing, thawing, riming, and smaller scale exchanges of energy. Aerosol can be reduced in number and increased in single particle mass size through hydrometeor collisions; aerosol that was externally mixed can become internally mixed in the droplets.

The gross thermodynamics of the cloud and the microphysics influence each other. For example, a rising parcel of air tends to cool, changing local supersaturations around droplets in the parcel, while condensational growth of droplets releases latent energy, warming the parcel and making it more buoyant. Of course the division between thermodynamics and microphysics is artificial. Release of latent heat energy due to condensation and "parcel warming" are actually one and the same, but are simply conceptualized on different time scales.

The third perspective involves the gas and aqueous phase chemistry. The presence of cloud can greatly enhance the conversion of gaseous precursors to solute/aerosol mass,

deplete important oxidants, and acidify precipitation in the course of heterogeneous chemical reactions (Lelieveld and Crutzen, 1991). Cloud chemistry and microphysics are not independent, and proceed in an interactive fashion. One example of this: droplet growth will cause a dilution of dissolved species, and the species concentration will affect droplet growth rate via the solute effect. Droplet size influences the rate of gaseous uptake by the droplet. The freezing of haze and other concentrated droplets will be dependent on concentrations of the "impurities", which depress freezing temperatures (Jensen, Toon, and Hamill, 1991; Heymsfield and Sabin, 1989; and Luo, Peter, and Crutzen, 1992). The mass increase of aerosol due to aqueous oxidation of SO_2 to SO_4^{2-} can also greatly affect subsequent clouds formed on the aerosol distribution, making them easier to activate (Hegg and Larson, 1989; Seidl, 1989).

It is the cloud microphysics and chemistry that this thesis focuses on, with the goal of linking models of these processes to a dynamical cloud model in an interactive way. The inclusion of explicit droplet size resolving microphysics adds greatly to memory demands for a numerical cloud model, and a simple cloud type is preferred. In the case that additional hydrometeor categories, such as snow, aggregates, and hail, are to be explicitly resolved, sometimes in both size and shape, these memory demands are greatly compounded. This thesis explores only warm cloud microphysics.

2.3 Approaches to modeling microphysics plus chemistry: simple dynamics.

Both microphysical and chemistry models have become more and more complex. Approaches to unite these typically involve a very simplified version of one coupled with a more rigorous version of the other. For example, many studies of cloud chemistry include explicit chemistry, but simple cloud dynamics and microphysics. The cloud is assumed to have one water phase (liquid water), a certain uniform droplet size, typically $10\text{ }\mu\text{m}$, constant temperature, and trace gas concentrations initialized in the ambient air. The incorporation of aerosol as cloud condensation nuclei (CCN) in the droplet is handled by entering an initial solution concentration for the droplets

which is uniform in composition and concentration. The chemistry is then "turned on" and observed. Examples of work using this approach include those by Jacob (1986) and Lelieveld and Crutzen (1991). Cloud microphysics and chemistry are decoupled. Such models are useful in the study of gas/aqueous chemistry, sensitivity of reactions to pH, temperature, LWC, droplet size, and ambient gas concentrations.

A compromise solution has been to use some simple chemistry with simple microphysics. Two recent examples are the models used in Ayers and Larson (1990) and Twohy, Austin, and Charlson (1989). These simulate droplet nucleation and growth in an adiabatic air parcel moving upward with a specified updraft velocity. Concurrent with the droplet growth is the exchange of soluble gases between ambient air and the droplet. Both of these models used an externally mixed aerosol (a sulfate aerosol and NaCl), and used the Lagrangian aerosol bin structure described above. Cloud dynamics were simulated by imposing a constant updraft speed for the parcel. Roelofs (1992) used a simple entraining parcel model with solute following microphysics to investigate sulfate production and concentrations in size defined droplets.

Many of the complexities of a multi-dimensional convective cloud (i.e., collision and coalescence) are ignored. These kind of models are useful in the determination of how parameters such as drop size, drop pH, and aerosol composition, can affect gas uptake and addition of aerosol mass. Many researchers have found that the rate of sulfate production varied greatly across droplet categories (of droplet size, nucleating aerosol type) (Ogren et al., 1989). These results are typical.

In the Ayers and Larson (1990) model gaseous species included were carbon dioxide (CO_2), ammonia (NH_3), ozone (O_3), SO_2 , and hydrogen peroxide (H_2O_2). The aqueous species associated with these were also included in the chemistry. The oxidation of aqueous SO_2 to SO_4^{2-} by O_3 and H_2O_2 is included. Initial concentrations of the gaseous species were taken from several references to be representative of typical conditions. Because of the high concentration of some species, activity coefficients were used to account for ionic interactions. Using the condition of electroneutrality in the droplet,

concentrations and pH in the droplet could be computed. Although the chemistry was kept rather simple as far as number of species included, the exchange of soluble gasses between the droplet and the surrounding air, and the aqueous chemical reactions, were handled by explicit rate equations. Equilibrium was not assumed. Sixty bins of aerosols were followed from their 99% relative humidity (RH) size through a 150m rise from cloud base. The bulk of the water at the end of the run is associated with the few smallest droplet classes containing the largest number of droplets, yet those drops that formed on sea salt count for a significant fraction, 21%, of total liquid water. Also, this 21% of the liquid water was associated with 40% of the total sulfate production. Sulfate production percentage associated with the different droplets are given in Table 2.1.

Table 2.1: Liquid water fractions and sulfate production (Ayers and Larson, 1990)

six smallest sulfate bins	71% of the liquid water	35% of total sulfate produced
next six sulfate bins	6.3%	23%
all sea-salt bins	21%	40%

The calculated sulfate production has a significant non-linear distribution with liquid water across the droplet size range and across the two nucleus types, largely due to pH dependent oxidation processes.

Hegg and Larson (1989) used an internally mixed aerosol in their simulation. Sulfate production differed drop to drop. The predicted overall sulfate production was from 3 to 30 times higher than that predicted using a bulk model. The difference in production is caused by a complex interaction of factors. The aerosol size distribution affects the droplet size distribution, which affects the pH distribution, which causes a distribution of rates of SO₂ oxidation. The difference that pH makes in the oxidation rate is large enough that only a few bins of high-pH droplets are needed to affect overall oxidation rates.

The model of Twohy, Austin, Charlson (1989), similar to the Ayers and Larson (1990) model, also uses an externally mixed sulfate and sea-salt aerosol distribution. They used a Lagrangian parcel model, with a vertical speed of 0.35 m/s which ran for 215 s,

focusing on initial growth and chemistry. Droplet molarities were calculated as a function of cloud droplet height in the cloud. Below the cloud, the concentration was determined by the CCN mass and its equilibrium size via the Köhler equation at the supersaturation at that position (in this case, 0.31%).

Many results paralleled those of Ayers and Larson (1990), finding a sulfate production dependence on droplet size, aerosol constituent, and pH. In addition, calculations of the Mie scattering of pre- and post-cloud aerosol (after cloud evaporation) were performed. The aerosol did increase in mass due to in-cloud sulfate production. The Mie scattering also increased significantly. The implication is that cycling of an aerosol through a cloud can cause an increase in short wave forcing. A 30% increase in aerosol can cause a forcing equal and opposite to the magnitude of longwave forcing due to doubling of CO₂ (Twohy, Austin, and Charlson, 1989).

2.4 Approaches to modeling microphysics plus chemistry: complex dynamics.

Another approach to investigating cloud effects on aerosol is to use a detailed dynamical model to simulate the cloud, with either explicit or parameterized microphysics and parameterized chemistry. Activation of CCN may be included as a parameterization, based on observation (Ghan et al., 1994; Twomey, 1977). When heterogeneous chemistry is included in such a multi-dimensional convective cloud model, the chemistry is typically decoupled from any dynamics and/or microphysics. Chemistry on "bulk" water in a grid of one or two averaged drop sizes, often cloud drops and raindrops, is common (Taylor, 1989a; Barth et al., 1992; Wang and Chang, 1993a; Gregoire et al., 1994).

The more complex, convective type cloud models that include some form of heterogeneous chemistry are rather inconclusive. The reaction rates will be very cloud specific and will depend on the local environmental characteristics and reactant concentrations. As these vary significantly from case to case, the results are hard to

compare. Taylor (1989b), for example, in a 1.5-dimensional model of a small, isolated cumulonimbus cloud, predicts that about 50% of the sulfate fraction in the cloud is produced by oxidation. Wang and Chang (1993b), found, for their three-dimensional case study of a thunderstorm, that aerosol scavenging provided 50 times the amount of sulfate produced via aqueous oxidation.

2.5 Explicit Resolution of water droplet distribution

Microphysical models which explicitly resolve drop size can either use an analytical distribution function for the drops, or resolve drops in separate size categories or bins. Bin storage can be accomplished in a Lagrangian manner in which droplets always stay in the same category and the category "size" changes with droplet growth. Alternatively, Eulerian bins with fixed sizes can be used, with the drop spectrum flowing through the grid as drops grow and coalesce.

Several reported studies have used adiabatic parcel models (Ayers and Larson, 1990; Twohy et al., 1989; Easter and Hobbs, 1974). In these, the Lagrangian representation of aerosol/droplet is used. This type of model has the advantage of no computational mixing between bins, but collision/coalescence of droplets is difficult to include. When collision/coalescence processes are ignored or deemed insignificant, this type of model can be advantageous for computing gas uptake, aqueous chemistry, or any process that might require knowledge of many variables stored for each droplet category.

The Eulerian bin type works well when collision/coalescence and breakup are important, allowing for convenient algorithms for the stochastic simulations (Feingold et al., 1993; Tzivion et al., 1989; Chen and Lamb, 1994). This representation can also simulate the structure of mean microphysical fields and is useful in representation of the coupling of dynamics and microphysics (Stevens et al., 1995b). However, condensational growth has been shown to be diffusive, broadening the water droplet spectrum unrealistically (Feingold et al., 1994). Additionally, when a solute category is added to these Eulerian-type sectional water bins, solute distributions can undergo

averaging as the droplet spectrum narrows into fewer water bins. This averaging was found to be quite significant in comparing sulfate production rates from the model by Roelofs (1992), and the model by Twohy et al. (1989), which followed each aerosol size independently in a Lagrangian-type aerosol representation. In these two studies, the explicit drop size dependent sulfate production was compared to a bulk run. Use of the explicit drop size representation resulted in a fourfold increase of sulfate production in the Roelofs (1992) simulation, and factor of three or more increase in the Twohy (1989) simulation. Use of the Eulerian-type bins did enable Roelofs to show some results of collision/coalescence processes on sulfate concentrations on effected drops.

Recent work by Stevens, et al. (1995a, 1995b) elucidate some of the problems in attempting to put drop size resolved microphysics in a grid type model. The use of grid-average thermodynamic fields in predicting microphysical evolution produces anomalous supersaturations at cloud edges, and can also cause a decrease in supersaturation maximum at cloud base. This brings about significant effects on the number of cloud condensation nuclei (CCN) activated, and hence on cloud droplet number.

2.6 The Hill Cap Cloud

Cloud types similar to the one chosen for this thesis, a simple hill cap cloud, have been chosen for study by others, as the flow of air over the hill is considered to approximate a laminar flow, and it has proven simple to model. In addition, observational data is easier to collect (Carruthers and Choularton, 1982; Hill et al., 1986; Bower et al., 1991). Carruthers and Choularton (1982) modeled the dynamics of the hill cap airflow with a three layer simulation similar to shallow water type models. A similar cloud type for its simplicity and its laminar flow is the wave cloud (Easter and Hobbs, 1974; Hegg and Hobbs, 1982). In these, a parcel flowing through a wave cloud is modeled using a sine curve for a spatial profile in order to simulate the wave cloud properties.

3. Aqueous Chemistry

3.1 The model

A chemistry module has been developed for quantifying the chemical cloud processing of aerosol. 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 to include multiple drop categories, where all rate equations involved are solved simultaneously using the Variable-coefficient Ordinary Differential Equation (VODE) solver (Brown et al., 1989).

Gases represented in the model were $\text{SO}_2(\text{g})$, $\text{O}_3(\text{g})$, $\text{H}_2\text{O}_2(\text{g})$, $\text{NH}_3(\text{g})$, and $\text{HNO}_3(\text{g})$.

Aqueous species were $\text{S(IV)} = \text{SO}_2(\text{aq}) + \text{HSO}_3^{-1} + \text{SO}_3^{-2}$; $\text{O}_3(\text{aq})$; $\text{H}_2\text{O}_2(\text{aq})$;

$\text{N(III)} = \text{NH}_4\text{OH} + \text{NH}_4^{-1}$; $\text{N(V)} = \text{HNO}_3(\text{aq}) + \text{NO}_3^{-1}$; and $\text{S(VI)} = \text{H}_2\text{SO}_4 + \text{HSO}_4^{-1} + \text{SO}_4^{-2}$. In this work, we added $\text{CO}_2(\text{g})$ and the aqueous species $\text{C(IV)} = \text{CO}_2(\text{aq}) + \text{HCO}_3^{-1} + \text{CO}_3^{-2}$.

HCl chemistry ($\text{HCl}(\text{g}) \rightarrow \text{H}^{+1} + \text{Cl}^{-1}$) was included for some runs, but deemed relatively unimportant for the intended purpose of the code and removed. In another addition, internal or external mixtures of the following aerosol species are also coded for direct input: ammonium sulfate $(\text{NH}_4)_2\text{SO}_4$, letovicite $(\text{NH}_4)_3\text{H}(\text{SO}_4)_2$, ammonium bisulfate NH_4HSO_4 , sulfuric acid H_2SO_4 , pure NaCl (includes seasalt alkalinity), and silicon. Upon drop evaporation, the size distribution of resuspended mater is computed.

An initial droplet spectrum is input by specifying values of water content in each droplet size category. Aqueous species concentration initialization for each category are determined using the associated aerosol component(s), and /or simply specified. Concentration values for each aqueous species are stored independently for each drop category. Initial gaseous species concentrations may be specified as constant

throughout the model run, or be depleted in the course of the run by uptake and chemical reaction.

The relative rates of several processes must be considered in modeling the in-cloud aqueous chemistry: diffusion of the gases to the droplet, mass transfer of species into or out of the droplet, diffusion of species inside the droplet, dissolution and/or ionization of soluble species in the drop, and chemical reactions of species in the drop. For the different species and/or in different conditions, what steps are rate determining vary. In this formulation, diffusion of the gases to the droplet, mass transfer of species into or out of the droplet, and chemical reactions of species in the drop will be calculated in a set of species mass balance equations. Ionization of soluble species in the drop are determined by assuming fast equilibrium.

The aqueous species mass balance equations have the general form (Pandis and Seinfeld, 1989):

$$\frac{d_i[C_i(aq)]}{dt} = k_{mi}[C_i(g)] - k_{mi} \frac{1}{K_{Hi}RT}[C_i(aq)] + R_i \quad \text{Equation 3.1}$$

where $[C_i(aq)]$ and $[C_i(g)]$ are the aqueous and gaseous concentrations, respectively, of species i in mol/L, R is the ideal gas constant (0.082058 L atm/ mol K), K_{Hi} is the effective Henry's law constant of species i , and T is the temperature in Kelvin. R_i is the source or sink term for aqueous chemical reactions. Brackets surrounding a chemical species, i.e. $[C_i(g)]$, represents concentrations in mol/L. The mass transfer rate coefficient, k_{mi} , combines both gas-phase and interfacial mass transport:

$$k_{mi} = \frac{3\eta D_{g,i}}{\alpha^2} \quad \text{Equation 3.2}$$

where $D_{g,i}$ is the diffusivity of species i in air, taken to be $0.1 \text{ cm}^2/\text{s}$, α is the droplet radius, and η is a correction coefficient for free molecular effects. The coefficient is approximated as:

$$\eta = \left\{ 1 + \left[\frac{1.33 + 0.71Kn^{-1}}{1 + Kn^{-1}} + \frac{4(1 - a_w(i))}{3a_w(i)} \right] Kn^{-1} \right\} \quad \text{Equation 3.3}$$

where Kn is the Knudsen number, the ratio of the mean free path of air to the droplet radius. The sticking coefficient for species i , $a_w(i)$, is taken to be 0.01 for all species.

The mean free path (mfp) is a function of temperature and pressure:

$$mfp = \frac{R \times T}{\sqrt{2} \times \pi \times d_c^2 \times Av \times P} \quad \text{Equation 3.4}$$

where P is the atmospheric pressure, Av is Avagadro's number, and d_c is the collision diameter. A mole weighted average of the collision diameters for atmospheric constituents N_2 and O_2 ($3.74\text{e-}10$ and $3.57\text{e-}10$ m respectively) is taken as the value for d_c , $3.67\text{e-}10$ m. The equation can be simplified in terms of $[air]$, the atmospheric concentration in mol/L:

$$mfp(cm) = \frac{3.738e - 25}{d_c^2(m) \times [air]} \quad \text{Equation 3.5}$$

where $[air]$ is calculated using the Ideal Gas Law.

The general form of the mass balance equations for the gaseous species are (Pandis and Seinfeld, 1989):

$$\frac{d[C_i(g)]}{dt} = k_{mi} w_l [C_i(g)] - k_{mi} w_l \frac{1}{K_{Hi} RT} [C_i(aq)] + R_i \quad . \quad \text{Equation 3.6}$$

where w_l is the liquid water content in L/L.

Two alternate formulations for representing seasalt aerosol have been implemented. Although in some cases pure NaCl has been used as an approximation to a coarse mode marine aerosol (Flossmann, 1991), this surrogate for a marine aerosol is a neutral, nonbuffering species. In this work, the method of Pszenny, et al. (1982), for computing seasalt alkalinity was implemented. In this method, total boron, B_T , is assumed to be proportional to the salinity in g/kg, sal :

$$[B_T] = [B(OH)_3] + [B(OH)_4^-] = sal \times 0.000012 \quad . \quad \text{Equation 3.7}$$

The equilibrium reaction is $[H_2O] + [B(OH)_3] \rightarrow [B(OH)_4^-] + [H^+]$, and the equilibrium constant, K , is defined as:

$$K = \frac{[H^+][B(OH)_4^-]}{[B(OH)_3]} = 5.01e-13 \cdot M \quad . \quad \text{Equation 3.8}$$

Equations 3.7 and 3.8 can be then used to determine the concentration of negative ions contributed by B_T in the electroneutrality equations:

$$[B(OH)_4^-] = \frac{0.000012 \times sal \times K}{[H^+] + K} \quad . \quad \text{Equation 3.9}$$

The net positive ion concentration due to seasalt alkalinity, A_T , in moles/liter, are approximated by (Pszenny et al., 1982):

$$A_T = 0.0000657 \times sal \quad . \quad \text{Equation 3.10}$$

Table 3.1: *Equilibrium reactions (Pandis and Seinfeld, 1989).*

Reaction number	Reaction	Equal. const. K_{298} M or M/atm	$-\Delta H/R$, K
1	$\text{SO}_2(\text{g}) \rightarrow \text{SO}_2 \cdot \text{H}_2\text{O}$	1.23	3120
2	$\text{SO}_2 \cdot \text{H}_2\text{O} \rightarrow \text{HSO}_3^{-1} + \text{H}^{+1}$	1.23e-2	1960
3	$\text{HSO}_3^{-1} \rightarrow \text{SO}_3^{-2} + \text{H}^{+1}$	6.61e-8	1500
4	$\text{H}_2\text{SO}_4 \rightarrow \text{HSO}_4^{-1} + \text{H}^{+1}$	1000	
5	$\text{HSO}_4^{-1} \rightarrow \text{SO}_4^{-2} + \text{H}^{+1}$	1.02e-2	2720
6	$\text{H}_2\text{O}_2(\text{g}) \rightarrow \text{H}_2\text{O}_2(\text{aq})$	7.45e+4	6620
7	$\text{HNO}_3(\text{g}) \rightarrow \text{HNO}_3(\text{aq})$	2.1e+5	
8	$\text{HNO}_3(\text{g}) \rightarrow \text{NO}_3^{-1} + \text{H}^{+1}$	15.4	8700
9	$\text{O}_3(\text{g}) \rightarrow \text{O}_3(\text{aq})$	1.13e-2	2300
10	$\text{NH}_3(\text{g}) \rightarrow \text{NH}_4\text{OH}(\text{aq})$	75	3400
11	$\text{NH}_4\text{OH}(\text{aq}) \rightarrow \text{NH}_4^{+1} + \text{OH}^{-1}$	1.75e-5	-450
12	$\text{H}_2\text{O} \rightarrow \text{H}^{+1} + \text{OH}^{-1}$	1.0e-14	-6710
13*	$\text{CO}_2(\text{g}) \rightarrow \text{CO}_2(\text{aq})$	3.4e-2	2420
14*	$\text{CO}_2 \cdot \text{H}_2\text{O} \rightarrow \text{HCO}_3^{-1} + \text{H}^{+1}$	4.5e-7	-1000
15*	$\text{HCO}_3^{-1} \rightarrow \text{CO}_3^{-2} + \text{H}^{+1}$	4.69e-14	1796
16*	$\text{B}(\text{OH})_3 \cdot \text{H}_2\text{O} \rightarrow \text{B}(\text{OH})_4^{-1} + \text{H}^{+1}$ (Pszenny et al., 1982)	5.01e-13	

(Asterisks designate reactions added to the Kreidenweis, 1992 code.)

In this representation of seasalt aerosol, the Pszenny (1982) method is employed for electroneutrality calculations. Salinity is calculated from the chlorinity ($\text{g Cl}^{-1} / \text{kg}$) of the droplet, using the relationship of salinity = $1.080655 \times \text{chlorinity}$ (Weyl, 1970). As seasalt is approximately 55% Cl^{-1} , the measure for salinity is then:

$$\text{sal} = 0.55 \times \text{seasalt mass} \times 1000 / \text{droplet total mass} \quad \text{Equation 3.11}$$

The use of a more explicit seasalt aerosol has been explored, to represent seasalt aerosol components ignored when using NaCl (or NaCl plus alkalinity) only. Major seasalt

components are Cl^- , Na^+ , Mg^{+2} , SO_4^{-2} , Ca^{+2} , K^+ , HCO_3^{-1} , Br^- . Including all of these species independently would not be worthwhile here; however, pure seasalt is approximately 8% sulfate by weight, not a negligible amount especially when the determination of sulfate sources is such a significant part of this type of modeling study. In addition, the bicarbonate ion can influence CO_2 equilibrium chemistry.

Seasalt bicarbonate and sulfate are not added to the electroneutrality equations, as they are accounted for in the Pszenny, 1982 modifications. Seasalt carbonate is considered in equations relating to $\text{CO}_2(\text{aq})$ ionization, and therefore CO_2 uptake. Seasalt SO_4^{-2} is included in final SO_4^{-2} concentrations, although not in the category of sulfate produced.

Two chemical reactions are considered: they are the oxidation of S(IV) by O_3 and the oxidation by H_2O_2 (Hoffmann and Calvert, 1985):

$$-\frac{d[S(\text{IV})]}{dt} = (k_0[\text{SO}_2 \cdot \text{H}_2\text{O}] + k_1[\text{HSO}_3^{-1}] + k_2[\text{SO}_3^{-2}]) \times [\text{O}_3] \quad \text{Equation 3.12}$$

$$k_0 = 2.4e4 \text{ M}^{-1}\text{sec}^{-1} \text{ at } 298 \text{ K}$$

$$k_1 = 3.7e5 \text{ M}^{-1}\text{sec}^{-1}$$

$$k_2 = 1.5e9 \text{ M}^{-1}\text{sec}^{-1}$$

and

$$-\frac{d[S(\text{IV})]}{dt} = \frac{k[H^+][\text{H}_2\text{O}_2][S(\text{IV})]\alpha_1}{1 + K[H^+]} \quad \text{Equation 3.13}$$

$$k = 7.45e7 \text{ M}^{-1}\text{sec}^{-1} \text{ at } 298 \text{ K}$$

$$K = 13 \text{ M}^{-1}\text{sec}^{-1}$$

Catalytic oxidation reactions and gas phase chemistry are not considered.

3.2 Representative model runs and discussion

3.2.1 Bulk vs. explicit aerosol size representation

Use of the droplet distribution capability is demonstrated in a comparison study between bulk and explicit representation of cloud droplets. Oxidation of SO_2 to SO_4^{2-} 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 mass (see Table 3.2). In both the bulk and the explicit runs, a total of 217 droplets, all with diameter of $10\text{ }\mu\text{m}$, were used. In both runs, the average aerosol size was the same. However, in the bulk run, all droplets had aerosol of this diameter, while in the explicit run, three aerosol size categories were represented. The number in each category is determined by an approximation to a lognormal type aerosol distribution.

The purpose of the comparison study was to determine if bulk representation using averaged aerosol size can well represent the aerosol concentration dependent explicit representation. In the explicit run, a significant increase in the amount of SO_2 oxidized over that for the bulk run was found initially. The dependence on aerosol mass decreased with time. As time increases, there is less dependence on the rate of the reactions and more on limiting factors such as oxidant depletion and the adjustment of droplet pH to the increased sulfate.

Additional runs using larger drop size showed a decrease in sensitivity for the more dilute drops, as expected.

Table 3.2

	Bulk run	Explicit run
total number	217 drops	217 drops
droplet diameter	10 μm	10 μm
bin 1 aerosol diameter		0.0163 μm (number =50)
bin 2 aerosol diameter		0.163 μm (150)
bin 3 aerosol diameter		1.63 μm (17)
average aerosol diameter	0.696 μm (number = 217)	0.696 μm (number = 0)
S(IV) oxidation, %	1.892% after 10 seconds	2.280% after 10 seconds
	61.36% after 1 hr	66.36% after 1 hr

4. The Model : Regional Atmospheric Modeling System (RAMS)

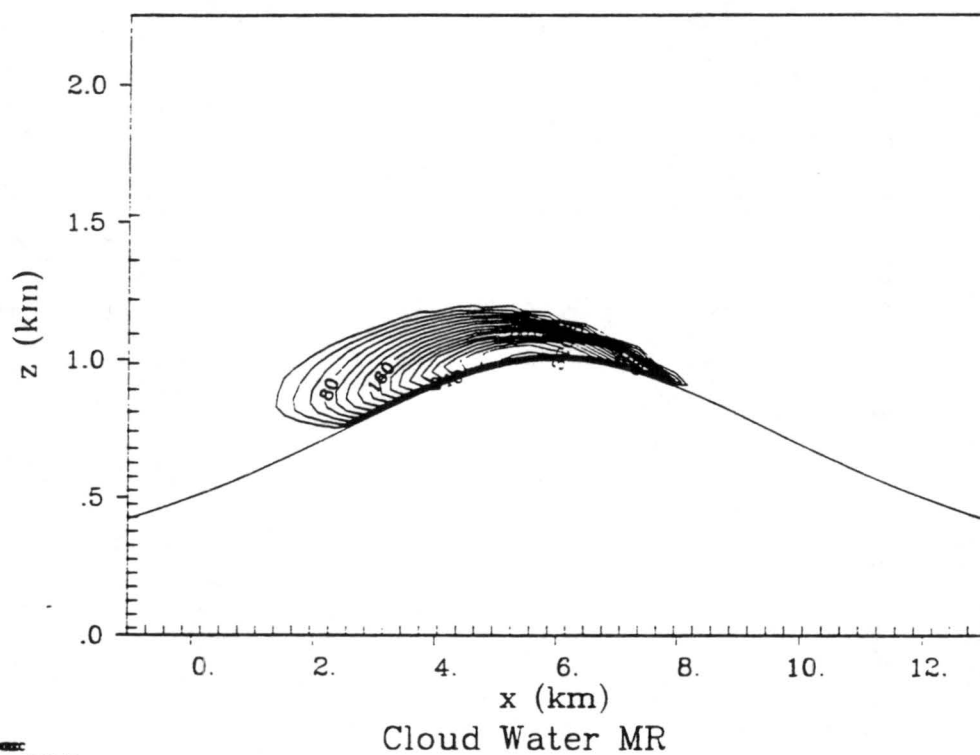
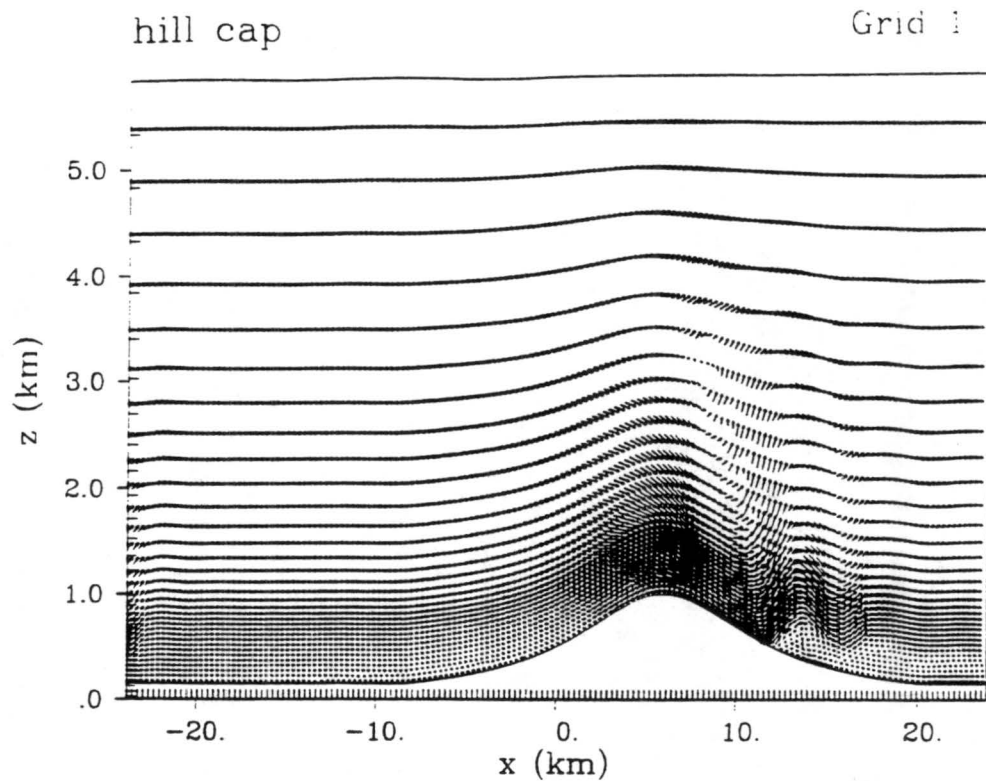
RAMS has been developed at Colorado State University. This model can be implemented as a large eddy simulation (LES) model in simulating a diverse array of mesoscale systems, including clouds of different forms (Pielke et al, 1992). This model has been adapted for use with the enhanced, solute following microphysics described in Chapter 4.2.

4.1 Dynamics

For the testing of this newly enhanced microphysics a simple two-dimensional hill top cloud was produced by initializing westerly winds flowing over a 1 km high hill in a stable atmosphere. Figure 4-1A is a schematic of the model domain. The model extent was 48 km in the west-east direction (x - direction) with a constant Δx of 300m. The vertical extent was approximately 6 km (z direction). Δz is constant at 50m from the ground through the depth of the cloud, and then is stretched at 115% for each z level above. Constant inflow winds of 5 m/s at the surface to 10 m/s at the upper boundary are input at the western boundary. The hill is located in the eastern half of the area to allow the inflow winds to equilibrate before the orographic forcing of the hill. Solar radiation is not used for the runs, making this a nighttime cloud. Coriolis forces are also not applied.

The nonhydrostatic option was used. For the upper boundary conditions, the Klemp-Durran (K-D) boundary condition is used, allowing internal gravity waves induced by the hill to propagate out of the top of the domain. Non-cyclic Klemp-Wilhelmson boundaries are used laterally, that is, normal velocity component at the lateral boundary is advected from the interior. To keep the winds at the inflow boundary constant, the phase speed of this advection from the interior is set at a speed smaller than the initial winds. Total water and aerosol distributions were also held constant at the inflow boundary.

After approximately 0.5 hours, the hill cap cloud has formed, and after 2 hours is relatively stable. Horizontal extent of the cloud is approximately 10 km, and vertical extent 4-5 km. For the bulk of the cloud, liquid water content is around 0.1 g water per kg of air. Airflow through and in the vicinity of the cloud is approximately laminar.



CONSEC
from .0000E+00
to .2000E-03
by .2000E-04
labels * .1000E+07

Figure 4-1: a: Wind vectors, full domain for 2-D hill cap cloud at time = 2 hrs. b: Cloud water mass mixing ratio r_i in kg/kg, time=2 hrs.

4.2 Microphysics

Earlier warm cloud versions of RAMS have included an option for parameterized microphysics (RAMS with level two microphysics). The simplest bulk parameterization computes liquid water by condensing all water above $RH = 100\%$; more complex parameterizations separate liquid water into cloud drops and rain drops (Cotton et al., 1986; Walko et al., 1995). An explicit microphysics module, which includes initiation activation of an aerosol spectrum and size dependent storage of water droplets, was recently developed and included in the RAMS model (Feingold et al., 1994). Water droplets were stored in 25 size categories or bins. The bins increased in size via mass doubling. The smallest bin (bin 1) had a lower size limit of diameter (d_w) of $3.125 \mu m$, which corresponds to a water mass, x_{w_1} , of $1.6 \times 10^{-11} g$, where x_w = water mass. The upper limit of this bin (which corresponds to the lower limit of bin 2) had mass $x_{w_2} = 2 \times x_{w_1} = 3.2 \times 10^{-11} g$. For all bins, $x_{w_{i+1}} = 2 \times x_{w_i}$. Mass doubling has been chosen because of its convenience in collision-coalescence calculations. A two moment scheme was used for the water bins, with a mass conserving scheme for all water mass bin-to-bin transfers due to condensation/evaporation, collision/coalescence or breakup processes (Tzivion et al., 1987; Feingold et al., 1988; Tzivion et al., 1989). The moments stored for each bin were the 0th mass moment, the number concentration in a bin, and the 1st mass moment, the mass concentration in a bin. Higher moments are calculated from these for the collision/coalescence scheme.

In the work of Feingold et al. (1995), aerosols were represented by number concentration of cloud condensation nuclei (CCN) in six bins, with limits determined by the minimum and maximum supersaturation needed to activate the CCN in each bin. The bins 1-6 were arranged in order of decreasing supersaturation needed for activation, bin 1 requiring the greatest supersaturation for activation, bin 6 the smallest. The initial distributions entered in these bins were obtained either from experimental data, where number concentration of activated particles were determined as a function of supersaturation, or from parameterizations which are fit to experimental data of this

type. The use of this type of bin structure precluded the need for information on the size and composition of the aerosol in each bin. When the ambient supersaturation was smaller than that of the upper limit of the bin, then no aerosol in that bin activated. When the supersaturation was larger than that of the lower limit of the bin, then all of the aerosol in that bin activated. For supersaturations lying within a bin, some fraction of aerosol from that bin was allowed to activate. When a portion of the aerosol were activated, that number of new cloud drops were formed, and the equivalent number of particles removed from the aerosol bin.

In this handling of the microphysics, the aerosol did not become solute, but was simply removed upon activation. Because this information was lost, it was impossible to know any properties of an aerosol regenerated to the atmosphere after droplet evaporation. Likewise, any aqueous chemistry effects could only be approximated using bulk chemistry (average drop sizes, average solute concentration), or based on some further assumptions. In reality, the soluble portion of activated aerosol becomes solute in the drops, leading to a spectrum of solute concentrations in cloud. The solute influences the rate of growth when solute concentration is relatively high, and may also modify rates of aqueous-phase chemistry. These effects depend strongly on droplet size and concentration (Ogren and Charlson, 1992).

For investigation of chemical effects and cloud processing, it is important to retain as much aerosol/solute information as possible. In this work, new aerosol and solute representations have been developed which can follow the aerosol mass throughout the evolution of the cloud: as aerosol, solute, and regenerated aerosol. The aerosol representation has aerosol composition and size information as its basis. The number of categories has also been increased to better follow the cloud processing of aerosol: as in the Feingold et al. (1994) version, this representation has a bin structure, but here, the bin limits are defined by particle size. For this initial study, use of a single composition aerosol has been imposed, either sodium chloride or ammonium bisulfate. Because of this, the aerosol bin limits can be interchangeably referred to as the mass limits for the bin or, when making an assumption of spherical aerosol shape, the diameter limits for the bin. For the limits in mass space, the lower limit of aerosol bin i is defined as x_{a_i} ,

which is the smallest mass value allowed for an aerosol in the bin and $x_{a_{i+1}}$, which is the largest mass value allowed for an aerosol in this bin. With knowledge of the aerosol density and an assumption of spherical shape for the aerosol, the diameter limits d_{a_i} and $d_{a_{i+1}}$ can be easily calculated, where these are the same bin limits in diameter space:

$$d_i = \left(\frac{6}{\rho\pi} x_i \right)^{1/3} .$$

Equation 4.1

4.2.1 Aerosol bin range and resolution

The choices of range of aerosol sizes, number of aerosol bins, and bin sizes, depend on the physics to be represented in the model and the aerosol distribution to be modeled. An aerosol distribution may, for example, have some significant number of aerosol smaller than some diameter d_c , but if these are too small to activate in the conditions of the model run, and impaction scavenging of these particles is not considered, it may be unnecessary to resolve the size distributions of these particles. One should be cautious, however. There may, for example, exist what appears to be some insignificant number of large aerosol, yet it is thought that these few particles can serve to initiate collision/coalescence processes that would otherwise not commence as quickly. When the model includes evaporation of drops formed by collision and coalescence, aerosol larger than that in the initial distribution can be regenerated into the aerosol spectrum, and care should be taken that the bin range encompasses these larger aerosol.

For the hill cap clouds modeled in this work, the supersaturations in preliminary runs did not exceed 0.2%. Because one goal of this model is to have the versatility to estimate cloud aerosol processing in a marine stratus cloud, the average supersaturation in Kogan's (1994) modeled stratus of 0.3% is also considered. For a sodium chloride (NaCl) aerosol having a diameter of $0.01\ \mu\text{m}$, the critical supersaturation (at average cloud temperature of $10\ ^\circ\text{C}$) was about 5%, and a $0.02\ \mu\text{m}$ particle has a critical supersaturation of about 2%. For the same diameter ammonium bisulfate ($(\text{NH}_4)\text{HSO}_4$) aerosol, the supersaturations required for activation were 6% and 2% respectively (see Table 4.1). Because it is possible to have pockets of high supersaturation in even a stratus cloud, the lower limit of the range has been set to $0.013\ \mu\text{m}$, which corresponds to a supersaturation of 3.2% for ammonium bisulfate aerosol. The initial number distribution used for these model runs is a log normal function whose geometric mean corresponds to a $0.1\ \mu\text{m}$ diameter particle, having a geometric standard deviation of 1.5, and a total number concentration of aerosol of $200/\text{cm}^3$ at 1 atmosphere. The number and mass mixing ratios are initialized constant

Table 4.1: Haze and equilibrium activation droplet sizes for ammonium bisulfate and for sodium chloride, 283 K

NH ₄ HSO ₄ :							
xa(g)	da(μm)	dropd(μm)	act_ss%	90%RH(μm)	95%RH	99%RH	100%RH
9.32E-19	1.00E-02	3.28E-02	4.74E+00	1.48E-02	1.63E-02	1.82E-02	1.89E-02
7.46E-18	2.00E-02	9.27E-02	1.68E+00	3.41E-02	3.94E-02	4.87E-02	5.35E-02
1.17E-16	5.00E-02	3.67E-01	4.24E-01	9.42E-02	1.14E-01	1.62E-01	2.12E-01
9.32E-16	1.00E-01	1.04E+00	1.50E-01	1.95E-01	2.41E-01	3.72E-01	5.98E-01
9.32E-13	1.00E+00	3.28E+01	4.74E-03	2.02E+00	2.54E+00	4.29E+00	1.89E+01
7.46E-12	2.00E+00	9.27E+01	1.68E-03	4.05E+00	5.10E+00	8.66E+00	5.35E+01
2.52E-11	3.00E+00	1.70E+02	9.12E-04	6.08E+00	7.65E+00	1.30E+01	9.83E+01
9.32E-10	1.00E+01	1.04E+03	1.50E-04	2.03E+01	2.55E+01	4.36E+01	5.98E+02
NaCl:							
xa(g)	da(μm)	dropd(μm)	act_ss%	90%RH(μm)	95%RH	99%RH	100%RH
1.13E-18	1.00E-02	4.14E-02	3.75E+00	1.80E-02	2.00E-02	2.28E-02	2.39E-02
9.07E-18	2.00E-02	1.17E-01	1.33E+00	4.08E-02	4.76E-02	6.03E-02	6.77E-02
1.42E-16	5.00E-02	4.63E-01	3.35E-01	1.11E-01	1.35E-01	1.97E-01	2.68E-01
1.13E-15	1.00E-01	1.31E+00	1.19E-01	2.30E-01	2.84E-01	4.44E-01	7.57E-01
1.13E-12	1.00E+00	4.14E+01	3.75E-03	2.36E+00	2.97E+00	5.03E+00	2.39E+01
9.07E-12	2.00E+00	1.17E+02	1.33E-03	4.74E+00	5.96E+00	1.01E+01	6.77E+01
3.06E-11	3.00E+00	2.15E+02	7.22E-04	7.11E+00	8.95E+00	1.53E+01	1.24E+02
1.13E-09	1.00E+01	1.31E+03	1.19E-04	2.37E+01	2.99E+01	5.10E+01	7.57E+02

xa=aerosol mass (grams); da=aerosol diameter (microns); dropd=equilibrium droplet activation diameter (microns); act_ss% = equilibrium activation supersaturation (in per cent); 90,95,99, and 100%RH=equilibrium haze diameter at 95,99, and 100% relative humidity sizes (microns).

with height. Initially, most of the aerosol particles lie between 0.01 and 1.0 μm in diameter, and less than 10⁻⁵ particles lie outside of this range. Based on this criterion alone, it is not deemed important to extend the range lower than 0.01 μm. In fact, a larger diameter for the lower limit may be in order. Impaction scavenging is not included in these runs as the aerosol mass scavenged by impaction scavenging has been shown to be orders of magnitude smaller than nucleation scavenging (scavenging by activation) (Flossmann, 1989; Barth, *et al.*, 1992). In these runs, the supersaturations are not expected to be very high, and there is no source of smaller aerosol (for example,

homogeneous nucleation, natural or anthropogenic primary sources, or breakup of droplets with subsequent evaporation). Therefore, any aerosol too small to be affected by activation need not be included, and bins small enough for these aerosol can be eliminated.

The upper limit of the aerosol bin range should be selected for the initial aerosol distribution to be represented and on the amount of collision/coalescence of cloud drops taking place, since these can subsequently evaporate during the model run and the solute returned to the aerosol spectrum. The effect of droplet collision/coalescence on the size of the regenerated aerosol can be shown as follows. A portion of the initial aerosol spectrum is activated. For simplicity, assume they each have formed on an average size aerosol of $0.1 \mu\text{m}$ in diameter. Assume these aerosol grow by vapor deposition to a monodisperse population of cloud droplets that are $10 \mu\text{m}$ in diameter. These then form larger drops by collision and coalescence. It would take 1000 of these to make a large cloud droplet of $100 \mu\text{m}$ and if this droplet evaporated, one particle of $1 \mu\text{m}$ would be regenerated. It would take 10^6 of the $10 \mu\text{m}$ cloud droplets to make a 1 mm small raindrop. If the drop were evaporated, a $10 \mu\text{m}$ particle would be regenerated. It becomes clear that the upper limit for the range of aerosol sizes accounted for will depend on the type of cloud and the range of cloud drop sizes formed by droplet collision/coalescence.

There are several considerations in deciding how to partition the aerosol bins within the selected range. An even distribution, where Δd_a (or Δx) is constant throughout the bins is one option. Another method is that of multiplicative bin limits, $x_{a_{i+1}} = c \times x_{a_i}$, where c is the constant of multiplication. For example, for a "mass doubled" bin structure, c would equal 2, and for "mass tripled", $c=3$. In these cases, Δx increases with increasing bin number. However, $\Delta(\log x)$, which is equivalent to $\log x_{a_{i+1}} - \log x_{a_i}$, will stay constant in log space.

$$\Delta(\log x) = \log x_{a_{i+1}} - \log x_{a_i} = \log(c x_{a_i}) - \log x_{a_i} = \log c \quad . \quad \text{Equation 4.2}$$

There are three advantages for using this latter structure in our model. First, for the lognormal initial distributions of aerosol used (see appendix) this allows for a more even distribution of aerosol mass and/or number in the bins. Second, mass doubling simplifies the mathematical formulation for collision and coalescence. Third, it shall be seen in the bin mapping section that this allows an easy transfer of activated aerosol into water droplet bins.

4.2.2 Activation and aerosol distribution in a bin

A new activation scheme has been developed for the explicit activation of aerosol, based on work of Köhler (1936). The critical mass, x_a^* , is the minimum mass aerosol that would activate under equilibrium conditions and is calculated as a function of the aerosol composition and the ambient temperature and supersaturation. All aerosol with mass greater than x_a^* activate, where

$$x^* = \frac{a^3 M}{29.0 \cdot (S^*)^2} \quad \text{Equation 4.3}$$

M is the molecular weight of the aerosol species, S is the ambient supersaturation, and a is part of the curvature term, numerically approximated by $a = 3.3 \cdot 10^{-5} / T$ [in cm], where T is the temperature in Kelvin. When x_a^* lies between bin limits x_{a_i} and $x_{a_{i+1}}$, it must be determined what portion of that bin will be considered activated : all, none, or part. This process of activation will therefore be important in determining aerosol bin structure. In the process of activation, aerosol leaves the aerosol distribution and is transferred to solute mass in the water droplets. In order to decide on a method of mathematically representing aerosol in a bin, a test was derived, and applied to four different bin representations.

Theoretically, a continuous aerosol distribution which activates at a given supersaturation would lose all of the aerosol above x_a^* to activated droplets. At later times, for equal or smaller supersaturation, no more aerosol should activate. In the model, the distribution of aerosol in the bin is unknown : only the prognostic variables of total aerosol mass and the total aerosol number are set. When x_a^* lies within a bin, it is not resolved what fraction aerosol in the bin would be larger than x_a^* . As with a true continuous distribution, it should hold that at later times, for equal or smaller supersaturations, no more aerosol should activate. A suitable test for the aerosol bin

representation might then be the repeated activation at a constant supersaturation. After the first timestep, no additional aerosol should activate.

In this work, four different bin structures were constructed and evaluated under the assumption of repeated activation at constant supersaturation. In the first formulation, all of the aerosol in a bin was assumed to be at some bin specific average mass. The moments are related by the following equation: $\overline{m}_{a,i} = \frac{M_{a,i}}{N_{a,i}}$. For this distribution, one prognostic variable is stored for each bin in what is deemed a "one moment" scheme, using either aerosol number, $N_{a,i}$, or aerosol mass, $M_{a,i}$. As only one variable for the aerosol has to be stored per bin, twice as many bins can be used for the same memory allotment in a two moment scheme, using both $N_{a,i}$ and $M_{a,i}$. In the tests reported here, $N_{a,i}$ was chosen to represent the aerosol and this bin structure is referred to as the "number only", or single moment bin representation. The remaining three bin structures tested were two moment schemes, and are referred to as "mass/number" bin representations.

For the number only representation, where all aerosol in a bin are of identical size, the number of aerosol activated at a given supersaturation are weighted according to where x_a^* falls between the bin limits. For example, if x_a^* is exactly halfway between $x_{a,i}$ and $x_{a,i+1}$, then half of the aerosol in the bin is considered activated. The number of aerosol activated is then: $N_{a_{act}} = \frac{x_{a,i+1} - x_a^*}{x_{a,i+1} - x_{a,i}} \times N_{a,i}$. The new mass in the bin $N_{a,i}(\text{next})$ will then be equal to $N_{a,i} - N_{a_{act}}$. When $x_{a,i} < x_a^* < x_{a,i+1}$, and therefor $x_{a,i+1} - x_a^* > 0$, repeated activation at the same supersaturation (therefore the same x_a^*) will tend to deplete the bin of aerosol: $N_{a_{act}}(\text{next}) = \frac{x_{a,i+1} - x_a^*}{x_{a,i+1} - x_{a,i}} \times N_{a,i}(\text{next}) > 0$. Also, for a given time period, shorter timesteps will cause a faster depletion of aerosol from the bin. Of course, after the initial activation at this supersaturation, there ideally should be no more activation and the aerosol should not deplete at all.

In the mass/number bin structures it is possible to assume some distribution of aerosol size within the bin. It is this possibility that encourages us to use this distribution at all. One distribution type is based on an assumption of linear distributions in both number and mass, such as that used for the water bins based on work by Tzivion et al., 1989 (see also chapter 4.2.4). The linear number and mass distributions as applied to aerosol are then:

$$f_n(x_a) = k \times x_a + b \quad \text{Equation 4.4}$$

$$f_m(x_a) = k' \times x_a + b' \quad \text{Equation 4.5}$$

where $f_n(x_a)$ is the number distribution as a function of aerosol mass, x_a , and $f_m(x_a)$ is the mass distribution function as a function of mass. The values k and b are the slope and intercept of the linear approximation to $f_n(x_a)$, and k' (not equal to k) and b' (not equal to b) are their respective counterparts for $f_m(x_a)$. The slopes and intercepts are determined by setting their integrals between the bin limits equal to the moments for number $N_{a,i}$ and mass $M_{a,i}$ respectively. The number and mass activated will equal the integrals of Equation 4.4 and 4.5 above integrated between x_a^* and $x_{a,i+1}$:

$$N_{a,act} = \int_{x^*}^{x_{i+1}} (kx + b)dx = \frac{1}{2}k(x_{i+1}^2 - x^{*2}) + b(x_{i+1} - x^*) \quad \text{Equation 4.6}$$

$$M_{a,act} = \int_{x^*}^{x_{i+1}} (k'x + b')dx = \frac{1}{2}k'(x_{i+1}^2 - x^{*2}) + b'(x_{i+1} - x^*) \quad \text{Equation 4.7}$$

Figure 4-2: Linear-linear bin distribution representations shows a simple pictorial representation of this linear-linear bin structure.

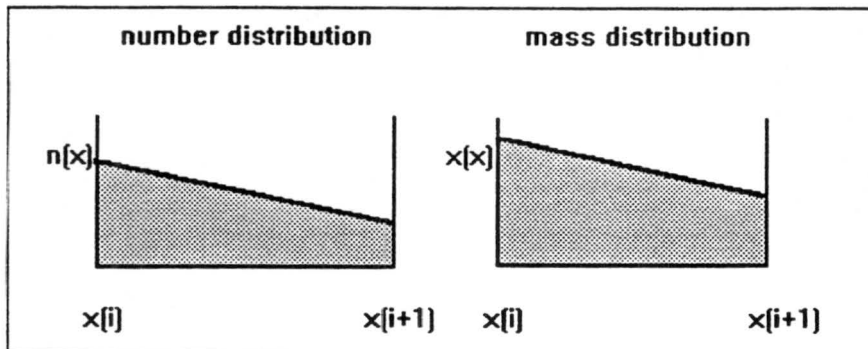


Figure 4-2: Linear-linear bin distribution representations

The problem with these linear distributions is that the physical relationship between the mass and number distributions, $f_m(x_a) = x_a * f_n(x_a)$, is only met at the bin limits, x_{a_i} and $x_{a_{i+1}}$. It is possible to find some x_a^* such that the activated aerosol number and mass actually represent some average sized aerosols $\bar{m}_{a_{act}}$ that are larger than $x_{a_{i+1}}$ or smaller than x_{a_i} : that is, the aerosol activated from bin i do not "fit" in bin i . It is also possible that the aerosols remaining are out of the bin bounds, $\bar{m}_{a_{rem}} < x_{a_i}$ or $\bar{m}_{a_{rem}} > x_{a_{i+1}}$.

A third method investigated assumes distributions in such a way as to preserve the physical relationship between the number and mass distributions. In this method, the distributions are:

$$f_n(x_a) = k * x_a + b \quad , \quad \text{Equation 4.8}$$

$$f_m(x_a) = x_a \times f_n(x_a) = k * x_a^2 + b x_a \quad , \quad \text{Equation 4.9}$$

$$N_{a_i} = \int_{x_i}^{x_{i+1}} (kx + b) dx = \frac{1}{2} k (x_{i+1}^2 - x_i^2) + b (x_{i+1} - x_i) \quad , \quad \text{Equation 4.10}$$

and

$$M_{a_i} = \int_{x_i}^{x_{i+1}} (kx^2 + bx) dx = \frac{1}{3} k (x_{i+1}^3 - x_i^3) + \frac{1}{2} b (x_{i+1}^2 - x_i^2) \quad . \quad \text{Equation 4.11}$$

These distributions have the problem that if the bin is heavily weighted to one side, that is if $\bar{m}_{a_i}$ is close to x_{a_i} or $x_{a_{i+1}}$, the distribution functions can be negative over part of the bin, even though the integrals over the whole bin are always positive. This is represented in Figure 4.3: Linear-nonlinear bin distribution representation.

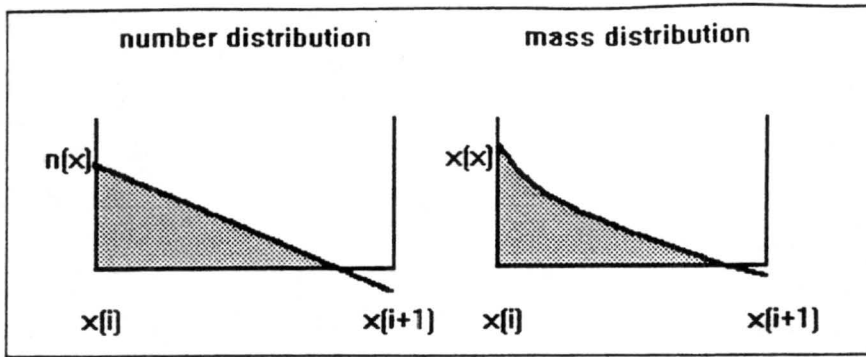


Figure 4.3: Linear-nonlinear bin distribution representation

In the case of activation, where integration between x_a^* and $x_{a_{i+1}}$ should give the amount of aerosol activated, it would be possible to arrive at negative values for activated mass and number when the negative portion of the distribution lies on the larger end of the bin. It is also possible to arrive at values for Na_{act} which are greater than the total mass and number in the bin. This can be easily avoided by setting activation to size zero if the integrated values are negative, or setting $Ma_{act} = Ma_i$ and $Na_{act} = Na_i$ when the calculated $Ma_{act} > Ma_i$ or $Na_{act} > Na_i$. In some ways this leads to interesting distributions: each essentially cuts off at some intermediate value. In the case of activation of a continuous distribution, this would be the case. However, because the number distribution is linear and the mass distribution is nonlinear, the cut off is not at the same position in the bin for the two distributions. Because of this, when x_a^* is in the vicinity of the cutoffs, the same problem as discovered in the previous (linear-linear) bin structure type can be found: Activated aerosol, and/or aerosol remaining in the bin does not necessarily lie within the bin limits.

The last bin structure type examined is the hybrid bin distribution introduced by Chen and Lamb (1994), and is similar to the linear-nonlinear distributions. However, in the case when some portion of the distribution functions are negative, the distribution functions are redefined in such a way that they only takes up a portion of the bin. For example, when the negative portion of the bin is at the high end of the bin ($f_n(x_{a_{i+1}}) < 0$), then the slope and intercept are defined such that:

$$f_n(x_{a_i}) = k \times x_{a_i} + b \quad \text{Equation 4.12}$$

$$f_n(x_{a_c}) = 0 \quad \text{Equation 4.13}$$

$$f_m(x_{a_i}) = k \times x_{a_i}^2 + b \times x_{a_i} \quad \text{Equation 4.14}$$

$$f_m(x_{a_c}) = 0 \quad \text{Equation 4.15}$$

where x_{a_c} is defined as the x cutoff value for x_a . In the remaining portion of the bin, the distribution functions are defined as zero everywhere. That is, for $x_{a_{i+1}} > x_a > x_{a_c}$ both $f_n(x_a) = 0$ and $f_m(x_a) = 0$. (See Figure). When the linear-nonlinear distribution functions are negative in the lower mass values (left side) of the bin, the hybrid distributions are defined in an analogous way to Equation 4.12 through 4.15, with the total mass and number in the bin being represented in the distribution functions between x_{a_c} and $x_{a_{i+1}}$. Because this method zeroes a portion of the bin when the average mass of the bin is near one of the limits, it helps to improve results for repeated activation at a constant supersaturation. In contrast, the linear-linear structure will always have non-zero distributions across the entire bin. One problem that can be accommodated easily is the existence of very steep slopes when the average mass lies near either bin limit. In this case, all of the bin aerosol can be activated when the average mass is greater than x^* , and none otherwise.

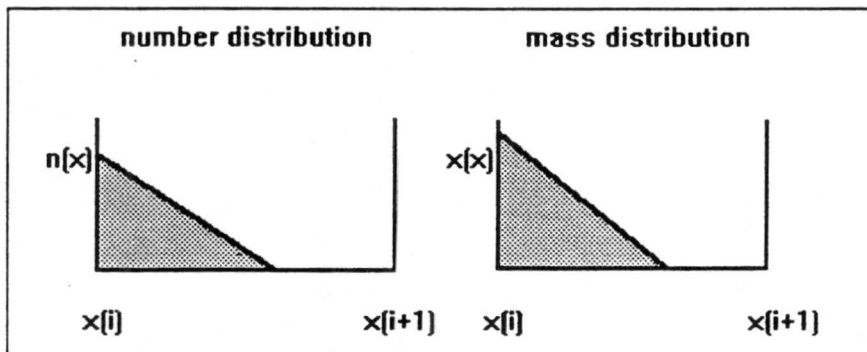


Figure 4.4: Chen and Lamb (1994) bin distribution representation

To test the four bin constructs for repeated activation, it is only necessary to examine the bin in which x^* resides, as all bins larger than this will activate completely, and all

bins smaller will have no activation. Because the number only structure needs only one saved variable ($M_{a,i}$ or $N_{a,i}$), as opposed to the other three bin structures which need two saved variables (both $M_{a,i}$ and $N_{a,i}$), a direct comparison of the bin structures should include, for the number only structure, twice as many number only bins as for the others. Therefore, two number only bins in the range of activations explored will be compared to one bin for the others.

The results of repeated activation can be seen in Figure 4.6. Figure 4.5 depicts the bin divisions assumed for repeated activation. Bin limits for the number/mass bin are x_{a_c} and $x_{a_{i+1}}$. Bin limits for the corresponding two number only bins are x_{a_c} , x_{a_c} , and $x_{a_{i+1}}$. Five positions across the bin(s) range from x_{a_c} to $x_{a_{i+1}}$ were selected by dividing the total linear range into sixths (see Figure 4.5).

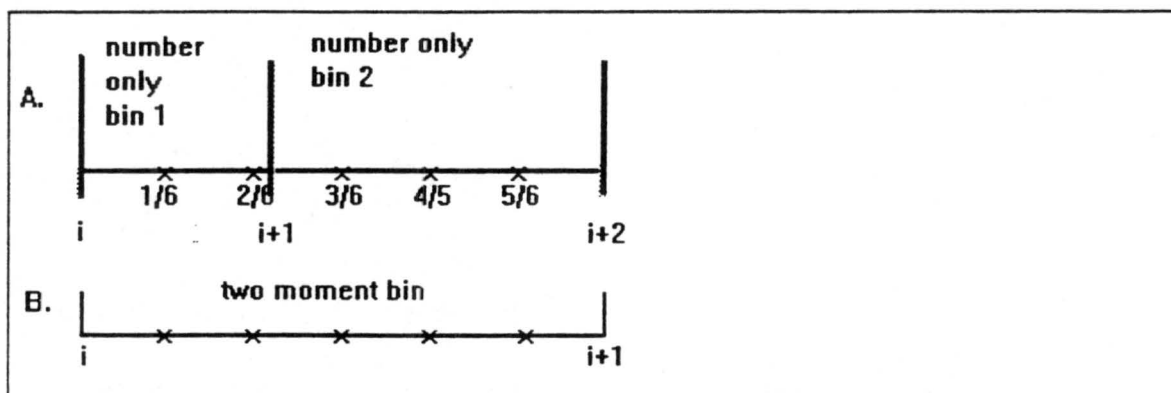


Figure 4.5: The test divisions for the one moment, number only bins (A) and the two moment bins (B).

Figure 4.6A, for example, represents repeated activation from position $x^* = x_{a_c} + 1/6$ range. The x-axis indicates the activation iteration, where iteration = 1 is the initial iteration, and iterations 2-10 are the repeated activations. The y-axis indicates the additional fraction of aerosol in the bin(s) which activated with subsequent iterations. Because x^* remains constant, this fraction should ideally remain at zero for all subsequent activations. However, it can be seen that there is rapid depletion of the unactivated aerosol in the bin in subsequent iterations. For three of the distributions tested, greater than 50% of the aerosol which remained in the bin after iteration 1 has been activated as of iteration 2. In contrast, use of the Chen and Lamb (1994)

distribution contained the additional activation to below 20%, even after 9 additional iterations.

Similar, though not always as pronounced results, can be seen in Figure 4.6A-E, for activations across the range of the bin. The Chen and Lamb (1994) distribution has therefore been chosen for representation of the aerosol mass and number distributions within a bin.

In summary, four separate schemes for describing the aerosol spectrum in size delineated bins were investigated. In one, aerosol in a bin was stored by number mixing ratio only, with all aerosol in a given bin at an average size. In the other three, a two moment method was used, storing aerosol in a bin by both number and total mass mixing ratios in the bin. Because the two moment storage method required two variables stored per bin as opposed to the one required for the number only method, it was necessary to allow twice as many number only bins for direct comparisons. A test of repeated activation at a constant supersaturation was invoked. Ideally, at a constant supersaturation there would be some initial activation of all aerosol larger than a critical mass (for one component aerosol), after which no more aerosol would activate. With all four aerosol distributions chosen, there was some continued activation with time. The distribution type chosen was that that would cause the least additional (unwarranted) activation. The two moment Chen and Lamb type distribution was chosen based on this criterion.

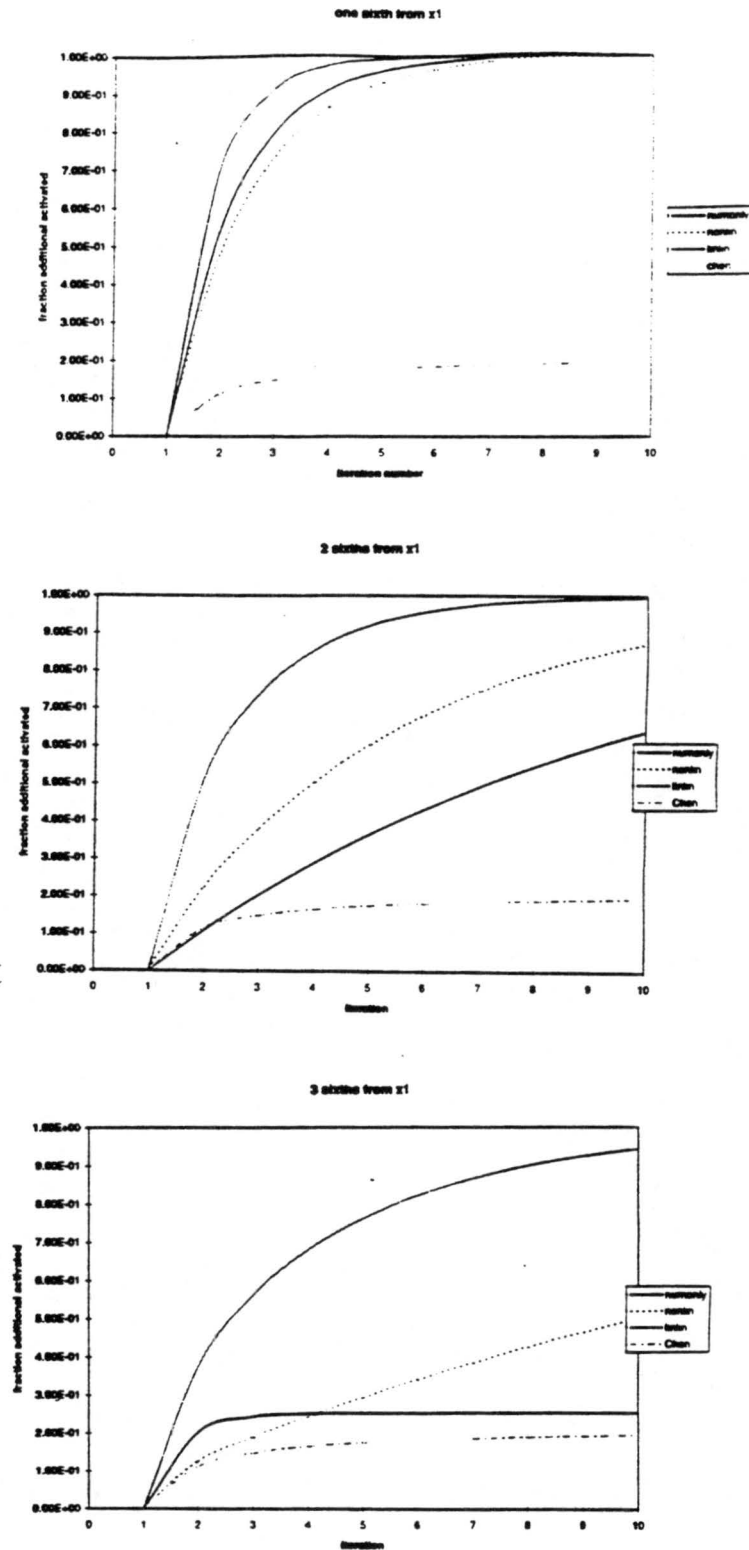


Figure 4.6: A,B, and C After initial activation, the fraction of additional aerosol that activates on further activation iterations at the same x^* . Number only bins, thick solid line. Linear-linear bin structure (see figure 4.2), thin solid line. Linear-nonlinear bin structure, dotted line. Chen and Lamb (1995) distribution, dot-dash line.

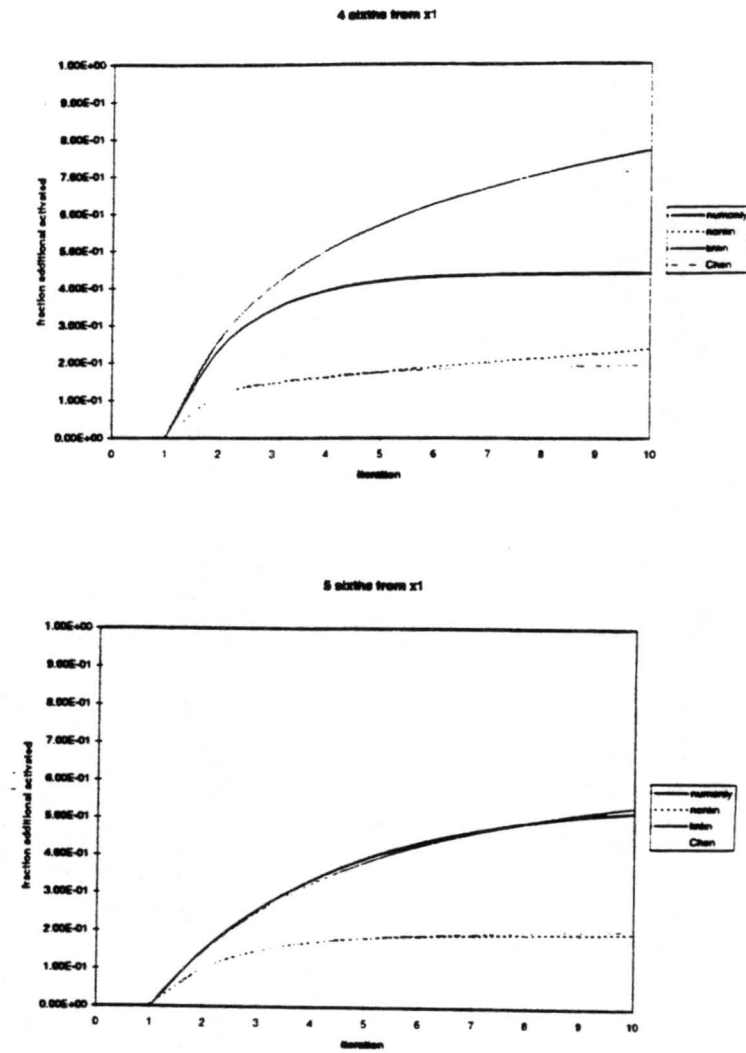


Figure 4.6 : D and E After initial activation, the fraction of additional aerosol that activates on further activation iterations at the same x^* .

4.2.3 Mapping: activated aerosol to water bins

The activation of aerosol produces new water droplets containing solute, where mass of the aerosol is converted to solute mass. In a microphysical model representing this process, the size of the newly formed water droplet must be determined. To a first approximation, each aerosol could be considered to be activated at its equilibrium activation droplet size, d_w^* , also called the critical water droplet size. However, equilibrium conditions are not often met in convective or orographically forced clouds. In addition, the d_w^* sizes for larger aerosol are inconsistent with typical observed cloud droplet sizes. For example, 1 and 2 μm NaCl aerosol have d_w^* of 28 and 80 μm respectively (Table 4.2). Droplets evolving from condensation processes alone would tend to be kinetically limited to an upper size limit of 10-20 μm , as there would be competition for the water vapor from other newly activated drops, and/or collision and coalescence of droplets would initiate and become the greater contributor to droplet growth. In the rare case that these larger haze sizes could evolve, they would have a very large fall velocity and be removed quite quickly.

Calculations based on the Köhler and growth equations can illuminate the rate dependence of haze growth. How quickly an aerosol can reach its equilibrium haze or activation size depends on the haze size at time zero, the solute concentration and composition, and on the total amount of forcing, which is greater for higher supersaturation and greater for a longer time spent in a supersaturated environment. For example, a 0.16 μm diameter ammonium sulfate aerosol will deliquesce to an equilibrium haze size of 0.55 μm at a relative humidity (RH) of 95%. It takes 28 seconds for the haze particle to then grow to its activation size d_w^* of 2.4 μm in a constant supersaturation field of 0.1%, at a temperature of 13 °C. Under the same conditions, a 0.35 μm ammonium sulfate aerosol at its 95% RH haze size of 1.2 μm will grow to its d_w^* of 7.14 μm in 58 seconds (Stevens, 1994).

Because the unreasonable equilibrium droplet sizes along with their slow rates of growth relative to the model microphysical timestep of 2s and to the time rate of change in supersaturation, it does not appear to be reasonable to immediately activate an aerosol to its d_w^* droplet size when the supersaturation reaches ss^* , the critical supersaturation for the aerosol.

One popular method for dealing with the activation of larger aerosol has been to choose one RH less than 100% (i.e., 95%) and activate aerosol larger than x^* at its equilibrium haze size for that RH. This somewhat alleviates the giant droplet problem, and takes into account rate limited growth of the haze (Roelofs, 1992; Flossman, 1989). The method adopted here is on that theme. It, furthermore, accounts for the disparity of deliquescence growth rates for the various size aerosol/haze (pre-activation).

At a given RH (where the RH is below that needed for activation), a larger aerosol will equilibrate to a larger haze size (for any internally mixed, soluble aerosol). The larger aerosol will have a slower rate of growth (in diameter space) because of the inverse dependence of the growth rate (dd/dt) on diameter. A method of generating activated water droplet sizes which can compensate for these different rates of growth is desired. These rate dependencies can be accounted for in a relative way by activating at a haze size corresponding to a lower RH for larger aerosol. For example, one might imagine aerosol of diameters 0.5, 1.5, and 3.0 μm . Instead of choosing the droplet size for the new droplets by equilibrium sizes at some RH, for example their 99% RH size, one could use the 99% RH size for the 0.5 μm aerosol, 98% RH size for the 1.5 μm aerosol, and 97% RH size for the 3.0 μm aerosol. These choices of RH are arbitrary, but the point is that in a relative way, they take into consideration the different growth rates for the different sized aerosol.

The bin structure and size limits for water droplet storage is already in existence, and for this preliminary code enhancement, will be accommodated. The lower diameter limit of the water bins is 3.125 μm , and the limits for the remainder of the bins are determined by mass doubling. (see Table 4.2). The divisions of choice for aerosol bins

would also be mass doubling or mass quadrupling, as noted in section 4.2.1. It would be most convenient if the aerosol bins could map directly into water bins on activation, that is, an aerosol bin that activates completely would create new water droplets that all fit into one water droplet bin. This would eliminate the need to integrate portions of the aerosol bin for mapping into more than one water bin.

Table 4.2: Mapping

Cloud temp=283.15K, aerosol density=1.78, mol. wt. =115.22

aer bin	xa(g)	da(um)	ddeq(um)	act_ss_ %	wat bin	xw(g)	dw(cm)	RHeq
1	2.07E-18	1.30E-02	4.88E-02	3.18E+00				
2	8.27E-18	2.07E-02	9.77E-02	1.59E+00				
3	3.31E-17	3.29E-02	1.95E-01	7.96E-01				
4	1.32E-16	5.22E-02	3.91E-01	3.98E-01				
5	5.30E-16	8.28E-02	7.81E-01	1.99E-01				
6	2.12E-15	1.32E-01	1.56E+00	9.95E-02				
7	8.47E-15	2.09E-01	3.13E+00	4.97E-02	1	1.60E-11	3.13E-04	1.00E+00
8	3.39E-14	3.31E-01	6.25E+00	2.49E-02	2	3.20E-11	3.94E-04	1.00E+00
9	1.36E-13	5.26E-01	1.25E+01	1.24E-02	3	6.39E-11	4.96E-04	1.00E+00
10	5.42E-13	8.35E-01	2.50E+01	6.22E-03	4	1.28E-10	6.25E-04	9.98E-01
11	2.17E-12	1.33E+00	5.00E+01	3.11E-03	5	2.56E-10	7.88E-04	9.96E-01
12	8.68E-12	2.10E+00	1.00E+02	1.55E-03	6	5.11E-10	9.92E-04	9.92E-01
13	3.47E-11	3.34E+00	2.00E+02	7.77E-04	7	1.02E-09	1.25E-03	9.84E-01
14	1.39E-10	5.30E+00	4.00E+02	3.89E-04	8	2.05E-09	1.58E-03	9.68E-01
15	5.55E-10	8.41E+00	8.00E+02	1.94E-04	9	4.09E-09	1.98E-03	9.37E-01

Aer bin = aerosol bin index, x_a = aerosol bin lower limit for mass, d_a = aerosol bin lower limit for diameter, d_{deq} (um) = lower bin limit for d^* (equilibrium conditions), act_ss_ % = lower bin limit ss, wat bin = water bin index, x_w = water bin lower limit for mass, d_w = water bin lower limit for diameter, RHeq = relative humidity corresponding to equilibrium haze size of d_w for the corresponding x_a .

The bin setup and mapping is accomplished as follows. The lowest water bin limit size is assumed to be the equilibrium activation size for some size aerosol which can be calculated by rearranging the equation for determining equilibrium drop size, d_w^* :

$$d_w^* = 2\sqrt{3b/a} \quad \text{Equation 4.16}$$

where $a \equiv 3.3 \times 10^{-5} T(cm)$, $b \equiv 4.3i \times x_{a,eq} / \mathcal{M} \text{ (cm}^3\text{)}$, T = temperature, i = Van't Hoff factor, and $\mathcal{M}$ = molecular weight of the solute. Rearrangement yields:

$$x_{a,eq} = \left(\frac{d_w}{2} \right)^2 \times \frac{a}{4.3i / \mathcal{M}} \quad \text{Equation 4.17}$$

This mass size is x_{a,eq_1} or the aerosol equivalent mass for water bin limit x_{w_1} . It should be noted that this equation has both temperature and aerosol component dependence. An average cloud temperature is chosen for use throughout the model, as one standard bin structure will be created for a model run. The aerosol distribution is then created about x_{a,eq_1} , using the desired range limits and the desired multiplicative relationship (aerosol mass doubling or quadrupling). Any restrictions pertaining to the number of aerosol bins, for example, limited memory space allocated, must also be considered. In this application, aerosol mass quadrupling has been applied, enabling encompassing the entire range desired in 14 aerosol bins. Here, six of the aerosol bins are smaller than x_{a,eq_1} , and eight are greater. The lower limit of aerosol bin 7 has mass defined by x_{a,eq_1} , that is, $x_{a_7} = x_{a,eq_1}$. The larger aerosol bins, bins 7-14, will map (activate) directly into water bins 1-8. That is, all aerosol activated from aerosol bin 7 activate into water bin 1. The water droplet size for new droplets is taken as some "average" size for the water bin. This is considered to be the log average of the water bin limits, where

$$\log(x_{w_{i,avg}}) = \frac{1}{2} (\log x_{w_i} + \log x_{w_{i+1}}).$$

Aerosol in aerosol bin 8 activate into water bin 2, and the other larger aerosol bins activate similarly.

The aerosol in bins smaller than x_{a,eq_1} will all activate directly into water bin 1. The droplet size chosen for activation of these smaller aerosol is at the "lower end" of water bin 1, specifically at $1.1 \times x_{w_1}$.

In summary, haze sizes are used for larger aerosol instead of unrealistic equilibrium activation droplet sizes. The relative humidity used for each bin haze size takes into account in a relative way the slower rate of condensational growth on larger particles.

4.2.4 Solute transfer ("advection") between water bins

In a given water bin, i , the stored variables in the pre-existing RAMS explicit microphysics are $M_{w,i}$, the total water mass in bin i , and $N_{w,i}$, the total droplet number in bin i . An additional variable is created for storage of total solute mass in the bin, $M_{s,i}$. With these values, one can determine the average droplet water mass,

$$\overline{m}_{w,i} = \frac{M_{w,i}}{N_{w,i}} ; \text{ the average solute concentration in the bin (and also in each drop),}$$

$$\overline{c}_{s,i} = \frac{M_{s,i}}{M_{w,i}} ; \text{ and the average solute mass per drop, } \overline{m}_{s,i} = \frac{M_{s,i}}{N_{w,i}} .$$

The average solute mass per drop can be thought of as a "virtual aerosol", since this would also equal the mass of an aerosol generated on evaporation of one of these average drops, $m_a = \overline{m}_{s,i}$. Although the average values above are all that can be recovered with confidence, it would be valuable if reasonable distribution functions of the water mass and solute mass could be constructed for each bin. Linear distributions for water mass and number can be formed based on the method of Tzivion et al. (1989), and have been implemented in RAMS explicit microphysics. A method of distributing the solute in a bin should be compatible with this existing format.

The linearized number and mass distributions in a water bin are then:

$$f_n(x) = k \times x + b \quad \text{Equation 4.18}$$

$$f_m(x) = k' \times x + b' \quad \text{Equation 4.19}$$

where x is water mass of a droplet, and the distributions are represented in mass space. The values k and b are the slope and intercept of the linear approximation to $f_n(x)$, and k' (not equal to k) and b' (not equal to b) are their respective counterparts for the $f_m(x)$.

An assumption is made that at the bin limits, x_i and x_{i+1} , the value for the mass distribution function is $x \cdot f_n(x)$. This follows from the definition of a mass distribution function, and are the only points on the line where the relationship is not an approximation, but the true relationship. At the bin limits, values for $f_m(x)$ are then:

$$f_m(x_i) = x_i \times f_n(x_i) \quad \text{Equation 4.20}$$

$$f_m(x_{i+1}) = x_{i+1} \times f_n(x_{i+1}) \quad \text{Equation 4.21}$$

In order to determine the slope and intercept of each of the two distribution functions, the equations are integrated over the extent of a bin from bin limits x_i to x_{i+1} and the integral set equal to the known values, $N_{w,i}$ and $M_{w,i}$, respectively, where

$$N_{w,i} = \int_{x_i}^{x_{i+1}} (kx + b)dx = \frac{1}{2}k(x_{i+1}^2 - x_i^2) + b(x_{i+1} - x_i) \quad \text{Equation 4.22}$$

and

$$M_{w,i} = \int_{x_i}^{x_{i+1}} (k'x + b')dx = \frac{1}{2}k'(x_{i+1}^2 - x_i^2) + b'(x_{i+1} - x_i). \quad \text{Equation 4.23}$$

These four equations can be solved to arrive at values for k , b , k' , b' , and then any portion of a bin's mass or number determined by integration over the distribution functions between the desired mass (x) limits.

$$k = \frac{f_n(x_{i+1}) - f_n(x_i)}{x_{i+1} - x_i} \quad \text{Equation 4.24}$$

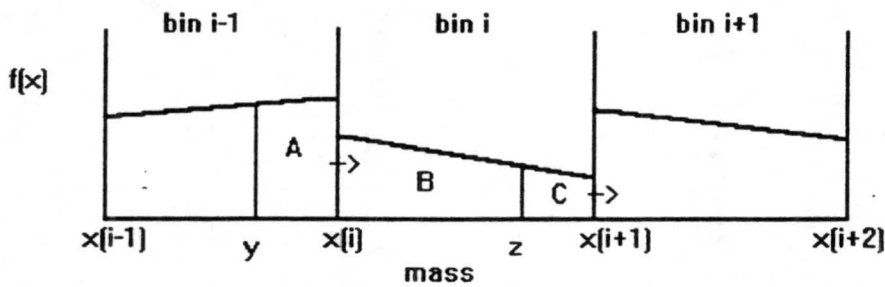
$$b = f_n(x_i) - kx_i \quad \text{Equation 4.25}$$

$$k' = \frac{x_{i+1}f_n(x_{i+1}) - x_if_n(x_i)}{x_{i+1} - x_i} \quad \text{Equation 4.26}$$

$$b' = x_if_n(x_i) - k'x_i \quad \text{Equation 4.27}$$

An example of the use of the water number and mass distributions is in the handling of condensation and evaporation in the RAMS code. Consider a water bin, i , having some $N_{w,i}$ and $M_{w,i}$. During condensation, the droplets in this bin will grow by vapor deposition. Some portion of these droplets might be expected to undergo a size increase large enough to cause them to no longer fit within the bin limits, but to belong in a larger water bin. If we were to only use an average drop size (no distribution information), all drops in a bin would reach this size simultaneously and the bin would empty. However, using a distribution allows growing only the drops above a certain size along the distribution to grow into the next bin, while the remaining drops simply grow to sizes remaining in bin i 's limits.

Figure 4.7: Bin to bin condensational advection of water



$f(x)$ can represent the number or mass mixing ratio distribution. See text.

The advection of water droplet number in bin (mass) space, and the advection plus growth of water mass were handled in the following way. For a given bin i , between limits x_i and x_{i+1} , a new set of mass limits were calculated, y and z . These are the limits that delimit all of the drops that will grow (or condense) into the limits x_i , x_{i+1} during a given timestep, Δt . Take for example the process of condensation. In the timestep, droplets of size y will grow to size x_i . Droplets of size z will grow to size x_{i+1} (see Figure 4.7). To determine the total number mixing ratio of droplets in bin i at $t+\Delta t$, the number distribution functions are integrated between y and z and would consist of A

and B in Figure 4.7, assuming $f(x)$ to be the number distribution function. The integrated number in C would move from bin i to bin $i+1$. To determine the total mass mixing ratio of droplets in bin i at $t+\Delta t$, the mass distribution functions are adapted for condensational growth and integrated between y and z . Assuming $f(x)$ to be the mass distribution function in this case, the resulting mass mixing ratio would consist of A and B in Figure 4.7 plus associated mass from condensational growth. The integrated number in C plus additional mass from condensational growth would move from bin i to bin $i+1$.

It may be possible to create a third linear equation, this one for the solute distribution, and solve this in conjunction with those for the water mass and droplet number. However, some assumption would have to be made about the relationship between the solute mass distribution and one of the other distributions. The assumption made for the mass distribution values at the bin limits (Equations 4.20 and 4.21) has a physical basis, as these values are related by definition. However, there is no analogous definitional relationship for the solute distribution. An intuitive relationship might be between droplet number and solute mass: five droplets will probably have more solute than 1 droplet. One method of weighting solute by some property of the water distributions might then be a simple weighting by number.

There is also incentive to consider weighting solute with water mass. Flossman (1989) and Roelofs (1992) have determined the general relationship associating the solute mass with the water mass. This association is also verified in observations: larger water droplets tend to have a greater solute mass than smaller water droplets (Ogren et al., 1989). There is no convenient way to generally quantify this relationship, as it would depend on too many factors such as solute composition and droplet history. However, weighting the solute mass transferred across bin boundaries by water mass transferred in direct proportion is also considered.

Simple schemes for weighting solute by either water droplet number (Graham Feingold, personal communication) or mass have been developed and tested for consistency in the stand-alone version of the microphysical module. The amount of

solute mass to be transferred from one bin to another during vapor deposition or evaporation processes is be scaled to the amount of water being transferred. For number weighting, the scaling is to the ratio of the number of droplets transferred (or "advected" in mass space) to the total number of droplets in the bin:

$$M_{s,adv} = M_{s,i} \times \frac{N_{w,adv}}{N_{w,i}} \quad \text{Equation 4.28}$$

where $M_{s,adv}$ is the mass of solute to be advected across the bin limit, and $N_{s,adv}$ is the number of drops to be advected across the bin limit. In the condensational growth example of Figure 4.7, $M_{s,adv}$ or $N_{s,adv}$ can be equated to A being advected into bin i.

Equation 4.28 can be rearranged to give

$$M_{s,adv} = N_{w,adv} \times \frac{M_{s,i}}{N_{w,i}} = N_{w,adv} \times \bar{m}_{s,i} \quad \text{Equation 4.29}$$

where $\bar{m}_{s,i}$ is the average solute mass per drop (virtual aerosol size) for the bin. The mass of solute transferred is then effectively the number of drops transferred times an average solute mass per drop. Each drop transferred from the bin would have the same size virtual aerosol, no matter what the size of the drop. This would result in drops on the larger end of the bin (near x_{i+1}) having less solute than expected, and drops in the smaller end of the bin having more solute than expected.

For mass weighting, the scaling of solute mass is to the ratio of the water mass of droplets transferred to the total water mass in the bin:

$$M_{s,adv} = M_{s,i} \times \frac{M_{w,adv}}{M_{w,i}} \quad \text{Equation 4.30}$$

where $M_{w,adv}$ is the mass of drops to be advected across the bin limit. This can be rearranged to give

$$M_{s,adv} = M_{w,adv} \times \frac{M_{s,i}}{M_{w,i}} = M_{w,adv} \times \bar{c}_{s,i} \quad \text{Equation 4.31}$$

where $\bar{c}_{s,i}$ is the average solute concentration in the bin. In this case, the solute concentration of all drops advected will be the same, no matter what the size of the drop. Although there has shown to be a tendency of increased solute mass with increased solute size, there has also been noted a tendency for smaller drops to have a greater concentration of solute than larger drops. So, in the case of solute weighting, the amount of solute in larger drops in the bin may well be over-predicted, while under-predicting solute in the smaller drops.

This, however, might not be so bad. Although it is not always the case, in most instances the average amount of solute per drop will increase with increasing water bin size. During condensation, droplets with smaller solute aerate mass are transferred to a bin where the average solute mass is larger. At that point in time, they are mixed to some intermediate size. Although some mixing is "real" there is a tendency to smear or smooth out size definition during the advection in mass space. Some over-prediction of solute mass in drops advected into larger bins (during condensation) may help to counterbalance that mixing. During evaporation, under-prediction of solute in the smaller drops in a bin may partially compensate for the analogous evaporational smoothing. In this case, drops from a bin where the average solute size is larger are transferred to a smaller mass size bin.

In order to determine which scheme will best transfer solute in a given cloud, tests must be developed which can mimic the vapor deposition processes that will be experienced by the droplets. Forcings will include both the supersaturations experienced as well as the time spent in a supersaturation and/or subsaturation field.

A simple spike regeneration is used in the comparisons of mass and number weighting schemes. In this, any droplets plus solute which evaporate to a size smaller than the lowest water bin limit are considered completely evaporated. The solute is returned to the aerosol spectrum at its average size while water is returned as vapor. Nine separate box-model runs based on different levels in the hill cap cloud, and therefore different thermodynamic histories, were used for the comparisons. These are the hybrid

LES/parcel runs described more fully in Chapter 4.3. In these runs, the mass mixing ratios of aerosol/solute and total water are kept constant, and the processes allowed are activation, condensation, evaporation, and aerosol regeneration only. Initially the aerosol mass is represented as aerosol only and no droplets are present. After the runs completion, there is complete evaporation of all droplets, such that the aerosol is once more represented as dry aerosol only. Because there is no grid-to-grid or parcel-to-parcel mixing, and because collision/coalescence processes are not allowed, the final aerosol distribution would ideally be identical to the initial distribution.

The deviation of the final from the initial aerosol number and mass mixing ratio distributions, is quantified for each run by calculation a sample standard deviation, s , for the 14 aerosol bins, calculated from the "true" or initial values. The deviation for the number and mass mixing ratio distributions (s_N and s_M) are then:

$$s_N = \frac{\left(\sum_{i=1}^{14} (N_{i,final} - N_{i,init})^2 \right)^{1/2}}{14 - 1} \quad \text{Equation 4.32}$$

and

$$s_M = \frac{\left(\sum_{i=1}^{14} (M_{i,final} - M_{i,init})^2 \right)^{1/2}}{14 - 1} \quad \text{Equation 4.33}$$

In both cases there is a great deal of computational mixing of the activated aerosol/solute. However, the mass weighted solute transfer scheme was found in all cases to have a much lower standard deviation than the number weighted scheme. The first two examples in Table 4.3 shows averaged results for s_M and s_N for a series of eight parcel runs using the number weighting scheme and the mass weighting scheme. The eight parcel runs are based on trajectories through the hill cap cloud, and are discussed more fully in Chapter 5. Ideally, the initial and final aerosol concentrations would be identical, and values for s_M and s_N would be 0. The smaller average s_M and s_N values for the mass weighting vs. number weighting infers a greater agreement between initial

and final aerosol distributions. Additional tests using sinusoidal and step function supersaturation curves for input also support these results, and in all cases the mass weighting reduced the amount of solute averaging occurring between drops. A method of compensating for this mixing is discussed in the following section.

Table 4.3: *Deviations of final to initial aerosol distributions.*

run	weight	regen	σ	average s_N	average s_{N^*}
1	number	spike		17610	.1317e-9
2	mass	spike		10368	.6436e-10
3	mass	lognormal	1.3	6526	.4203e-10
4	mass	lognormal	1.4	5077	.2816e-10
5	mass	lognormal	1.5	4604	.1637e-10
6	mass	lognormal	1.6	5075	.8504e-11
7	mass	lognormal	1.7	5994	.1031e-10

8 parcel runs were used in determining s_M and s_N averages. These correspond to parcel-SB representations for trajectories 1-8 as described in chapter 5.

In summary: once the aerosol is activated, it then exists as solute in the droplet spectrum. As droplets undergo condensational growth and evaporational reduction in size due to environmental super and subsaturations, the droplets are considered to be advected through mass space. As they advect in mass space the solute must follow along with the water droplets. Two methods of correlating the advection of solute with water were investigated. In one method the solute was weighted with the number of droplets advected, effectively advecting the average solute mass per drop multiplied by the number of droplets advected. In the other, the solute was weighted with the droplet mass advected, effectively advecting the average solute mixing ratio (with droplet water mass) multiplied by the water mass advected. In the simple cases studied (hill cap cloud, sine wave and step function supersaturation trials) the mass weighting scheme always produced less computational mixing of solute between water bins. This will not necessarily be the case in more complex situations: the distribution of solute mass with drop size in one water bin could well be quite elaborate. For example, the entrainment of larger aerosol in an area of cloud with existing droplets formed on smaller aerosol could result in a distribution that was the inverse of the

expected relationship of larger drops associated with larger solute mass. Because of lack of complete information, some general rule must be assumed, and the mass weighting of solute is chosen for solute advection in the droplet spectrum.

4.2.5 Aerosol Regeneration

During condensation and evaporation, there is computational mixing of solute concentrations in the water bins. In condensation, water plus solute from a water bin below (i-1) typically having an average solute mass per drop smaller than that of the destination bin (i) is mixed into the destination bin. This "distribution information" is immediately lost, as only the total solute mass mixing ratio in the bin is stored, and only the average solute mass per drop is known. During evaporation, the process is reversed, and water plus solute from a water bin above (i+1) typically having an average solute mass per drop larger than that of the destination bin (i) is mixed into the destination bin. Overall, in the course of both condensation and evaporation, some distribution of solute mass per droplet including some smaller than and larger than the average size represented can be imagined, although the information about the particulars of that distribution is lost.

Complications due to mixing are compounded when there is grid to grid mixing, where drops from neighboring grids contained in a certain bin size and having different histories and different solute concentrations are mixed. There is certainly some distribution that cannot be represented by the few values stored.

One method of compensation for this mixing and averaging of information is to assume some distribution about the average of solute per drop in the bin. Upon evaporation of water droplets in one bin, the aerosol regenerated is assumed to be at some distribution about the average instead of all at the average. The distribution type chosen for regeneration was a lognormal. This method of regeneration of aerosol was tested against a simple "spike" (no distribution, average value only) regeneration. One cycle

of activation, condensation, evaporation, and regeneration was simulated: no other processes (i.e. collision/coalescence) were allowed. An initial aerosol distribution should in this case be exactly returned at the cycle's completion. In full LES model runs, box model runs, and LES/parcel hybrid model runs the lognormal regeneration scheme gave far superior results. One would assume that the breadth of the distribution used in aerosol regeneration might be dependent on many factors including the amount of in-cloud time and the overall amount of forcings encountered by aerosol in its processing. Different choices of the lognormal standard deviation do have an effect on reproducibility of the initial spectrum in these tests, with a broader distribution giving better reproducibility for the runs with longer time in-cloud.

Testing of regeneration schemes using the hybrid LES/parcel model illustrate both the general applicability of the lognormal regeneration and the dependence of the scheme parameters on the thermodynamic environment (overall forcing). The simple spike regeneration described in the previous section is compared to a lognormal generation using several values for the lognormal sigma (see appendix A). Average standard deviations of initial to final aerosol distributions can be calculated to quantitatively compare the effects of using the different regeneration schemes. In Table 4.3, run number 2 has been performed using spike regeneration, while runs 2-7 use the lognormal regeneration. In all cases, the use of the lognormal regeneration scheme results in lower average standard deviations.

The difference in resulting aerosol spectrum can be seen in Figures in 5-5 and 5-6. These figures are described more fully in Chapter 5.2. However, the bottom left hand plot in each of the figures shows a final aerosol distributions in mass and number, where the lognormal distribution with $\sigma=1.5$ was (solid line) and was not (dotted line) applied. For comparison, initial distributions are in the first row for each figure. When the lognormal distribution is not applied, the aerosol returns over a much narrower spectrum, illustrating the extent of solute mixing in the water bins. Use of the lognormal simply distributes this aerosol over a broader spectrum.

4.3 Hill Cap Cloud with Hybrid LES/Parcel model: model runs

In the full RAMS LES model used, the water droplets in the bins are subject to transfer between grids due to advection, sedimentation, turbulence, and precipitation processes. In addition, within one grid there can be water mass transfer between bins as a result of condensation and evaporation, collision-coalescence, and breakup processes. A comparison of the full RAMS Eulerian-type model run with a corresponding parcel or Lagrangian model run can help to identify the different effects on the model predictions due to inter- and intra-grid effects. It is expected that the grid-to-grid mixing should influence the aerosol and water distributions through diffusion, turbulent mixing, entrainment, and detrainment processes. Additional distortions due to computational effects may be encountered.

A trajectory-based method of directly comparing the Eulerian and Lagrangian models is used. During an Eulerian run, a number of parcels can be initialized ("released") and allowed to flow with the wind. Their trajectories can be determined, and the time dependent thermodynamic conditions experienced by those parcels compiled. These thermodynamic quantities can then be used as input to stand-alone Lagrangian parcel model runs. The parcel runs can include some microphysical processes (evaporation/condensation, collision/coalescence) but not directly others (grid-to-grid interactions such as drizzle, entrainment, and detrainment).

Trajectories can be determined by two methods. In the first method, a "snapshot" of the cloud can be saved for some instantaneous point in time, and the parcel is assumed to be driven in space over some period of time by those instantaneous wind vectors. The wind vectors at the grid points surrounding the parcel are used to determine a weighted average wind vector for the trajectory. The stored thermodynamic variables along the trajectory are used to drive the stand alone model. The second method of determining a trajectory is to do so interactively as the model run progresses. At some initial timestep, a parcel is "released" at some point in space. The simulated wind vectors at the grid points in this model surrounding the parcel are used

to determine a weighted average wind vector that operates on the parcel for the timestep. The thermodynamic variables are also determined by weighted averaging and stored. The average wind vector is used to update the parcel position, and the process is repeated at the next timestep.

If the dynamics of the cloud are in a steady state, the two methods are equivalent. However, in an LES type model the cloud evolves much in the same way a "real" cloud does, and a trajectory determined by a snapshot of the evolving cloud would not be the same as that determined in the time-dependent method. Even in the hill clouds of these simulations, in which the cloud does settle into approximately steady state with regard to maximum liquid water content in a grid and maximum wind speeds, the cloud never stops changing moment by moment. The time dependent method has been chosen for the parcel runs here.

A parcel trajectory module (Stevens, 1995b) was adapted to be compatible with RAMS simulations using topography. This module both recovered the thermodynamic variables required, and wrote out aerosol and water droplet distributions designated spatially or temporally as the model progressed. In this application, nine time-evolved parcel trajectories were determined for each model run. The parcels are initiated in a model run after two hours of simulation time has elapsed, to allow for equilibration of winds and formation of the cloud in the domain. Initial positions for eight passive tracers were approximately 1 km upwind of the cloud (at $x=-1000$ m), and distributed vertically at 100 m intervals (see Figure 4.8). At each 2 second timestep following parcel initiation, the thermodynamic variables are recovered for each parcel's environment and stored for use in the stand-alone Lagrangian parcel runs. Variables recovered were time, wind vectors, spatial coordinates, total water mixing ratio, r_t , liquid water potential temperature, θ_l , and pressure, P . In addition, both total liquid water and supersaturation along the trajectory were recovered for comparison to parcel model runs.

The aerosol and water droplet distributions are collected at 1 km horizontal intervals along the trajectory paths. These distributions, computed in the full Eulerian LES run,

were used for direct comparison to the analogous distributions determined from the Lagrangian parcel model runs, parcel-SB and parcel-LA, at the corresponding times and positions.

The hill cap cloud described above was run with a RAMS model run utilizing both explicit solute following microphysics (the RAMS+ simulations) and a RAMS "level two microphysics" with parameterized microphysics (RAMS2). In the RAMS2 simulations, a simple microphysical parameterization is used in which the presence of cloud and liquid water is designated as any position where the relative humidity goes above 100%. Differences between the results of the parcel-SB run based on RAMS+ and RAMS2 simulations are compared, and differences found to be minor for this cloud simulation.

In summary, four model runs are directly compared.

1. RAMS+
2. Parcel-SB model driven by trajectory thermodynamic profiles from RAMS+ simulations
3. Parcel-LA model driven by trajectory thermodynamic profiles from RAMS+ simulations
4. Parcel-SB model driven by trajectory thermodynamic profiles from RAMS2 simulations

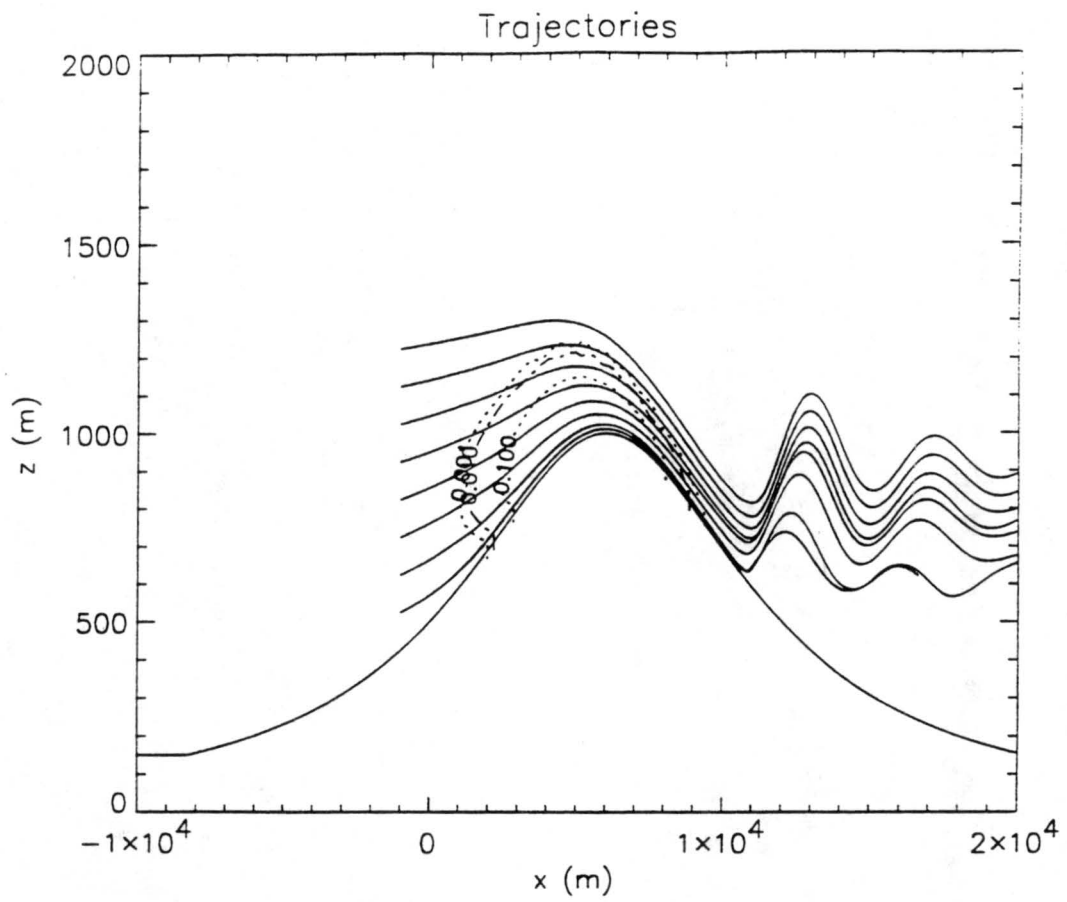


Figure 4.8: Hill cap cloud with trajectories 1-8.

5. Results of Hill Cap Cloud Model Runs

5.1 *Thermodynamics in model runs*

As described in Chapter 4.3, eight passive tracers are initiated in pre-cloud air during the RAMS+ simulation. These tracers flow with the winds and are used to determine time and space dependent thermodynamic profiles along each of their trajectories. The spatial position of the passive tracers can be initialized at any x, z position for the 2-d representation. The thermodynamic variables in RAMS+, however, are defined only at gridpoints at stationary x, z positions. A method must be defined to assign a wind vector to the tracer position.

To update the tracer position each timestep, wind vectors at the momentum gridpoints surrounding the passive tracer position are used to drive the tracer. The surrounding wind vectors are weighted according to their proximity to the tracer position. The weighted average wind vector at the tracer position, a velocity in meters per second, is then used to update the tracer position during the timestep.

When identifying the thermodynamic values to associate with each tracer position along a given trajectory, two techniques can be envisioned. In one method, the procedure is to use the grid-averaged thermodynamic values, as it is these values that drive the microphysics in the RAMS+ simulations. In this case, the one closest thermodynamic grid point to the tracer position is used to associate thermodynamic values with the tracer trajectory. These profiles, based on grid-average values, will be designated RAMS+. In Figure 5-1 to 5.2, A designations correspond to the RAMS+ simulation thermodynamic profiles. In an alternative method, the procedure is to take the weighted average of the thermodynamic values at the surrounding gridpoints, which is the same methodology used in determining the wind vector used to drive the tracer as described in the paragraph above. These weighted average profiles for the prognostic variables r , θ , and P , are used to drive the parcel models. In Figure 5-1 to

5.2, *B* and *C* designations correspond to the parcel simulations thermodynamic profiles, and these weighted average profiles for the prognostic variables can be seen here. Making a cursory comparison of the *A*, *B*, and *C* figures, one can see a the smoothing influence of using the RAMS+ weighted-average representation (*B* and *C*). In the RAMS+ grid-averaged view (*A*), grid resolution can be quite apparent where the tracer crosses from one grid point value to the next.

Graphic representations of the thermodynamics of representative trajectories 1 and 6(T1 and T6) can be seen in Figure 5-1 to 5.2. Designations *B*, *C*, and *D*, represent the various parcel model runs. Respectively they are: parcel-SB, parcel-LA, and parcel-SB driven by RAMS with bulk microphysics based thermodynamics. *B* and *C* figures represent the parcel-SB and parcel-LA thermodynamics. RH , s , and r_i depend on the partitioning of r_T into r_i and r_v , and are model dependent.

It should be noted that the thermodynamic variables plotted are intensive variables, not associated with a particular parcel "size" in mass or volume. If one prefers, an assumption of a unit mass can be assigned to the parcel, and the parcel would have some air-density dependent volume associated with it as well. This type of association will become more important when attempting to quantify the total processing of the cloud.

5.1.1 Representation of Mixing

In the figures showing the trajectory path and topography (upper left, Figure 5-1 to 5.2) the parcel initially rises, increasing in z , where z is the height defined with respect to sea level. The passive tracer trajectory is at the same time decreasing in its distance from the hill's surface. Inspection of the wind vectors over the hill in Figure 4.1 also show this pattern. In fact, it can be seen that as pre-cloud air rises over the hill, the air both converges vertically, and its velocity increases. This increase in flow within a

constriction, here, a constriction due to orographic forcing, is expected. In the LES domain representation used, this vertical convergence can cause a lower level grid at hilltop to have sources of air at some pre-cloud value of x which encompass a greater number of vertical grids. The hilltop grid will then have thermodynamic values that embody a mixture of the air in all source grids. Additionally, the pre-cloud source air tends to come from higher level grids, as the convergence in grid space tends toward lower level grids. In short, upper level air tends to mix downward into the lower grids at hilltop. In all three figures, r_T is seen to decrease along the uphill portion of the trajectory, and θ_i increases in the same locations where r_T decreases. This is consistent with the mixing in of the upper level air, which is dryer and has higher θ_i .

A sharp decrease is seen in the r_T profile as the trajectory again rises at around $x = 11000$ m, corresponding to a rise in the parcel position due to the downstream wave motion initiated by the orographic forcing of the hill. θ_i increases in the same location where r_T decreases. This is an indication of the mixing of environmental air.

The variation in the r_T and θ_i profiles is greater for T1 (Figure 5-1), where wind vector vertical convergence is greater than in T6 (Figure 5.2). Because these r_T and θ_i profiles are used to drive the parcel models, the effects of the convergence and grid resolution are fundamentally included in the parcel simulations. To eliminate this effect, one could simply hold r_T and θ_i constant throughout the parcel simulations. In this thesis, the direct comparisons attempted necessitate use of the diagnosed, varying profiles which encompass mixing.

Diagnosed aerosol and droplet variables are not included with the thermodynamics used to drive the parcel models. Initial values are used in the parcel. Therefore, variations in these variables due to diffusion processes cannot be represented in the parcel runs. One possible consequence of this can be a misrepresentation of the microphysics. For example, a scenario can be imagined where droplets along a trajectory could be depleted by detrainment or by precipitation. In RAMS+, both r_T and droplet number will adjust. However, in the parcel models, the decreasing r_T is

represented, but the change in number is not. Here, because there is no precipitation allowed, and because total number of aerosol + droplets is initialized identically in all grids, these effects are minimal.

5.1.2 Liquid water mixing ratio

The B figures for Figure 5-1 and 5-2 depict the thermodynamics of the parcel-SB model driven by the RAMS+ thermodynamic profiles of r_T , θ_1 , and P . It is first noted that RH , S , and r_l differ from the profiles in the RAMS+, "A" figures, in spite of both the use of identical subroutines to calculate the microphysics, and identical values for the prognostic variables r_T , θ_1 , and P . First compare Figure 5-1 A and Figure 5-1 B, which are the T1 profiles for the RAMS+ and the RAMS+ driven model, parcel-SB. Notice that for these, the supersaturation is positive through the left, or upwind, portion of the cap cloud, where activation and condensation take place. Liquid water, in the plot below, increases here, and reaches a maximum at the point where supersaturation changes sign. The negative supersaturation in the region that follows induces a decrease of liquid water until complete evaporation. The S profiles seem closely related. Recall that the RAMS+g profiles are not based on an averaged value of surrounding gridpoints, but on one gridpoint value.

There is strong evidence of the influence of grid-to-grid advection of liquid water, which can be seen in Figure 5-2, representing T6. In Figure 5-2 B, the parcel-SB model, the presence of liquid water and of positive supersaturation initiate at approximately same position. In Figure 5-2 A, RAMS+, it is seen that supersaturation values become positive, although not with a consistent duration that could account for the r_l profile.

The RAMS+ profile of r_l do show both a larger r_l than in the corresponding parcel-SB model (for some positions), as well as a greater persistence of positive r_l in the evaporative regime of the cloud. In spite of the inconclusive comparisons of the

supersaturation profiles, it does seem the r_l in the plots supports the notion of the advection into the T6 area from neighboring grids.

A complication is noted here. In RAMS+ current format, both r_T and r_l , the liquid water mass mixing ratio for the water bins, are influenced by model advection routines. The water vapor mass mixing ratio, r_v , is calculated by their difference. It is possible to develop inconsistencies. An exaggerated scenario would be drizzle processes increasing or decreasing liquid water, and also total water in a grid. Total water, r_T , may not follow suit, causing unlikely responses in the vapor field.

5.1.3 Supersaturation profiles

The supersaturation profile shapes exhibit quite a different nature for the RAMS+g vs. the parcel models. The parcel models (B and C figures) have a pronounced supersaturation maximum at the inflow edge of the cloud. For the parcel-SB model runs, this can be a local or absolute maximum, while for the parcel-LA runs, the maximum is absolute. This feature, comparable to the supersaturation maximum seen near cloud base of a convective cloud, is expected. Inspection of the A figures show no evidence of this feature. The grid averaging of thermodynamic data has, perhaps, diminished this feature, and maximum supersaturation values are not reached until further into the bulk of the cloud.

When directly comparing the parcel-SB and parcel-LA supersaturation profiles, the S maximum at the cloud inception edge is seen to be approximately a factor of three greater for the parcel-LA model runs. This difference is due to the use of different microphysical schemes, specifically for droplet growth, used in the two parcel models. Both the parcel-SB and RAMS+ microphysical modules use an averaged droplet growth forcing over all drops, based on pre- and post-supersaturation values for the timestep (2 s). Post timestep supersaturation values are based on dynamical forcings and adjusted for projected condensation/evaporation approximated for the timestep. The parcel-LA

model, on the other hand, uses explicit solute dependent growth equations for each droplet category. Use of the variable timestep differential equation solver VODE (Brown et al, 1989) allows for competitive vapor deposition onto all droplets, adjustable timesteps within the overall 2 s model timestep, and, in general, growth equations having a much greater agreement with theoretical predictions for droplet growth. It seems that at the cloud's inception edge, where grid-to-grid mixing has yet to become a great influence, the parcel-LA supersaturation profile may more reliably emulate theoretical predictions than either the RAMS+ or parcel-SB models. However, this is not to say that current theory necessarily encompasses all the processes required to approximate observational findings.

5.1.4 Driving parcel models with bulk microphysics RAMS

The last feature which is illustrated here is the applicability of using a bulk microphysics RAMS model run to determine trajectory profiles for driving the parcel models. In the RAMS bulk microphysics, RH has a limit of 100%, and any water vapor beyond that is immediately defined as liquid water. The representation of the detailed interaction between dynamics and microphysics is lost. However, with some cloud types, this level of detailed interaction may not be necessary. When this is the case, a great savings in time can be accomplished using the bulk microphysics to produce fields from which trajectories are diagnosed. In the case of the hill cap cloud modeled, the full RAMS+ runs, the CPU time needed to simulate the model is approximately four times the time simulated. For the RAMS-bulk runs, this factor is 0.1 times the time simulated.

Figure 5-1 D and 5-2 D are for the parcel-SB model driven by RAMS with bulk microphysics trajectories for T1 and T6, respectively. The r_T , θ_1 , and P profiles have the same shape as those from the RAMS+ simulations (A figures). The resulting r_i and S profiles are also very similar to those of the parcel-SB model driven by RAMS+ trajectories. The aerosol and droplet distributions derived (not shown) are also very

similar to their counterparts derived from RAMS+ thermodynamic profiles. Although this agreement cannot be taken for granted, it may be possible to use RAMS with bulk microphysics to drive parcel models at a great savings of time in certain cases, especially non-precipitating clouds.

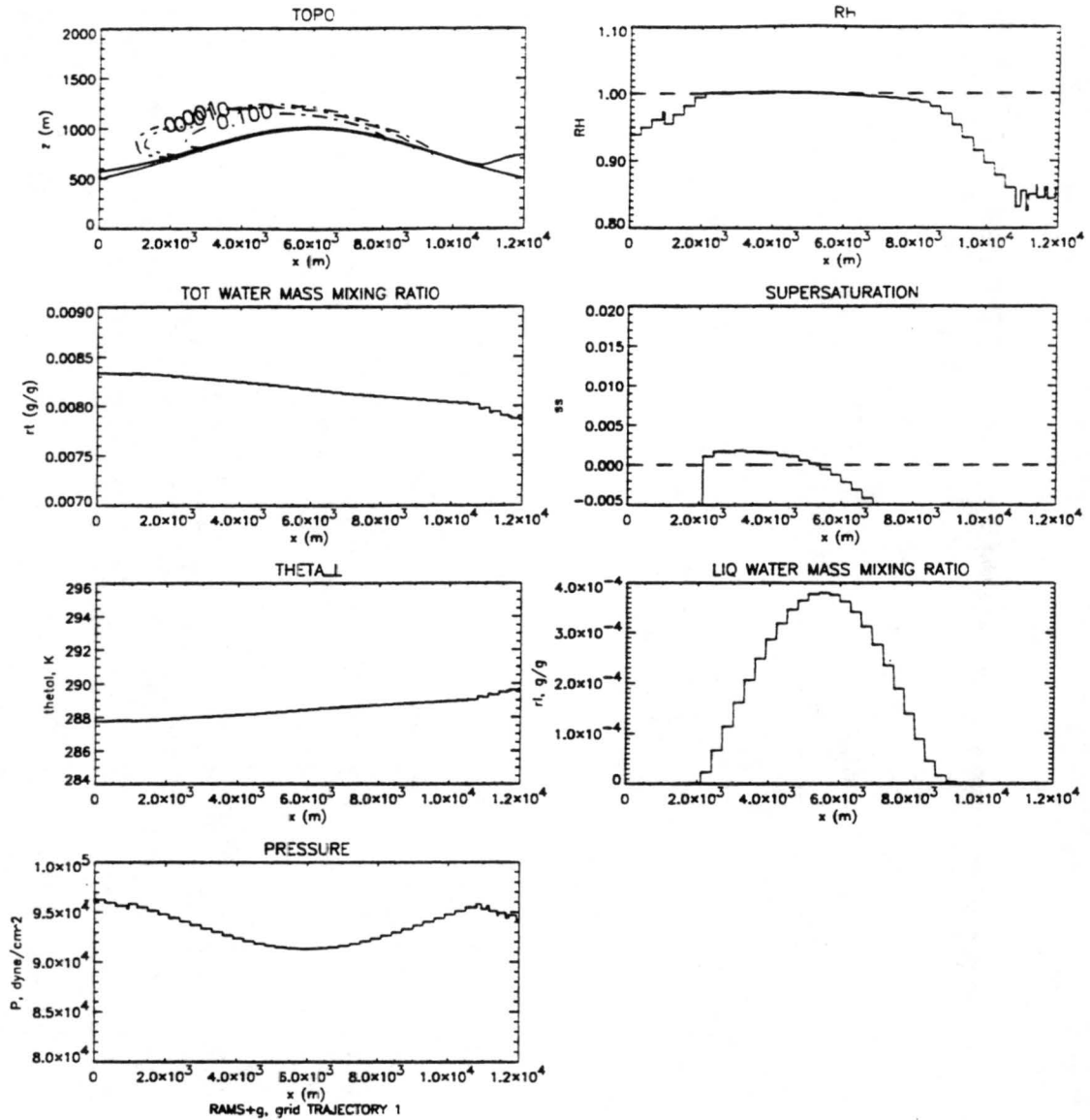


Figure 5-1 A: RAMS+g, trajectory 1, thermodynamic profiles. The first plot in each figure depicts the section of the model domain where the cloud resides, including the hill, the hill cap cloud (defined by the cloud liquid water mixing ratio contour plot), and the trajectory. The three plots below this are for r_t , total water mixing ratio; θ_L , liquid water potential temperature; and P , pressure. The other variables plotted in these figures are RH , relative humidity; S , supersaturation; and $LWMR$, liquid water mass mixing ratio.

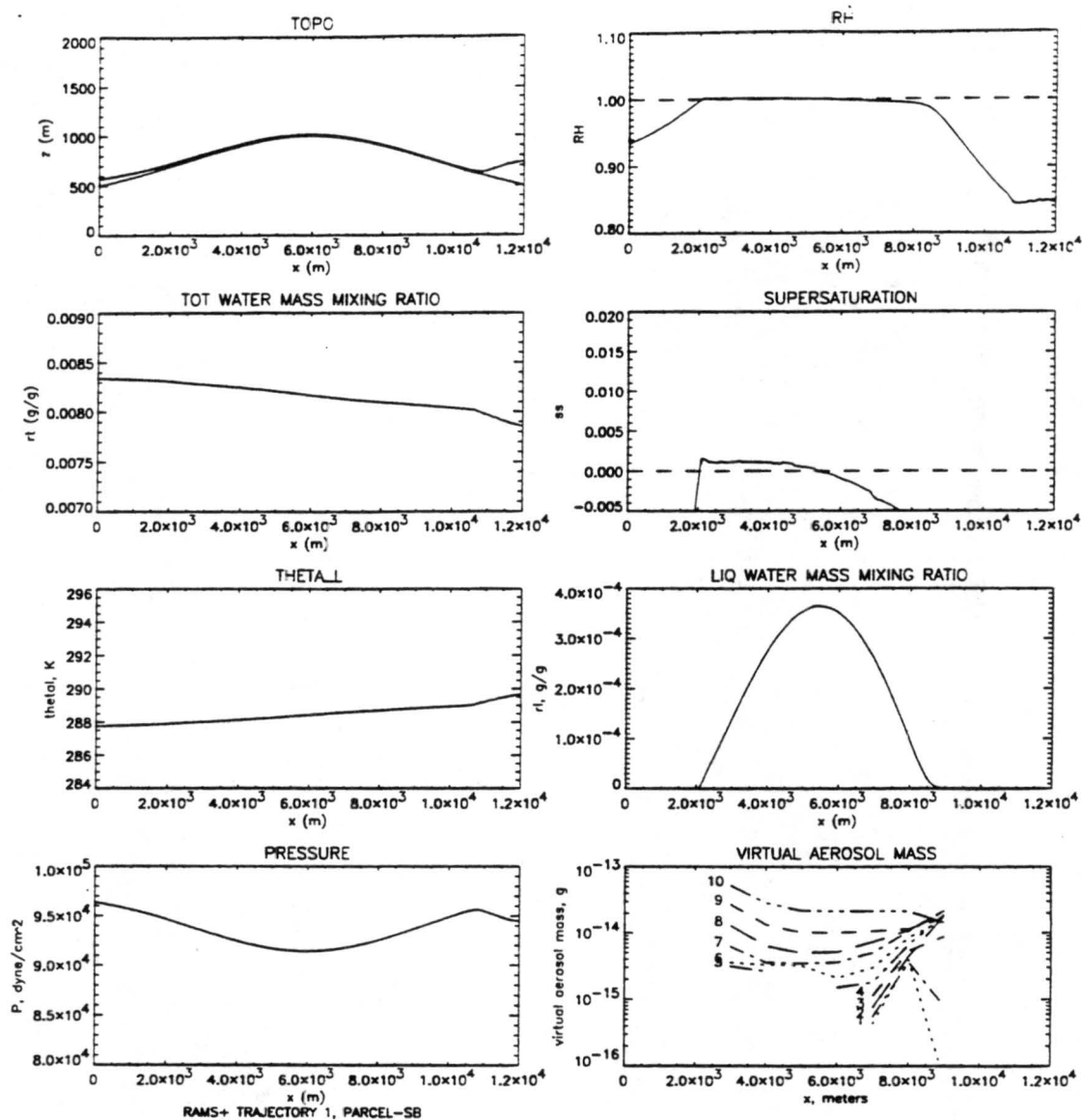


Figure 5-1 B: Parcel-SB, trajectory 1, thermodynamic profiles. See text, Figure 5.1 A.

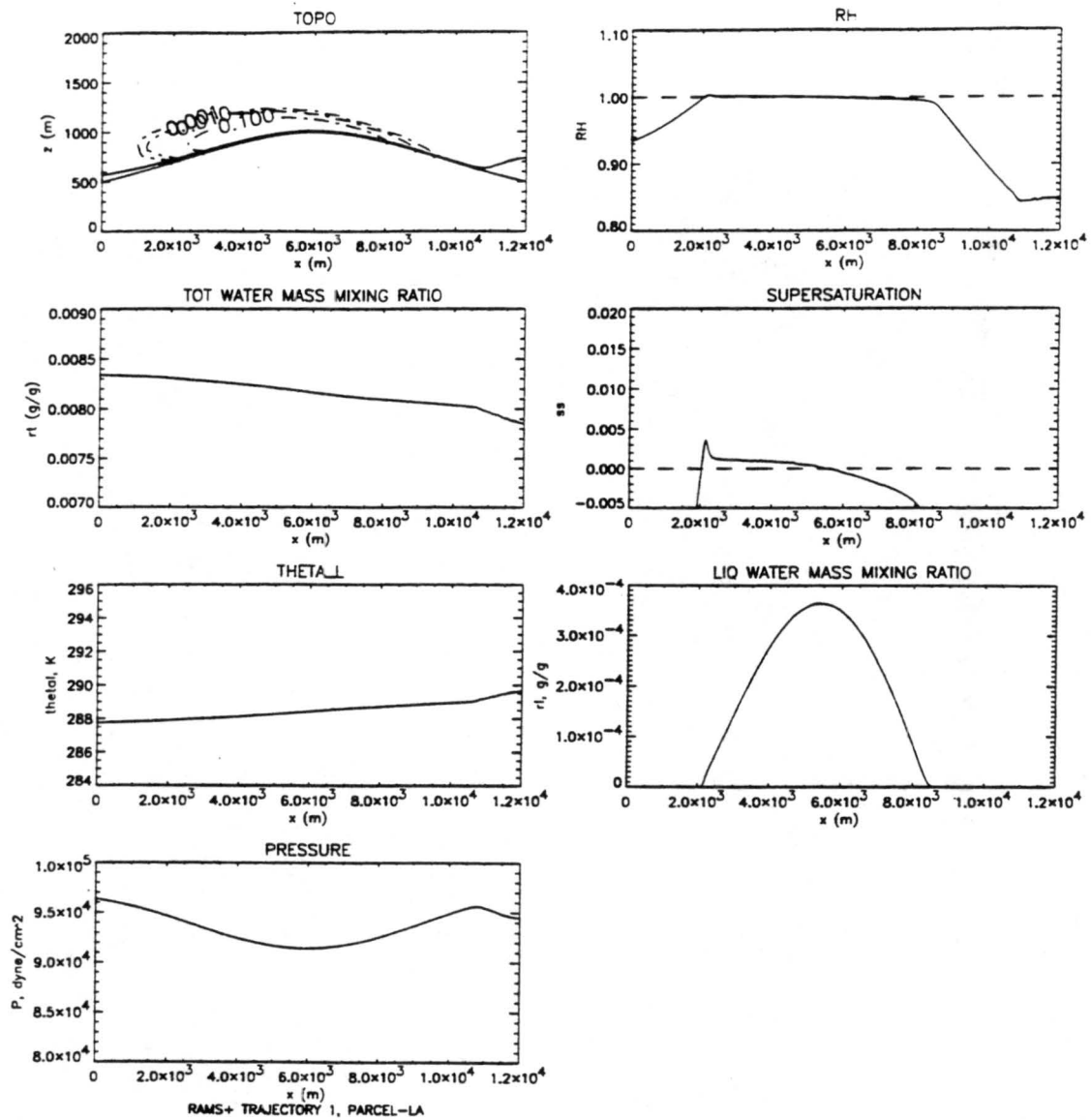


Figure 5-1 C: Parcel-LA, trajectory 1, thermodynamic profiles. See text, Figure 5.1 A.

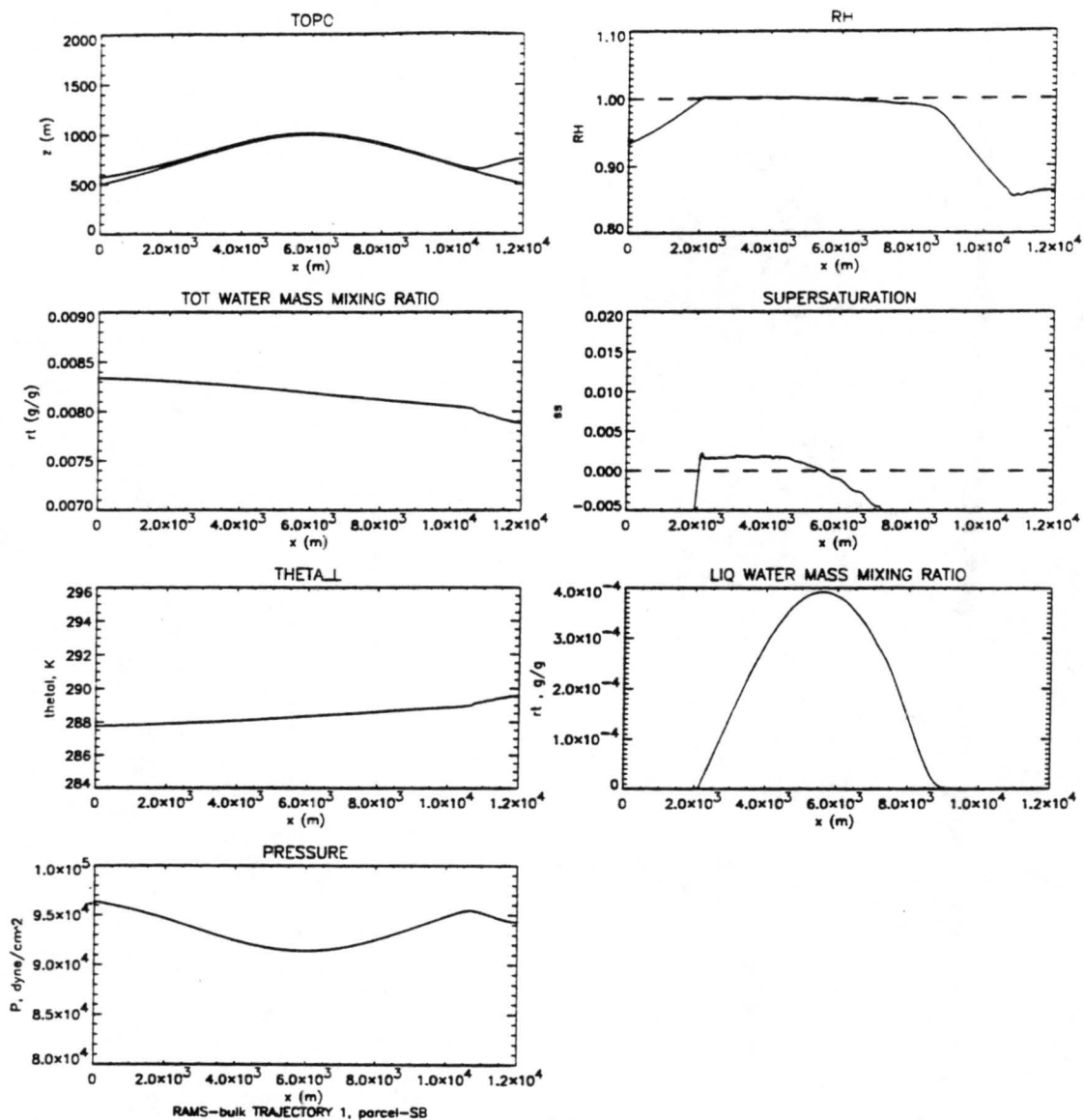


Figure 5-1 D: Parcel-SB, RAMS-bulk, trajectory 1, thermodynamic profiles. See text, Figure 5.1 A.

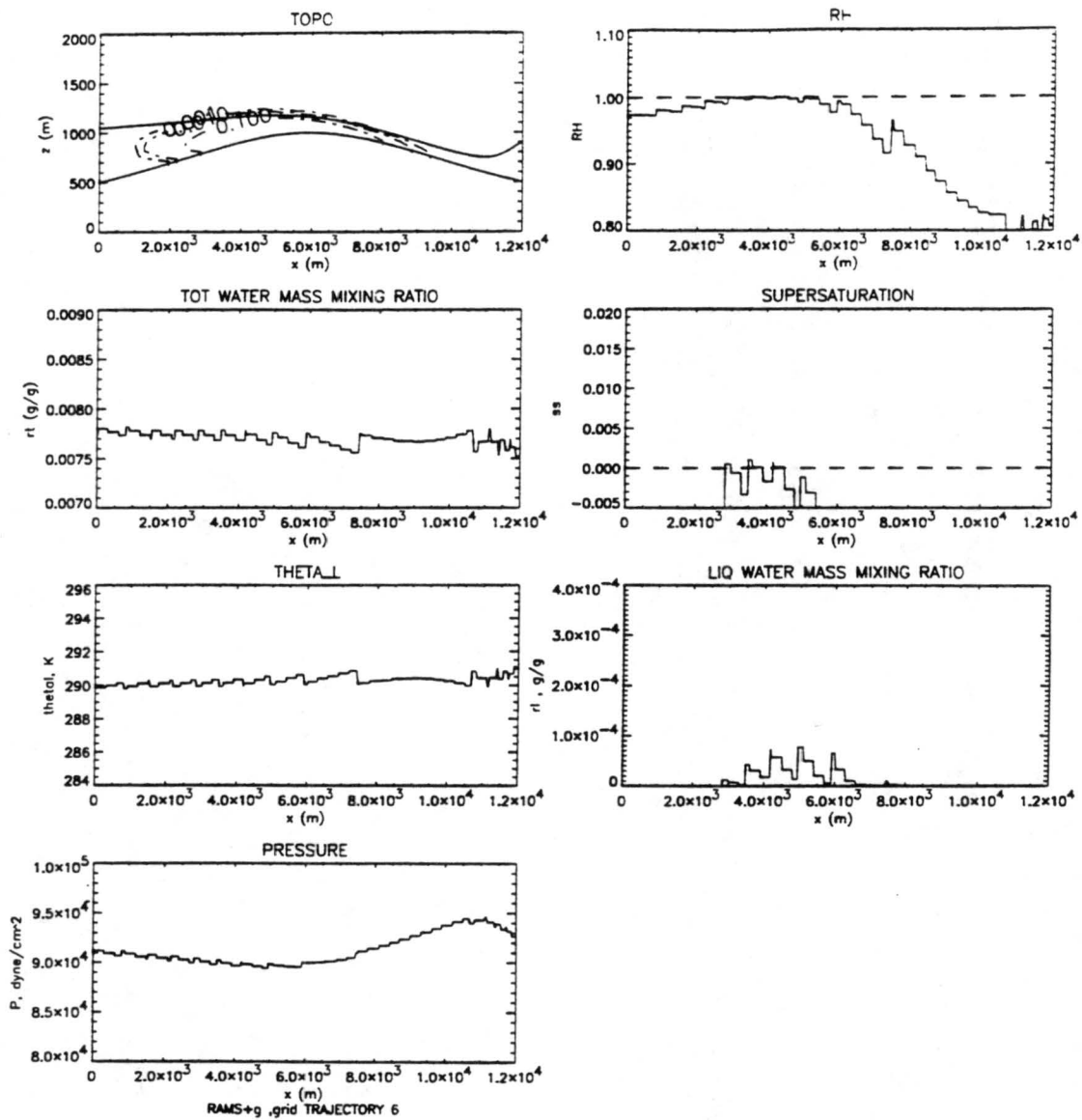


Figure 5-2 A: RAMS+g, trajectory 6, thermodynamic profiles. See text, Figure 5.1 A.

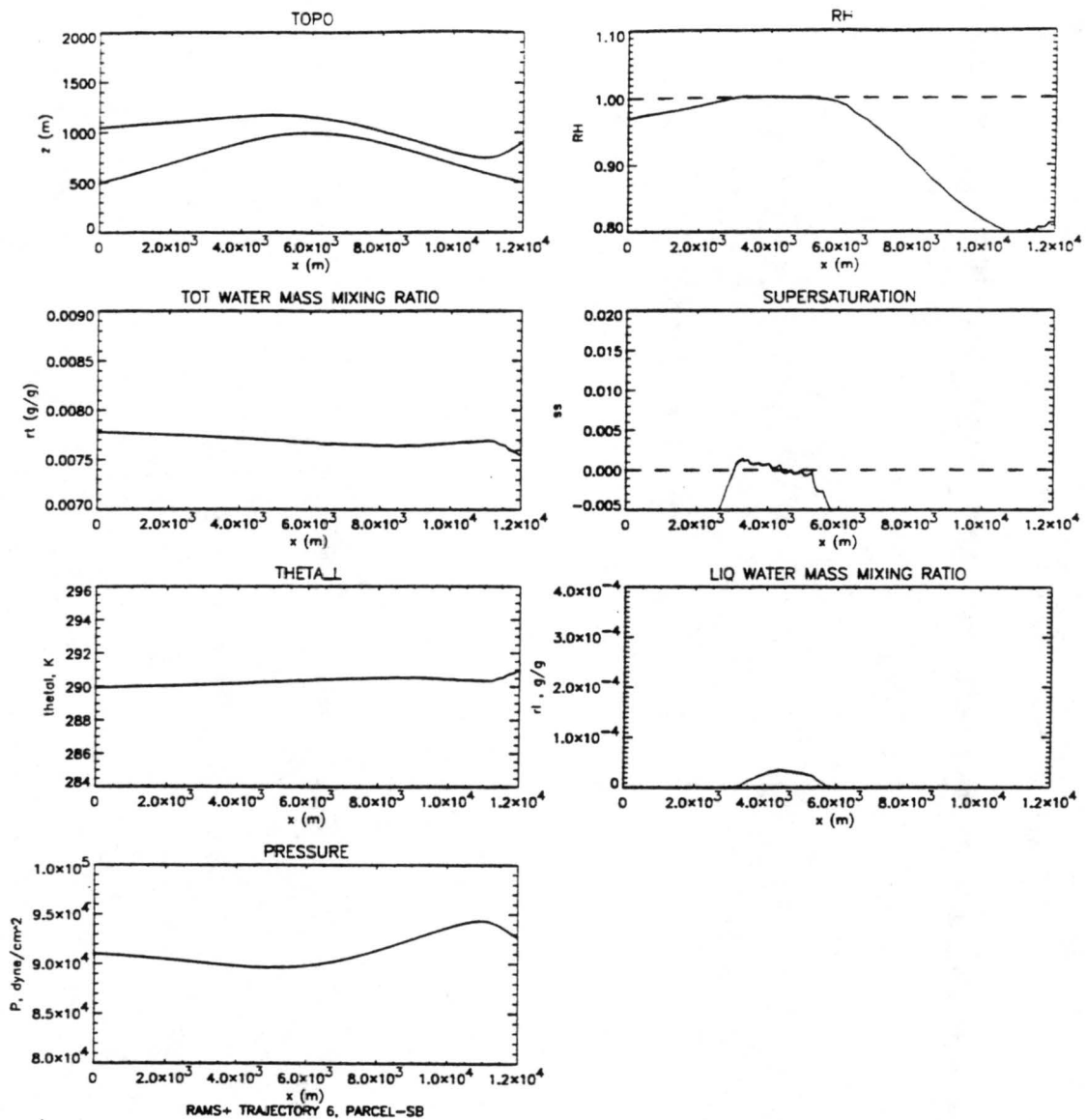


Figure 5-2 B: Parcel-SB, trajectory 6, thermodynamic profiles. See text, Figure 5.1 A.

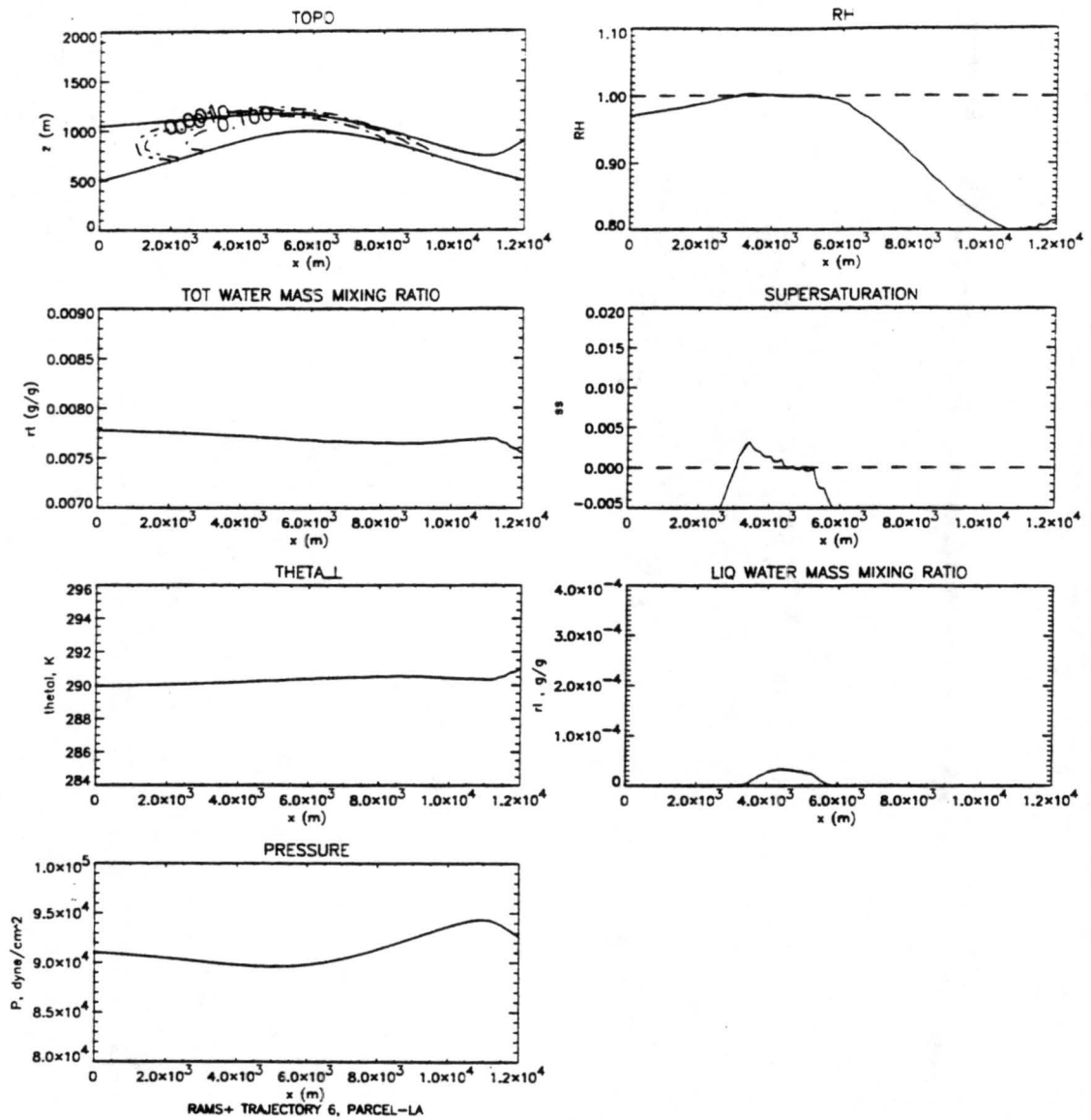


Figure 5-2 C: Parcel-LA, trajectory 6, thermodynamic profiles. See text, Figure 5.1 A.

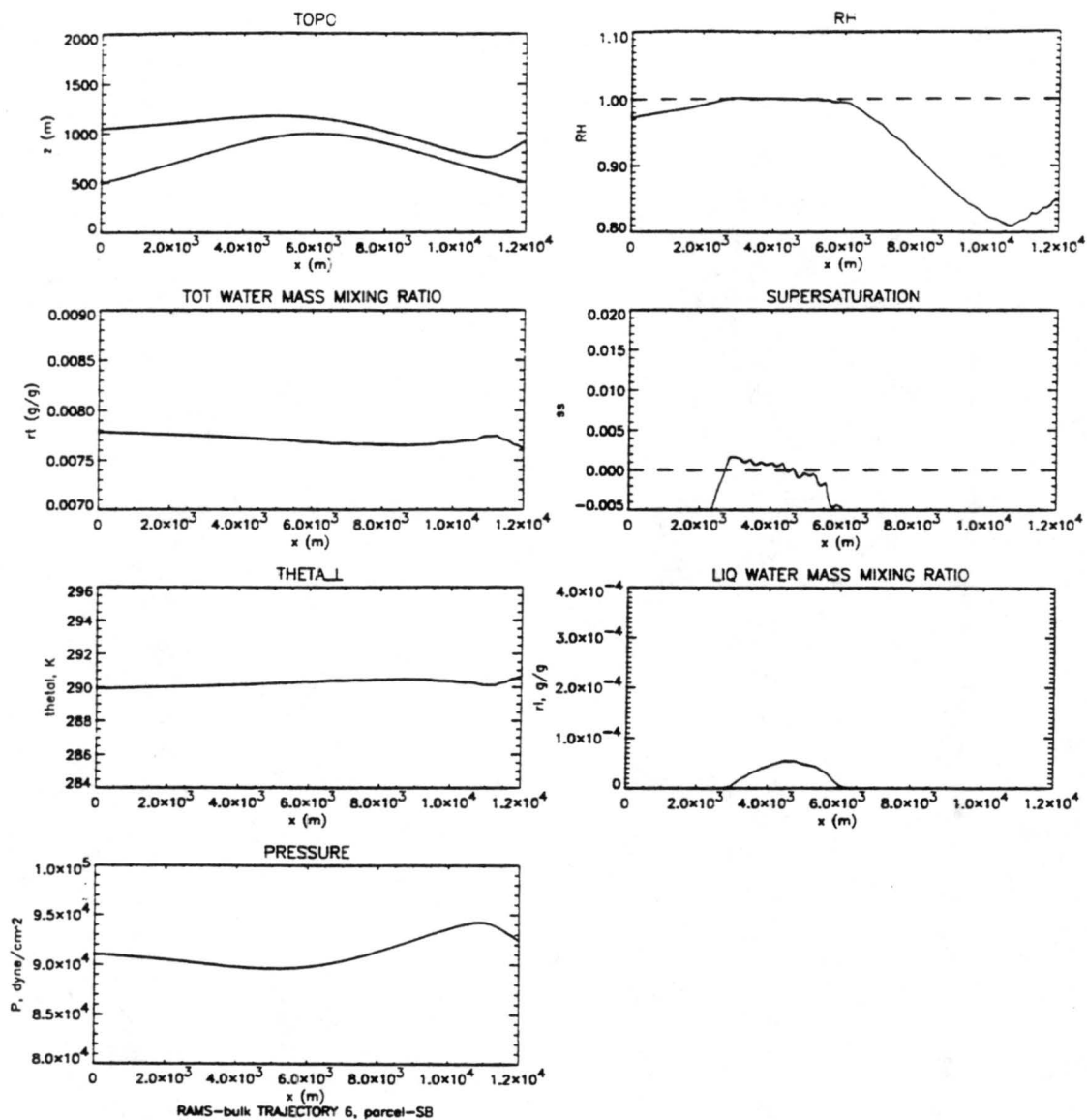


Figure 5-2 D: Parcel-SB, from RAMS-bulk, trajectory 6, thermodynamic profiles. See text, Figure 5.1 A.

5.2 Aerosol and water droplet distributions

Examination of the aerosol and water droplet distributions can help to illustrate findings of the previous section. Figure 5-3 through Figure 5-5 represent the distributions associated with T1 for the RAMS+, parcel-SB, and parcel-LA model runs, respectively. The first column in each figure shows the aerosol number mixing ratio in an aerosol bin. For reference, the right hand scale refers to number per bin per cm^3 . The second column is the aerosol mass mixing ratio. The analogous mixing ratios for number and mass of droplets in a water bin are in columns three and four. Scales above refer to particle diameter, and scales below to bin indices. Each row represents a different spatial x position along the trajectory where the distributions are retrieved. The first row is at $x=1000$ m, and the second through fifth row are incremented at 2000 m intervals: $x=3000$, 5000, 7000, and 9000 m. Notice that in the first and last rows, there is no liquid water present, and these correspond to pre- and post-cloud positions.

In the parcel-LA model, the aerosol/droplets are not represented in sectional bins. For the purpose of plotting and direct comparison to the other models, aerosol/droplets that have activated and grown to sizes greater than $3.125 \mu\text{m}$ in diameter are mapped into water bins, and unactivated aerosol/droplets are mapped into their corresponding dry aerosol size bins.

5.2.1 Number of aerosol activated

In comparing the aerosol number mixing ratio distributions for the three models it can be seen that a greater number of aerosol has been removed by activation in the parcel model runs than in the RAMS+ model run. That is, there is a greater number of aerosol remaining in the aerosol bins for RAMS+, in the 2nd-4th rows of Figure 5-3, than in corresponding parcel run bins, Figure 5-4 and Figure 5-5. The minimum size aerosol that activates is a function of the supersaturation. It is the greater S maximum, seen in the cloud incipient edge in the parcel models, that causes the greater overall number of

aerosol to be activated in the parcel runs. It follows that because this S maximum is greatest in the parcel-LA model, there should be a greater number of aerosol activated in this vs. the parcel-SB model. Inspection of the distributions in Figure 5-4 and Figure 5-5 show this to be the case.

An additional feature of the activation and supersaturation relationships can be noted in these figures. Recall that the position of S maximum for the RAMS+ simulation was much further into the cloud than for the parcel models simulations. Consequences of this positioning of the S maximum can be seen in the progressions of the aerosol distributions in rows 2-4 of the figures. For RAMS+, additional activation has taken place between the row 2, or the $x=3000$ m position, and row 3, the $x=5000$ m position: the aerosol remaining has decreased. In both of the parcel models, where S maximum is near the cloud incipient edge, there is no additional activation taking place. Water droplet number mixing ratios relate directly to the aerosol number mixing ratio, as each aerosol activated becomes a water droplet with solute. In terms of the water droplet mixing ratio, the RAMS+ simulation produces a total droplet number mixing ratio that increases across the initial portion of the cloud, where for the parcel models, that number stays constant beyond the S maximum near cloud inception. The latter situation, relatively constant droplet number after cloud incipient edge, or cloud base, is in better agreement with observational data.

5.2.2 Breadth of water droplet distribution

The processes controlling the breadth of the droplet distribution include both dynamical grid-to-grid mixing (for RAMS+) and microphysics. As RAMS+ and parcel-SB have identical microphysical representations in their model code, it would seem that these could be directly compared to ascertain the effects of grid-to-grid mixing in the model. This must be qualified, however. As was seen in the previous section, number of aerosol activated and number of cloud droplets are different in the corresponding trajectories for the two models. Additionally, distributions recovered from RAMS+ simulations pertain to one grid, a point representing an area in the two-dimensional

cloud. In the parcel-SB model, the distribution pertains to one point along the trajectory.

The droplet distributions in Figure 5-3 and Figure 5-4 differ in their breadth, the parcel-SB distribution being noticeably narrower. The greater number of droplets in the parcel-SB model causes the droplet size to be slightly smaller than those sizes at comparable positions in the RAMS+ simulation, as the liquid water available is distributed over a greater number of droplets. Along most trajectories, there is a noticeable narrowing of the droplet distribution between row 2 and row 3 distributions for a given model. This can be seen clearly in the droplet number mixing ratios for RAMS+, Figure 5-3.

One notion might be that a sampling of parcel-SB distributions in the spatial extent of the corresponding grid might better represent the overall distribution in a grid. Within the spacing of one grid, there could be slightly different S profiles in the vertical, and varying total time in a condensational regime in the horizontal. A test case across a grid area was performed. In looking at these distributions, it was found that any broadening they contribute is quite small, as all parcel-SB distributions calculated appeared quite similar.

Water droplet distributions for the parcel-LA model are narrower than those for the parcel-SB model. These two models are driven by identical thermodynamic variables, so the cause is undoubtedly in the microphysics, specifically in the growth equations, as discussed in the previous section on S profiles.

Total number of particles (aerosol plus droplets) stays constant at initial values for both parcel models. In the RAMS+, there is the possibility of this total changing. However, because this is initialized with one value for all grids, and because precipitation, drizzle, and collision/coalescence are not allowed, this number changes minimally along the trajectory, by less than 1%.

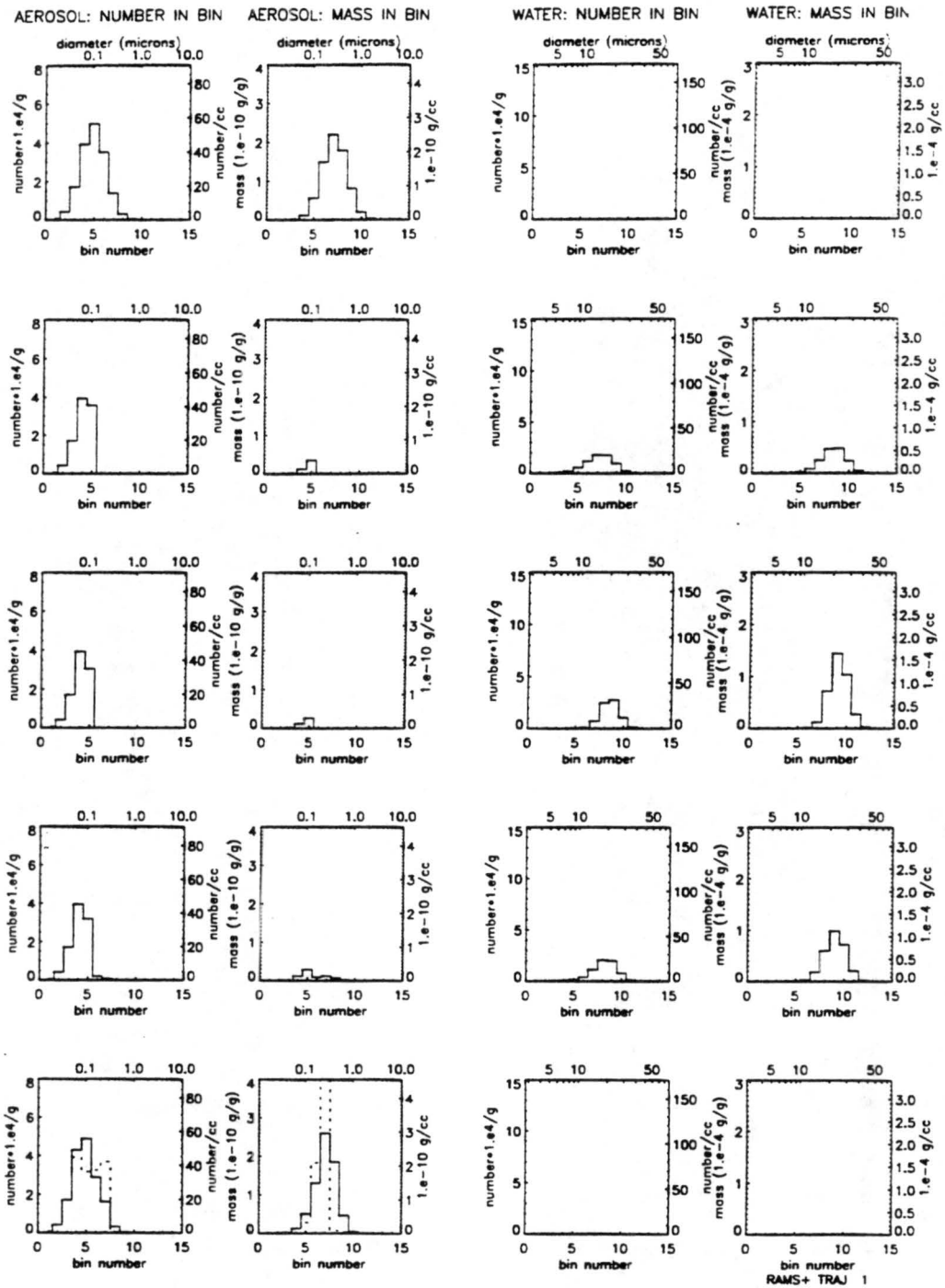


Figure 5-3: T1, RAMS+ aerosol and water droplet distributions. Rows 1-5 designate distributions along T1 at $x=1000, 3000, 5000, 7000$, and 9000 m. Dotted line in lower left distributions refers to final aerosol distribution when lognormal regeneration is not applied.

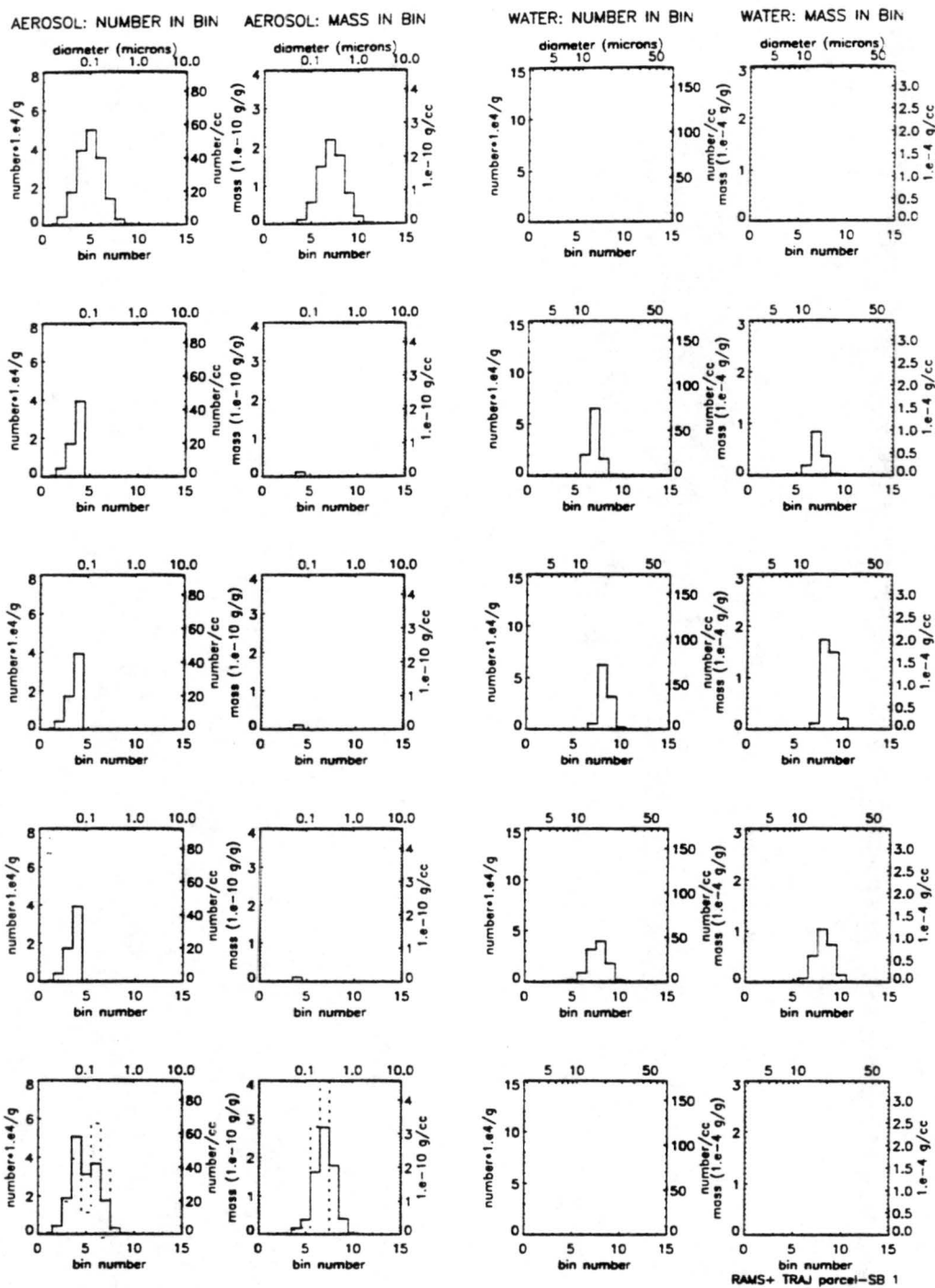


Figure 5-4: T1, Parcel-SB aerosol and water droplet distributions. See text for Figure 5-3.

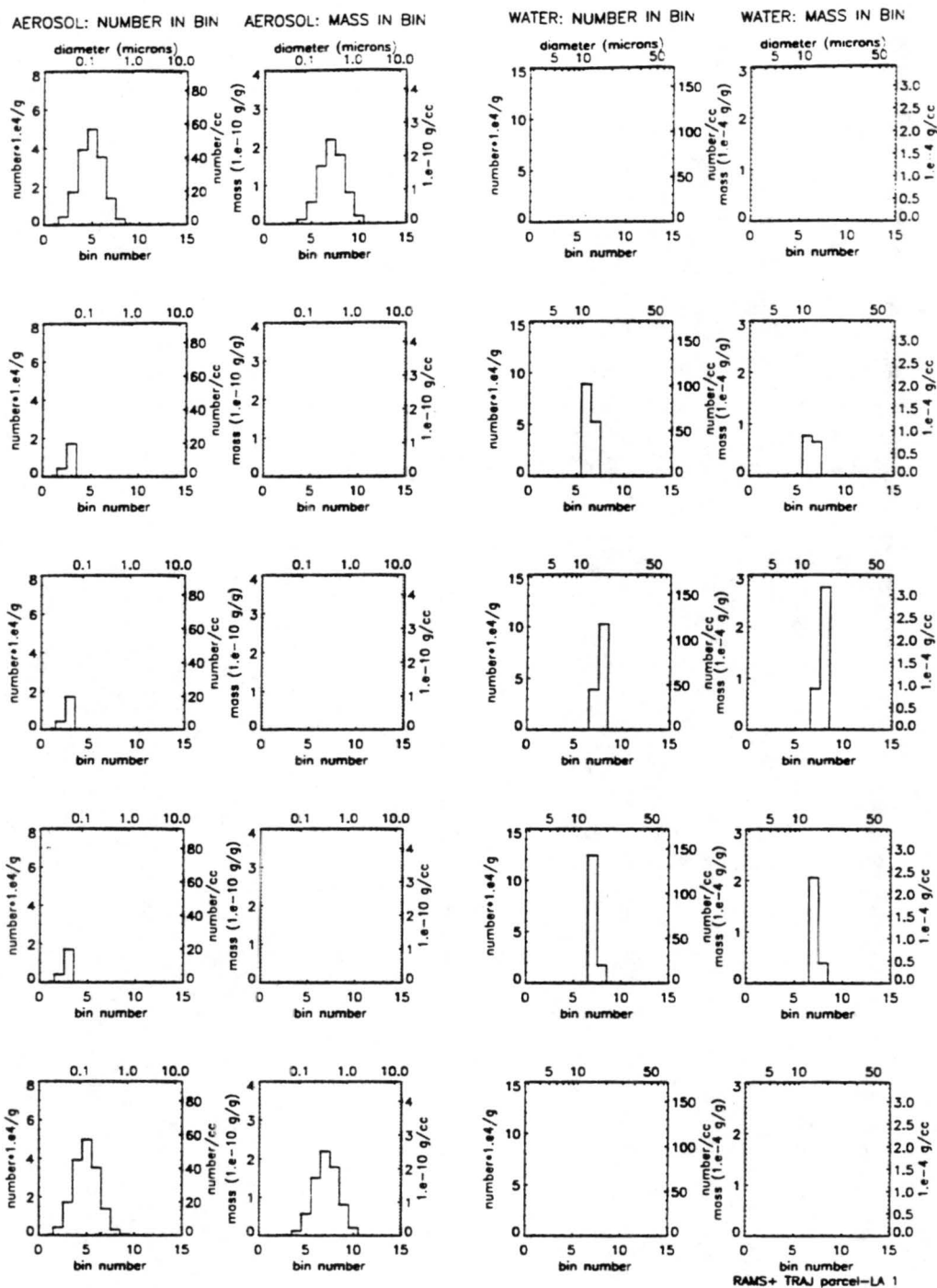


Figure 5-5: T1, Parcel-LA aerosol and water bin droplet distributions, as mapped to a sectional bin representation. Rows 1-5 designate distributions along T1 at $x=1000, 3000, 5000, 7000$, and 9000 m.

6. Conclusions

Comparisons were made of three model representations of a two-dimensional hill cap cloud:

1. RAMS+, a cloud resolving model with explicit, solute following, sectional bin microphysics. 14 aerosol and 25 water droplet bins are represented.
2. Parcel-SB, a sectional bin parcel model with the same microphysical representation as 1. above. The thermodynamics used to drive the parcel model were derived using passive tracers flowing through the RAMS+ simulation, or a similar RAMS simulation with bulk microphysics.
3. Parcel-LA, a parcel model with a Lagrangian aerosol representation. Each aerosol/droplet size category is followed through deliquescence, activation, and condensation/evaporation, without separate aerosol and droplet designations. There is no mixing between categories. This parcel model is driven by the same thermodynamics as 2. above.

Notable differences in these simulations involved the supersaturation fields, number of activated aerosol, droplet distributions, and solute mixing/redistribution.

The strengths of the LES model lie in its ability to simulate the dynamical processes driving the gross evolution of the cloud: inflow, vertical and horizontal winds, mixing, and cloud base and extent. The inclusion of explicit microphysics in these models, in the form of a sectional bin resolved droplet distribution, is thought to allow for a more realistic coupling between cloud dynamics and microphysics. However, as shown by Stevens et al. (1995a,b), there are limits to the integrity of both supersaturation and droplet number activated.

The use of LES type model trajectory thermodynamics in conjunction with a parcel model can result in a great savings of time and memory. More complex chemistry and microphysics can be then represented in the parcel model and applied only along the

trajectories, as opposed to at every gridpoint and for every timestep in the LES simulations.

6.1 Supersaturation fields and distributions

Supersaturation profiles for both parcel models showed a sharp S maximum near the cloud incipient edge. In all cases, this maximum was greater for the parcel-LA simulations. Aerosol distributions exhibit activation occurring concurrently with positive S , followed by relatively constant droplet and aerosol number mixing ratios along a trajectory until droplet evaporation initiates.

The RAMS+ simulation did not have an analogous S maximum at the incipient edge, but had a more smoothly varying S profile with its maximum further into the cloud. With this profile, aerosol continued to activate until the S maximum was reached, producing an increasing droplet number along the trajectory in this regime.

Water droplet distributions were broadest in the RAMS+ simulations, and narrowest in the parcel-LA simulations. A smaller mean drop size is seen in the parcel models, a result of the distribution of liquid water over an increased number of aerosol activated.

6.2 Solute distribution in the drops

Upon aerosol activation in both the RAMS+ and parcel-SB, the activated aerosol is redefined as solute and stored as a solute category in each droplet bin. As the droplets undergo growth by vapor deposition and narrowing of the droplet spectrum, both solute and liquid water are transferred from droplet bin to droplet bin. This causes a computational mixing of solute, averaging out the virtual aerosol sizes associated with each droplet bin, a result also seen by Roelofs (1992). This finding of the SB behavior is disconcerting. Major reasons for including solute following microphysics are to obtain the capacity to quantify cloud processing: to explicitly regenerate an aerosol

distribution upon cloud or parcel evaporation, and to include solute concentrations in both droplet growth equations and aqueous chemistry equations.

The approach used here was to regenerate aerosol in a lognormal distribution about the mean of the solute mass and number in an evaporating drop. This may prove useful in cases where repeated activation and regeneration of aerosol is occurring, as in updrafts and downdrafts in a stratocumulus cloud. In the case where there is no solute-following microphysics, aerosol is regenerated using a best guess approach (Kogan et al., 1994). Here, even in a worst case scenario of complete mixing, the average size information of aerosol regenerated is known, and can be used in estimating the distribution of the returning aerosol. However, aerosol regeneration schemes do not meet concerns involving the degradation of reliable solute concentration information in a drop.

In these runs, where collision/coalescence is not permitted, the LA representation of microphysics may be preferred. There is no mixing between categories, so none of the averaging present in SB representations can take place. When solute concentrations are important or explicit aerosol regeneration is desired, this is the representation of choice. When collision/coalescence plays a major role, the SB representation has major computational benefits.

6.3 *Mixing in parcel runs*

A. It was found that using the thermodynamic data from the RAMS+ and RAMS with bulk microphysics in driving the parcel models, some mixing information was intrinsically included. This included both the mixing resulting from the vertical convergence of the air as it passed over the hill, and turbulent diffusional mixing represented in the model. This was very noticeable in the r_T and θ_1 profiles, which gave indication of convergence of dryer, higher θ_1 upper level air into the grids from which trajectories were diagnosed, decreasing r_T and θ_1 along the trajectories.

7. Future work

Obvious continuations to the work of this thesis are:

1. Completion of chemistry in RAMS+
2. Inclusion of collision/coalescence, precipitation, and other processes previously "turned off"
3. Perform three-dimensional simulations of this and other cloud types
4. Quantification of cloud processing
5. Parameterization of entrainment of environmental atmosphere including aerosol and ambient gases

1. Chemistry has been applied to the parcel-LA model, and initial work done in adding chemistry into the RAMS+ and parcel-SB models, although results are not reported in this thesis. To add a complete aqueous chemistry package into the RAMS+ model, the chemistry can and must be simplified. One example is to consider species that partition quickly between gaseous and aqueous phases (on the order of a timestep or less) to be in equilibrium, instead of calculating differential equations for determining the partitioning. These include O_3 , H_2O_2 , and SO_2 . Preliminary investigations show that running the chemistry module in RAMS+ only along the trajectories may be a very reasonable way to incorporate chemistry into a LES model.
2. Currently, collision/coalescence and precipitation has been "turned off" in the RAMS+ and parcel-SB simulations. This processing can be investigated and tested in the framework of these models.
3. The work in this thesis has been performed using a simple two-dimensional hill cap cloud. This is now ready for use in three-dimensional models. The current model was devised with the goal of implementation into a simulated marine stratus cloud, and SB representations and even initial aerosol distributions were arranged with

this end in mind. This aerosol processing model should be next implemented in a three-dimensional marine stratus cloud.

4. A method for quantitatively assessing the cloud processing of aerosol and atmospheric gases is needed. Parcels are evaluated using mixing ratios. It may be enough to simply add a total mass component to each parcel according to its initial position and its proximity to neighboring parcels.
5. A simple parameterization of environmental air for parcel models may be possible using the θ_i and r_i information available in a Paluch diagram (Paluch, 1979) to approximate the source location of entrained environmental air.

My own long term goals for this work are to develop parameterizations of cloud processing for use in models of local and global change, using initial data such as chemical source terms, aerosol distribution, and cloud type, duration, and fraction. The processing of aerosol by physical and chemical processes, as well as physical redistribution of atmospheric components by cloud formation and evolution would be approximated. Work in the area of quantification is required.

Development of a scheme to represent collision/coalescence processes in the LA representation is another desire. A mapping scheme to associate the LA bins to SB representation could be possible. However, an addition of a few variable size "raindrop" bins may enable a parameterized collision/coalescence process, similar to that used in RAMS warm cloud parameterized microphysics. The SB model with chemistry can be used in determining what size drops and how many drops might be expected to move from the aerosol/cloud-drop category to the larger raindrop category.

Appendix A : Lognormal distributions

The probability density function for normally distributed u , is the Gaussian distribution:

$$f(u) = \frac{N}{(2\pi)^{1/2} \sigma_u} \exp \left[-\frac{(u - \bar{u})^2}{2\sigma_u^2} \right] \quad \text{Equation A.1}$$

where N , the total number, is defined as

$$N = \int_{-\infty}^{\infty} f(u) du \quad \text{Equation A.2}$$

Distributions in terms of a particle diameter, D_p , are required. The probability density function for normally distributed $u = \ln D_p$, is the Gaussian distribution of $\ln D_p$:

$$f(\ln D_p) = \frac{N}{(2\pi)^{1/2} \ln \sigma_g} \exp \left[-\frac{(\ln D_p - \ln \bar{D}_{pg})^2}{2 \ln^2 \sigma_g} \right] \quad \text{Equation A.3}$$

This distribution is a lognormal distribution of D_p . $\bar{D}_{pg}$ is the median diameter, and half of the particles are smaller than, and half are larger than, this diameter. σ_g is the geometric standard deviation.

To describe the function in terms of D_p (instead of $\ln D_p$) and to relate the independent variables, it must be noted that in any size range D_p to $D_p + dD_p$, the quantity dN does not change, and therefore:

$$f(D_p) dD_p = f(\ln D_p) d \ln D_p \quad \text{Equation A.4}$$

Solving for $f(\ln D_p)$ results in:

$$f(\ln D_p) = f(D_p) \frac{dD_p}{d \ln D_p} = D_p f(D_p) \quad \text{Equation A.5}$$

Using Equation A.5 in Equation A.3, the conventional form of the lognormal distribution is determined:

$$f(D_p) = \frac{N}{(2\pi)^{1/2} D_p \ln \sigma_g} \exp \left[-\frac{(\ln D_p - \ln \bar{D}_{pg})^2}{2 \ln^2 \sigma_g} \right] \quad \text{Equation A.6}$$

The relationship between the volume distribution function and the number distribution function is:

$$f_v(D_p) = \pi/6 D_p^3 f(D_p) \quad \text{Equation A.7}$$

Number mean (or average) diameter, $\bar{D}_p$, is determined from:

$$\bar{D}_p = \bar{D}_{pg} \exp(\ln^2 \sigma_g / 2) \quad \text{Equation A.8}$$

A more general form of this equation is:

$$\bar{D}_{p\mu} = \bar{D}_{pg} \exp(\mu \ln^2 \sigma_g) \quad \text{Equation A.9}$$

where $\mu=0$ for the geometric mean, $\bar{D}_{pg}$, $\mu=1$ for the surface area mean, and $\mu=1.5$ for the volume and for the mass mean.

Distributions with respect to surface area and volume are represented in the following equation (Seinfeld, 1986):

$$f_\gamma(D_p) = \frac{N}{(2\pi)^{1/2} D_p \ln \sigma_g} \exp \left[\gamma \ln \bar{D}_{pg} + \frac{\gamma^2}{2} \ln^2 \sigma_g \right] \exp \left[-\frac{(\ln D_p - (\ln \bar{D}_{pg} + \gamma \ln^2 \sigma_g))^2}{2 \ln^2 \sigma_g} \right]$$

$$\text{Equation A.10}$$

where $\gamma=1, 2$, and 3 for the diameter, surface area, and volume respectively and $a_\gamma = 1, \pi$, and $\pi/6$ for $\gamma = 1, 2$, and 3 respectively.

The cumulative number of particles with diameters of size D_p and smaller is (Seinfeld, 1986):

$$F(D_p) = \frac{N}{2} + \frac{N}{2} \operatorname{erf} \left(\frac{\ln \frac{D_p}{\bar{D}_{pg}}}{\sqrt{2} \ln \sigma_g} \right) \quad \text{Equation A.11}$$

where erf is the error function operator.

For aerosol initialization, a lognormal distribution is chosen to represent the aerosol number distribution function. Lognormal parameters N , $\bar{D}_{pg}$, and σ_g must be designated. The number and mass concentrations (or mixing ratios) in each bin i with bin limits $D_{p,i}$ and $D_{p,i+1}$ is desired. Equation A.11 is used to find the total number between the bin limits, where $N_i = F(D_{p,i+1}) - F(D_{p,i})$.

For the lognormal distribution of aerosol regenerated from evaporating droplets, the N regenerated particles, each with an average mass $N/M=m$, must be redistributed. A σ_g appropriate to the model must be arbitrarily specified. $\bar{D}_{pg}$ is determined from $\bar{D}_{pg,3}$ using Equation A.10. The new lognormal distribution is then distributed into the aerosol bins as in initialization, that is, using the cumulative function to determine N_i in each bin.

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