TOWARD ASSESSING THE EFFECTS OF AEROSOLS ON DEEP CONVECTION: A NUMERICAL STUDY USING THE WRF-CHEMISTRY MODEL

BY

WENDILYN J. KAUFELD

THESIS

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Adviser:

Assistant Professor Stephen W. Nesbitt
ABSTRACT

As the formative agents of cloud droplets, aerosols play an undeniably important role in the development of clouds and precipitation. Few meteorological models have been developed or adapted to simulate aerosols and their contribution to cloud and precipitation processes. The Weather Research and Forecasting model (WRF) has recently been coupled with an atmospheric chemistry suite and is jointly referred to as WRF-Chem, allowing atmospheric chemistry and meteorology to influence each other’s evolution within a mesoscale modeling framework. Provided that the model physics are robust, this framework allows the feedbacks between aerosol chemistry, cloud physics, and dynamics to be investigated.

This study focuses on the effects of aerosols on meteorology, specifically, the interaction of aerosol chemical species with microphysical processes represented within the framework of the WRF-Chem. Aerosols are represented by eight size bins using the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) sectional parameterization, which is linked to the Purdue Lin bulk microphysics scheme. The aim of this study is to examine the sensitivity of deep convective precipitation modeled by the 2D WRF-Chem to varying aerosol number concentration and aerosol type. A systematic study has been performed regarding the effects of aerosols on parameters such as total precipitation, updraft/downdraft speed, distribution of hydrometeor species, and organizational features, within idealized maritime and continental thermodynamic environments.

Initial results were obtained using WRFv3.0.1, and a second series of tests were run using WRFv3.2 after several changes to the activation, autoconversion, and Lin et al. microphysics schemes added by the WRF community, as well as the implementation of prescribed vertical levels by the author. The results of WRFv3.2 runs contrasted starkly with WRFv3.0.1 runs. The WRFv3.0.1 runs produced a propagating system resembling a developing squall line, whereas the WRFv3.2 runs did not. The response of total precipitation, updraft/downdraft speeds, and system organization to increasing aerosol concentrations were opposite between runs with different versions of WRF. Results of the WRFv3.2 runs, however, were in better agreement in timing and magnitude of vertical velocity and hydrometeor content with a WRFv3.0.1 run using single-moment Lin et al. microphysics, than WRFv3.0.1 runs with chemistry. One result consistent throughout all simulations was an inhibition in warm-rain processes due to enhanced aerosol concentrations, which resulted in a delay of precipitation.
onset that ranged from 2-3 minutes in WRFv3.2 runs, and up to 15 minutes in WRFv.3.0.1 runs. This result was not observed in a previous study by Ntelekos et al. (2009) using the WRF-Chem, perhaps due to their use of coarser horizontal and vertical resolution within their experiment.

The changes to microphysical processes such as activation and autoconversion from WRFv3.0.1 to WRFv3.2, along with changes in the packing of vertical levels, had more impact than the varying aerosol concentrations even though the range of aerosol tested was greater than that observed in field studies. In order to take full advantage of the input of aerosols now offered by the chemistry module in WRF, the author recommends that a fully double-moment microphysics scheme be linked, rather than the limited double-moment Lin et al. scheme that currently exists. With this modification, the WRF-Chem will be a powerful tool for studying aerosol-cloud interactions and allow comparison of results with other studies using more modern and complex microphysical parameterizations.
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CHAPTER 1
INTRODUCTION

1.1 Background and Motivation

The effects of aerosols, both natural and anthropogenic, on climate processes are one of the outstanding challenges facing scientists as described in the Intergovernmental Panel on Climate Change (IPCC, 2001). Under consideration in the IPCC report is the magnitude and sign of changes in the energy budget due to radiative effects of the presence of aerosol. Not under consideration are issues of even higher uncertainty: secondary effects of aerosols, such as modification of cloud properties or precipitation due to differences in aerosol type, concentrations, and size distributions. Numerous modeling and observational studies are tackling these difficult problems with a wide variety of tools and techniques.

Adequate observation and modeling of aerosols and their interactions within the earth system is difficult, and often expensive both financially and computationally due to the vast range scales of space and time involved in measurements, retrievals, and model simulations. These microscopic particles can have global consequences on time scales of seconds to centuries, and number concentrations vary in orders of magnitude depending on type and distance from source of the aerosols. Because aerosols act as a site for water vapor to condense upon, they are intrinsically crucial to processes involving the redistribution of water and energy in Earth’s atmosphere.

Aerosols are particularly important in some of the heaviest precipitation-producing events: deep convective thunderstorms. Understanding the role aerosols play in these systems is important not only because of their role in the redistribution of water and energy in the atmosphere, but also because of their resultant influences on the atmosphere’s wind circulations. Strong updrafts in the cores of these storms loft aerosol high into the troposphere, and changes in size and phase of associated water influences the strength of the storm itself through radiative and thermodynamic effects. Results of complex meteorological models that include aerosol-cloud interactions have shown that changes in aerosol characteristics can have significant impacts on the entire dynamical structure of these storms (e.g., van den Heever and Cotton, 2006). Nailing down just what effects the type, number concentration, and size distribution of aerosol have on deep convective processes and resultant precipitation has become a controversial and active area of research over the past few years, especially in the modeling community, as
computational resources begin to allow for researchers to directly model aerosol-cloud interactions within meteorological models.

Properties of aerosols and interactions of aerosols with their environment can be modeled, and this is arguably the most convenient and least expensive method to expand the current state of the science. In a field such as this, with relatively sparse observations, convergence of theories amongst different studies is necessary in order to our understanding to advance. Therefore, the most reasonable approach to solving these problems has been to seek a quorum amongst different model types and parameterizations. Through a meta-analysis of these studies we can ascertain what the most important factors are – for example, aerosol number concentration, height of freezing level, relative humidity – in governing increased or decreased storm invigoration and resultant precipitation. One of the goals of this study is to clarify and add to understanding addressed by this body of research.

The simulation of the coupling among aerosol, chemistry, and cloud processes is a fairly recent capability, especially when integrated within a meteorologically viable framework that allows for both mass and number of aerosol and hydrometeors to vary independently (Jacobson 2006). One such model is the Weather Research and Forecasting (WRF) model, which has been linked with atmospheric chemistry modules (WRF-Chem, Grell et al. 2005, Fast et al. 2006, Gustafson et al. 2007). The WRF-Chem allows meteorological and chemical fields to evolve together, which makes investigation of feedbacks between aerosol chemistry, cloud physics, and dynamics tractable. Parameterizations of microphysics within clouds, such as growth of cloud droplets, rain, and snow, typically are represented only by their mass. Parameterizations of this kind are termed “single-moment” schemes. Developers of the WRF-Chem built double-moment capability (consideration of mass, and number, independently) into a preexisting single-moment microphysics parameterization, but for cloud droplets only. This addition is the most crucial step in the connection between aerosols and the formation of cloud droplets, but is it sufficient to produce results similar to those from studies employing more sophisticated microphysics?

This study considers the influence of varying aerosol type, concentration, and size distribution on deep convection within the framework of the WRF-Chem. Two different thermodynamic environments are tested, varying aerosol concentrations in each environment to examine the response in precipitation and storm invigoration. The goal is to investigate the capability of this
state-of-the-art model in understanding changes in cloud and precipitation structure due solely to changes of aerosol characteristics.
CHAPTER 2
BACKGROUND AND LITERATURE REVIEW

2.1 Aerosols: theory
The formation of precipitation ultimately is rooted to the presence of water vapor, particulates in the air, and a mechanism that causes air to cool, usually caused by rising motion. In order to model each of these aspects correctly, especially for a model that seeks to represent aerosol as accurately as is possible given computational constraints, special consideration should be given to the first step in the process of rain production: representation of aerosol and the activation of aerosol to form cloud droplets.

Aerosols are tiny liquid, solid, or mixed phase particulates suspended in the air, generally ranging in size from 0.001\,\mu m to 100\,\mu m (Slinn 1975). Clouds would be quite rare, or might not even form in an atmosphere without aerosols because relative humidities in excess of several hundred percent are required for droplets to form homogeneously (Rogers and Yau 1989). Aerosols therefore play a crucial role in the transfer of water and energy throughout the atmosphere, acting as surfaces for water vapor to condense upon at frequently obtainable relative humidities of near or slightly exceeding 100%.

The chemical composition, total number concentration, and size distribution of aerosols each have an impact on the behavior of water associated with the aerosols. These factors can vary greatly in time and space, depending on the sources of aerosols and atmospheric conditions. Aerosols are often typified into two regimes, maritime (clean) and continental (polluted). The main source of marine aerosols is sea spray (sodium chloride) and aerosols are relatively few, with total number concentrations of $6.0 \times 10^8 \text{ m}^{-1}$ in more pristine environments (Junge and McLaren 1971). Continental aerosols originate from wind-generated dust, forest fires, and anthropogenic sources (mainly sulfates), and total number concentrations can be as high as $1.0 \times 10^{11} \text{ m}^{-1}$ in heavily polluted cases (Rogers and Yau 1989).

Chemical properties of aerosols [and their size] determine how readily water will condense upon them. By Raoult’s law, the presence of solute in a solution can act to lower the vapor pressure over the droplet as compared to the vapor pressure of pure water. This can be expressed as:

$$\frac{e'}{e_s(\infty)} = \frac{n_0}{n + n_0}$$
where $e'$ is the equilibrium vapor pressure over a solution consisting of $n_0$ molecules of water and $n$ molecules of solute (Rogers and Yau 1989). This property allows for droplets in solution to exist at relative humidities below 100%.

Also important to the theory of droplet formation is Kelvin’s formula, which describes the equilibrium state between a curved surface of liquid, and air. The Clausius-Clapeyron equation is an expression for an equilibrium state between a flat surface of water and air – not a curved surface. The added force due to surface tension of the curved droplet’s surface makes this correction necessary. Kelvin’s formula is stated as

$$S = \frac{e'_s(r)}{e_s(\infty)} = \exp\left(\frac{2\sigma r}{\rho L R_v T}\right)$$

where $\sigma$ is the surface tension of water, $\rho_L$ is the density of water, $R_v$ is the gas constant for water vapor, $T$ is the air temperature, and $e_s(\infty)$ is the saturation vapor pressure over a flat liquid surface given by the Clausius-Clapeyron equation.

Consideration of Raoult’s law and Kelvin’s formula together allows one to describe droplet growth at a constant temperature through a range of relative humidities. These are known as Köhler curves, and are typically calculated given a specific chemical species and temperature. The generic equation for a Köhler curve is

$$S = \frac{e'_s(r)}{e_s(\infty)} = 1 + \frac{a}{r} - \frac{b}{r^3}$$

where $e'_s(r)$ is the equilibrium vapor pressure of a solution droplet, $r$ represents the radius of the droplet, $a = 2\sigma/\rho L R_v T$, and $b = 3i m_v M / 4 \pi \rho_L m_s$ (here, $i$ is the degree of ionic dissociation, $m_v$ is the molecular weight of vapor, $M$ is the mass of the solute, $m_s$ is the molecular weight of the solute). Models that represent aerosols can use this relationship to simulate droplet activation and initial growth, depending on the aerosol chemical species, associated water content, and temperature.

Winkler (1969) and Junge and McLaren (1971) found that water preferentially condenses onto soluble chemical species with a high affinity for water. These aerosols are termed as hygroscopic, and are potential sites upon which water can condense, so they are considered to be cloud condensation nuclei (CCN). Aerosol with a low affinity for water are termed as hydrophobic and though they may also function as CCN within strong updrafts, due to their chemical structure they are often better sites for ice nucleation and therefore more likely to be considered as ice nuclei (IN) than hygroscopic aerosol (Pruppacher and Klett, 1997). Junge and
McLaren (1971) show that in the presence of varying supersaturations, the size distribution of aerosols is the primary factor contributing to a resultant CCN size distribution. Solubility only becomes a significant factor for aerosols whose mass consists of less than 10% soluble material.

Nenes et al. (2002) argue that in some cases, aerosol chemical composition is much more important than size distribution. More complicated aerosol chemistry, such as water-soluble organic compounds and dissolution of soluble gases to form droplets, can impact droplet activation enough to overwhelm dependence on aerosol size distribution and concentration. In these cases, chemical effects can outweigh the first indirect effect. For most meteorological applications, and for the purposes of this study in particular, the Dusek et al. (2006) argument is adopted as it applies to the simple aerosol chemistry implemented with the model configuration.

The findings by Junge and McLaren (1971) were important for establishing a laboratory-proven correlation between aerosol and CCN, and has held up over time. Similar results were found by Dusek et al. (2006), with 84-96% of variation in cloud nuclei concentrations due to differing aerosol size distributions. Evidence of the importance of size, rather than chemistry, can be traced back to studies predating even Junge and McLaren (1971) by 13 years, as Squires (1958) observed that droplet size distributions in maritime or continental clouds mirror their respective aerosol size distributions. Dusek et al. (2006) therefore recommends that efforts be put toward more accurately representing aerosol size distributions rather than chemistry.

These conclusions are significant because identification of aerosol composition is more costly and involves more instrumentation than identification of CCN. Rosenfeld (2006) notes that this relationship could be quite useful from the perspective of remote sensing from space: data on a global scale from instruments such as Moderate Resolution Imaging Spectroradiometer (MODIS), which can detect coarse and fine aerosol and optical depth, could be used to estimate effective radius. Unfortunately, the calculated cloud drop nuclei concentrations also have high error bars associated with them, and due to the requirements for the method of estimation, could only be retrieved over water. Capabilities of new satellite missions such as the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) platform and Polarization and Directionality of Earth’s Reflectances instrument (POLDER) may help to decrease error regarding these measurements, however, and will certainly be the subject of future studies.

Concentrations of aerosol, which have been linked to CCN concentrations, also influence cloud droplet concentrations, which has far-reaching implications. Twomey (1977) was the first
to effectively link changes in the microphysical properties of a cloud with macrophysical cloud properties. Termed the “first indirect effect,” Twomey found that differences in cloud droplet concentrations alter the radiative properties of the cloud. An increase in aerosol concentrations implies that more condensation nuclei exist for water vapor to condense upon. Considering a given liquid water content, a cloud with higher concentrations of condensation nuclei will have a higher concentration of cloud droplets, with smaller radii as compared to a cloud with lower concentrations of condensation nuclei. The presence of a higher number of small drops results in a higher reflectivity to incoming solar radiation as compared to a cloud with fewer, larger drops.

Though Albrecht (1989) is often credited with the discovery of the “second indirect effect,” precipitation suppression with increased aerosol concentrations. This idea had been around for at least 30 years at the time Albrecht published. Squires (1956, 1958) was the first to demonstrate evidence of differences in droplet number concentration and size distribution for maritime and continental cumulus clouds, presumably due to differences in the respective aerosol concentrations. Squires hypothesized that differences in resultant precipitation from these clouds is also linked to CCN concentrations. Other pioneering work by Gunn and Phillips (1957) connected increased aerosol concentrations due to pollution with decreased precipitation efficiency. Albrecht modeled drizzle-producing marine stratiform cumulus clouds to determine the effects of increasing CCN on precipitation efficiency. He was able to show that the presence of increased CCN concentrations lead to decreased coalescence efficiencies, and therefore decreased precipitation. While Albrecht established this relationship for shallow stratocumulus, the relationship between increased aerosol or CCN and precipitation in deep convection is much more difficult to ascertain and continues to be an active area of research.

Within deep convection, updraft cores accelerate air parcels quickly, and as they ascend, the parcels experience reductions in pressure. The parcels adiabatically expand to compensate for the lowering pressure and work is done by the parcel, which requires energy. The source of energy for this process is the kinetic energy, represented by the temperature of the parcel itself. Significant additional energy for parcel ascent comes from the latent heat of condensation upon CCN. The importance of accurately representing aerosols becomes clearer here: for a given water vapor content and updraft, increased aerosol or CCN presence will result in increased droplet concentration, but decreased droplet size. If the temperature remains above the freezing point of water, the convective process is termed a “warm-rain” process. In warm-rain processes,
aerosol activation, droplet growth, and collision coalescence are considered to be the primary mechanisms that lead to the formation of rain, and precipitation.

In most cases of deep convection, even within the tropics, updrafts ascend higher and the temperature of the parcel will continue to decrease. At subfreezing temperatures, the energy required to allow the parcel to continue to expand can come at the expense of another phase change of the water in the parcel. With the changeover of droplets to ice particles comes a secondary, rapid “boost” of energy, which helps accelerate the parcel upward even more (Zipser 2003). The gain in elevated convective available potential energy (CAPE) due to this release of latent heat is estimated to add as much as 1000 J kg\(^{-1}\), which can be an increase in updraft invigoration by approximately 20% in areas of the Amazon that typically see surface-based CAPE values of 1000 – 1500 J kg\(^{-1}\) (Williams et al. 2002; Khain et al. 2005, 2008). Again, the importance of representing aerosol concentrations and size distributions becomes clearer: an abundance of small droplets will be able to ascend higher than few, larger droplets, meaning that in the case of increased aerosol, updrafts can be stronger.

2.2 Theory on deep convection and aerosols

Deep convection is loosely defined as convection that occurs on vertical scales that encompass most of the troposphere. All mesoscale convective systems (MCS) can be considered to be deeply convective, as they exist over the entire vertical span of the troposphere and often horizontally cover hundreds of kilometers (Houze 1993), and are fueled by moist convective processes. Updrafts within the convective cores are quite strong, on the order of several tens of m s\(^{-1}\), and penetrate the freezing level. Uptake of water by aerosol or CCN as described in the previous section is just the first step in the journey of the aerosol and water in the case of deep convection. As water ascends and experiences changes of phase, water and energy are redistributed vertically.

Within the updraft, latent heat release occurs at low levels due to condensation of water vapor onto aerosol or CCN, and occurs aloft due to freezing of droplets and ice. The net energy change due to microphysical processes in the updraft region above cloud base of a deep convective system would therefore be a gain in diabatic heating (Houze 1989). As the system matures, a cloud shield typically develops and stratiform precipitation falls within a broad area of weaker vertical motion. Within the stratiform region, the net energy change due to microphysical processes is more of a balance between heating by vapor deposition aloft and cooling by melting
and evaporation at lower levels. Though this theory applies to mesoscale convective systems (MCS), it can be extended to squall lines (Houze 2004), which are a related subclassification of MCSs. Studies of MCS (and squall line) organization and structure are important for determining the redistribution of energy and moisture within the storm environment, as well as how these processes affects surrounding environment (Houze 2004).

The controversy over the influence of aerosols on deep convection has surfaced in part due to numerous observational studies of highly variable aerosol conditions and precipitation resulting from storms over Indonesia, India, and the Amazon basin (e.g. Rosenfeld 1999; Rosenfeld and Woodley, 2003; Andreae et al. 2004). In some of these cases of tropical convection with tops of under 6 km, high aerosol concentrations prohibited any precipitation from falling to the ground. Satellite and in-situ observations show that high concentrations of aerosols within smoke plumes effectively reduce cloud droplet size and decrease efficiency of droplet coalescence due to narrowing of the CCN size distribution. Andreae et al. (2004) report that storms invigorated with polluted air produced large hail and latent heat release higher in the atmosphere than storms under a cleaner air mass. These findings present a number of challenges such as radiative effects of the “venting” of aerosol from the boundary layer into higher levels, effects of these aerosol on the water budget, and redistribution of energy within the atmosphere, at local and global scales.

Rosenfeld (2008) presents a synthesis of many of the recent observational and modeling studies and attempts to make conclusions regarding the influence of aerosol on deep convective systems (Figure 2.1). Not only is the theory by Zipser (2003) of enhanced invigoration noted, but a secondary (outflow-based) updraft invigoration is also recognized as a characteristic of high aerosol concentration deep convective cases. Higher cloud water mixing ratios present in cases of high aerosol concentrations are argued to lead to enhanced cooling by evaporation within downdrafts. Through the enhanced cooling and downward momentum, these more invigorated downdrafts can spawn secondary updrafts as they push ambient air upward, leading to more precipitation (Rosenfeld 2008).

The effects that have been described thus far are solely due to what are termed “microphysical” mechanisms, and “radiative” effects of aerosol are also undoubtedly intertwined within the dynamical and thermodynamical processes that occur within not just deep convection, but the atmosphere at large. Aerosol effects are also highly dependent upon the environmental conditions in which they exist, as rates of conversion are dictated by factors such as the presence
of moisture, temperature, and wind velocities. Because of these inherent complexities, it is incredibly difficult to separate the microphysical and radiative effects using observational datasets. Therefore, models are an appropriate tool to use to disentangle and quantify contributing factors.

2.3 Relevant numerical studies on aerosols and deep convection

A few meteorological models have incorporated aerosols and aerosol chemistry (e.g. WRF-Chem, CRM), but aerosols are so chemically diverse and computationally expensive to represent that several studies skip the step of representing aerosols, and take advantage of the findings of Junge and McLaren (1971) and Dusek et al. (2006) in representing CCN instead (e.g. van den Heever 2004, van den Heever et al. 2006, Seifert and Beheng 2006a, 2006b). The issue of aerosol effects on precipitation has therefore been indirectly studied by using CCN or CCN droplet spectra as a starting point, instead of aerosol themselves. This is an important intermediate step that has to be taken before confidence can be gained in linking of these two as-of-yet-separate components of the atmosphere: atmospheric chemistry, and meteorology. It is a step removed, however, as not all aerosol are CCN, and not all CCN are aerosols.

In many meteorological models, such as the Regional Atmospheric Modeling System (RAMS) aerosol are not represented explicitly, but rather an aerosol “activity spectrum” is used to approximate the number of aerosol activated for a particular supersaturation. This is implemented as a power law relation, a form of which was first used by Twomey (1959):

\[ N_c = C s^k \]

with \( s \) being the percent supersaturation, \( s = (S-1) \times 100\% \), \( N_c \) being the number of nuclei per unit volume activated at supersaturations less than \( s \), and \( C \) and \( k \) being parameters that are representative of conditions for different airmass types. Some skepticism is expressed by Hobbes (1993), who states that the dependence of numerical cloud models on the value of \( k \) can be quite weak. This may not be applicable to cloud models, however, as resolutions of 1 km or less can more accurately resolve convective updrafts (Bryan et al. 2003), and therefore, values of \( S \) and estimated number of activated nuclei. Typical numbers for a continental case would be \( C=1.26 \times 10^9 \) m\(^{-3}\) and \( k=0.308 \), and for a maritime case \( C=1.0 \times 10^9 \) m\(^{-3}\) and \( k=0.462 \) (Khain et al. 2001).

Banta and Hanson (1987) were the forerunners of examining the influence of aerosols on convection within modern meteorological model. They realized the importance of adequately
capturing the process of collision/coalescence in cloud microphysics, represented as a statistical “autoconversion” rate. Choice of a proper autoconversion method allows for a more accurate representation of the size and number of raindrops, and therefore the evolution of the thermodynamic environment. Using a 2D cloud model of Tripoli and Cotton (1986) which included ice microphysics and a Manton and Cotton (1977) autoconversion scheme sensitive to a threshold cloud droplet number, Banta and Hanson bypassed explicit representation of aerosol or CCN by varying in-cloud droplet concentrations from representative maritime to continental numbers in a thermodynamic environment that produced a thunderstorm. The model was run at 250 m horizontal resolution and 200 m vertical resolution. Results showed that warm-rain processes were suppressed for increased droplet concentrations, and graupel and other ice particles were produced primarily through cold processes (such as riming, ice aggregation, and vapor deposition). Decreased droplet concentrations yielded a lower cloud base and graupel formed mainly by freezing of raindrops.

The spectral microphysics 2D cloud model used within the Hebrew University Cloud Model (HUCM) is first described within Khain and Sednev (1999) and is used, with modifications, for many other ensuing studies. This scheme was quite advanced when it was first introduced, and still is quite advanced when compared to other current schemes, featuring 33 bins for mass and number of several hydrometeor species: cloud drops, ice crystals (plate, dendrites, and columns), snowflakes, graupel, and hail/frozen drops. A unique aspect of this model is the consideration of aerosol. 33 mass bins are set up for aerosol particles, the number of which become activated are dependent upon ambient supersaturation and critical radii of the particles, as determined by Köhler theory. The microphysical processes of CCN nucleation, ice nucleation, condensation, evaporation, deposition, sublimation, freezing, melting, collisions of drops and ice both independently and together, and breakup of drops and snowflakes. Scavenging of aerosol particles by uptake of water and collision with hydrometeors is included, but particles are assumed “lost” thereafter and cannot be activated again.

The meteorological scenario under study in Khain and Sednev (1999) was a typical Mediterranean wintertime sea breeze convection case, carried through with varying aerosol types and concentrations. The simulations were 2 hours in duration, and featured a horizontal resolution of about 1.5 km and 41 vertical levels. They used an empirically determined distribution of aerosol typical for maritime aerosol environments common to the eastern
Mediterranean and ran the model for experiments contrasting runs with total number concentrations of 100 cm$^{-3}$, 500 cm$^{-3}$, and 1000 cm$^{-3}$; alternating salt (NaCl) and sulfate [(NH$_4$)$_2$SO$_4$]. Strong microphysical responses were elicited in each of their cases. They found that lower aerosol concentrations are much more conducive for rain formation, and precipitation occurs in the vicinity of development of the cloud. Higher aerosol concentrations were found to delay the formation of rain, and more water mass was lofted above the freezing level. In the simulations, precipitation was diminished, and tended to fall further downwind (as a function of ambient wind speed). Changes in chemical composition were observed to alter the concentration of activated droplets, and resulted in precipitation differences of 25-30%.

This study formed a very comprehensive and simple experiment of modeling aerosol-cloud interactions, and set a sufficient platform for following work. Khain et al. (2001) used their model to show that high mixing ratios of liquid water at temperatures of up to -38°C can occur due to higher concentrations of CCN within convective environments, and that the number of CCN can influence important environmental factors such as the height of the glaciation level. Thus far, the emphasis of these modeling studies had been deciphering microphysical differences due to CCN concentrations and types, but it was becoming clear that the simultaneous effects on dynamics of convective systems was just as important, and the tools they had developed were suitable for such investigations.

van den Heever and Cotton (2004) was perhaps the first study to uncover through modern numerical simulations how increased aerosol concentrations could invigorate convection. A subsequent study by van den Heever et al. (2006) tested the effects of aerosol size distribution on precipitation in deep convective storms by altering the CCN, giant CCN (GCCN), and IN concentrations independently. A version of the Regional Atmospheric Modeling System (RAMS, Pielke et al. 1992, Cotton et al. 2003) is used with the two-moment bulk microphysics scheme of Meyers et al. (1997) updated to include two categories for cloud droplets, and activation of CCN, GCCN and IN based on a lookup table formulated from a more complicated bin model. The model setup employed four grids with a horizontal resolution of 500 m in the innermost domain and 36 vertical levels. CCN, GCCN, and IN were initialized with idealized profiles based on data from the CRYSTAL-FACE field campaign on clean and polluted days. Typical concentrations for clean cases were in the range of 300 cm$^{-3}$ for CCN (here, particles with diameters of 1 μm or less), 0.5 cm$^{-3}$ for GCCN (here, particles with diameters of 1-50 μm),
and 0.1 m$^{-3}$ for IN. Typical concentrations for polluted cases were in the range of 1000 cm$^{-3}$ for CCN, 500 cm$^{-3}$ for GCCN, and 15 m$^{-3}$ for IN; the highest concentrations were found within the boundary layer except for the case of IN, for which the highest concentrations were found at $\sim$3 km.

The effects of varying CCN on dynamical and microphysical properties of the simulated thunderstorms in Florida were the most pronounced. Updraft velocities in the developing stages of the storms consistently increased with increased CCN concentrations, while surface precipitation decreased due to inhibition of the warm-rain process and enhanced upward transport of liquid relative to the clean case. During the mature and dissipating stages of the storms, higher concentrations of GCCN and IN were observed to have more influence on updraft velocities by a hypothesized “second mode” of convection which occurs higher in the storm, where IN and larger drops from GCCN have more effective ice-phase interactions which resulted in a stronger latent heat release.

Lynn et al. (2005a) incorporated a simplified version of the HUCM spectral microphysics scheme within the 3D fifth-generation Pennsylvania State University – NCAR Mesoscale Model (MM5) and contrasted simulations using the bulk Reisner microphysics scheme (Reisner et al. 1998) against the spectral scheme. As with the HUCM, 33 mass and number bins represent cloud drops, ice crystals (plate, dendrites, and columns), snowflakes, graupel, and hail/frozen drops. The microphysics setup from the HUCM was retained except for a reduction in the number of size distributions representing ice processes (from six to three). Aerosol in the spectral microphysics simulations was assumed to be all NaCl, and a low concentration and high concentration case were run. Nested grids were employed for all three simulations, with a horizontal resolution of 1km on the innermost domain, and 35 vertical levels.

The spectral microphysics cases with low aerosol concentrations and high aerosol concentrations both produced small convective clouds, which dissipated and initiated new convection along the outflow boundaries of the previous convection. The secondary convection in the low aerosol case had smaller updrafts and did not ascend as high in the atmosphere, but exhibited more rapid cloud growth with respect to the high aerosol case. This corroborates well with the findings of Khain and Sednev (1999). The Reisner scheme’s primary and secondary convection had stronger updrafts and shorter cloud lifetime than either of the other cases. The authors attribute this result to method of parameterization of terminal fall speed.
A subsequent study by Lynn et al. (2005b) used the same modeling platform to simulate a squall line over Florida. The spectral microphysics scheme performed better in terms of radar reflectivity, total precipitation, storm structure and shape, and maximum precipitation amounts, though precipitation was overpredicted. Bulk schemes within this study included Reisner (Reisner et al. 1998), the Goddard Space Flight Center parameterization (Tao et al. 2003), and another scheme of Schultz (1995). Overall, the studies of Lynn et al. (2005a, 2005b) successfully used a highly complex spectral microphysics scheme within a widely used 3D meteorological model to show that for deep convective clouds with mixed-phase regions, increased CCN leads to lower drop collision efficiency, lower total precipitation amounts, and a delay in onset of precipitation.

Khain et al. (2005) used the 2D HUCM model with 33-bin microphysics to show effects of varying CCN on deep convection, in particular, secondary convection formed from outflow of primary convection as in Lynn et al. (2005a, 2005b). Based on the results of previous studies, the microphysics within HUCM were updated to a new collision scheme, autoconversion parameterization, and a treatment of droplet-droplet and graupel-droplet collisions in turbulent flow. Two aerosol environments (low and high concentrations) were tested in two different types of thermodynamic conditions: an unstable summertime Texas case, and a more stable oceanic case. Aerosol concentrations and size distributions did not vary with height, and were all assumed to be NaCl. The high case and low case had distributions according to Twomey (1959) of the same settings as Khain et al. (2001) (see beginning of Chapter 2.3). The results of this study provide evidence of reduced precipitation efficiency with increased aerosol concentrations due to greater rates of evaporation and sublimation.

Several studies since these have focused on testing the influence of aerosol, and other environmental factors such as wind shear, humidity, and turbulence on deep convective storms, a summary of which can be found in Tao et al. (2007). An important addition to the literature from Tao et al. (2007) study of three deep convective systems using a 2D model with bin microphysics is the importance of evaporative cooling and low-level convergence on secondary convection.

On the topic of dimensionality dependence, Phillips et al. (2007) showed that larger vertical velocities and mass fluxes in updraft cores and higher mass fluxes in downdrafts are simulated in 3D deep convective simulations as compared to 2D. Lee et al. (2008) notes that for this reason,
secondary convection induced by downdrafts may not be as accurately simulated when using a 2D model instead of a 3D model. Interestingly, the same thermodynamic environment was used for the 3D simulation by Phillips and Donner (2006) and a 2D simulation by Lee et al. (2008), and similar results with regard to precipitation were arrived at. Both cases of increased aerosol led to increases in precipitation despite differences in model microphysics and resolution. This lends strength to the concept that processes leading to enhanced precipitation in cases of increased aerosol can be represented by models with less sophisticated, bulk microphysics and 2D simulations, such as the present study performs with the WRF-Chem.

Khain et al. (2008) introduces an attempt at classifying different types of aerosol effects on surface precipitation. Their classification schematic incorporates wind shear, humidity, and aerosol effects, the combinations of which lead to either a net loss or gain in condensate. Also, it should be noted that their scheme is appropriate only for convective systems reaching at least 4 km, and include ice and mixed-phase precipitation. This schematic, while highly simplified, addresses the issues core to the influence of aerosol on deep convective precipitation and provides a good system of orientation for past, current, and future studies.

Three studies have been carried out recently that include atmospheric chemistry within the meteorological model: Ekman et al. (2007), Lee et al. (2008), and Ntelekos et al. (2009). An interesting commonality between two of these studies is the use of meteorological conditions as ascribed by model or reanalysis data, rather than an idealized environment.

The first of these is a simulation of a cumulonimbus cloud was carried through by Ekman et al. (2007) using a 3D cloud resolving model (CRM) that incorporates aerosol in the form of CCN (hygroscopic particles), IN (hydrophobic particles), and double-moment bulk microphysics as formulated by Wang and Crutzen (1995). An uncommon feature of this model is its sophisticated aerosol chemistry module. This module represents aerosol species of sulfate, organic carbon, black carbon, and mixtures thereof using a multi-modal approach. Aerosol chemistry processes include advection, mixing, dry deposition, nucleation, scavenging by hydrometeors, and aerosol-microphysics interactions. Both number and mass of all five aerosol modes are prognostic. The number of aerosols that form cloud droplets is determined by the critical radius and saturation calculated according to Köhler theory, but only the hygroscopic particles are allowed to function as CCN. The threshold-based autoconversion scheme of Berry (1967) is used. IN are allowed to loft upward with the updrafts, where they can function as sites
for heterogeneous nucleation. Once activated, CCN and IN are assumed to be scavenged and no interstitial state is accounted for (for potential re-activation).

Ekman et al. systematically varied concentrations and vertical profiles of CCN and IN, separately, from 250 cm$^{-3}$ to 3000 cm$^{-3}$ over a domain with 2 km horizontal resolution and vertical resolution of 400 m. The responses of the model to increasing CCN concentrations were all non-monotonic with respect to average updraft velocity, precipitation rates, and ice formation. The model showed similar sensitivities to the range of IN tested. This study emphasizes the importance of performing systematic, stepwise tests with several increments of aerosol concentrations, rather than the tendency other modeling studies have shown to represent more limited testing scenarios of clean or polluted conditions.

In Lee et al. (2008), two idealized experiments, representing high and low aerosol concentration profiles, were carried out using the WRF-ARW with the goal of replicating results like those of van den Heever et al. (2006) and Lynn et al. (2005a, b). The double moment bulk microphysics scheme of Phillips and Donner (2007) was used, and limited chemistry with several types of particles (sulfate, sea salt, dust, organics, and black carbon). Only hygroscopic aerosol such as sulfate and sea salt were allowed to function as CCN, while the hydrophobic aerosol types were assumed to act only as IN. By analyzing microphysical terms of the precipitation budget, and examining dynamical terms associated with near-surface convergence, this study helped clarify processes that lead to increased precipitation in cases of increased aerosol. They found that higher values of CAPE and wind shear resulted in more precipitation for scenarios with high aerosol concentrations. A delay in warm-rain onset with increased aerosol concentrations was also observed.

Ntelekos et al. (2009) was the first experiment to use the WRF-Chem with fully coupled, online chemistry to simulate the influence of different aerosol conditions on meteorology. This implementation of the WRF-Chem is the same as is used in the current study, so the reader is referred to Chapter 3.1.2 for details on the chemistry module and microphysical parameterizations. To initialize the aerosol environment, “low” and “typical” anthropogenic emissions datasets based on a national emissions inventory were used rather than a prescribed, homogeneous initializations as is common for other experiments. The model was run over two domains, with resolution of 3 km on the innermost domain. Vertical levels were set such that the lowest levels (beneath 1 km) had a resolution of 100 m.
A significant disagreement of this experiment compared with other aerosol-microphysics interaction experiments was the absence of the suppression of warm-rain processes, including the absence of a delayed onset of precipitation with increased aerosol concentrations. The authors offer possible explanations that fault an insufficient model vertical resolution or microphysics scheme not commensurate with the complexity of the meteorological scenario. Other important points of this study include the recognition that metrics commonly used for idealized scenarios such as precipitation efficiency may not be appropriate for real-world practice. Instead, they recommend adoption of a threshold-based areal precipitation gauge or the metric of maximum accumulation within a certain area.

Results of the WRF-Chem runs indicated both enhancement and suppression of rainfall due perhaps to the lack of control within the study. The open-source WRF model and publicly available chemistry module provide an opportunity for the current study to delve into the code and set up cases in a more idealized manner.
CHAPTER 3

METHODOLOGY

The physics and implementations of the models and parameterizations used will not be
detailed herein; rather, the reader should refer to the original publications on these items.
Relevant details of the models and parameterization, however, are outlined.

3.1 Modeling System

3.1.1 WRF

The Weather Research and Forecasting (WRF) modeling system is a state-of-the-art
numerical weather prediction and simulation framework developed as a community effort,
involving several governmental, university/research, and operational agencies. A few of the
more prominent members of this community are National Center for Atmospheric Research
(NCAR), the National Oceanic and Atmospheric Administration’s (NOAA) National Centers for
Environmental Prediction (NCEP), and the Department of Defense’s Air Force Weather Agency
(AFWA). WRF was constructed to be highly portable between different computational
platforms, and to be modular in code structure to facilitate adaptability (Skamarock et al. 2008).

WRF is a fully-compressible, nonhydrostatic modeling system that includes options for
“online” chemistry, physics packages for radiation, microphysics, cumulus parameterizations,
and land-surface models, just to name a few. Two dynamics solvers are available for WRF: the
Advanced Research WRF (ARW) solver developed by NCAR, which integrates nonhydrostatic
Eulerian equations, and the Nonhydrostatic Mesoscale Model solver developed by NCEP. A
positive definite scheme was added to WRF version 3.0.1, and options for monotonic advection
were added with WRF version 3.2. Both versions of WRF used within this study allow for
conservation of mass and scalar quantities, including chemical species. The user has a choice of
running “real” simulations initialized and updated with preexisting datasets, or “ideal”
simulations initialized simply with a thermodynamic profile and built-in triggering mechanism.

Some of the specifications common to WRF studies and used within the current study are
listed here. The ARW solver within WRF employs Arakawa C-Grid staggering for the
horizontal grid, and is run with a third-order Runge-Kutta time integration scheme. Prognostic
variables include wind velocity (components u and v in the Cartesian coordinate system), vertical
velocity w, perturbation potential temperature, perturbation geopotential height, perturbation
surface pressure of dry air, and hydrometeor mixing ratios of water vapor, cloud water, rain,
snow, and graupel. An additional prognostic variable used in this study is number of cloud droplets. More information regarding the structure of the model can be found in the technical note by Skamarock et al. (2008). Specifics of the model setup for the current study can be found in Section 3.1.4.

### 3.1.2 WRF-Chem

The WRF Version 3.0 (WRFv3.0.1, used in the current study) was the first public release of WRF to include the chemistry module for simulating atmospheric chemistry (WRF-Chem, Version 3.0.1, released April 2008). An updated version of chemistry (WRF-Chem, Version 3.1.1, released July 2009) is included in the WRF Version 3.2 (hereafter WRFv3.2). For simplicity, the modeling systems will be referred to from the base WRF Version employed, as results from WRFv3.0.1 is compared with results of WRFv3.2 later in this study. Historically, the linking of atmospheric chemistry and meteorological models was typically implemented in an “offline” manner, in which predetermined meteorological forcings would be used in atmospheric chemistry and aerosol transport calculations. Independent evaluation of meteorology and chemistry is not realistic, however, and mutually important parameters such as aerosol activation, droplet growth, precipitation, and even wind speed and direction, cannot be accurately represented in an “offline” simulation. “Online” implementations are the key to providing valuable insights into how atmospheric chemistry and meteorology fields co-evolve.

Though this model is certainly not the first of its kind to actively couple atmospheric chemistry and meteorology “online” (Jacobson 2006), it is perhaps the most accessible model of its kind to date as over 6000 users have registered for WRF, and thus have access to this code, at the time of writing of the WRF Version 3 technical note (Skamarock et al. 2008). The integration of atmospheric chemistry modules with this well-established meteorological model allows for an unprecedented look at interactions between these two fields of study. The computational cost of an “online” model can be quite intimidating, however. Meteorological models include up to 11 prognostic variables, depending on the physics settings, but when aerosol and gas chemistry are included the maximum number of variables increases to 323 (Gustafson et al. 2005). Such an increase in prognostic variable count is significant and can easily result in at least a tenfold increase in computation time.

The task of composing or modifying chemistry modules to be compatible with WRF began as a project at the Forecast Systems Lab (FSL) under the direction of Georg Grell (Gustafson et al.
WRF-Chem was adapted largely from the MM5/Chem (Grell et al. 2000), which has been rigorously evaluated from the standpoint of air quality meteorology (Eder et al. 2005, McKeen et al. 2003). More recently, adaptations to WRF-Chem contributed by others such as researchers at PNNL have extended the capabilities largely in the addition of cloud chemistry modules and treatment of cloud-aerosol interactions (Gustafson et al. 2005, 2006). A graphical summary of processes, including those involving cloud-aerosol interactions, is seen in Figure 3.1.

The transport of chemical species, including aerosols, is done online – with the same methods that apply to meteorological parameters as described above. The same vertical and horizontal coordinates are used for chemistry and meteorology, as well as time, so no errors result from interpolations of these fields in any dimension. This allows for both meteorological and chemical parameters to be evaluated explicitly; directly at the gridpoints of interest. This is especially important for predicting aerosol activation and evaporation, as well as scavenging of aerosol by precipitation. Advection of aerosol is also important for dispersion and air quality forecasting.

An aerosol module termed the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) (Zaveri et al. 2005a, 2005b, 2008) allows for a sectional representation of aerosol size distribution. This module has been integrated into the WRF-Chem with a choice of either four or eight bins to represent the aerosol mass and number concentrations, according to dry radius. The range of dry radii covered with these bin setups span from 0.039 µm to 5 µm (subdivisions of which can be seen in Fig.3.2). Bins are assumed to be internally mixed, meaning that the chemical composition within a bin is the same throughout the bin. Eight bins are used in the current study.

A crucial step in the direction of integrating the chemistry modules with the microphysics modules within WRF is the inclusion of aerosol activation. Descriptions of the linkages of microphysics and radiation schemes with chemistry can be found in Gustafson et al. (2007) and Chapman et al. (2008). For each type of aerosol and each size, two bins related to mass exist, and one related to water exists. One of the mass bins contains the dry aerosol mass, and the other mass bin contains aerosol mass that has been activated by water. The water is not stored in this bin, but is stored in a separate bin.
It is important to keep activated and dry (termed interstitial) aerosol separate so that uptake of water does not influence the mass of the aerosol itself and cause it to “jump” bins. Transfer of aerosol between bins is permitted through chemical processes (e.g. chemical reactions, uptake/release of gases) and physical processes (e.g. coagulation).

It is important to note that the method of aerosol activation has been grandfathered into MOSAIC from the MIRAGE general circulation model (Ghan et al. 2001, Zhang et al. 2002). Therefore, it is advisable to be wary that some of the aspects of activation parameterized with a large horizontal and vertical resolution in mind may not be directly applicable to mesoscale simulations. Examples of this include supersaturation determined by a Gaussian spectrum of updrafts (the upper limit of which was altered from 10 m s$^{-1}$ to 50 m s$^{-1}$ for use in WRF), and activation dependent upon change of cloud fraction within the grid box from the previous to current timestep. The number and mass fractions of aerosols activated at each time step are calculated for each bin individually according to Köhler theory. It is of some comfort, however, that the range of aerosol hygroscopicities, number concentrations, radii, masses, and updraft velocities chosen for this study were generally within the range tested for use in MOSAIC by Abdul-Razzak and Ghan (2002). Also, this interpretation of aerosol activation allows for activated particles to “lose” their water through evaporation and return to an interstitial state for the chance at re-activation. This is a quality unique to the WRF-Chem that does not exist in the other reviewed studies.

To complete the aerosol-cloud link, values calculated from the chemistry modules are used in a modified version of the microphysics within WRF. The number of activated droplets and water mass from each bin is summed, and acts as the basis of cloud droplet number and cloud droplet mass for use in the microphysics scheme. The addition of a new autoconversion scheme with dependence on droplet number (Liu et al. 2005) to the microphysics allows the chemistry module to add value to the meteorological model. Because of these important modifications, the Chen and Sun (2002) adaptation of the Lin et al. (1983) scheme (see Section 3.1.3) can be regarded as a limited double-moment scheme and may be used to study aerosol-cloud interactions, including direct and indirect effects.

### 3.1.3 WRF Microphysics

Though several microphysics parameterizations are available within WRF, the only one currently linked with the chemistry module is the Purdue Lin scheme (Chen and Sun 2002),
hereafter referred to as the Lin et al. scheme. Based upon the single-moment parameterizations of Lin et al. (1983) and Rutledge and Hobbs (1984), the traditional implementation of this scheme calculates the time-evolution of mass of six hydrometeor species: water vapor, cloud water, rain, graupel, snow, and cloud ice. Cloud properties are directly modeled instead of derived statistically, making this an “explicit” scheme. The shapes of the hydrometeor size distributions are assumed to be inverse exponential; with rain parameters given by Marshall and Palmer (1948), snow by Gunn and Marshall (1958), and graupel by Federer and Waldvobel (1975). Because the parameterizations account for the transfer of water among many pathways between both water and ice species, the Lin et al. scheme was regarded as a complex scheme for many years (McCumber 1991), and has been used for a wide variety of applications.

The current state of the Lin et al. scheme is detailed by Chen and Sun (2002) and is summarized here. Microphysical interactions between hydrometeor species include evaporation, sublimation deposition, condensation, aggregation, accretion, Bergeron–Wegener–Findeisen processes, freezing, melting, and melting evaporation. Cloud water is allowed to exist at temperatures as low as -40°C, after which homogeneous nucleation occurs. Cloud water and ice species can coexist between 0° and -40°C.

The Lin et al. scheme has experienced a few updates throughout the years, some of which are worth mentioning for the sake of relevance to the current study. The parameterizations that were developed for the original Lin et al. (1983) microphysics scheme were tuned to the high plains region of the United States. The applicability of this scheme to tropical regions is therefore called into question. In a review of microphysical parameterizations, McCumber et al. (1991) tested several single-moment schemes against each other in 2D simulations of a tropical squall line. A simple swap of the densities, intercept parameters, and coefficients for fall speed from those of hail (Lin et al. 1983) to those of graupel (Rutledge and Hobbs 1984) produced patterns of reflectivity and precipitation more similar to observations, perhaps because graupel is thought to be more common in the tropics than hail.

Another adaptation is the saturation adjustment scheme. With components taken from Lord et al. (1984) and Tao et al. (1989), the Lin et al. saturation adjustment scheme incorporates transactions of water across all hydrometeor species and compensates for supersaturation left over from microphysical processes at the end of the microphysics routine. In the case of a surplus supersaturation, the necessary amounts of condensation (deposition) are calculated to
remove the excess vapor and apply it to pre-existing species. In sub-saturated conditions, evaporation (sublimation) is calculated if cloud droplets (or cloud ice) are present. In this way, supersaturations are resolved in an iterative fashion, every timestep and are not permitted to cross grid cells. It should be noted that the saturation adjustment scheme has not yet been updated to include aerosol activation, however, the modifications to the microphysics within Lin et al. make this scheme an appropriate one for the current study.

New changes added to the Lin et al. scheme not included in Chen and Sun (2002) are specifically for the linking of the chemistry and microphysics modules, and are partially described in Chapman et al. (2008). Table 3.1 depicts key differences in steps of the Lin et al. scheme transition from single moment to double moment with chemistry. The author altered the activation code to test its double-moment capabilities without including full chemistry and ran some comparative simulations, which are not shown here. The developers of WRF-Chem likely built this capability into the model for testing purposes, but do not advertise it as part of the WRF modeling package or a feature of the Lin et al. microphysics scheme. This “intermediate” configuration was run with both aerosol properties and the same range of aerosol concentrations, but results are not shown here. Magnitudes of total precipitation, updraft speeds, and other meteorological metrics were close to the range of results of the single-moment Lin scheme.

Perhaps the most important addition, other than droplet number, is the new autoconversion scheme of Liu et al. (2005). The autoconversion scheme originally within Lin et al. was a simple threshold scheme similar to Kessler (1969), based on an empirically derived constant. In the previous scheme, the trigger for cloud droplets to collide/coalesce into raindrops was based upon an amount of liquid present – not any physical aspects of the droplets themselves. The new Liu et al. (2005) scheme is based upon the droplet number, the droplet size spectrum, droplet mass, and cloud liquid water content. This allows the autoconversion rate to be calculated at each timestep based on physical features of the cloud droplets, and is much more realistic. This autoconversion scheme is automatically chosen if droplet number concentrations exist within the model – otherwise the Lin et al. scheme will behave as usual, as a single-moment scheme.

3.1.4 WRF-Chem Configuration (model setup)

Tables (Table 3.2) and (Table 3.3) show model configurations for WRFv3.0.1 and WRFv3.2 as used within this study. Some of the desired configurations for chemistry were not available,
however, and were built into the model by the author. Some modifications to the model will be described at the end of this section, as well as in Section 3.2.2.

Initial simulations (Sections 4.1 and 4.2) were performed with WRFv3.0.1 according to the configuration in Table 3.2. Idealized 2D cases with a horizontally homogeneous oceanic (Section 4.1) and continental (4.2) thermodynamic environment are considered, initialized by a warm bubble (3 K) in the center of the domain. Within the idealized scenarios, insolation and surface fluxes are omitted in order to better isolate microphysical processes. The microphysics parameterization chosen is the limited double-moment Lin et al. scheme as previously described. Horizontal grid spacing is set to 500 m to resolve processes on a convective scale, so a sub-grid cumulus parameterization, boundary layer model, and surface layer model are not required. A free-slip condition is set at the lower boundary. Coriolis terms are not included due to the small size of the domain. The domain encompasses 40 km, and 61 vertical levels as determined by a stretching algorithm that automatically assigns levels within WRF are represented. A positive definite advection scheme is implemented for all prognostic variables including chemistry. Boundaries are open in the y-direction and periodic in the x-direction. All timesteps within the model are set to 1.0 s, data is output every 60 s, and the simulation is allowed to run for 90 minutes. 178 model runs were performed with WRFv3.0.1, including additional sensitivity tests (not shown) for aerosol number concentration.

The degree to which chemistry is included in this study is quite limited, as we seek to represent aerosol populations as simply as possible. For example, to approximate oceanic aerosol, atomic sodium was chosen (Na), and for continental aerosol, sulfate was chosen (SO$_4$). The Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) currently implemented within WRF-Chem includes eight aerosol types, however, and distributes their mass and number over either four or eight bins. For this study, aerosol initialization for number concentration and mass in these idealized cases is determined by a user-defined power law for the desired aerosol type (see section 3.2.2), and sets other aerosol types to zero. The WRF-Chem includes many chemistry-specific mechanisms and processes, but we preserve only the processes that are necessary for aerosol-cloud interaction, such as aerosol activation, water uptake, aerosol resuspension, and wet scavenging (Fig. 3.1).

Later simulations (Section 4.3) were performed with WRFv3.2 over a more limited range of aerosol and only for the oceanic case, for the purpose of comparison against WRFv3.0.1 results.
Updates to the model in WRFv3.2 include a choice for a monotonic, rather than positive definite advection scheme for all prognostic variables, and modifications to the Lin et al. scheme for supersaturation calculation and some sublimation terms. Recommendations for other settings relevant to the more current WRF-Chem options such as horizontal diffusion and zeroing out of microphysics variables less than a threshold value of $1.0 \times 10^{-12} \text{ g kg}^{-1}$ were found in tutorial presentations found at the WRF Working Group 11 website (http://ruc.noaa.gov/wrf/WG11). The author added a routine to the initialization code to specify vertical levels within the model, termed “sigma levels.” Though the overall number of vertical levels is reduced to 50, more of them exist within the low levels of the atmosphere in an attempt to capture important interactions within and near the boundary layer such as vertical transport and activation of aerosols.

3.2 Simulation design

This study tests the capabilities of WRF-Chem in simulating two 2D idealized deep convective cases with a range of simplified aerosol treatments. The chosen thermodynamic environments were taken from cases producing squall lines: one over the western Pacific Ocean, and another over the Amazon. For each thermodynamic environment, two sets of runs were completed: one with a stepwise range of sodium aerosol concentrations, and another with a stepwise range of sulfate aerosol concentrations. Results of each case were analyzed separately, then a new battery of runs was performed with an updated version of the model.

3.2.1 Meteorology

Squall lines are a type of deep convective mesoscale system that exhibits a linear radar horizontal reflectivity appearance (Houze 1993, Markowski and Richardson 2010). Environments with deep-layer wind shear and moist conditions at low levels are particularly conducive for these types of MCSs to develop. The direction of the shear with respect to the line of initiating convection determines, to a large extent, the type of structure the MCS will take on and where the characteristic features will be found when the system matures. In the case of squall lines where the vertical wind profile is orthogonal to the initiating line, dominant features include a convective region in which the convective cores form, ascending front-to-rear flow, descending rear inflow, and a stratiform region trailing the convective region (Houze 1989). Convective rain is found directly under the updraft cores, while stratiform precipitation tends to fall behind the updraft due to ascending front-to-rear flow. This case is known as trailing stratiform, and is the dominant squall-line type, but other cases also exist: a leading stratiform
case can develop if the deep-layer shear has a strong rear-to-front component, or a parallel stratiform case can develop if the deep-layer shear is aligned parallel to the orientation of updrafts (Markowski and Richardson 2010).

Cases of squall lines with both trailing (e.g. Rotunno et al. 1988) and leading (e.g. Parker and Johnson 2004a, b) stratiform regions have been modeled extensively with both 2D and 3D models, and the theory of squall line development and maintenance is well developed from the observational side (Rutledge et al. 1988, Rotunno et al. 1988, Johnson and Hamilton 1988, Houze et al. 1989). Because the dynamic and thermodynamic forcings behind squall lines are relatively well identified, they are an attractive testbed for investigation of the influence of “external” variables such as aerosols. Recently, a few studies (e.g. Khain et al. 2005, 2008; Lynn et al. 2005b) have explored this topic, as does the current study.

Choice of model dimensionality (2D vs. 3D) can impact convective simulation results, and the differences are worthy of investigation. Rogers and Yau (1989) state that wind speed shear and wind directional shear cannot be sufficiently represented using a 2D model, but this is relevant for studies that seek to model rotational circulations such as mesocyclones and tornadoes within deep convective storms. Tompkins et al. (2000) showed through systematic tests of deep convective systems in both 2D and 3D cloud-resolving modeling systems that highly organized two-dimensional systems such as squall lines can be sufficiently modeled in 2D. This lends credence to the argument that a 2D model is sufficient to simulate squall lines, but the case of airflow around convective cells in a 2D simulation is still of concern. Because air is limited to moving essentially in two dimensions, air is not permitted to flow around the storm, possibly leading to intensified upshear development. For the purposes of the current study, this may be relevant only in consideration of cross-comparing effects of aerosol on different thermodynamic environments. Two different soundings were therefore chosen for this study, based on cases supporting highly organized two-dimensional deep convection (squall lines) for oceanic and continental regimes.

The oceanic squall line case (Fig. 3.3a) is represented using a sounding obtained over the western Pacific Ocean, on 22 February 1993 during the Tropical Ocean Global Atmosphere (TOGA) Coupled Ocean-Atmosphere Response Experiment (COARE). The focus of this experiment was to capture characteristics of organized mesoscale convective systems over the ocean, and to observe the influence of fluxes between the ocean surface and the atmosphere in
deep convective cases. Trier et al. (1996) provides a sounding of height, pressure, potential temperature, water vapor mixing ratio, and \( u \) and \( v \) wind components in tabular format, which they used in numerical simulations with and without surface fluxes and ice microphysics. To construct the sounding, observations were compositied from P-3 aircraft and rawinsonde data, and low-level aircraft data in the immediate vicinity of the convective system within a 6-hour period that encompassed the event (Trier et al. 1996). The environment features significant low-level wind shear directed nearly parallel to the leading edge of the squall line, high CAPE of \( \sim 3000 \) J kg\(^{-1}\), and moist conditions through the depth of the troposphere. For the purposes of preventing the system from traveling out of the domain in the 2D simulation in the current study, the shear was reduced by subtracting 10 m s\(^{-1}\) from the \( u \) component of the wind. From the sounding, the top of the boundary layer was deduced to exist at approximately 930 mb, or 670 m. In terms of the Khain et al. (2008) proposed classification scheme, the experiments with this sounding would be considered as a case of tropical squall lines (upper right of their schematic).

The continental squall line case (Fig. 3.3b) is based upon a sounding from 26 January 1999 during the Tropical Rainfall Measuring Mission Large-Scale Biosphere-Atmosphere Experiment (TRMM LBA), which was situated over Rondonia, Brazil (Lang et al. 2007). This experiment set out to examine the dynamical, microphysical, electrical, and diabatic heating aspects of continental convection. The sounding was created using data from a nearby rawinsonde launch approximately two hours prior to the development of the squall line, as well as tethersonde data for the lowest 1 km. For use within the current study, the tabulated sounding data was acquired from Steve Lang. Features of this meteorological setup include strong low-level shear, moist low-level conditions, and high CAPE of \( \sim 2500 \) J kg\(^{-1}\). The sounding indicated that the boundary layer height was at approximately 900 mb, or 670 m. In terms of the Khain et al. (2008) proposed classification scheme, the experiments with this sounding would be considered as a case of continental squall lines (far upper right of their schematic). Model runs with this scenario did not produce significant convection after the initial pulse triggered by the temperature perturbation, but future work will seek to resolve this issue. Therefore, the author chose to not include model runs of the continental case within this document.

3.2.2 Chemistry

A point was made from the outset of this study to simplify the chemistry as much as possible in order to isolate the cloud-aerosol interactions within the WRF-Chem and increase control.
The choice of aerosol within this study is similar, but not identical to, Khain and Sednev(1999). This study is also similar in the way we represent aerosol: one species at a time. To represent maritime aerosol, sodium (Na) was chosen, and to contrast the maritime with a continental aerosol, sulfate (SO$_4$) was chosen. The approach to this study is to initialize the meteorological model with one of two thermodynamic environments, and perform systematic, stepwise tests with a range of aerosol according to total number concentration, for both sodium and sulfate.

For total aerosol number concentrations within the boundary layer, appropriate numbers were generated according to a power-law relationship using the distributions shown by Patterson (1982) (Fig. 3.4). Extensions on either side of the number distributions were added to encompass lower or higher values seen in field studies of observations of aerosol (Fitzgerald 1989, Fitzgerald 1991, O’Dowd et al. 1997, Porter and Clarke 1997, Andreae, et al. 2000, Heintzenberg et al. 2000, Quinn et al. 2000).

\[
\frac{dN}{d(\log r)} = Ar^k
\]

Values of $A$ (intercept on log-log scale) and $k$ (slope on log-log scale) were determined using a MATLAB routine for power-law fits. The values of $A$ ranged from $1.929 \times 10^{12}$ to $1.929 \times 10^{9}$, while the $k$-value of -2.518 was not altered through the course of the study. Fixing the $k$ value effectively proportionalizes the both number and mass bins relative to each other (at initialization), and provides an extra measure of control in the simulations. Aerosol concentrations above the boundary layer were around a factor of 100 less than the boundary layer concentrations for each case tested.

The range of total number concentrations covered by this study are $1.50 \times 10^6$ m$^{-3}$ to $1.50 \times 10^6$ m$^{-3}$. In terms of total number concentration, the values used in this study compare well with other studies of CCN/aerosol-cloud interactions, which range between $0.50 \times 10^8$ m$^{-3}$ and $1.50 \times 10^9$ m$^{-3}$ (see Chapter 2.3). Model runs were also performed with lower than typical concentrations (as low as $1.50 \times 10^6$ m$^{-3}$) to test the boundaries of the microphysics parameterization. Because of the wide range of aerosol conditions tested, they are referred to in terms of “per cubic meter” instead of “per cubic centimeter” as is the norm for other studies of this nature.
Each of the eight size and number bins used in MOSAIC (e.g. Fig. 3.2) was initialized according to the power law shown above. First, total number concentrations were calculated for each bin after integrating the power law (below).

\[
\frac{\#}{bin} = \frac{A}{k \cdot \ln(10)} [(r_2)^k - (r_1)^k]
\]

For each mass bin, aerosols were assumed to be spherical, allowing their mass to be calculated given the density and total number concentration. These calculations were incorporated within the chemistry initialization module and linked with the external configuration list for WRF, so adjustments could be made on-the-fly. Masses ranged from 0.0079-9.745 µg for Na. The density used for Na is 2.20 g cm\(^{-3}\), and the density used for SO\(_4\) is 1.80 g cm\(^{-3}\). Hygroscopicities for these aerosol species are 1.16 and 0.5, respectively. It should be noted that hygroscopicities for Na and NaCl are identical within MOSAIC, as are SO\(_4\) and (NH\(_4\))\(_2\)SO\(_4\) (the latter of which were used in Khain and Sednev 1999).
CHAPTER 4
RESULTS

4.1 Oceanic Case, WRF Version 3.0.1

Vertical distributions of aerosol used for the 32 test runs contrasted here can be seen in Fig. 4.1. Number concentrations summed over all eight bins (Fig. 4.1a) of sodium shown for time of initialization of the model are also valid for sulfate. Each line denotes a separate condition for initialization of the model; so this figure (and many of the following figures) show the results of 16 model runs simultaneously. Note that the model includes two levels within the boundary layer and that aerosol concentrations above the boundary layer are reduced by a factor of 10-100. The runs were set up in groupings of six such that all six runs within a group would possess the same aerosol concentrations above the boundary layer – hence the “forked” appearance of these profiles. Mass concentrations summed over all bins are shown for sodium (Fig. 4.1b), but would look similar for sulfate.

Total precipitation over the 40-km domain with time is shown in Fig. 4.2a for all runs with sodium, and in Fig.4.2b for sulfate. These plots look nearly identical, presumably due to the high hygroscopicity values of both aerosol under consideration. Because of their high hygroscopicities, all aerosol are effectively acting as CCN. A close examination of sulfate and sodium results reveals slight differences in the cases of intermediate total aerosol concentrations. Sulfate is slightly more sensitive to the median range of this study (~1.23 x 10^6 m^-3) than sodium. In both cases, the lower aerosol concentration runs produce more precipitation initially than the high aerosol cases, but the high cases overtake the low at approximately 78 min. Low aerosol cases produce precipitation that reaches the surface by 20 minutes, while the high cases tend to cluster around 30 minutes. The convection in all cases of aerosol concentration and type carries through the full 90 minutes of the simulation. Clustering tendencies of low and high aerosol concentration results are beginning to appear so will be referred to LA (low aerosol total number concentrations) and HA (high aerosol total number concentrations) hereafter.

The differences in precipitation characteristics between LA and HA cases becomes starker upon considering the conditional rain rate (Fig. 4.3). The consideration of conditionality of precipitation shows that LA cases not only begin to experience precipitation 10-15 min. earlier than the HA cases, but the maximum rain rate is lower, and takes longer to achieve. The HA cases cluster upon a precipitation start time of about 30 min., after which an intense rainout
occurs approximately five minutes later. A secondary maximum occurs in LA cases around 63 min. and in HA cases at 73 min. Timing of precipitation onset, timing of maximum rain rate, and even the values of rain rate are nearly identical once again between sulfate and sodium cases. If any microphysical differences exist between cases of different aerosol types, they should manifest in domain-total precipitation or rain rate. Because these two metrics are nearly identical, for simplicity of analysis only the sodium case will be considered for the remainder of Section 4.1.

A sense of the overall hydrometeor profile with time can be achieved by looking at the domain-total liquid, snow/ice, and graupel content in Figure 4.4. The five plots show a gradual succession of model runs, stepwise through the range of total aerosol concentrations. No notable change is observed in initial cloud height, but mass of liquid water builds to higher values and heights in the cloud as time progresses for HA cases. Graupel and cloud ice increase are observed to increase in value and height with increasing aerosol concentrations, but take longer to build vertically. By the end of the model run, substantially higher values of all hydrometeor species exist over the domain. To determine the relative dependence of hydrometeor presence by aerosol activation, maximum droplet concentration per grid cell is shown in Figure 4.5. Similar patterns are seen in all different total aerosol concentration cases and are magnified with increasing concentrations. The peak in maximum droplet concentration of $6.0 \times 10^8 \text{ m}^{-3}$ within the first 5 min. of the simulation is due to activation within the updraft generated by the initial temperature perturbation. Other peaks at 18 min. and 25 min., and more diverse peaks after 35 min. are indicative of possible secondary updrafts.

Microphysical metrics of domain total cloud water, rain, graupel, and combination of ice and snow can be found on Figs. 4.6-4.9. Domain total cloud water peaks earliest for the lowest LA cases, while the HA cases tend to cluster with a very dominant peak at 25 min. Total rainwater peaks for HA cases at 33 min. – earlier than the LA cases, which first reaches a maximum at 37 min. The HA cases are seen to take on more liquid in the form of cloud droplets, which is converted to rain very rapidly, while LA cases take on lower amounts of cloud water and converts it to rain water more gradually. The onset of precipitation in the LA case is most likely faster due to proximity of rain to the ground, as cloud and rain water was observed to be lofted higher in Figure 4.4. A snapshot of the average cloud liquid water profile at 25 min. is shown in Fig. 4.10, which indicates all HP cases contain the same average amount at this point, while LP
cases contain lower averages by a factor of 3. Ice species do not appear until after 20 min, and the first relative maximum of these that occurs is a direct effect of the initial updraft.

For 40 min. and beyond, distinct secondary maxima are evident for LA and HA cases in all hydrometeor concentrations. Maxima in domain-totals of cloud liquid water are seen to precede maxima in total rain water, which in turn precede maxima in graupel and ice species. This sequence is most likely due to a succession of updrafts. Figs. 4.11 and 4.12 provide support for this observation, as total graupel and ice species also lag peaks in maximum updraft speed by approximately 10 minutes. HA cases follow a definite trend, with maximum updraft speeds on the order of 6-7 m s\(^{-1}\), while LA cases follow a different trend of gradually diminishing updrafts. Updraft maxima in HA and LA cases do not coincide in time, either, with the exception of the initial updraft. This could be due to differences in precipitation loading between cases, including the vertical distribution of hydrometeors, as well as microphysical effects.

Downdrafts also follow two distinct patterns for HA and LA cases. LA cases achieve a less intense “maximum” downdraft speed of -4.0 m s\(^{-1}\) at approximately 23 minutes, which coincides with the first precipitation maximum. This downdraft is therefore due to precipitation loading, with a possible contribution by evaporative cooling. HA cases exhibit two noteworthy maxima in downdraft speeds, one at approximately 25 min. and another at 65-70 min. Precipitation occurs at both of these points, but at 65-70 min the conditional rain rate is significantly lower than the initial peak. Therefore, it is possible that the secondary stronger updrafts and downdrafts seen within the HA cases may be due to microphysical effects.

The observations thus far have largely not considered the location of features in the vertical. Cross-sectional plots of vertical velocity, temperature perturbation, and the freezing level through time (Figs. 4.14-21) depict the evolution of the most highly contrasting cases in this study: the lowest \((1.50 \times 10^6 \text{ m}^{-3})\), to the highest total aerosol concentration cases \((1.50 \times 10^9 \text{ m}^{-3})\). At 10 min. (Fig. 4.14), both simulations look identical with heating due to condensation occurring within the updraft, heating to the east due to compressional warming, and cooling above the updraft due most likely to expansion as the freezing level has not yet been reached. By 25 min. (Fig. 4.15), some differences begin to appear. The initial updraft, aided by condensational heating, ascends to a higher level in the LA case than the HA case, which is due to the unloading of some hydrometeor mass in the form of surface precipitation. At 35 and 45 min. (Figs. 4.16-17) the remnants of the initial updraft can be seen continuing to rise, while new
updrafts form near the surface. Strongest updraft speeds and positive temperature perturbations occur above the freezing level in the LA case, yet remain below the freezing level in the HA case. The developing updraft in the HA case appears more cohesive and is oriented more upright than its LA counterpart. At 55 and 65 min., (Figs. 19-20), updrafts are seen to intensify above the freezing level as microphysical processes releasing latent heat take over, such as freezing of cloud droplets, or rain freezing to become graupel. Compressional warming intensifies to the east of the updraft in the HA case, while the LA case becomes more chaotic looking and forcings weaken. The last two time periods shown (Figs. 4.19, 4.20) are the most disparate of the simulations, showing continuing development of updrafts within the HA case along what may be the leading edge of a developing squall line, while activity in the LA case becomes minimal and constrained to below the freezing level.

Another look at what occurs in the vertical with time is depicted by Figs. 4.22-4.25. In Figure 4.22, each plot shows a maximum vertical velocity that occurs according to each model vertical level with time. The structure of the LA case updraft is dominated by the initial temperature perturbation, which travels rapidly upward. While the magnitudes of vertical velocities within the first 25-30 minutes of all cases are similar, their vertical position is not. The HA cases are likely increasingly suppressed by the high amounts of cloud liquid water they contain relative to the LA cases (Fig. 4.10). This helps explain the longevity of the initial updraft in the HA cases, which can be seen extending up to near 11 km by the end of the model run. Cloud droplets contained within this initial upward burst continue to add energy to the updraft through freezing, as postulated by Zipser et al. (2003). The first maximum downdraft speeds (Fig. 4.23) occur with the strongest conditional rain rates (Fig. 4.3) and are stronger in the HA case. Through time, HA cases show that the stronger updrafts aloft have accompanying stronger downdrafts above the freezing level.

The final plots shown for results of the oceanic sounding compare horizontal winds for the extreme LA and HA cases at initialization, 25 min., and 85 min., in order to look for features that indicate squall line development. The initial wind field is seen in Figure 4.26a, which is to the west at low levels (perpendicular to the propagation of our updrafts), to the east at mid-levels, and to the west at upper levels. The developing updrafts essentially block and redirect the horizontal wind, causing the easterly winds to be entrained upward within the updraft and into
higher levels of the system. Westerlies at mid-levels are observed to intensify near the updrafts. These features are suggestive of a developing squall line.

4.2 Oceanic Case, Comparison of WRF Version 3.0.1 with Version 3.2

Profiles of the initial aerosol conditions for all WRFv3.0.1 and WRFv3.2 runs are depicted in Figures 4.27 and 4.28. The resolution of the vertical resolutions is immediately apparent, as WRFv3.0.1 with stretched vertical levels capture three points under 850 mb, while WRFv3.2 with prescribed levels capture nine points under 850 mb. Because of the improved resolutions at low levels, the WRFv3.2 initialization appears to have a lower boundary layer, but in fact this is just an optical illusion due to the method used to visualize the profiles. For the comparison of WRFv3.0.1 and WRFv3.2 runs, an abbreviated range of concentrations were used to reflect concentrations closer to those observed in field studies, and to highlight the range of sensitivity of the model. As in Section 4.1, total aerosol number concentration profiles (Fig. 4.28) are valid for both sodium and sulfate, while total mass concentrations are depicted for sodium only. Also as in Section 4.1, due to negligible differences in results due to aerosol types, sodium will be the type of aerosol depicted in all future discussion and figures. In WRFv3.2 cases, a single total aerosol concentration was assigned to heights above the boundary layer in order to increase control within the study.

Total precipitation over the domain can be seen in Figure 4.29, with added metrics of results of single-moment Lin et al. test runs overlain. Severe contrasts between WRFv3.0.1 and WRFv3.2 runs are immediately apparent, as precipitation in the WRFv3.0.1 runs falls more consistently and at a higher rate throughout the 90 minute run. The onset of precipitation in all WRFv3.2 runs occurs near 20 minutes, in stark opposition to the highly segregated precipitation start times seen with increasing aerosol concentrations in WRFv3.0.1 runs. WRFv3.2 runs preserve the characteristic of LA cases producing precipitation earlier than HA cases, but the delay is only a matter of 1-2 minutes. Rainfall from the LA cases remain higher than from HA cases for the WRFv3.2 runs, also in opposition with the WRFv3.0.1 output. Comparisons with single-moment Lin et al. output (no chemistry) suggest that WRFv3.2 results may be more rational.

Comparison of conditional rain rates (Fig. 4.30) between the two sets of runs reveal just how much more intense rainfall was for the WRFv3.0.1 runs. Here, conditional rain rates reached a maximum at 36 min. of 19 mm hr\(^{-1}\), while in WRFv3.2, the maximum was reached ten minutes
earlier and was a factor of ten lower. In all runs a secondary maximum in conditional rain rate was observed, which was higher for the HA cases in WRFv3.0.1 and higher for LA cases in WRFv3.2. The initial precipitation occurs much earlier in all cases in WRFv3.2 as compared with WRFv3.0.1 runs. Conditional rain rates begin at 10 minutes for LA cases in WRFv3.2 runs and increase rapidly, while in HA cases the onset occurs as much as 5 min. later. The magnitudes of conditional rain rate in the WRFv3.2 runs are more consistent with output seen from the WRFv3.0.1 Lin et al. (no chemistry) run (black line).

Hydrometeor maxima over the domain with time also contrasted strongly between WRFv3.0.1 and WRFv3.2 runs, as shown in Figure 4.31 and Figure 4.32. Consistent contours and shading at the same levels in each figure call attention to the high levels of liquid water occurring in WRFv3.0.1 runs relative to WRFv3.2 runs. Interestingly, values of cloud ice and graupel are comparable between the two sets of runs, though the frozen species are lofted higher into the atmosphere in WRFv3.2 runs. A substantial amount of hydrometeor still resides in the atmosphere at the end of the WRFv3.0.1 run (90 min.), while at 90 min. the majority of hydrometeors have either precipitated or are below threshold levels for contouring in the figure.

Onset of cloud droplet formation occurs at approximately three minutes for both WRFv3.0.1 and WRFv3.2 sets of runs (Fig. 4.33). Despite the same aerosol concentrations used in initialization, more droplets appear in WRFv3.2 than WRFv3.0.1. Droplet concentrations after the initial burst of formation are similar thereafter. Domain-total cloud water (Fig. 4.34) in WRFv3.2 runs reach a peak at 10-15 min., followed by subsequent maxima presumably from conversion to rain and precipitation unloading. WRFv3.0.1 runs instead tend to continually build up cloud water, perhaps due to stronger updrafts, until a critical mass is reached and cloud water is converted to rain and released (Figs. 4.30, 4.35). This sequence of build-up and release of cloud water continues through the run for WRFv3.0.1, and may be contributing to continual regeneration of updrafts through latent heat release. The single-moment Lin et al. scheme results fall somewhere in between of these two chemistry run suites.

Figures 4.35-37 show further disparate results from WRFv3.0.1 and WRFv3.2 runs. Amongst WRFv3.0.1 hydrometeors, there is a definite trend of rising mixing ratios through the run period, while in WRFv3.2 runs a definite maximum is achieved, followed by a steep downward trend. HA cases in the WRFv3.2 runs consistently produce lower mixing ratios of rain, graupel, and cloud ice, while this trend in WRFv3.0.1 is reversed after approximately 70 min. At 25 min., the
height of the cloud water maximum in the WRFv3.0.1 HA cases, the average cloud liquid water present is a factor of ten greater than the maximum average cloud liquid water content in any of the WRFv3.2 cases. The vertical distribution of liquid is also strikingly different: instead of a maximum at 600 mb, the maximum is seen at much lower levels (900-800 mb) in the WRFv3.2 case.

Maximum and conditionally averaged updraft speeds (Figs. 4.39-40) are initially the same across all runs, as they are forced by the same initial temperature perturbation. As the bubble of warm air rises and saturation occurs, differences due to the microphysics begin to manifest. The WRFv3.0.1 runs experience the same changes in vertical velocity until the first maxima is reached at 21 min., while in WRFv3.2 runs the maximum updraft speeds are reached at 19-20 min., and are lower in magnitude for HA cases than LA cases. This indicates that the increased efficiency of warm-rain processes within LA WRFv3.2 cases are better represented as rainout begins earlier and as a result, the updraft is more buoyant. The inhibition of warm-rain processes in WRFv3.0.1 may be part of the explanation of the greater maximum updraft speeds, as the enhanced cloud water mixing ratios may be contributing to updraft speeds through the release of latent heat. While WRFv3.0.1 runs go through cycles of updrafts and downdrafts (Figs. 4.39, 4.41), all cases in WRFv3.2 runs see a gradual decrease in updrafts and downdrafts after the initial plume rises.

Profiles of maximum and minimum vertical velocities with time (Figs. 4.42-45) and temperature perturbations with time (Figs. 4.46-49) depict the evolution and reveal some of the processes occurring that cause these distinct differences between WRFv3.0.1 and WRFv3.2. The initial updraft and temperature perturbation seen at 0-10 min. appears identical across all runs, though is better resolved with the increased low-level vertical resolution in WRFv3.2. Differences in the intensity and height of the updraft from time are quite striking, as a more consistent updraft remains in place over 15-25 min. in the WRFv3.0.1 cases while a more narrow updraft travels upward from approximately 750 mb to 600 mb over the same time period. Similar values are seen in the maximum temperature perturbation with these runs, which changes character around approximately 30 min. After this, more latent heat release occurs at increasing heights in WRFv3.2 cases, while higher temperature perturbations remain constrained to below the freezing level (~500 mb) in WRFv3.0.1 runs.
Updrafts and downdrafts in all cases of WRFv3.2 runs (Figs. 4.43, 4.45) reach maximum values as a direct consequence of the initial temperature perturbation that triggers convection in the model runs. Temperature perturbations (Figs. 4.47, 4.49) that occur at low levels after the initial perturbation rises are likely due to evaporation and sublimation of hydrometeors in weak subsidence that occurs in an attempt to resolve the initial perturbation. Instead of diminishing, the updrafts in all WRFv3.2 cases (Figs. 4.42, 4.44) are observed to go through cycles of build-up and release of hydrometeors, with bursts of latent heat both at lower and upper levels assisting the continued development of what may be the beginnings of a squall line. Distributions of the u wind component at different points in the system evolution for LA and HA WRFv3.01 cases (Figs. 4.50a, b) show the development of strengthening rear-to-front flow on the west side of the system at mid-levels, which is stronger in the HA case, and the development of a venting front-to-rear flow at mid to upper levels, also stronger with in the HA case. The WRFv3.2 LA and HA cases are notably devoid of these features, as it appears the initial temperature perturbation is absorbed or resolved by the environment.
5.1 Oceanic Case, WRF Version 3.0.1

Trier et al. (1996)’s 3D simulation of this system transitions to a squall line within 2 hours of initiation. Analysis by Trier et al. (1996) begins after the system evolves into a squall line, so the current study can unfortunately not be examined side-by-side with this analysis. Overall, it was perhaps unrealistic to expect that a squall line would develop to maturity within the 90-minute time frame of this study, and future work will have to consider a larger domain and model run time of 5-6 hours at a minimum. Despite the lack of development of a squall line, the model runs did effectively simulate cases of deep convection. Maximum vertical velocities reached 7 m s$^{-1}$ within the WRF-Chem due to forcing by the initial temperature perturbation. Vertical velocities of this magnitude were present within the squall line modeled in Trier et al. (1996) as well. Since the temperature perturbation responsible for initiating convection is the dominant forcing that occurs within the time period of this study, the oceanic case was analyzed in terms of a sensitivity study to changes in initial aerosol concentration and type.

Very little in the way of differences was seen with simulations varying aerosol types of sodium and sulfate. These findings counter the findings of Khain and Sednev (1999), who reported a change of 20-30% in total precipitation due the change of aerosol type of salt or sulfate (using the same hygroscopicities as the current study). The Khain and Sednev (1999) study was one of sea-breeze convection, whereas the current study is of a squall line with strong forcing produced by the strong temperature perturbation in the initialization. The discrepancy between models may be understandable within this context.

The delay in precipitation onset time of ~10-15 min. between low and high aerosol cases is due to an inhibition of warm-rain processes as determined by activation routine and the autoconversion parameterization. The magnitudes of domain-total rainfall over the 90 min. model seem rather high (over 300 mm for all cases) and total precipitation is observed to rise steadily rather than undergo periods of rainfall from sequential cells.

The sequence of total hydrometeor species with time (Fig. 4.4) shows an increase of liquid water with increasing aerosol concentrations. LA cases achieve similar values as compared with HA cases until 10 min. into the simulation, but due to either the increased efficiency of warm-rain processes or a weaker updraft, is not able to retain the water in-cloud. Figure 4.5 reveals
that the activated droplets are most likely dominated by larger aerosol concentrations. Because the shape of the aerosol size distribution is fixed, the relationship between number and mass of aerosol in each of the eight bins is fixed, and the magnitude of maximum droplet concentration of $6.0 \times 10^8 \text{ m}^{-3}$ even in the highest HA case indicate that not all aerosol are being activated. If this observation is valid it should also appear in comparisons with WRFv3.2 (Section 4.3).

Examination of the domain totals of hydrometeor species together (Figs. 4.6-4.9) with vertical velocity information (Figs. 4.11-13) allows for a fairly intuitive explanation of events within the 90 minutes of this simulation. Strong updrafts due to the warm-bubble initialization loft aerosol particles upward, achieving supersaturations high enough to activate many of the available aerosol in each case. Timing of updrafts coincide well with maxima in domain-total cloud liquid water, and delays of 5-10 minutes are observed in the other hydrometeor species as microphysical processes such as autoconversion and freezing take place.

Differences in maximum and conditionally averaged vertical velocities among the simulations may have microphysical or dynamical explanations. The secondary updraft seen at 45 min. in HA cases occurs below the freezing level and appears to be a new (secondary) updraft nearly as strong as the initial, artificial updraft. Figure 4.22 confirms this and also depicts two other low-level vertical velocity maxima that are stronger in HA cases than LA cases (at 60-65 min and 80-85 min).

Temperature perturbations with height (Figs. 4.23-24) overall show more warming (up to 3 K) than cooling (up to -1.5 K). It is not possible to disassociate contributions from microphysical processes from dynamical processes in these figures, but through cross-analysis with other figures (esp. Figs. 4.14-21) some conclusions can be drawn. For LA cases, latent heating is dominated for the first 30 minutes by condensation within the updraft and sinking motions to the east of the updraft. After this, a burst in heating occurs above the freezing level (near 500 mb), which decreases with time and travels out of the domain. Through the rest of the LA run, some compressional warming occurs, peaking near 700 mb. Figures 4.24a-b are considered as the HA cases here, and both show maxima near an updraft near the freezing level at 60-65 min, and are likely due solely to microphysical processes.

All cases have potential to evolve into squall lines if a longer simulation run were allowed, however, consistency of strong updrafts developing in HA cases and upright orientation (Figs.4.19-21) indicate that this case looks more favorable for development. Fig. 4.26a supports
this idea, as some identifiable features of squall lines can be seen. Rear inflow winds strengthen over the 90 minute run, and front-to-rear winds located just above this area (dark blue) are concurrent with positive temperature perturbations. Similar features are seen in the LA case but are weaker, and appear more disorganized. No definite cold pool has developed in either case, but negative temperature perturbations exist in areas where evaporative cooling is likely taking place, around 16-18 km and between 680-750 mb.

The convergence of hydrometeor values in HA cases, particularly for domain-total cloud liquid water and rain, and the profile of average cloud liquid water at 25 min. indicate that a threshold maximum value of water transport between vertical levels has been reached within the microphysics. One of the purposes of this study was to test the limits of this microphysics parameterization, and these observations are good indications that this has goal has been reached. On the low side of aerosol concentrations, not as much clustering was observed. It is interesting that seemingly unrealistic total aerosol number concentrations still produced hydrometeor species and precipitation within the model. It is very surprising to the author that such a range of total aerosol concentrations was necessary to elicit differences in the meteorology. The lack of microphysical sensitivity to commonly observed aerosol concentrations (approximately $1.50 \times 10^8$ m$^{-3}$ to $1.50 \times 10^9$ m$^{-3}$) is concerning, and will need to be addressed more thoroughly in future studies.

The gradual changeover in hydrometeor and total precipitation characteristics suggests that our battery of runs were very well controlled, the response of the system to the initial perturbation dominates the results, that the cautions of Ekman et al. (2007) are not valid for this case, or a combination of these possibilities. Additional cases using pre-set aerosol conditions present within the WRF-Chem would shed light on these issues.

The bulk microphysics scheme used within Li et al. (2009a,b) is quite similar to the scheme used in the current study, and their simulation of a squall line exhibited similar properties to the current study such as rearward tilt of leading convection and rearward-propagating discrete convective cells within the convective region. Despite some significant organizational differences on the cellular level between their test runs contrasting a bulk scheme and much more complex bin scheme, however, overall characteristics such as magnitude and structure of time-averaged potential temperature and pressure perturbations compared well with each other, as well as with observations.
Differences in the ABL concentrations were assumed to play a minor role within these cases because the system is dominated by the initial instability. ABL concentrations will be critical for entrainment and activation above the initial perturbation. The clustering behavior observed between LA and HA cases might be linked to the ABL concentrations. Figure 4.1 shows that LA cases are dominated by unrealistically small ABL concentrations, while HA cases all possess a more realistic ABL concentration. The ABL aerosol populations could be having more of an effect than expected. To test for this possibility, a different ABL scenario was set up for the WRFv3.2 runs (Sections 4.3 and 5.2).

5.2 Oceanic Case, Comparison of WRF Version 3.0.1 with Version 3.2

Updates to WRFv3.2 relevant to this study include change in the number and packing of vertical coordinates made by the author, and changes to the Lin et al. microphysics scheme (Section 3.1.4), and changes to the chemistry modules not made by the author. Additional WRF runs with chemistry turned off were performed to help ascertain differences between WRF versions, vertical level settings, and changes to the Lin et al. scheme. Output from WRFv3.0.1, which was run with 61 stretched vertical coordinates (hereafter referred to as stretched levels) was compared with output from WRFv3.2 with 50 assigned vertical levels (hereafter referred to as stretched levels) and output from WRFv3.2 with stretched levels. The results of total precipitation from these runs are compared in Figure 4.29a (black lines only). The most important feature is the trend of precipitation with time near the end of the run: in the case of WRFv3.0.1, the precipitation appears to asymptotically approach a value of 110 mm, while the precipitation in both runs of WRFv3.2 continues to increase. Also shown is a later onset of precipitation (by appx. 3-5 min.) and a reduced rate of rainfall (as compared to WRFv3.0.1 runs) over the first 60 min. These features are solely due to changes in the Lin et al. scheme and WRFv3.2 code, and not to the differences in vertical levels, as precipitation in the contrasting WRFv3.2 cases does not bifurcate until 30 minutes. Additional observations of updraft speeds, horizontal velocity, and vertical distribution of hydrometeors (not shown) indicate that changes made to the Lin et al. scheme favor the development and propagation of the squall line under consideration.

The adaptation of sigma levels to this 2D scenario also appears to have influenced the simulation. In Figure 4.29a (dashed black lines), the results from WRFv3.2 runs diverge around 30 min., with the run with stretched levels achieving higher precipitation rates for the remainder
of the run than the run with sigma levels. This might be attributed to the updraft and mass fluxes between vertical levels being better resolved at lower levels within the WRFv3.2 run with sigma levels than with the coarse levels offered in the stretched levels case.

Changes between the chemistry modules from WRFv3.0.1 to WRFv3.2 were not documented, but comparing the subroutines revealed some significant differences, particularly with regard to the activation routines. Constants used within these subroutines were corrected or made more precise. Perhaps the most serious example of this was an incorrect setting of the constant, “twothird,” equal to 2.0/6.0 in the original code. This constant had been used as an exponent at various points within the code to calculate activation fractions, but not for the 8-bin setup used in the current study. Another undocumented change is a new method of activation based upon a flux-weighted average activation fraction rather than a more simple calculation. Presumably this modification was implemented to compensate for turbulent vertical mixing within the smaller resolutions of the WRF-Chem as compared to the larger resolutions the activation scheme was initially designed for. Changes such as these undoubtedly account for differences in model output from WRFv3.0.1 to WRFv3.2 for the current study. These significant changes also cast doubt upon results from other studies using the previous versions of WRF-Chem such as Ntelekos et al. (2009).

A comparison of total precipitation for the chemistry cases with the single-moment Lin et al. cases seen in Figure 4.29 highlight the differences between all model runs within this study. It is important to note that though the single-moment Lin et al. scheme may not be the ideal microphysics parameterization to use for simulation of mesoscale systems, it has been used with satisfactory performance in the past (McCumber et al. 1991) and serves as a worthy metric against which to judge the performance of the WRF-Chem. The changes to the activation routines within WRF-Chem combined with changes to microphysics had the most impact overall, as the character of the total precipitation is completely different from the WRFv3.0.1 case (Fig. 4.29a) to the WRFv3.2 case (Fig. 4.29b). The structure of the current study was controlled sufficiently to make conclusions about the microphysical effects of aerosol within the implementation of WRFv3.0.1, however. It is interesting that the output of WRFv3.2 runs with chemistry are nearly constrained within the bounds of the WRFv3.2 runs without chemistry (dashed black lines), which shows that the developers of the WRF-Chem were aware of, and actively resolved, problems related to the aerosol-cloud interface within WRFv3.0.1. In
sensitivity studies of cloud-resolving models to resolution, Khain et al. (2004) found that a decrease in grid resolution led to more “maritime” characteristics. Figure 4.30b does not corroborate this finding: with time, the LA runs are closer to the single-moment Lin et al. runs with poorer low-level resolution, while the HA runs are closer to the single-moment Lin et al. runs with greater low-level resolution. Perhaps the most significant observation of Figure 4.29b, and perhaps within the entire study, is that for the most currently available version of WRF-Chem, the distribution and resolution of vertical levels has more impact on precipitation than a range of total aerosol concentrations larger than that which has been observed in nature.

Apparent in both WRFv3.0.1 and WRFv3.2 runs was the initial sensitivity to total aerosol concentrations within the boundary layer, to varying degrees. Higher aerosol concentrations consistently produced more cloud droplets due to the activation scheme that links the chemistry modules with the microphysics modules in the WRF-Chem. Warm-rain processes were delayed for increased aerosol concentrations, resulting in a delay in precipitation of only 2-5 min. in the WRFv3.2 cases to as much as 15 min. in WRFv3.0.1 cases. These results directly counter the conclusions of Ntelekos et al. (2009). Instead, they did not observe any delay in warm-rain processes with increased aerosol concentrations. The coarseness of their model (3 km) as compared to the current study (500 m) may partly explain this, as might differences in vertical resolution.
CHAPTER 6
CONCLUSIONS

6.1 Summary

A systematic series of model runs using the WRF-Chem model were performed with varying initial aerosol total number concentrations and types in order to determine impacts of aerosol on deep convective systems. The experimental design was set up to simplify aerosol chemistry and increase control within the study. To represent two different, but typical, aerosol environments, sodium was chosen to represent oceanic conditions and sulfate was chosen to represent continental conditions. Oceanic and continental thermodynamic environments supporting deep convection were likewise chosen, and according to each thermodynamic environment, WRF-Chem was run with idealized vertical profiles of sodium or sulfate, with stepwise increases in total aerosol concentrations for each run. The first series of tests, completed with WRFv3.0.1, showed high sensitivity to aerosol total number concentration but not to type. Higher aerosol concentrations in the case of the oceanic sounding resulted in the development of a squall line, while lower aerosol concentrations exhibited similar, but weaker, characteristics such as a strengthening rear inflow jet at low to mid levels and a front-to-rear flow at mid to upper levels. In these model runs with WRFv3.0.1, increasing aerosol concentrations lead to a storm with stronger, recurring updrafts and downdrafts, and higher total precipitation.

A new battery of tests using only the oceanic thermodynamic environment was run using a more current version of WRF, Version 3.2, with updates to microphysics and chemistry modules, and enhanced resolution at low levels. The results of WRFv3.2 runs, in stark contrast with the WRFv3.0.1 runs, did not produce a propagating system and had an opposite response of decreased updrafts and downdrafts and total precipitation with increasing aerosol concentrations. Results of the WRFv3.2 runs, however, were in better agreement in timing and magnitude of vertical velocity and hydrometeor content with a WRFv3.0.1 run using single-moment Lin et al. microphysics, than WRFv3.0.1 runs with chemistry. One result consistent throughout all simulations was an inhibition in warm-rain processes due to enhanced aerosol concentrations, which resulted in a delay of precipitation onset that ranged from 2-3 min. in WRFv3.2 runs, and up to 15 min. in WRFv3.0.1 runs. The changes to microphysical processes such as activation and autoconversion from WRFv3.0.1 to WRFv3.2, along with changes in the packing of vertical levels, had more impact than the varying aerosol concentrations even though the range of aerosol
tested was greater than that observed in field studies. In order to take full advantage of the input of aerosols now offered by the chemistry module in WRF, the author recommends that a fully double-moment microphysics scheme be linked, rather than the limited double-moment Lin et al. scheme that currently exists. With this modification, the WRF-Chem will be a powerful tool for studying aerosol-cloud interactions and allow comparison of results with other studies using more modern and complex microphysical parameterizations.

6.2 Conclusions and Discussion of Limitations

The fact that the results of this study do not coincide with another study using the same modeling framework (Ntelekos et al. 2009) is a cause for concern, but may be due to differences in horizontal and vertical resolution between the two studies. In a series of sensitivity tests, Khain et al. (2004) found that microphysical processes are highly dependent to model resolution, both vertical and horizontal. High vertical resolutions, especially at low levels, are crucial because updrafts forming at low levels will activate aerosol, which have highest concentrations at low levels, and droplet formation and subsequent hydrometeor production and latent heat release will be driven by the representation of those updrafts. This ultimately affects the evolution of the microphysical and dynamical processes within the remainder of the system’s lifetime. Bryan et al. (2003) showed through simulations of a squall line at varying horizontal resolutions that the character and evolution of the system’s features such as total precipitation, cloud depth, and propagation speed.

In both WRFv3.0.1, and especially WRFv3.2 cases, the sensitivity of the model to the range of total aerosol concentrations tested was underwhelming. The range initially chosen for the study, which are typical values used in other studies (e.g. van den Heever et al. 2006; Khain et al., 2005, 2008) elicited so little response that the range was expanded to $1.50 \times 10^6 \text{ m}^{-3}$ to $1.50 \times 10^9 \text{ m}^{-3}$. The results of WRFv3.0.1, while interesting from a modeling standpoint, were able to be described from a microphysical standpoint but appear unphysical when compared with output from a single-moment scheme that has been in use for almost 30 years (Fig. 4.29). With updates to the activation and autoconversion in WRFv3.2, output looked more reliable as it compared well with various renditions of WRF runs with single-moment microphysics. Despite the increased quality of output, the sensitivity of the model to the wide range of total aerosol input was again less than expected. The author attributes this lack of sensitivity to the limits of the microphysical scheme: in particular, the extent to which the scheme is truly double-moment.
The limited double-moment scheme currently implemented within the WRF-Chem allows only for prediction of mass and number for cloud droplets. After this point, important information regarding the mass and number distribution is thrown out as the mass values are adapted to the single-moment, prescribed distributions within the Lin et al. scheme. No consideration can be given to important attributes such as the broadening or narrowing of rain or other hydrometeor spectra, or the impacts that a changing spectra will have on terminal velocity of hydrometeors such as graupel and rain. Li et al. (2009a) argue that even fully double-moment bulk schemes only partially address the problems encountered, due to gross simplifications in assumed distributions of mass within single-moment schemes. For this reason, even relatively sophisticated double-moment bulk schemes may be insufficient for deriving meaningful results for studies of aerosol-cloud interaction. Particle activation, shapes of size distributions of particles and hydrometeors, and terminal fall speeds still are subject to assumptions that will dictate distributions of energy and moisture within a system. These unaccounted-for microphysical factors have significant influences on the dynamics of deep convective systems (Khain et al., 2004; Lynn et al. 2005a,b; Li et al, 2009a,b).

One of the most important lessons learned throughout the course of this study is to not blindly trust a model. When performing an idealized study, especially in a sensitivity study of a mesoscale phenomenon, it is not enough to simply run a model and trust it to perform without examining “what’s under the hood.” Several sensitivity studies should be performed to determine the response of the model to changes in vertical and horizontal resolution, and to microphysical routines. In many cases, microphysics schemes were developed to be suited to a specific meteorological scenario, and therefore are likely not globally applicable. Another item to be wary of with regard to a community-maintained model is changes made to the model from release to release. In general, changes that are critical to the phenomena under study may not be publicly documented thoroughly.

6.3 Recommendations for Future Work

In both the oceanic and continental cases, forcings within the simulated storms were arguably dominated by the artificial initial perturbation. Within the 90-minute time period of the study, a feature of both cases was a significant weakening of the systems with time, along with propagation of hydrometeors out of the 40 km domain. An increased domain and altered sounding with reduced wind velocity and shear would allow the systems to remain more
coherent, and would add more confidence to the results of this study. Also worthy of further testing are responses of more varied aerosol input that are offered as pre-determined case studies within WRF-Chem.

An adaptation or development of a double moment bulk or bin microphysics scheme compatible with the chemistry module is the next logical step on the meteorological side for the WRF-Chem. Double moment microphysics parameterizations have been developed and used with the WRF but have not been linked with WRF-Chem, or in some cases have not been made publicly available (e.g. Lee et al. 2008, Li et al. 2008). Studies as McCumber (1991), now twenty years old, recommend that fully double-moment microphysics schemes be developed, especially for simulations of convective systems where hydrometeor number and size distributions can vary between different parts of the storm. The current, very limited double-moment implementation of Lin et al. with chemistry falls short of this recommendation and therefore, results of the chemistry-meteorology connections must be treated with extreme caution. The author acknowledges that the results of this study are simply a snapshot in time of what the model is capable of, and that all models being used by the scientific community are, and should be considered as, works in progress. In the same sense, perhaps this study can help guide its future direction.

Despite the rather disparaging results from this study and Ntelekos et al. (2009), WRF-Chem looks to be a promising tool for the future. All simulations of convection covered within the scope of the literature review for this study were constrained to using highly idealized or pre-determined horizontally homogeneous aerosol distributions. As Tao et al. (2007) succinctly states, “Since the real atmosphere is 3D, further 3D cloud-resolving simulations are needed to address aerosol-precipitation interactions.” As theories for aerosol-cloud interactions continue to develop, eyes will turn toward modeling platforms with the ability to handle the integration of aerosol and emissions data. The WRF-Chem already operates within these terms. If crucial limitations such as lack of multiple-processor capability and a rapidly aging microphysics parameterization are resolved, WRF-Chem may become a forerunning tool for future aerosol-cloud interaction studies.
REFERENCES


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**Figure 2.1.** Evolution of deep convective clouds in the clean (top) and polluted (bottom) atmosphere as depicted by Rosenfeld et al. (2008). In the clean case, droplets coalesce into raindrops and fall as rain from the clouds. In the polluted case, more numerous, smaller droplets are lofted above the freezing level, where they become supercooled and freeze onto ice hydrometeors which fall and melt on their descent. Additional latent heat is released higher in the atmosphere and is absorbed in the lower atmosphere in the polluted case. Secondary updrafts formed in response to the initial updraft in the polluted case are therefore more invigorated with respect to the pristine case.
Figure 3.1. A schematic of chemical and meteorological processes involving aerosol within WRF-Chem. Processes used in this study are denoted by black text; those omitted are denoted by gray text. Adapted from Gustafson et al. (2006).
Figure 3.2. An example of how aerosol number and mass are allocated into eight size bins as implemented with MOSAIC in the WRF-Chem. Bins are initialized according to a power-law relationship. This example demonstrates an initialization of “oceanic” aerosol comprised solely of sodium (Na), with a total number concentration of $1.50 \times 10^9$ m$^{-3}$. Aerosol mass and number concentration are allowed to evolve with time.
Figure 3.3. Meteorological initialization supporting deep convection for simulations: a) Oceanic case from TOGA-COARE field experiment; b) Continental case from TRMM-LBA experiment
Figure 3.4. Aerosol size spectra, as functions of particle radius $a$, inside planetary boundary layer and aloft for a) continental environment and b) marine environment. From Patterson (1982). Overlain are linear approximations.
Figure 4.1. Vertical profile of initial aerosol conditions prescribed for WRFv3.0.1 runs: a) total number concentration over all bins; b) total mass concentration over all bins. Each line color from blue to red indicates a separate model run using different aerosol total number concentration. Shown for sodium.
Figure 4.2. Accumulated precipitation over the entire domain for the oceanic case; a) run with sodium aerosol, b) run with sulfate aerosol.
Figure 4.3. Conditional rain rate for the oceanic case; a) run with sodium aerosol, b) run with sulfate aerosol.
Figure 4.4. Profiles of total hydrometeor mass concentration over the horizontal domain with time, for the oceanic case. Each plot shows the results of a WRFv3.0.1 model run initialized with a different aerosol total number concentration. Shades of gray indicate liquid hydrometeor (cloud and rain) every 6 g m$^{-3}$, red dashed lines indicate frozen hydrometeor (ice and snow) every 0.5 g m$^{-3}$, and blue lines indicate graupel every 1 g m$^{-3}$.
Figure 4.5. Maximum cloud droplet number concentration over the domain for the oceanic case and sodium aerosol.
Figure 4.6. Domain total cloud liquid water per timestep (1 min) over the domain for the oceanic case and sodium aerosol.
Figure 4.7. Domain total rain water per timestep (1 min) over the domain for the oceanic case and sodium aerosol.
Figure 4.8. Domain total graupel per timestep (1 min) over the domain for the oceanic case and sodium aerosol.
Figure 4.9. Domain total cloud ice and snow per timestep (1 min) over the domain for the oceanic case and sodium aerosol.
Figure 4.10. Domain average cloud liquid water at 25 min over the domain for the oceanic case and sodium aerosol.
Figure 4.11. Maximum vertical velocity per timestep (1 min) over the domain for the oceanic case and sodium aerosol.
Figure 4.12. Vertical velocity averaged over points in the domain greater than 1.0 m s\(^{-1}\) in each (1 min) timestep, for the oceanic case and sodium aerosol.
**Figure 4.13.** Maximum downward vertical velocity per timestep (1 min) over the domain for the oceanic case and sodium aerosol.
Figure 4.14. Comparative plots of vertical velocity, temperature perturbations, and height of freezing level (orange). The plots show the results of very low aerosol concentrations (left; $1.50 \times 10^6 \text{ m}^{-3}$); and very high aerosol concentrations (right, $1.50 \times 10^9 \text{ m}^{-3}$). Vertical velocity greater than $1 \text{ m s}^{-1}$ is colored in red, less than $-1 \text{ m s}^{-1}$ is shaded in blue, and both are contoured every $1 \text{ m s}^{-1}$. Solid black contour lines indicate positive temperature perturbations and dashed black lines indicate negative temperature perturbations, every $0.5 \text{ K}$. All plots were created using the oceanic sounding and sodium as the aerosol type.
Figure 4.15. As in Fig. 4.13, 25 minutes into the simulation.
Figure 4.16. As in Fig. 4.13, 35 minutes into the simulation.
Figure 4.17. As in Fig. 4.13, 45 minutes into the simulation.
Figure 4.18. As in Fig. 4.13, minutes into the simulation.
Figure 4.19. As in Fig. 4.13, 65 minutes into the simulation.
Figure 4.20. As in Fig. 4.13, 75 minutes into the simulation.
Figure 4.21. As in Fig. 4.13, 85 minutes into the simulation.
Figure 4.22. Profiles of maximum vertical velocity over the horizontal domain with time, for the oceanic case. Each plot shows the results of a WRFv3.0.1 model run initialized with a different aerosol total number concentration. Contours begin at 0.2 m s$^{-1}$ and are at 0.5 m s$^{-1}$ and every 0.5 m s$^{-1}$ thereafter.
Figure 4.23. Profiles of maximum downward vertical velocity over the horizontal domain with time, for the oceanic case. Each plot shows the results of a WRFv3.0.1 model run initialized with a different aerosol total number concentration. Contours begin at 0.2 m s\(^{-1}\) and are at 0.5 m s\(^{-1}\) and every 0.5 m s\(^{-1}\) thereafter.
Figure 4.24. Profiles of maximum positive temperature perturbation over the horizontal domain with time, for the oceanic case. Each plot shows the results of a WRFv3.0.1 model run initialized with a different aerosol total number concentration. Contours are every 0.5 K.
Figure 4.25. Profiles of maximum negative temperature perturbation over the horizontal domain with time, for the oceanic case. Each plot shows the results of a WRFv3.0.1 model run initialized with a different aerosol total number concentration. Contours are every 0.5 K.
Figure 4.26. Profiles of horizontal wind ($u$ component) and temperature perturbation at 0, 25, and 85 minutes into the simulation for a run with total initial aerosol concentration of a) $1.50 \times 10^6$ m$^{-3}$ and b) $1.50 \times 10^9$ m$^{-3}$. Contours in blue and orange indicate horizontal wind speed, every 0.5 m s$^{-1}$. Solid black contour lines indicate positive temperature perturbations and dashed black lines indicate negative temperature perturbations, every 0.5 K. All plots were created using the oceanic sounding and sodium as the aerosol type.
Figure 4.26 (cont.)
b)

Initial aerosol total number concentration = 1.50e9 m$^3$

1) 0 min

2) 25 min

3) 85 min

-4.5 -4 -3.5 -3 -2.5 -2 -1.5 -1 -0.5 0.5 1 1.5 2 2.5 3 3.5 4 4.5
Fig. 4.27. Vertical profiles of initial aerosol total number concentrations prescribed for a) WRFv3.0.1 runs; b) WRFv3.2 runs. Each line color from blue to red indicates a separate model run using different aerosol total number concentration.
Fig. 4.28. Vertical profiles of initial aerosol total mass concentrations prescribed for a) WRFv3.0.1 runs; b) WRFv3.2 runs. Each line color from blue to red indicates a separate model run using different aerosol total number concentration.
Fig. 4.29. Accumulated precipitation over the entire domain for the oceanic case and sodium aerosol; a) WRFv3.0.1 run, b) WRFv3.2 run. Black lines show the results of single-moment (no chemistry) WRF runs: Black line indicates results from the WRFv3.0.1 run (with 61 vertically stretched levels), the long-dashed line is from the WRFv3.2 run with stretched vertical levels, and the short-dashed is from the WRFv3.2 run with sigma levels (50 assigned vertical levels).
Fig. 4.30. Conditional rain rate for the continental case and sodium aerosol; a) WRFv3.0.1 run, b) WRFv3.2 run. Black line indicates results from WRFv3.0.1 run with the single moment Lin scheme.
Fig. 4.31. Profiles of total hydrometeor mass concentration over the entire domain with time. Each plot shows the results of a WRFv3.0.1 model run initialized with a different initial aerosol total number concentration. Shades of gray indicate liquid hydrometeor (cloud and rain) every 1 g m$^{-3}$, red dashed lines indicate frozen hydrometeor (ice and snow) at 0.1 g m$^{-3}$ and every 0.5 g m$^{-3}$ thereafter, and blue lines indicate graupel every 1 g m$^{-3}$. 
Fig. 4.32. Profiles of total hydrometeor mass concentration over the entire domain with time. Each plot shows the results of a WRFv3.2 model run initialized with a different initial aerosol total number concentration. Shades of gray indicate liquid hydrometeor (cloud and rain) every 1 g m$^{-3}$, red dashed lines indicate frozen hydrometeor (ice and snow) at 0.1 g m$^{-3}$ and every 0.5 g m$^{-3}$ thereafter, and blue lines indicate graupel every 1 g m$^{-3}$. 
Fig. 4.33. Maximum cloud droplet number concentration over the domain for the oceanic case and sodium aerosol; a) WRFv3.0.1 run, b) WRFv3.2 run.
**Fig. 4.34.** Domain total cloud liquid water per timestep (1 min) over the domain for the oceanic case and sodium aerosol; a) WRFv3.0.1 run, b) WRFv3.2 run.
Fig. 4.35. Domain total rain water per timestep (1 min) over the domain for the oceanic case and sodium aerosol; a) WRFv3.0.1 run, b) WRFv3.2 run.
Fig. 4.36. Domain total graupel per timestep (1 min) over the domain for the oceanic case and sodium aerosol; a) WRFv3.0.1 run, b) WRFv3.2 run.
Fig. 4.37. Domain total cloud ice and snow per timestep (1 min) over the domain for the oceanic case and sodium aerosol; a) WRFv3.0.1 run, b) WRFv3.2 run.
Fig. 4.38. Domain average cloud liquid water at 25 min over the domain for the oceanic case and sodium aerosol; a) WRFv3.0.1 run, b) WRFv3.2 run.
Fig. 4.39. Maximum vertical velocity per timestep (1 min) over the domain for the oceanic case and sodium aerosol; a) WRFv3.0.1 run, b) WRFv3.2 run.
Fig. 4.40. Vertical velocity averaged over points in the domain greater than 1.0 m s\(^{-1}\) in each (1 min) timestep, for the oceanic case and sodium aerosol; a) WRFv3.0.1 run, b) WRFv3.2 run.
Fig. 4.41. Maximum downward vertical velocity per timestep (1 min) over the domain for the oceanic case and sodium aerosol; a) WRFv3.0.1 run, b) WRFv3.2 run.
Fig. 4.42. Profiles of maximum vertical velocity over the horizontal domain with time. Each plot shows the results of a WRFv3.0.1 model run initialized with a different aerosol total number concentration. Contours begin at 0.2 m s$^{-1}$ and are at 0.5 m s$^{-1}$ and very 0.5 m s$^{-1}$ thereafter.
Fig. 4.43. As in Fig. 4.42, but with output from WRFv3.2.
Fig. 4.44. Profiles of maximum downward vertical velocity over the horizontal domain with time. Each plot shows the results of a WRFv3.0.1 model run initialized with a different aerosol total number concentration. Contours begin at 0.2 m s\(^{-1}\) and are at 0.5 m s\(^{-1}\) and every 0.5 m s\(^{-1}\) thereafter.
Fig. 4.45. As in Fig. 4.44, but with output from WRFv3.2.
Fig. 4.46. Profiles of maximum positive temperature perturbation over the horizontal domain with time. Each plot shows the results of a WRFv3.0.1 model run initialized with a different aerosol total number concentration. Contours are every 0.5 K.
Fig. 4.47. As in Fig. 4.46, but with output from WRFv3.2.
Fig. 4.48. Profiles of maximum negative temperature perturbation over the horizontal domain with time. Each plot shows the results of a WRFv3.0.1 model run initialized with a different aerosol total number concentration. Contours are every 0.5 K.
Fig. 4.49. As in Fig. 4.48, but with output from WRFv3.2.
Fig. 4.50. Profiles of horizontal wind \((u)\) component and temperature perturbation at 0, 25, and 85 minutes into the simulation for a run with total initial aerosol concentration of a) \(1.50 \times 10^6\) m\(^{-3}\) and b) \(1.50 \times 10^9\) m\(^{-3}\) using WRFv3.0.1. Contours in blue and orange indicate horizontal wind speed, every 0.5 m s\(^{-1}\). Solid black contour lines indicate positive temperature perturbations and dashed black lines indicate negative temperature perturbations, every 0.5 K. All plots were created using the oceanic sounding and sodium as the aerosol type.
Figure 4.50 (cont.)

b)

Initial aerosol total number concentration = 1.23e9 m\(^{-3}\).

- 1) 0 min
- 2) 25 min
- 3) 85 min
Fig. 4.51. Profiles of horizontal wind ($u$ component) and temperature perturbation at 0, 25, and 85 minutes into the simulation for a run with total initial aerosol concentration of a) $1.50 \times 10^6$ m$^{-3}$ and b) $1.50 \times 10^9$ m$^{-3}$ using WRFv3.2. Contours in blue and orange indicate horizontal wind speed, every 0.5 m s$^{-1}$. Solid black contour lines indicate positive temperature perturbations and dashed black lines indicate negative temperature perturbations, every 0.5 K. All plots were created using the oceanic sounding and sodium as the aerosol type.
Figure 4.51 (cont.)

b) Initial aerosol total number concentration = 1.23e9 m$^{-3}$

1) 0 min

Pressure (mb)

Distance (km)

2) 25 min

Pressure (mb)

Distance (km)

3) 85 min

Pressure (mb)

Distance (km)
Table 3.1. Model configuration for WRFv3.0.1

<table>
<thead>
<tr>
<th><strong>Domain Setup</strong></th>
<th><strong>Description</strong></th>
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</thead>
<tbody>
<tr>
<td>Model run type</td>
<td>2D (em_squall2d_x)</td>
</tr>
<tr>
<td>Horizontal grid spacing</td>
<td>500 m</td>
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<tr>
<td>Total domain size</td>
<td>40 km</td>
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<tr>
<td>Number of vertical levels</td>
<td>61</td>
</tr>
<tr>
<td>Timestep</td>
<td>1.0 s (for all processes)</td>
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<tr>
<td>Data output frequency</td>
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<tr>
<td>Integration time</td>
<td>90 minutes</td>
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<table>
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<tr>
<th><strong>WRF Option</strong></th>
<th><strong>Selection</strong></th>
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<tbody>
<tr>
<td>Time integration scheme</td>
<td>Runge-Kutta 3rd Order</td>
</tr>
<tr>
<td>Microphysics</td>
<td>Purdue Lin (limited double moment)</td>
</tr>
<tr>
<td>-- Autoconversion scheme</td>
<td>Liu et al. (2005)</td>
</tr>
<tr>
<td>-- Double-moment species</td>
<td>Cloud droplets</td>
</tr>
<tr>
<td>Positive Definite Advection</td>
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</tr>
<tr>
<td></td>
<td>* for moist, scalars, and chemistry</td>
</tr>
<tr>
<td>Prognostic aerosol</td>
<td>On</td>
</tr>
<tr>
<td>Chemistry Driver</td>
<td>CBMZ</td>
</tr>
<tr>
<td></td>
<td>* most chemical processes turned off</td>
</tr>
<tr>
<td>Aerosol Driver</td>
<td>MOSAIC (8 bins)</td>
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<tr>
<td>Wet Scavenging</td>
<td>On</td>
</tr>
<tr>
<td>Vertical Mixing</td>
<td>On</td>
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**Physics options disabled:**

-- Longwave, shortwave radiation
-- Surface layer, land-surface model, boundary layer, cumulus parameterization

**Chemistry options disabled:**

-- Biogenic emissions, cloud chemistry, aqueous chemistry, aerosol-cloud-radiation interactions
**Table 3.2. Model configuration for WRFv3.2**

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<tr>
<th>WRFv3.2 Configuration</th>
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<td>Model run type</td>
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<tr>
<td>Horizontal grid spacing</td>
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<tr>
<td>Total domain size</td>
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<tr>
<td>Number of vertical levels</td>
<td>50 -- sigma levels</td>
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<tr>
<td>Timestep</td>
<td>1.0 s (for all processes)</td>
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<tr>
<td>Data output frequency</td>
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<td>Time integration scheme</td>
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<tr>
<td>Microphysics</td>
<td>Purdue Lin (limited double moment)</td>
</tr>
<tr>
<td>-- Autoconversion scheme</td>
<td>-- Liu et al. (2005)</td>
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<tr>
<td>-- Double-moment species</td>
<td>-- Cloud droplets</td>
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<td>Chemistry Driver</td>
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<td>* most chemical processes turned off</td>
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<tr>
<td>Aerosol Driver</td>
<td>MOSAIC (8 bins)</td>
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<tr>
<td>Wet Scavenging</td>
<td>On</td>
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<tr>
<td>Vertical Mixing</td>
<td>On</td>
</tr>
</tbody>
</table>

**Physics options disabled:**
- -- Longwave, shortwave radiation
- -- Surface layer, land-surface model, boundary layer, cumulus parameterization

**Chemistry options disabled:**
- -- Biogenic emissions, cloud chemistry, aqueous chemistry, aerosol-cloud-radiation interactions
Table 3.3. Comparison of the single-moment Lin scheme, double-moment Lin scheme without chemistry, and double-moment Lin scheme including chemistry, as implemented in WRFv3.0.1 and WRFv3.2.

<table>
<thead>
<tr>
<th></th>
<th>Lin 1M</th>
<th>Lin 2M (offline chemistry)</th>
<th>Lin 2M with Chemistry (online chemistry)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Advection</td>
<td>Mass (Qv, Qc, Qr, Qs, Qg, Qi)</td>
<td>Mass (Qv, Qc, Qr, Qs, Qg, Qi) and number of drops (Qn)</td>
<td>Mass (Qv, Qc, Qr, Qs, Qg, Qi) and number of drops (Qn), and mass, number of each chemical species for each size bin</td>
</tr>
<tr>
<td>Activation</td>
<td>From prescribed CCN (1.0 x 10^8 m^-3)</td>
<td>From prescribed mass, predicted number of drops</td>
<td>From predicted mass, predicted number of drops</td>
</tr>
<tr>
<td>Autoconversion</td>
<td>Threshold</td>
<td>Variable threshold based on predicted quantities</td>
<td>Variable threshold based on predicted quantities</td>
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</tbody>
</table>