THESIS

REGIONAL AEROSOL EFFECTS ON PRECIPITATION: AN OBSERVATIONAL STUDY

Submitted by

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ABSTRACT

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There have been a multitude of studies on the effects increased amounts of aerosols may have on clouds. The connection between increased cloud condensation nuclei (CCN) and cloud microphysics has been established by in situ observations as well as modeling studies. However, the impact on precipitation is less well established. Of the studies that have assessed aerosol effects on precipitation most have been limited to modeling studies or global studies using satellite data. The few observational studies that have examined these relationships have been mainly limited to data from short-lived field campaign, such as oceanic stratocumulus decks or biomass burning areas.

This study attempts to examine regional aerosol effects on precipitation in areas not previously examined in field campaigns, using data from two different sites, one from an Atmospheric Radiation Measurement (ARM) Program permanent facility in Oklahoma and the other from a mobile facility located in the Azores. These two sites were chosen in order to illustrate the differences between a marine and a continental location. Meteorological conditions were taken into account in both locations through surface and sounding data and trends in precipitation were found with increasing aerosol concentrations. The marine site witnessed a suppression of precipitation, consistent with

past studies and proposed theories of aerosol effects. This was not true for clouds with liquid water paths exceeding 200g/m^2. These clouds appear to contain sufficient amounts of water to overcome the aerosol effect. The continental site, however, experienced an opposite trend, with enhancement of precipitation witnessed in all clouds examined in this study. This is thought to be due to a buffering mechanism in these types of clouds, as introduced by Stevens and Feingold (2009). Results were separated by season and cloud type using the horizontal variability of radar reflectivity at cloud top height. The seasonal results generally either were in line with the year round results or were too noisy to interpret. The results separated by cloud type give a concrete result, illustrating the fact that differing cloud dynamics may lead to opposing trends in precipitation with increasing aerosols. Competing effects of aerosols within clouds appear to dampen any effect on precipitation to the point that it is not detectable from the in-situ observations considered here.

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CHAPTER 1

INTRODUCTION

1.1 Indirect Effects of Aerosols

1.1.1 Motivation

The possible changes in global precipitation patterns are one of the more challenging aspects of climate change to predict. Nonetheless, it is also one of the most essential for humans. Changes in precipitation can produce major shifts in available fresh water, needed for human and agricultural applications. It has been theorized that increased air pollution due to anthropogenic activity may have a profound effect on the nature and initiation of precipitation (Gunn 1957 and Warner 1968). Many studies have investigated the effects of increased amounts of aerosol on clouds and precipitation; however, most of these have been limited to either modeling or observational studies using global satellite data (or a combination of these, such as Suzuki et al. 2004 and 2008), while the few regional observational studies that have been performed were limited to certain cloud regimes and areas (i.e. oceanic studies of stratocumulus clouds) (Cotton and Levin 2009). Therefore, regional effects of aerosols on precipitation are still unknown or at least uncertain in most areas of the globe. Some modeling studies have shown that regional differences in land use and background aerosol concentrations (e.g. Carrio et al. 2010, Warner 1968 and van den Heever and Cotton 2007) and terrain (e.g. Saleeby et al. 2011 and Givati and Rosenfeld 2004) can alter the effect of aerosols on clouds and precipitation. Other modeling studies have found that the aerosol type, cloud type, cloud dynamics and the meteorological conditions in which these clouds form may also cause differing effects on the cloud processes due to aerosols (e.g. Khain et al. 2008, Albrecht 1989, Wang and Feingold 2009a&b, Comstock et al. 2007, Savic-Jovcic and Stevens 2008, Fan et al. 2009, Storer et al. 2010, van den Heever et al. 2006, van den Heever et al. 2011). While global observational studies on the aerosol effect exist (e.g. L'Ecuyer et al. 2009, Lebsock et al. 2008 and Sekiguchi et al. 2003), these studies are limited to oceanic regions and have only crude information available on the atmospheric state as well as aerosol concentrations. More regional observational studies are necessary in order to determine the magnitude of the response of aerosol indirect effects to meteorological conditions and cloud type.

The past studies on aerosol effects have shown that the effect of increased pollution on precipitation is very complex, making it difficult to even predict the sign of the response, i.e. whether precipitation will be enhanced or suppressed. Before one can begin to examine these complexities and the existing theories of aerosol effects on precipitation, it is necessary to understand the cloud microphysical processes that lead to precipitation and which may be influenced by increased amounts of aerosols.

1.1.2 Basic Theories of Indirect Effects

Cloud droplets require a particle to act as a nucleus upon which to condense because observed atmospheric conditions generally do not reach cold enough temperatures or high enough supersaturations to allow for homogeneous nucleation. When an aerosol particle is used in this process, it is called a cloud condensation nucleus (CCN). The theory of the first indirect effect states that as the aerosol population increases, so does the number of possible CCN, leading to more nucleating surfaces for water vapor to condense upon and, consequently, more cloud droplets. If the liquid water content of the cloud remains the same, there is less water available for each droplet; hence the droplets will be smaller in size. The optical depth of a cloud can be related to its liquid water content and the size of the cloud droplets through the relation (Sekiguchi et al. 2003)

$$\tau = \frac{3}{4}\frac{Q_{ext}LWC}{r_e} \qquad 1.1$$

where r_e is the effective radius of the cloud droplets (a weighted mean of the size distribution of cloud droplets), Q_{ext} is the extinction efficiency (about 2 for wavelengths much shorter than r_e) and LWC is the liquid water content of the cloud. From equation 1.1, if LWC remains constant, one can see that as r_e decreases, the optical thickness of the cloud will increase. This is known as the Twomey effect (Twomey, 1974, Warner and Twomey 1967). Cloud albedo is primarily a function of cloud optical depth; therefore it follows that an increase in CCN concentrations should be associated with brighter clouds. This would have an effect on the global albedo and radiation budget and it has been suggested that this affect would be similar in magnitude and opposite in sign to the forcing of anthropogenic greenhouse gases (Twomey et al. 1984).

Following this theory comes another, the second indirect effect of aerosols. This states that as the cloud droplets become more numerous and smaller in size due to increased CCN concentrations, the resulting size distribution is narrower. This hinders warm rain formation processes, the most dominant of which is collision-coalescence because growth by pure condensational processes is not very efficient. This process relies upon the fact that as droplets are redistributed in the cloud due to air motions, they will collide with other droplets, sometimes resulting in a collection event, where a larger droplet collects a smaller droplet and grows. These coalescence events produce ever-larger droplets and eventually lead to precipitation-sized drops that fall out of the cloud as rain. This process is more efficient with a broader size distribution of droplets (Albrecht 1989), since smaller droplets have lower terminal velocities and collection efficiencies compared to large droplets (Warner 1968). If increases in CCN concentration lead to increases in smaller droplets, this narrows the droplet size distribution and therefore decreases the efficiency of the warm rain process (Warner 1968). If precipitation is suppressed, it is theorized that this will increase the lifetime of warm clouds, since rain out will be reduced (Albrecht 1989). Also, the vertical extent and LWC of the cloud will be increased, since (as has been discussed), there are more droplets and less rain out of these droplets occurs. Without precipitation, the cloud will continue to undergo its microphysical processes, which allow it to grow more than if it precipitated. An illustration of these processes is included in figure 1.1, however, please note that this depicts warm clouds only; effects on mixed phase clouds are discussed in section 1.2.1.

Increases to the global fractional cloudiness through this mechanism (Albrecht 1989), along with the increase of cloud albedo predicted in the first indirect effect theory, could lead to an increase in the global albedo and therefore a counteracting force to the global warming predicted due to increased emissions of greenhouse gases. In recent years, however, the aerosol effect on precipitation has been postulated to be more complex than these theories suggest. Specifically, recent studies have found dynamical causes as well as aerosol properties to play key roles in the albedo effect. Examples of modifications to these indirect effect theories are presented in the next section.

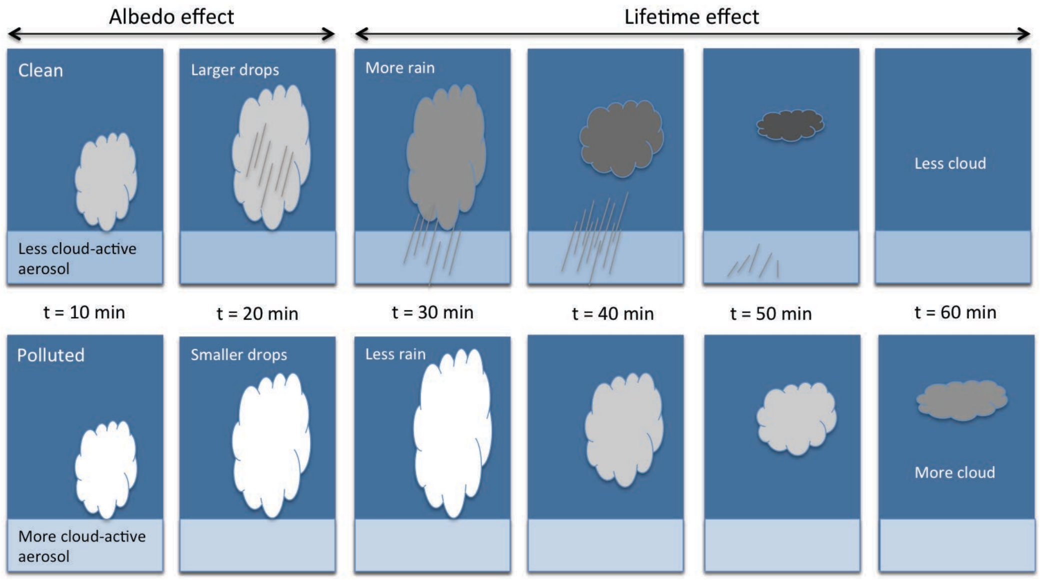

Figure 1.1
A conceptual model of indirect aerosol effects described in this chapter (specifically, the lifetime and albedo affects as originally proposed). Following figure 1 from Stevens and Feingold (2009). In polluted air masses, clouds consist of more droplets that coalesce into raindrops less effectively, leaving longer-lived clouds.

1.2 Other aerosol effects on precipitation

A large portion of aerosol research has focused on marine stratocumulus cloud types. This was partly due to the fact that these clouds have fairly stable dynamics (relatively similar meteorological conditions throughout) and also due to their large coverage of areas of the earth, which helps them to be a major contributor to the global albedo and radiation budget. In investigating these types of clouds, it was determined that there are 2 regimes of stratocumulus clouds, open and closed cellular convection. It has been generally observed that the open cellular structure is associated with relatively high drizzle rates, while the closed cellular structure did not exhibit this characteristic (e.g. Lu et al. 2009, Wood et al. 2008 and Comstock et al. 2007). It has also been shown that increasing amounts of aerosol concentration lead to the suppression of drizzle in these clouds, hence a shift to the closed cellular regime (Wang and Feingold 2009). However, once the open cellular regime is established, model studies have shown that increasing CCN concentrations does not reestablish the closed cellular regime (Wang and Feingold 2009). While there are subtle complexities, precipitation is generally found to decrease with increasing aerosol in this cloud regime, consistent with the aerosol indirect effect theories.

Another type of cloud in which aerosol effects have been investigated both through observational and modeling studies is the trade wind cumulus. Heymsfield and McFarquhar (2001) determined that drizzle was observed significantly less frequently in more polluted regions compared to pristine regions using data collected in the Indian Ocean. However, cloud resolving model experiments on trade wind cumuli (initialized with data from the Western Atlantic) showed that along with a reduction in precipitation for polluted conditions, an enhancement of precipitation amounts can also be seen if the amount of very large aerosol particles, also known as Giant Cloud Condensation Nuclei (GCCN) is increased along with the CCN concentration (Cheng et al. 2009). GCCN can enhance precipitation through its effect on the collision-coalescence process. A few large particles among the many smaller ones can lead to a wider cloud droplet size distribution, increasing the efficiency of the warm rain process. This effect has also been observed in modeling studies of stratocumulus clouds (Feingold et al. 1999) as well as deep convection (van den Heever and Cotton 2007). Both of these studies also found that GCCN tends to have a larger influence on precipitation processes when the CCN concentration is higher. Drizzle development is most hindered in polluted conditions and therefore GCCN have the greatest potential for enhancing the collection process (Feingold et al. 1999).

Along with GCCN, some studies have found other alternative reactions of clouds to aerosols that were not predicted by the traditional theories previously mentioned. One of these, the aerosol-induced enhancement of evaporation, was investigated in a modeling study by Xue and Feingold (2006). Since an increase in CCN leads to more cloud droplets within the cloud, these smaller droplets are more easily evaporated than larger droplets would be, especially near cloud edges where drier air is entrained. The entrainment that occurs at cloud tops, especially in the shallow cumulus cloud type investigated by Xue and Feingold (2006), has a significant effect on the cloud dynamics and can alter the cloud fraction. Although this effect still leads to a suppression of precipitation, it also can cause a decrease in cloud coverage and cloud depth (Xue and Feingold 2006), in opposition to the original theories of indirect effects of aerosols. They also found that this affected the LWC through multiple mechanisms; lower in the cloud, LWC was increased due to increased condensation rates of the smaller cloud droplets, while it was decreased higher in the cloud due to the increased evaporation rates. This might alter the magnitude of the response within the cloud, along with increasing the difficulty of observing aerosol effects. However, the decrease in LWC is consistent with an observational study done by Han et al. (2002), who found that in the mid-latitude regions the liquid water path (LWP) generally decreased with increasing column droplet number concentration (which was derived from satellite data of effective radius and optical thickness of the cloud). Specifically, they tended to observe neutral or slightly negative liquid water sensitivities for continental clouds. For maritime clouds, the sensitivities had a strong seasonal dependence; negative sensitivities were more common in the summer hemisphere (Han et al. 2002), hence they argued that this relation was seen more often in warm areas than cold areas. They cite results from a Del Genio and Wolf (2000) paper to explain this as a cloud microphysical effect on the boundary layer temperature, arguing that the decreases in droplet sizes would enhance cloud-base cooling due to evaporation and weaken decoupling between clouds and boundary layer (since cooler temperatures are often associated with stratified and convective boundary layers). Therefore, it appears that aerosol effects are much more complex than the original theories proposed and that cloud dynamics as well as meteorological conditions (including seasonal variations) may play important roles.

1.2.1 Clouds with Ice

If ice processes are allowed, the complexities grow rapidly. Through observational studies (e.g. Givati and Rosenfeld 2004) as well as modeling studies (Saleeby et al. 2011), orographic precipitation appears to experience what has been called a 'spillover effect'. This term refers to the fact that precipitation appears to be suppressed on the windward side of topography, while being enhanced on the leeward side. This process is hypothesized to be due to the reduction in riming of ice particles by cloud water, leading to lighter rimed hydrometeors with slower fall speeds and longer horizontal trajectories that travel over the mountain barriers more easily.

Recent studies investigating the effects of aerosols on deep convection have been mostly limited to modeling studies. They have generally found that initially the warm rain process is reduced due to higher concentrations of smaller cloud droplets. However, due to the complex dynamical and microphysical feedbacks in these types of storms, the resulting influence on precipitation has varied (van den Heever et al. 2006, Storer et al. 2010 and van den Heever and Cotton 2007). These studies all showed decreases in surface precipitation with increasing CCN concentrations, unless GCCN concentrations were also increased, in which case precipitation was enhanced (van den Heever and Cotton 2007). They also noticed that later in the simulation, secondary convection could have enhanced precipitation. In contrast, Carrio et al. (2010) found an initial enhancement of precipitation and a general invigoration of the storm cell with increasing CCN up to a point after which increases in the aerosol concentrations led to reduced precipitation. The non-monotonic response of the convection was thought to be due to the increase in supercooled liquid water transported to higher levels in the cloud (after initial suppression of warm rain processes), which freezes, releasing extra latent heating and invigorating convection (Carrio et al. 2010). However, with even larger CCN concentrations, the riming growth of ice particles is reduced and larger amounts of condensate were transported upwards into the storm anvil, contributing to a reduction in precipitation efficiency. Similar results from the modeling studies have been observed in a field campaign done by Andreae et al. (2004). Biomass burning creates large amounts of aerosols and precipitation was observed to increase as these aerosol concentrations increased due to an overall enhancement of cloud dynamics.

All of the previous studies mentioned here found varying magnitudes in the general response of precipitation to increased aerosol but for different reasons. Storer et al. (2010), as well as Carrio and Cotton (2011) found that variations in stability (e.g. CAPE) produced differing magnitudes on the variation in precipitation response. In general, low values of CAPE produced the largest aerosol effect. This illustrates the fact that meteorological conditions are important for determining the magnitude of precipitation responses to increased pollution. Carrio et al. (2010) found that land use changes and temperature variations due to the increased size of an urban region produced larger affects on convection than changes to aerosol concentration. Van den Heever and Cotton (2007) found that with increasing background CCN concentrations, the magnitude of the aerosol influences was reduced, illustrating that continental clouds (where background aerosols concentrations are relatively high compared with marine environments) may have reduced aerosol effects compared to those observed and modeled over the ocean, where most of the focus has previously been. Lastly, the orographic cloud modeling study of Saleeby and Cotton (2011) introduced earlier discussed how the interseasonal variability of precipitation was larger than any aerosol effect seen, showing the possible seasonal impact of aerosol effects.

Observational and modeling studies have produced a complex picture of the aerosol indirect effects. It seems that although some studies (with certain cloud types in certain environments) behave as expected, meteorological conditions and other factors such as the presence of ice can greatly complicate the resulting effect of increased aerosol concentrations on precipitation. In fact, in some cases, these other factors produce larger effects than the aerosols, making it difficult to quantify the aerosol effect. The following sections introduce global satellite studies of aerosol effects and a modeling study in an attempt to organize the differing effects of aerosol on precipitation.

1.3 Global Effects

Along with the more regional scale and cloud resolving model studies, the aerosol effect has also been studied on global scales. Incorporation of the aerosol indirect effects into general circulation models (GCMs) have led to the attempt at quantifying these effects, or at least the magnitude of the first indirect effect on the entire climate system. The International Panel on Climate Change (IPCC) estimated the indirect cloud albedo effect to be -0.7 (-0.3 to -1.8) Wm^{-2} (Meehl et al. 2007). However, the report also mentions that some models that incorporate more aerosol species or are constrained by satellite observations tended to find a reduced first indirect effect. A climate modeling study done at the UK Meteorology Office using the Hadley Centre Climate Model found that inclusion of sea salt aerosol led to a slight decrease in the global mean indirect forcing (Jones and Slingo 1997). This study also found a decrease in the albedo effect when they changed the sensitivity of droplet concentration over land to be less than that over the ocean (assuming there would be a weaker correlation between continental cloud droplet concentrations and sulphate aerosol concentrations than marine cloud droplet concentrations), illustrating, as van den Heever and Cotton (2007) showed, that perhaps the background concentrations are important to the magnitude of aerosol effects. Although the first aerosol indirect effect is being examined in GCMs, there is less emphasis on the second aerosol indirect effect and the IPCC AR4 report specifies the cloud lifetime effect (referred to as the 2nd indirect effect in this paper) to be part of the climate response, not a radiative forcing on the climate system and so does not provide a magnitude for this effect (Meehl et al. 2007). Along the same lines, the effect of aerosols on precipitation is not mentioned nor quantified.

Another way to investigate global aerosol effects through modeling is with the use of global cloud resolving models. GCMs do not resolve cloud scale processes, hence parameterizations are used to simulate clouds and their dynamics. However, Suzuki et al. (2008) have used global cloud resolving models in order to more accurately simulate the affects of increased aerosols on clouds on large scales. They were able to produce similar results as have been observed using satellite data, specifically that the LWP decreased with increasing aerosol index.

The only way to observationally analyze aerosol indirect effects on a global scale is through satellite observations, since they provide the only truly global coverage of aerosol and cloud data. There have been a few recent studies that specifically looked at the effects of aerosols on precipitation, mostly using data from NASA's A-train constellation of satellites. However, due to the limitations of these satellite retrieval algorithms over land, specifically lack of the LWP from the radiometer measurements, the following analyses are limited to warm clouds over the ocean. L'Ecuyer et al. (2009) found a suppression of precipitation with increasing sulphate aerosol index for both stable and unstable environments and all liquid water paths. The effect was larger for lower liquid water paths (i.e. higher LWPs are required for precipitation to develop in regions of high aerosol index). An increase in precipitation was observed when sea salt aerosols were present in high concentrations in unstable environments and this trend increased with increasing LWP. They also found some evidence for increased cloud depth with increased aerosol loading, particularly in unstable environments.

Lebsock et al. (2008) found a reduction in LWPs in high aerosol environments for non-precipitating clouds, suggesting a reduction in the albedo enhancement predicted by equation 1.1. They estimated the first indirect effect at -.042 W/m^{-2}, which is lower than the IPCC reports. This reduction in LWP was greater in unstable environments than stable ones, which indicates a greater sensitivity of the LWP of cumulus clouds than of stratus clouds to aerosol. They also found a reduction in precipitation with increasing aerosol, the magnitude of which depended on the thermodynamical state of the environment, but the reduction was approximately 5% in all LWP bins over $80gm^{-2}$ (Lebsock et al. 2008). The study also looked at the regional trends of these effects and found that in general, the magnitude of the indirect effect was larger in the extra tropics and subtropical stratus regions. Along these lines, the susceptibility to changes in LWP was larger in the tropics, where the environments tend to be thermodynamically unstable, suggesting that stable clouds would be more vulnerable to the albedo affect since the LWP effect can act as a compensator for the unstable clouds.

Global satellite studies provide insights into the large-scale magnitude of aerosol indirect effects that may otherwise be unknown. However, there are also some problems with satellite studies and the data used in them. For example, the only satellite measurement of aerosols comes in the form of the aerosol optical depth from MODIS. Although this quantity can be related to aerosol concentrations, it is not a direct measurement of these properties. Along with this, vertical profiles of aerosols cannot be obtained from satellite data, which can be an important measurement. Also, some satellite studies may even use model or reanalysis data for several quantities, including aerosol properties as well as meteorological variables such as stability. Another possible issue has to do with satellite retrievals of certain variables. For example, LWP is often retrieved using microwave radiometer brightness temperatures. These retrievals do contain biases, especially for high LWPs. Therefore, it is beneficial to check the results from satellite studies, necessitating regional studies using ground-based data.

1.4 Suppression vs. Enhancement and Cloud Type

Khain et al. (2008 and 2009) have attempted to classify aerosol effects through several comprehensive model simulations (including modeling of maritime and continental clouds as well as several differing thermodynamic conditions), which is presented in figure 1.2. They designate the Δ G (generation) as the difference in hydrometeor condensate mass formed by the drop condensation and ice deposition and Δ L as the difference in the precipitation loss due to evaporation and ice sublimation. Therefore, the actual change in precipitation (Δ P) due to increased aerosol is determined as the difference between these competing mechanisms, or

$$\Delta P = \Delta G - \Delta L \tag{1.2}$$

The simulations used for this classification scheme included several different types of maritime and continental clouds, as well as differing CCN concentrations, although it should be noted that only single clouds were modeled, not organized cloud complexes. The thermodynamical conditions were altered in the marine cloud experiments, providing a relatively complete set of meteorological conditions upon which the aerosol effect could be tested and observed. A table listing the experiments is included in Khain et al. 2008 (table 1). Referencing figure 1.2, an increase in aerosol (where point A denotes an initial condition characterized by a relatively low aerosol concentration) can lead to an increase in both condensate production (Δ G) and condensate loss (Δ L). If the condensate loss is greater than the production, it leads to a decrease in precipitation and

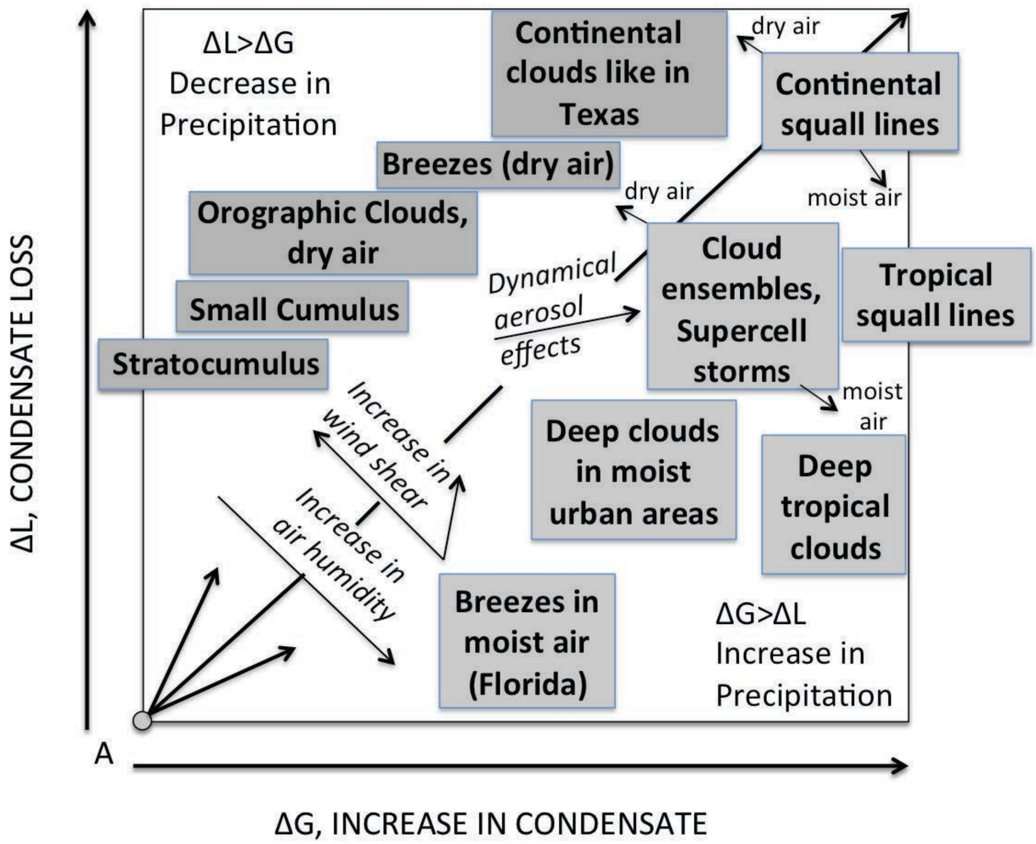

Figure 1.2
A schematic diagram of the aerosol effects on different types of clouds and cloud systems. Following figure 17 from Khain et al. (2008). The region above the diagonal line corresponds to a decrease in precipitation with an increase in aerosol concentration. The region above the diagaonal line corresponds to an increase in precipitation.

the opposite scenario leads to an increase in precipitation. The main factor controlling this designation is the cloud type, because both Δ G and Δ L depend on the convection structure. Small isolated clouds experience intense mixing with the environment, increasing condensate loss. However, larger cloud clusters, supercell storms and squall lines have large condensate production and relatively low Δ L because humidity is high within the convection zone (Khain et al. 2008). Note that the relationship between Δ G and Δ L also depends on other thermodynamic parameters, namely atmospheric instability, relative humidity and vertical wind shear. Khain et al. (2008) point out the 'crucial role of relative humidity in determining the sign of the precipitation response to aerosols' which tends to increase the condensate production. They also mention the importance of vertical wind shear for increasing condensate loss, as can be seen in figure 1.2. Instability can increase the vertical velocity in clouds, transporting more drops to upper levels and thus increasing both G and L, similar to aerosol effects. This makes the problem of separating aerosol and instability effects more difficult, but according to the authors, the net effect depends on the relative humidity.

A more recent modeling study, which also investigated aerosol forcing on a wide range of cloud types, was performed by van den Heever et al. (2011). However, in this study, the clouds were all allowed to develop within a large domain under differing environmental conditions. Specifically, they were modeling aerosol effects on tropical convection, allowing shallow as well as deep convection to develop. Unlike Khain et al. (2008), this allowed for modeling of multiple clouds rather than individual clouds. Similar to Khain et al. (2008), they did observe large differences between cloud types. In general, they found suppression of precipitation due to aerosols in the shallower clouds, while precipitation enhancement was observed in the deep convection. This was mostly attributed to the role ice plays, as has been discussed in this chapter, and is in agreement with figure 1.2.

The examination of the Khain et al. (2008) study and its classification technique as well as van den Heever et al (2011) provide further motivation for the current study. Ground based observational data from both maritime and continental locations will be assessed in order to validate the Khain et al. (2008) hypothesis presented in figure 1.2. It uses a number of well-instrumented surface observations that allow different cloud regimes to be compared and contrasted to the aforementioned global observational and modeling studies.

1.5 Buffering

A recent result that will be invoked in this study involves buffering of aerosol effects through cloud dynamics. Stevens and Feingold (2009) introduced a theory, which argues that the aerosol indirect effects are smaller than originally anticipated in theory due to a process they call 'buffering'. They argue that there are several mechanisms, both microphysical and macrophysical, which can weaken the expected aerosol effect by absorbing the impact of the original forcing. They present examples of both microphysical and macrophysical buffers, including the fact that an increase in wind speed over the ocean can produce more sea-salt aerosol, inhibiting precipitation development, while also producing more sea-spray and large cloud condensation nuclei which can help to enhance the efficiency of rain production. Another example presented was that through recent observations, it has been suggested that the precipitation susceptibility of shallow clouds is less than predicted by simple scaling arguments and what is currently used in most cloud resolving model microphysical schemes. This would mean that the sensitivity of rain to droplet number is weaker than theorized. It could be argued that the results presented in this study are consistent with these ideas, and this will be discussed further in chapter 5.

An example of a microphysical buffer, as discussed in Stevens and Feingold (2009) has to do with the fact that the flux of liquid water through a cloud layer is thought to be important for the growth of that layer. For example, in shallow convective type clouds, this would mean that a precipitation suppression results in more liquid water being lofted to the cloud-top region, where it evaporates. The cooling associated with this evaporation then destabilizes the environment further, making it conducive to the growth of deeper clouds. These deeper clouds tend to produce more rain than shallower clouds; hence increases in aerosols in these cases could produce precipitation enhancement, rather than suppression. This idea is similar to the theory of aerosol effects on deep convection (reference section 1.2), except that in this case, the evaporation leads to the cooling rather than the freezing of the supercooled water.

The results of the study by van den Heever et al. (2011) discussed in section 1.4 are an example of a case that is consistent with this theory. They modeled multiple cloud types and depths in a single domain and found that the aerosol effects on precipitation in shallow clouds tended to offset those in deep convection, with opposite trends. Therefore, they found that the buffering operated over complexes of clouds, rather than individual clouds, observing a muted response when analyzing the entire model domain compared to each type of cloud. This buffering idea is crucial to understanding the results presented in chapter 4 and will be discussed further in chapter 5.

As has been discussed in this chapter, there have been many modeling studies on aerosol effects as well as observational studies using global satellite data, with differing results as to the magnitude and sign of the precipitation response. There have also been studies looking at aerosol effects on specific cloud types. Some of these have even been regional observational studies, although they have focused on marine clouds. The current study was performed in an attempt to observe aerosol effects on precipitation on a regional scale in different climatological conditions and for different cloud types. It was designed to fill the gaps that have not been addressed in these previous studies and provide the first observational examination of the effect of aerosols on the onset of precipitation at a regional scale in a marine as well as a continental location. It is focused on warm clouds in order to avoid the complexities that ice processes can introduce on these aerosol effects, as was discussed in this chapter.

CHAPTER 2

INSTRUMENT AND DATA DESCRIPTION

In order to classify the aerosol effects on clouds and precipitation in two distinct environments as described in chapter 1, observational data must be employed. The study is limited to warm clouds for reasons described in chapter 3. The best instrumentation for determining cloud and precipitation properties are radars and microwave radiometers. Therefore, these instruments must be collocated in order for a site to provide sufficient data for this study. The Atmospheric Radiation Measurement (ARM) Program provided these, as well as other meteorological instrumentation that proved important for this study, at several sites. This program, as well as the data supplied, is discussed in this chapter.

2.1 ARM Program and Site Locations

2.1.1 ARM Program

The ARM Program was created in 1989 with funding from the United States Department of Energy (DOE). It was formed in order ‘to develop several highly instrumented ground stations to study cloud formation processes and their influence on radiative transfer’ (US DOE 2011a). In fact, its primary objective remains the improvement of ‘scientific understanding of the fundamental physics related to interactions between clouds and radiative feedback processes in the atmosphere’ (US

DOE 2011a). The mission statement describes the ARM Climate Research Facility as a national user facility for the study of global change by the national and international research community. The program has set up several permanent sites and now also has two mobile facilities in order to fulfill its mission. The three permanent sites were chosen for their broad range of weather and climate conditions with one located in Oklahoma, another in Alaska and the third has three sites of its own in and around Northern Australia. The ARM mobile facilities contain much of the same instrumentation as the permanent sites and can support short-term experiments in different locations.

2.1.2 Site Locations

Data was analyzed from several of the ARM permanent and mobile facility locations; however, only two will be used for the final analysis presented in chapter 4. These are the Southern Great Plains site in Oklahoma and the Graciosa Island deployment of the AMF (ARM Mobile Facility) in the Azores. These sites were chosen due to the length as well as the completeness of their data records.

The other sites that were examined but are not being used are the AMF deployment in Shouxian province in China in 2008 and the AMF deployment in Niamey, Niger in 2006. The Niger location was not used because it was determined that the Oklahoma site was sufficient for observing continental clouds with a more convective nature (i.e. shallow cumulus) and because it had an order of magnitude fewer cases of warm clouds than either the Oklahoma or Azores sites (15,000 and 18,000 cases respectively compared to 1600 at Niger), rendering the few trends that could be observed insignificant and weak compared with the sites used.

The China site was not used because of difficulty with the instrumentation during the deployment, which led to a reduced data record for those instruments. The radar was only operational at the site for two months of the eight-month deployment, during which time the microwave radiometer had issues and was not working for days at a time. Also, the two months of radar operation were during the fall, which is not a very active time for warm clouds in this region. Since the focus of this study will be on warm clouds, all of these factors produced only a few cloud events that could be analyzed and greatly reduced the robustness and usefulness of the results.

2.1.2.1 SOUTHERN GREAT PLAINS SITE

This site has been operational in Oklahoma since 1992, however additional instrumentation and data processing capabilities have been added in the successive years. According to the ARM website (US DOE 2011f), it was chosen as a permanent ARM site because of its ‘relatively homogeneous geography and easy accessibility, wide variability of climate cloud type and surface flux properties and large variation in temperature and specific humidity’.

The site was chosen for this analysis because of its relatively long data record and its continental location in the mid-latitudes. The location was also not near a major urban area, so that the possible effects of an urban region (as was observed in Carrio et al. 2010) on clouds and precipitation did not influence the results. Data from this site was analyzed from June 2005-September 2008 to correspond to the availability of the W-band radar. In order to keep the results consistent between sites, the Southern Great Plains

(SGP) data was only used when the W-band radar was operational at this site since the Azores site also used a W-band cloud radar.

2.1.2.2 AZORES SITE

The site at Graciosa Island was established in order to study ‘processes controlling the radiative properties and microphysics of marine boundary layer clouds’ (US DOE 2011b). Marine boundary layer (BL) clouds are very important to the global climate system particularly because they span large areas in the ocean and changes to them (e.g. due to increasing pollution) can alter the global radiative budget (as was discussed in section 1.2). These clouds are also important for controlling sea surface temperatures and can influence the strength of the trade winds (US DOE 2011b). The main objective of this AMF deployment was to create the first climatology of cloud and precipitation properties of low clouds at a remote subtropical marine site.

This site was chosen for this study because of this climatology and its marine location. It is a relatively long record of aerosol, clouds and precipitation data and at similar latitude as the SGP site (see figure 2.1). The ability to examine data from two sites with different climatology and cloud types is crucial for this project. The data from this site were collected from May 2009-December 2010 and although the data record for this site is not as long as the SGP data record, there is a similar magnitude in number of cases that can be used from each site. This is because there are more periods of warm clouds in the Azores than at SGP, which experiences more mid-latitude cyclones as well as deep convective storms, while the marine environment at the Azores allows for stratocumulus and other long lived warm clouds.

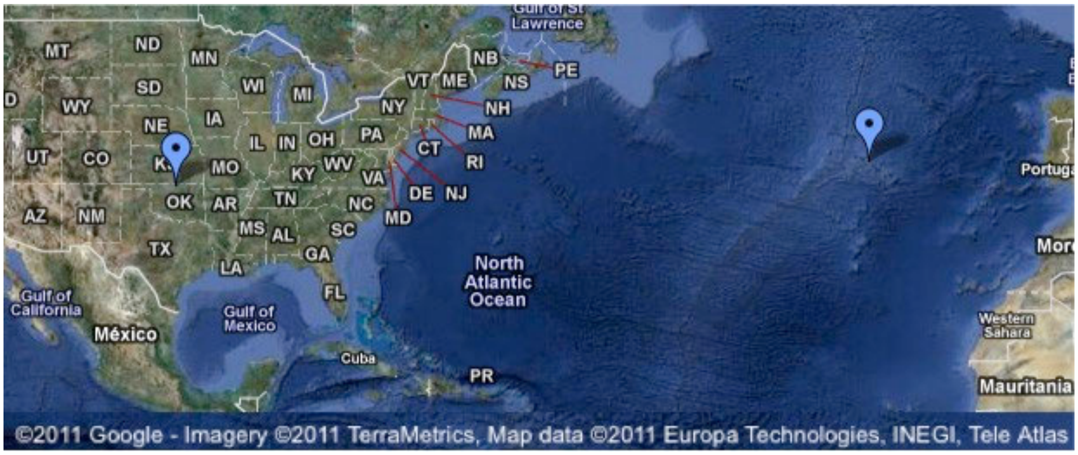

Figure 2.1
Map of the locations of the ARM sites used in this study. Azores is on the left in the Atlantic, while the Oklahoma site is shown on the right, over the United States.

2.2 Cloud Properties

2.2.1 W-Band Cloud Radar

The W-band ARM Program Cloud Radar systems are ‘zenith pointing Doppler radars that probe the extent and composition of clouds at 95.04 GHz’, up to 15km (US DOE 2011e). Millimeter wavelength radars are much better at detecting non-precipitating clouds than traditional weather radars (usually centimeter or longer in wavelength). Cloud droplets are too small to be detected by other radars, which are designed to detect precipitation-sized drops. The smaller wavelength of these radars allows detection of drizzle, which is not possible with larger wavelength radars, and they are less susceptible to Bragg scattering, which are backscattered returns caused by variations in refractive index associated with turbulence (Kollias et al. 2007).

Although the millimeter wavelength radars are helpful for studying clouds, their short wavelength also produces some issues that are not observed with centimeter wavelength radars. The first of these is signal attenuation. At 94 GHz attenuation from gases, especially in the tropics, is significant (Kollias et al. 2007). Signal attenuation induced by liquid hydrometeors is also significant at these frequencies, hence millimeter wavelength radars (such as the ARM W-band cloud radar) are heavily attenuated by precipitation and cannot provide useful information about rain when they scan (Kollias et al. 2007). Also, when these radars are used to look at large hydrometeors, the size parameter of the particles approaches unity or higher. This means that the Rayleigh approximation is no longer valid, causing the total extinction coefficient of raindrops at 94 GHz to be up to 1000 times that at 3GHz (Kollias et al. 2007). This suppresses the dynamic range of observed radar reflectivity values in rain and limits the penetration of millimeter-wavelength radar signals to a couple of kilometers into the precipitation layer at high rainfall rates (Kollias et al. 2003). However, these issues should not present a large problem in this study because it does not investigate the properties of the rain itself, but instead investigates only the probability of the onset of precipitation. Also, the limitation to warm cloud cases limits the distance over which this vertically pointing radar can be attenuated. Since the vertical profile of the rain is not studied and the vertical extent of the clouds examined is limited, neither the attenuation of the radar beam nor the non-Rayleigh scattering of the radiation are thought to impact the current study.

For this study, the main determination of aerosol impacts on precipitation was through the probability of precipitation. In order to determine if a cloud was precipitating, the maximum reflectivity was found between the surface and 1 km above the freezing level (discussed in section 3.2.1) at each time step. The cloud was considered to contain drops large enough to precipitate (even if this was just drizzle) if the maximum reflectivity was greater than -15dBZ (Haynes and Stephens, 2007).

2.2.2 Microwave Radiometer

The microwave radiometer provides measurements of column-integrated amounts of water vapor and liquid water. It is essentially designed as a microwave receiver that is tuned to measure the specific microwave emissions of the vapor and liquid water molecules in the atmosphere at 23.8GHz and 31.4GHz (US DOE 2011d). ‘Cloud liquid water in the atmosphere emits in a continuum that increases with frequency, dominating the 31.4GHz observation, whereas water vapor dominates the 23.8GHz-channel’ (Morris 2006). Therefore, observing at these two frequencies can separate the water vapor and liquid water signals. This instrument provides measurements of the brightness temperatures at each of the two frequencies as well as total water vapor and total liquid water along a line-of-sight path.

The radiometer data included a wet flag that identified if the instrument was wet or not. This is a useful quantity since the radiometer data should not be used when the instrument is wet, as it can greatly affect and bias the data (it measures the liquid on the instrument instead of in the cloud). However, this is also true with the radar data, since rain heavily attenuates its signal. Therefore, times when the radiometer was wet were filtered out of the data for the final results presented in chapter 4 by using the wet flag.

2.2.3 VAPs/ARSCL Data

Along with the data from their instrumentation, the ARM program also produces some additional data products, which essentially use algorithms developed by ARM scientists to process the data collected into supplementary variables of interest to those using the data. One of these ‘Value Added Products’ (VAPs) is the Active Remote

Sensing of CLouds (ARSCL). This VAP uses data from the millimeter wavelength cloud radar, laser ceilometer, microwave radiometer and micropulse lidar to produce a time series of vertical distributions of cloud hydrometeors over the ARM sites (US DOE 2011g). This VAP was used for determination of the cloud base and top heights. Specifically, it uses a cloud mask algorithm developed by Clothiaux (1995) to determine the vertical distribution of cloud hydrometeors from millimeter radar reflectivity. It defines cloud layers by using the different modes of the radar to decide if the backscatter power return is above the noise level of the receiver, hence detecting particles in the radar sample volume. The cloud base height itself was determined mainly from the ceilometer and lidar instruments located at the site. Generally, the ceilometer is used in the lower part of the troposphere due to its high resolution. However, it is also used if it is within 600m of a cloud detected by one of the two micropulse lidar cloud-detection algorithms. Otherwise, the height detected by either the Campbell et al. (1998) or the Clothiaux et al. (1998) algorithm is used. If these algorithms detect no cloud base height, either the clear sky or a no data value is reported, depending on if the laser systems produce no data or if no cloud is detected by the algorithms.

The data from the ARM ARSCL VAP provided cloud base and top heights for up to 10 layers of clouds. The SGP ARSCL dataset had two different algorithms for retrieving cloud base and top heights, the Clothiaux et al. and the Campbell et al. algorithms (the Azores site ARSCL data had only one set of cloud base and top heights). Since these were not very different, the Clothiaux et al. algorithm was used for consistency with a previous study (Feng et al. 2010 and personal communication). For the current study, since it was limited to warm clouds (discussed in section 3.2.1) and these tend to remain relatively low in the atmosphere, only the first (lowest) cloud layer was used. The cloud thickness was then found by subtracting the cloud base height from the cloud top height.

2.3 Aerosol Properties

The aerosol observing system (AOS) is the primary ARM instrumentation for observing aerosols at the surface and the AOS instrumentation at both the sites used in this study are identical. 'The principal measurements are those of the aerosol absorption and scattering coefficients. Additional measurements include those of the particle number concentration, size distribution, hygroscopic growth, and inorganic chemical composition' (US DOE 2011c). The instruments included in the AOS are two nephelometers, a light absorption photometer, a condensation nuclei counter, a cloud condensation nuclei counter and a continuous light absorption photometer. The data for this study come from the condensation nuclei counter, which measures the total number concentration of condensation particles of diameter in the size range of 10nm to 3μm (Jefferson 2005). The CCN counter, on the other hand, measures the cloud droplet number concentration at 7 supersaturations as well as the droplet size distribution from 1μm to 10μm in 20 size bins (Jefferson 2005).

In order to compare CN (Condensation Nuclei) and CCN (Cloud Condensation Nuclei) concentrations, the CCN values were calculated from the raw ARM data (at a single supersaturation value). The CCN counter works by stepping up and down through the 7 supersaturations every 5 minutes in a pyramid scheme (Kreidenweis & Jefferson, personal communication). The CCN concentration was calculated at a certain supersaturation (as an average over the time the instrument spent in that supersaturation), depending on the location. A slightly higher supersaturation was used for SGP than the Azores (1.0% vs. 0.8%, respectively), since it generally has more convective type clouds. A higher supersaturation corresponds to more convectively active clouds that will activate more of the CCN while lower supersaturations correspond with more stable or stratiform clouds (Kreidenweis, personal communication). The CN values were binned and the average CCN value for each bin was plotted in order to observe the resulting trend (refer to section 3.3.1 for more information about this process). It was determined from figure 2.2 that the increasing CN concentrations would provide a reliable proxy for increasing CCN concentrations, even though it was not a one to one relationship, since there was a strong trend between the two quantities.

The CN concentration was used in this study instead of the CCN concentration for several reasons. First, the CCN counter only produces a measurement at a certain supersaturation every half hour or so because of its need to step through the supersaturations. Since the other data related to the cloud properties is collected every minute, it was difficult to match comparable quantities. Also, since the two locations experience quite different cloud types, it was difficult to determine which of the supersaturations would be most appropriate for determining the CCN concentration. Lastly, as previously mentioned, the two quantities (CN and CCN) seem to at least correlate with each other in both locations (figure 2.2). As such, it was concluded that CN concentrations would provide a relatively good proxy for CCN concentrations. The focus of this study is on how increasing pollution can affect the onset of precipitation, not on the specific effects of increased CCN on cloud microphysics, so the CN proxy should be sufficient for these purposes.

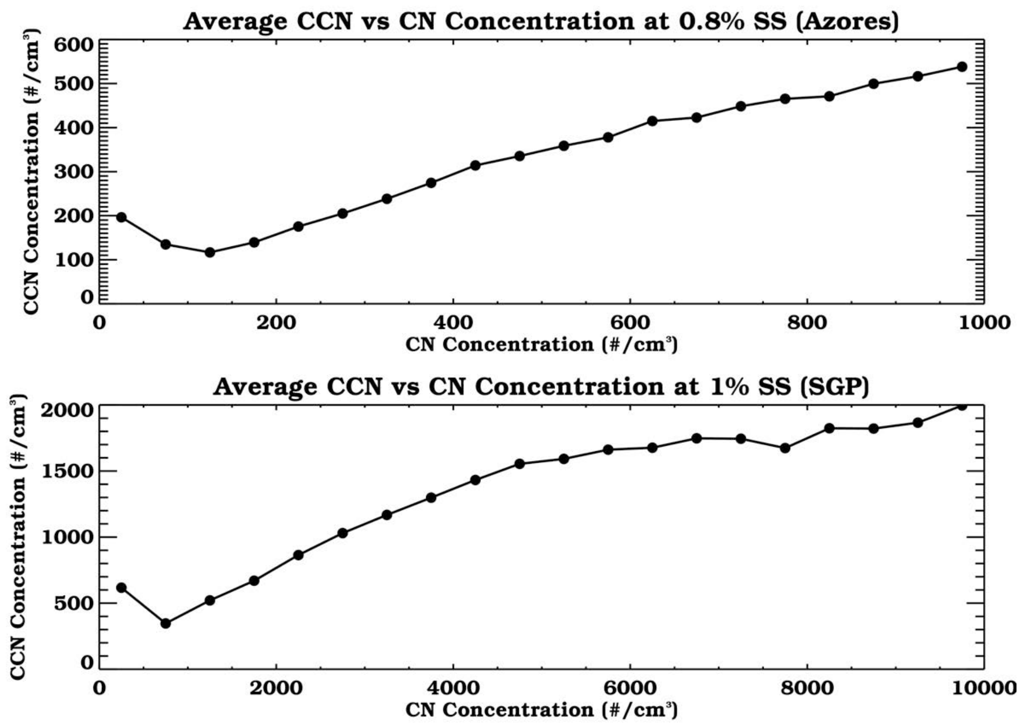

Figure 2.2
Average CCN concentration binned by CN concentration. Upper panel shows the trend at Oklahoma, while the lower panel shows the trend at the Azores. General trend is for the CCN concentration to increase with increasing CN concentration. SS is the supersaturation value used.

2.4 Surface Measurements

The ARM surface meteorology instrumentation includes measurements of surface wind speed, wind direction, air temperature, relative humidity, barometric pressure and rain-rate. All of these parameters were used at some point in this analysis, except for pressure. At both site locations, wind speed and direction are both measured by a propeller anemometer, while a digital barometer measured pressure. However, at SGP an electrically heated tipping bucket precipitation gauge measured the rain-rate while at the Azores it was measured by an optical rain gauge. In Oklahoma, a Thermistor and Vaisala RH were used to measure temperature and relative humidity (RH), respectively. The

Azores site temperature and RH probe is listed as a platinum RTD and RH (ARM MET handbook 2011).

2.5 Sounding Data

The balloon-borne sounding system provides measurements of the vertical profiles of both the thermodynamic state of the atmosphere as well as the wind speed and direction. These balloons were launched at each site 4 times per day, at approximately 0, 6, 12 and 18 UTC. These soundings were used to determine the atmospheric stability and the freezing level so that cold clouds could be removed from the analysis. The method used for this screening is introduced in chapter 3, section 3.1. Although the rest of the cloud data is collected at least every minute, the sounding data are still considered to be valid for use in this study despite their 6 hourly time resolution. This is because the large-scale meteorological properties such as the freezing level and stability do not change as rapidly in time as cloud microphysical properties, so the sounding data should still prove relatively accurate despite their lower time resolution. Also, the sounding data provides information on the general dynamic background state, rather than the thermodynamic state of individual clouds; therefore it should be suitable for the purpose of measuring the background meteorological conditions.

CHAPTER 3

METHODOLOGY

3.1 Sounding Data Procedures

The sounding data were used for several calculations of large-scale meteorological conditions. If the vertical temperature profile that the radiosonde measured dropped below 0^{o}C, then that was determined to be the freezing level. The overall stability of the atmosphere was calculated using the potential temperature profile, which was calculated from the temperature and pressure profiles. The lower tropospheric static stability (LTSS) is then calculated using the method of Klein and Hartmann (1993), by taking the difference of the potential temperature between 700hPa and the surface. This stability measure was developed specifically for oceanic conditions and so provides a good estimate at the Azores site. However, the general measure of stability used over land is Convective Available Potential Energy (CAPE) and although this can be calculated from soundings and would be the best measure of continental atmospheric stability, it is not used in this study because of the nature of the clouds. The limitation to warm clouds precludes the inclusion of deep convection, which is often the only type of cloud for which there will be a positive and useful value of CAPE. However, there can still be shallow convection and cumulus clouds or more stratiform clouds in a location, so stability is still a necessary measure of the atmosphere for a gauge of the meteorological conditions. Because of this, the LTSS is still used at Oklahoma, however, it was calculated using the difference of potential temperature between 600hPa and the surface. This was done because the atmospheric boundary layer generally extends higher over land than over the ocean and so the stability proxy should take this into account (van den Heever, personal communication). However, it is still not a perfect measure of stability in this location and this should be noted while analyzing the study results. Another method attempting to separate convective- from stratiform- type clouds will also provide a check into this measure of stability (refer to section 3.2.2).

3.2 Cloud Limitations and Algorithms

3.2.1 Warm Cloud Limitation

This study was focused on warm clouds in order to try to limit the possible effects of ice on precipitation processes and distinguish the effect of aerosols on warm rain processes. This limitation was also imposed because another recent publication has already examined the aerosol effect in a similar manner on all clouds at the SGP site. Filtering of warm rain cases was done by requiring at each time step that the cloud top height from the ARSCL data be less than or equal to 1km above the freezing level as determined by the radiosonde. An example day from the Azores is shown in figures 3.1 and 3.2, first with all the data points and then with the cold cloud cases removed, as well as times when the radiometer was wet (see section 2.2.2). The warm cloud limitation on cloud top height was extended to 1km above the freezing level due to the fact that supercooled water and warm rain processes can still exist above the freezing level.

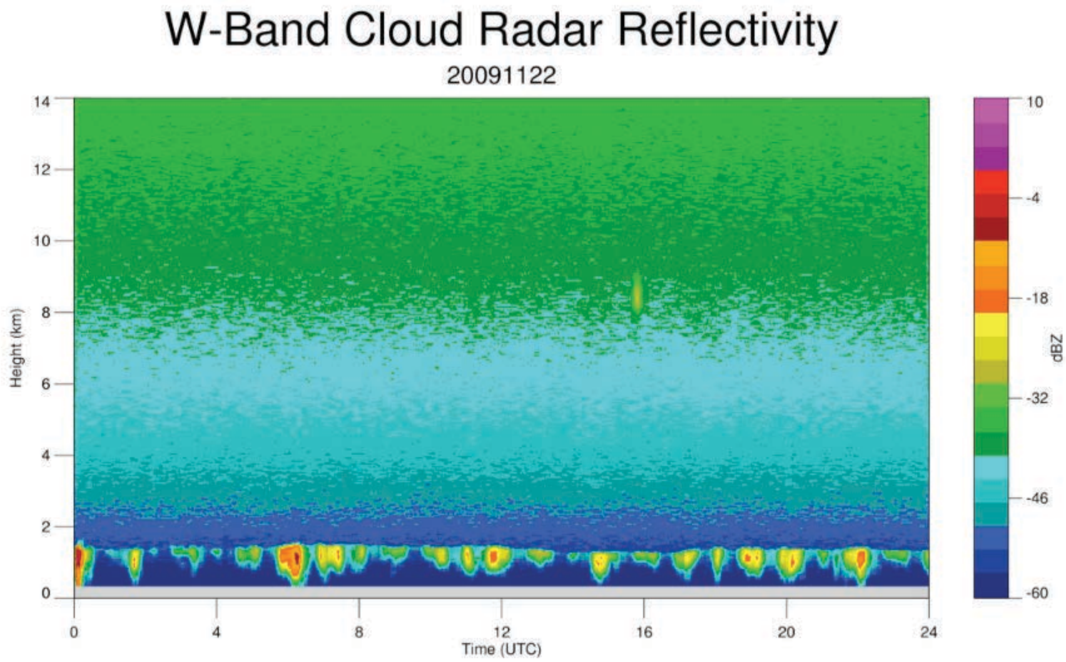

Figure 3.1
W-band radar reflectivity from a single day at the Azores (November 22, 2009).

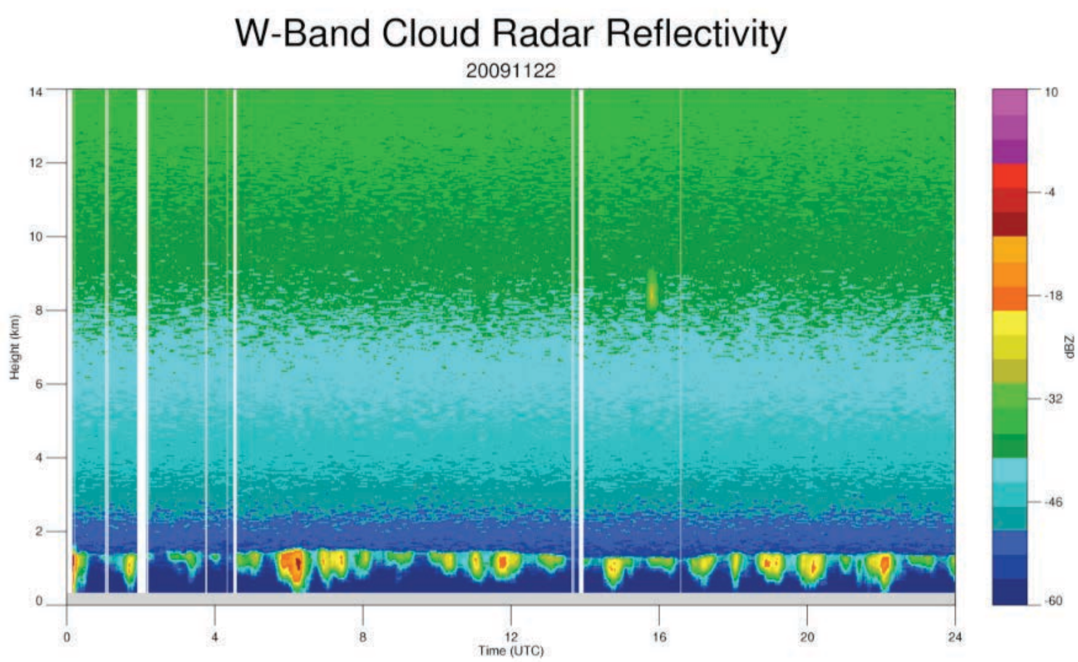

Figure 3.2
W-band radar reflectivity from a single day at the Azores (November 22, 2009). Times with rain at the surface, cold clouds and missing or bad data have been removed as is discussed in this chapter.

3.2.2 Stratus vs. Convective Clouds

As was mentioned in section 1.4, one of the main factors controlling aerosol effects on precipitation is the cloud type. In an attempt to separate out and observe the effects on different cloud types, an algorithm for classifying clouds was developed. This algorithm attempted to split the clouds into more ‘convective’ type clouds and more ‘stratiform’ type clouds by using the root mean square (RMS) of the radar reflectivity at cloud top. In general, it was determined that the lowest RMS (after being split into thirds at each location) would correspond to what will be called ‘stratiform’ clouds; although these are not necessarily stratus clouds, they do have the lowest horizontal variance in radar reflectivity at cloud top and low horizontal variability of cloud parameters is a general characteristic of stratiform clouds. The highest third of RMS will then correspond to ‘convective’ type clouds, as they tend to have less homogeneity.

When stratus and convective clouds are referenced in this study, it generally corresponds to these definitions based on the RMS; if other definitions are used, it will be distinguished.

3.3 LWP Binning and Plotting methods

Figure 3.3 shows the LWP for each location of this study (Azores site shown in the upper panels, SGP in the lower panels) plotted against CN concentration with error bars indicating the standard deviation of each bin. As one can see, the standard deviation at each site is quite large, indicating the its large variability. LWP, however, correlates fairly well with the probability of precipitation. As such it was necessary to separate this relationship out in order to observe the aerosol effect without bias from the difference in available cloud liquid water.

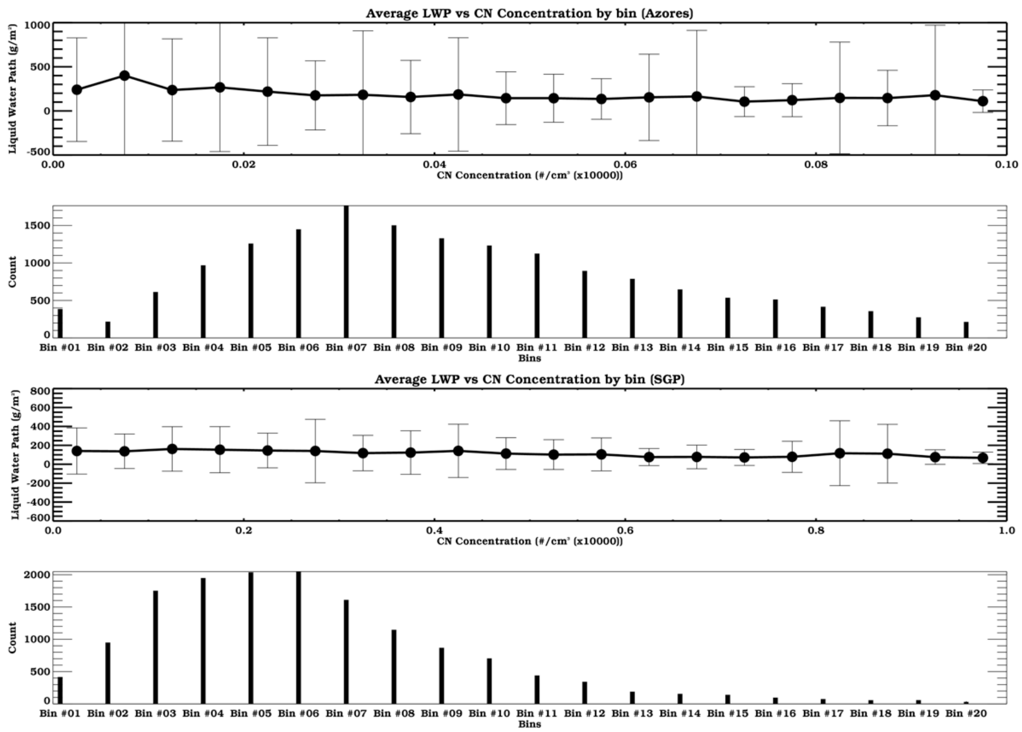

Figure 3.3
Average LWP of each bin, separated and plotted by CN concentration. Azores site is shown in the upper panels, while the SGP site is shown in the lower panels. Error bars indicate standard deviation of each bin.

In subsequent plots, low LWPs have been grouped into a single bin and the very high LWPs have also been grouped into their own separate bin. These groupings were determined by the fact that LWPs greater than about 300 g/m^2 always had a high probability of precipitation (generally greater than 0.6) while LWPs less than about 80 g/m^2 always observed a very low probability of precipitation and through a similar finding by Sorooshian et al. (2009). This allows for a more detailed view of the middle range LWPs, which is the range in which precipitation is the most susceptible to aerosol perturbations (Sorooshian et al. 2009). Clouds with low LWP generate little rain to begin with and are therefore less susceptible to aerosol, while clouds with high LWP also have a relatively low susceptibility to aerosol because the precipitation process is efficient due to the abundance of liquid water (Sorooshian et al 2009). Therefore, the focus of this study has been on the mid-range of LWPs.

CHAPTER 4

RESULTS

The following results are all sampled at 10-minute intervals. The only data set for which this is not true is the sounding data; the time resolution of these data is discussed in section 2.5. The results are also limited to times when warm rain clouds are present and the radiometer is not wet, as was discussed in chapters 2 and 3 (sections 3.2.1 and 2.2.2, respectively). The only exception is section 4.2 because this deals with precipitation at the surface, measured by the rain gauge and is therefore not subject to the bias experienced by the radiometer and radar when wet. This method does cause times with raining clouds to be left out of the final analysis. However, it is thought that this will not bias the precipitation trends due to the fact that precipitation-sized droplets can form in the cloud without hitting the ground. Therefore, analyzing times with clouds and no precipitation at the surface yet (through the wet flag) should be sufficient for observing trends in the precipitation. Another reason that the statistics should still be robust is that the screening was applied consistently across the datasets. As will be discussed in section 4.2, it is actually advantageous to limit the results to precipitation observed in the cloud rather than at the ground, due to the limited cases of precipitation reported at the rain gauges as well as the bias wind direction and speed can cause in the rain gauge measurements.

4.1 Probability of Precipitation

The probability of precipitation was calculated using the criteria mentioned in chapter 2 (section 2.2.1) and has been plotted in the figures included in this section. The probability of precipitation has been separated by LWP and also by the column water vapor (CWV) in order to compare results with Lebsock et al. (2008) and to determine if the effect of aerosols was dependent on cloud water or atmospheric moisture variables. It has also been stratified by stability in order to determine if the larger scale meteorological conditions alter the observed trends.

4.1.1 Azores

4.1.1.1 GENERAL RESULTS

The probability of precipitation was plotted as a function of LWP and stratified by aerosol and stability as was discussed in section 3.3.2. The plots for these variables at the Azores site are included in figure 4.1. In general, one can observe a suppression of precipitation with increasing aerosol content up to LWPs of 200 g/m^2 and a slight enhancement of precipitation at higher LWPs (panels a and b of figure 4.1). The suppression observed is consistent with past theories and studies presented in chapter 1, while the enhancement is not. Large LWP values may allow for efficient precipitation formation processes, therefore making these clouds less susceptible to the aerosol effect, as was discussed by Sorooshian et al. (2009). Where analyzed by environment, stable environments show strong suppression of precipitation up to 200 g/m^2 while unstable environments show little or no trend (panels c and d of figure 4.1). This is thought to be due to the fact that more convective clouds (which form in less stable environments) have

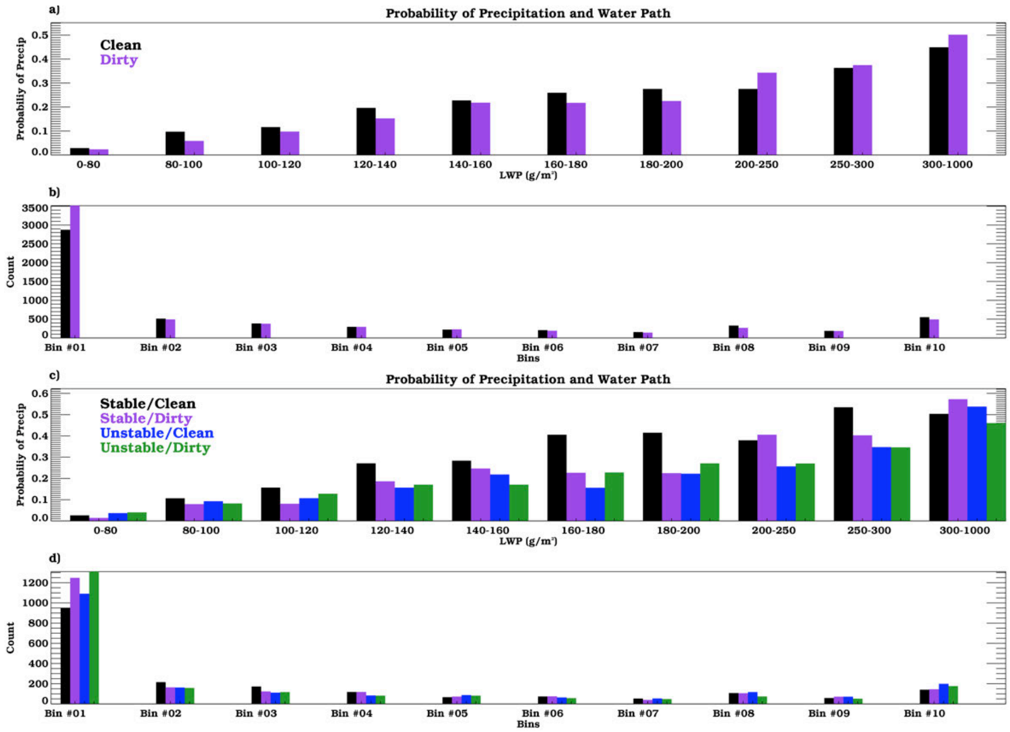

Figure 4.1
Probability of precipitation binned by LWP and histogram of bin populations at the Azores site. Probability of precipitation is determined by the radar threshold of -15dBZ. Panels a and b are separated by aerosol (top third and bottom third of the CN concentration dataset make up the 'dirty' and 'clean' distinctions, respectively). Panels c and d are separated by aerosol (same distinctions as upper figures) and stability (again, top third and bottom third of stability dataset represent the 'stable' and 'unstable' categories, respectively).

stronger updrafts and other dynamical factors that can overcome the aerosol suppression effect. The suppression of precipitation observed in most LWP bins is also consistent with the results of Lebsock et al. (2008), which was a similar study performed on satellite data at the global scale. The higher LWP values were not included in figure 8 presented in that study, hence it is unknown if the trend of increased precipitation with increased CN concentration observed in this study at those LWP bins is seen at the global scale as well.

Figure A1 shows the probability of precipitation binned by aerosol and relative humidity instead of stability, in order to test the hypothesis of Khain et al. (2008). In opposition to what would be expected according to figure 1.2, the moist and dry cases at the Azores behave similarly to observed trends in figure 4.1, with precipitation enhancement observed at high LWP ranges and precipitation suppression observed at lower LWP ranges. This shows that if relative humidity does play role in precipitation, it is more likely important only for the evaporation of drops below cloud base rather than the formation of precipitation processes. Although this would affect the response of precipitation reaching the ground, the focus of this study is on the onset of precipitation and not the hydorological effects. When separated by relative humidity, results for the following sections were also observed to be similar to the general trends and so it was determined that, at least for the Azores, water vapor was not as important of a controlling factor as originally hypothesized in section 1.4. Therefore, results stratified by relative humidity instead of stability are not included in sections 4.1.1.2 and 4.1.1.3. Results stratified by wind shear, as was also suggested by Khain et al. (2008) were not performed because wind shear has a strong effect only on deeper convection, as was observed by Fan et al. (2009).

The results were also stratified by CWV in order to see if this variable moderated the aerosol effects (since it is often well correlated with the probability of precipitation, Lebsock et al. 2008). However, the probability of precipitation does not show a consistent trend with aerosols when separated by CWV at the Azores site, as can be seen in figure 4.2 (panels a and b), in fact the results are very noisy. In contrast, Lebsock et al. (2008) found a decreasing trend of the probability of precipitation with increasing aerosol index in all water vapor regimes using satellite observations only over the oceans. In order to compare the current results to Lebsock et al. (2008), the probability of precipitation for a

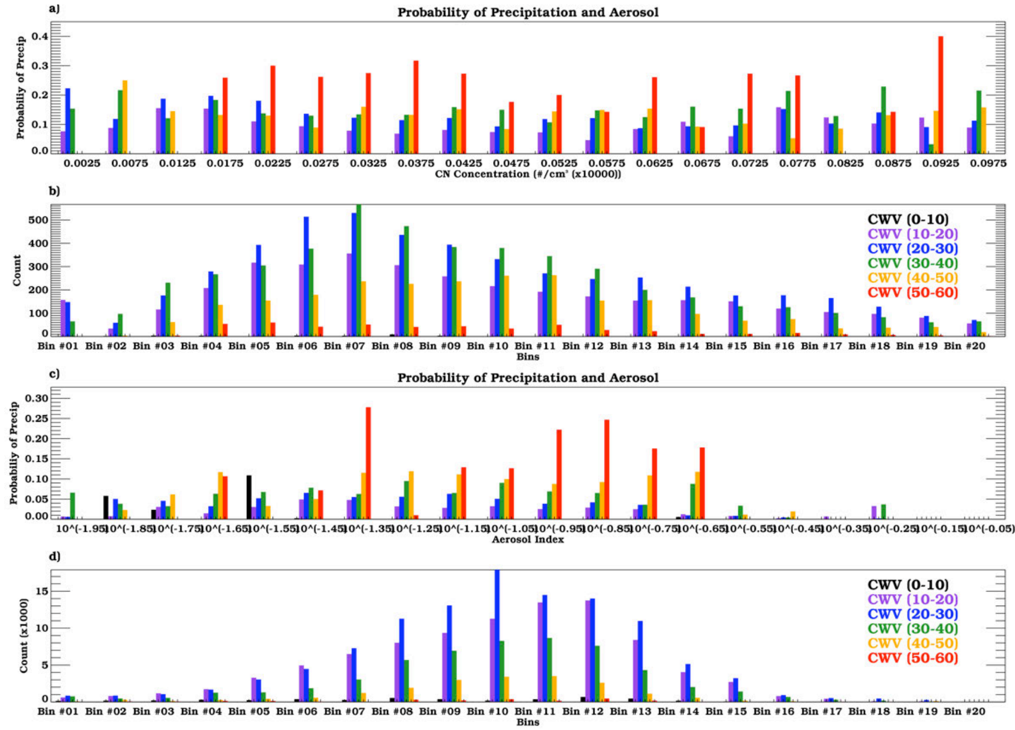

Figure 4.2
Probability of precipitation binned by CWV (color-coded) and CN concentration and histogram of bin populations. Panels a and b show the data from the Azores site, while the panels c and d show data from multiple satellite sensors, including Cloudsat (probability of precipitation), MODIS (aerosol index) and AMSR-E (CWV). This data was collected from July 2006 through October 2007 and was limited to region 10^oN-60^oN latitude and 10^oW-50^oW longitude (a box centered around the Azores site). This data came from Lebsock (personal communication).

smaller region around the Azores (using CloudSat data) was provided by Lebsock through personal communication. This was plotted in the same manner and it illustrated that the global results do not hold up at the regional level, as can be seen in figure 4.2 (panels c and d). Hence, even when an aerosol effect is observed on a global scale, the regional effect is still very uncertain.

It is noteworthy that the global scale aerosol effects were not seen at the regional scale in either of these studies, especially considering that the trends held up at the regional scale when the probability of precipitation was separated by LWP instead of CWV. The explanation for this is most likely due to the fact that CWV does correlate reasonably well with precipitation on larger scales, while LWP tends to relate to precipitation better on smaller, more regional scales.

4.1.1.2 SEASONAL RESULTS

Figure 4.3 (panels a and b) shows that the summer season reflects the same trends as was observed in the total data set, however, the other seasons do not show the same trends. Winter, for example, tends to show precipitation enhancement with aerosols in every LWP bin (figure 4.4, panels a and b), while the transitional seasons show almost opposing effects (figures A5 and A6, panels a and b). In spring (March, April and May, or MAM, figure A5), the precipitation tends to be suppressed with increasing aerosol in

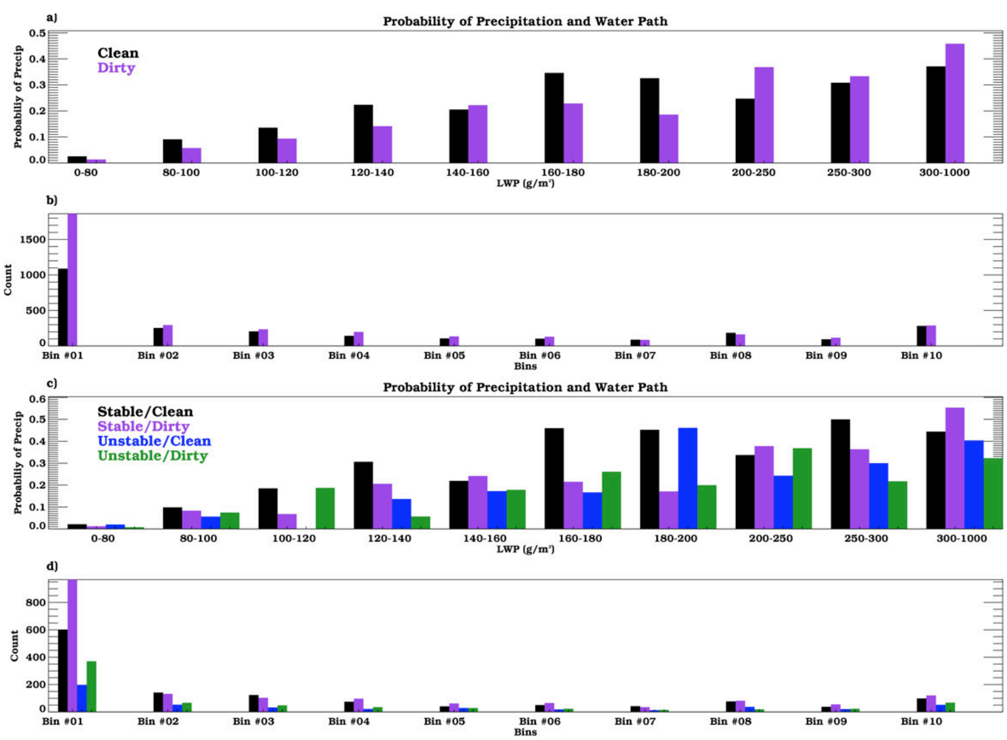

Figure 4.3
Probability of precipitation binned by LWP and histogram of the bin populations at the Azores site for summer (JJA) only. Panels a and b are separated by aerosol, while panels c and d are separated by aerosol and stability.

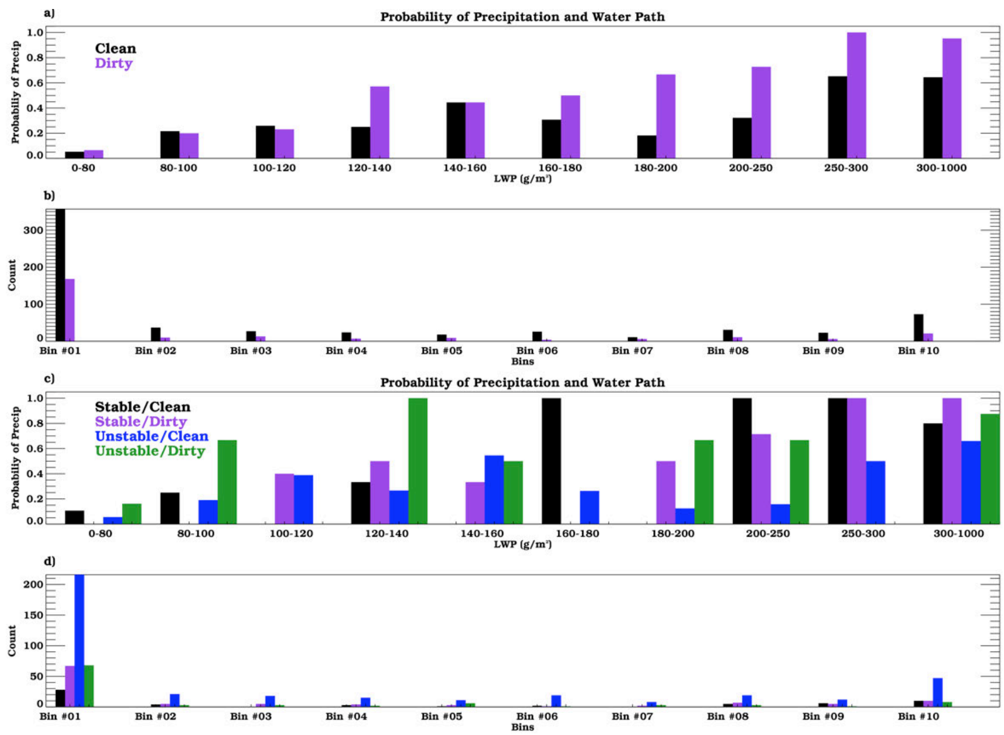

Figure 4.4
Same as figure 4.3, except for winter (DJF).

most LWP bins. On the other hand, in fall (September, October and November, or SON, figure A6) precipitation enhancement and suppression by aerosols are both observed, with suppression observed most often at the lowest and highest LWP ranges. However, these trends do not generally hold up once stability is introduced (panels c and d of previous figures). In fact, the seasonal effects of aerosols on the probability of precipitation are quite complex due to the fewer number of cases and do not seem to indicate a general trend.

4.1.1.3 RESULTS BY CLOUD TYPE

As was expected, examination of figure 4.5 (panels a and b) shows that the stratus type clouds at the Azores site have a similar effect of aerosols on the probability of

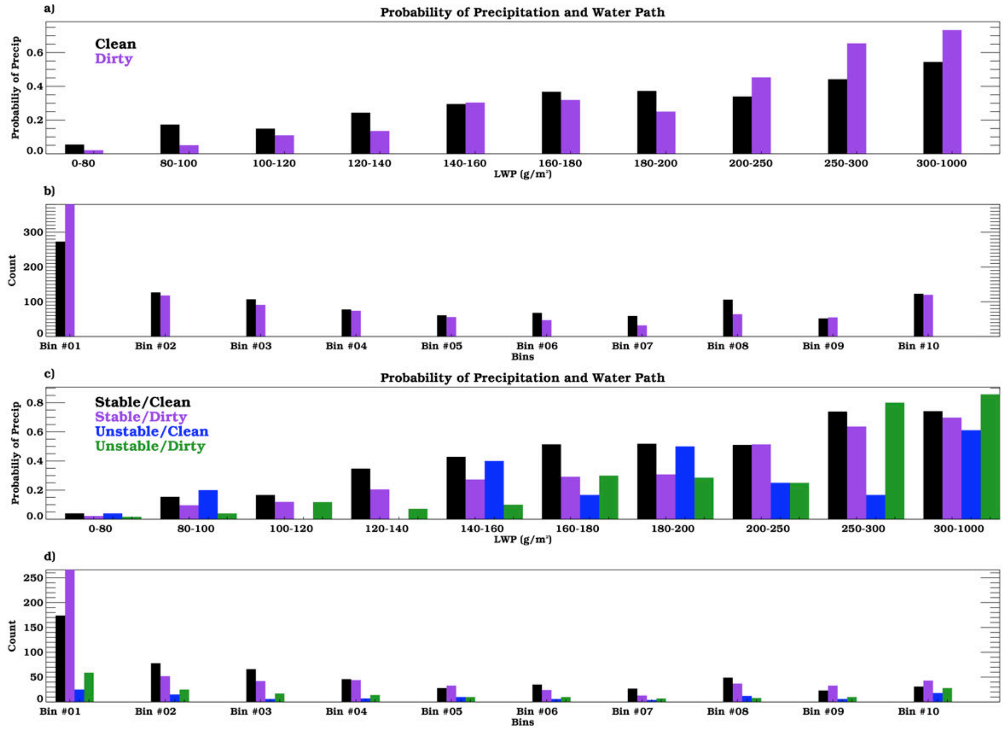

Figure 4.5
Probability of precipitation binned by LWP and histogram of the bin populations at the Azores site for stratus type clouds only. Panels a and b are separated by aerosol, while panels c and d are separated by aerosol and stability.

precipitation as the entire data set. Suppression of precipitation at low LWPs is observed while the three highest ranges of LWP seem to show an enhancement of precipitation. It was hypothesized in section 4.1.1.1 that this enhancement was due to more active cloud dynamics; however, stratus-type clouds usually do not have very active dynamics that could overcome the suppression of warm rain processes. In this case, it could instead be argued that the large amount of liquid water might enhance the warm rain processes itself. The idea behind this is that the presence of more plentiful small droplets would be easier for large drops to collect, assuming that a large drop could be formed somehow. In contrast, the probability of precipitation for the convective type clouds in this region do not have precipitation enhancement at large LWPs, and instead show a suppression of precipitation at most LWP ranges (figure 4.6, panels a and b). This is in approximate agreement with what was hypothesized in this study for this location based on the work of Khain et al. (2008), presented in section 1.4 and figure 1.2. These results illustrate that the two different cloud types do react differently to increases in aerosols, especially at high LWPs, and also that the stratocumulus and small cumulus clouds most likely present in this region are susceptible to precipitation suppression at lower LWPs. The different response between cloud types is also in agreement with other past studies, including van den Heever et al. (2011) and Seifert and Beheng (2006). Although the precipitation suppression of the convective cloud types at large LWPs (what we might assume to be larger cumulus clouds) is not consistent with the hypothesis of figure 1.2, perhaps an increase in evaporation of smaller droplets at cloud entraining edges is affecting cloud

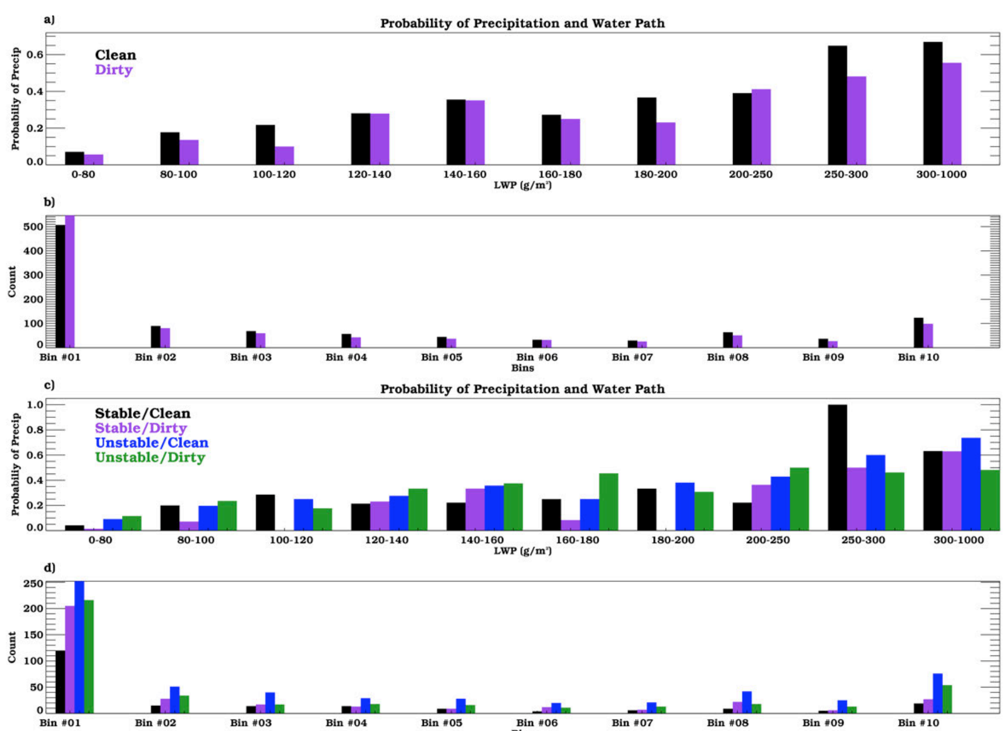

Figure 4.6
Same as figure 4.5, except for convective type clouds.

dynamics as well as sizes and suppressing the expected warm rain enhancement at large LWPs in the convective cloud case (as has been similarly observed in modeling studies by Xue et al. 2006 and Jiang et al. 2006).

The trends in probability of precipitation observed in panels a and b of figures 4.5 and 4.6 do not hold up as well as they did for the entire dataset when the cloud type data is separated by stability (figures 4.5 and 4.6, panels c and d). One can observe that at the highest LWP ranges (where the two cloud types behaved differently in panels a and b), the more unstable stratus clouds are the ones undergoing precipitation enhancement, while the more stable stratus clouds, as well as both stability regimes of the convective clouds remain in the precipitation suppression regime. Although this is not in complete agreement with the results of panels a and b, it is in general agreement with some theory. The enhanced cloud dynamical processes in the unstable stratus clouds might be able to overcome or ‘buffer’ the aerosol suppression effect (refer to section 1.5 for more discussion of this idea). In the convective type clouds, the suppression effect could be dominant because they have more cloud edges that experience entrainment. This could cause the evaporation of smaller cloud droplets to have a larger effect on cloud dynamics in convective type clouds compared to stratus type clouds, possibly suppressing dynamics through stabilization of the boundary layer. However, it is important to point out that the meaning behind the different stability regimes in this context (for each cloud type) is not really well defined. Although the stratus clouds do contain more cases that would be considered stable, while the convective dataset contains more unstable cases, it is still difficult to determine exactly what kind of cloud dynamics a stable convective cloud would have. Hence, this stratification may not be the best indicator of what role dynamics plays compared to aerosols in these cloud types.

4.1.2 Southern Great Plains

4.1.2.1 GENERAL RESULTS

At the Oklahoma site, the picture is quite different. When the probability of precipitation is plotted by LWP and stratified by aerosol content, precipitation is enhanced with increasing CN concentration for almost all LWP bins (figure 4.7, panels a and b). This pattern generally holds at this site when also stratified by stability, although it is not as obvious or smooth in all LWP bins. In general, the trend holds better for stable

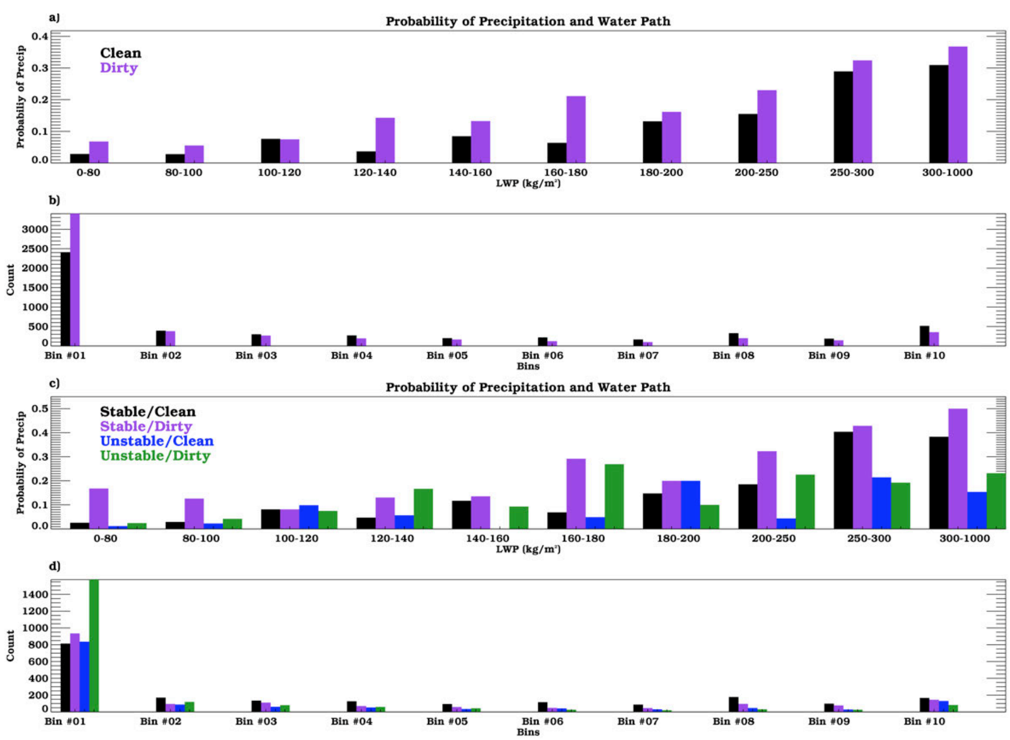

Figure 4.7
Probability of precipitation binned by LWP and histogram of the bin populations at the SGP site. Panels a and b are separated by aerosol, while panels c and d are separated by aerosol and stability.

environments than unstable ones (figure 4.7, panels c and d). This is counter to the theory presented in chapter 1 and in section 4.1.1.1, which states that the unstable environments should have precipitation enhancement, as the more active cloud dynamics are more likely able to overcome the suppression effect. The reasons for these relationships are still unclear, however, it is worthwhile to note that it could be biased by the stability measure, which is not as reliable over land. Nonetheless, this still does not explain why the results without stability (figure 4.7, panels a and b) still show an enhancement of precipitation, as do the cases with high stability (figure 4.7, panels c and d), when they are filtered by the warm cloud criteria introduced in section 3.2.1, and especially why there is this enhancing trend instead of a purely noisy relationship.

Since there have been few other observational studies examining this phenomena over a land location that were not part of a short-lived field campaign, it is difficult to compare these results to any others. However, one possible explanation is one of the buffering mechanisms introduced by Stevens and Feingold (2009) and discussed in section 1.5. This has to do with the liquid water flux through the cloud in more convective type clouds, which are more common in Oklahoma. Similarly to deep convection, when shallow convection experiences an initial suppression of precipitation, it can later be enhanced by the fact that more liquid water will be lofted to the cloud top and evaporate, invigorating the convection and enhancing precipitation. This would be difficult to verify with the data considering that evaporation rates cannot be measured in clouds, however it will be shown in section 4.3.1 that it is not substantiated by the cloud thickness trends, which are relatively constant rather than increasing.

When constrained by RH instead of stability, one can see similarly complex results as were observed in figure 4.7, but in general, increased aerosol led to enhancement of precipitation for dry conditions and more mixed results for moist conditions (figure A1, panels c and d). This does not help explain why the precipitation enhancement is occurring in this location. Considering that increased moisture should, according to figure 1.2, push the cloud types studied in this location into the enhancement regime, one would expect the precipitation enhancement in the moistest bins, but this is not observed in figure A1. Therefore relative humidity must not be a hugely controlling factor on the effect aerosols have on precipitation in this location. Similar to the Azores site, results stratified by relative humidity instead of stability are not included in the following sections. This is again because the complexity of those results did not provide any further insight into the aerosol problem and therefore, relative humidity was determined not to be a controlling factor as originally hypothesized by Khain, et al. (2008). Also not included in this section is the plot of the probability of precipitation stratified by CWV, as it is still a complex relationship and has no discernible trend, similar to the Azores.

4.1.2.2 SEASONAL RESULTS

Figures 4.8-4.9 illustrate the seasonal breakdown of the probability of precipitation for the Oklahoma site. In general, the precipitation enhancement that was prominent in the total data at this site is observed in most LWP bins in 3 of the 4 seasons, while summer (June, July and August, or JJA) instead shows a suppression of precipitation with aerosols at higher LWPs and only slight enhancement in lower LWP bins (figure 4.8, panels a and b). These trends are not as obvious, but tend to hold up when they are

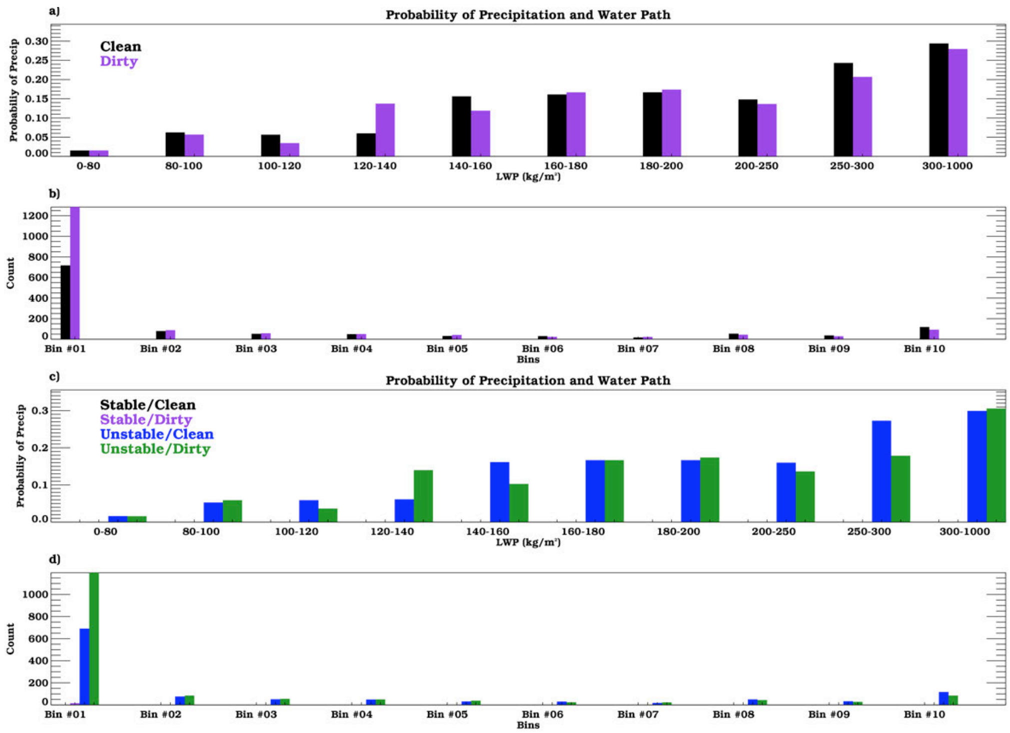

Figure 4.8
Probability of precipitation binned by LWP and histogram of the bin populations at the SGP site for summer (JJA) only. Panels a and b are separated by aerosol, while panels c and d are separated by aerosol and stability.

separated by stability (figure 4.8, panels c and d). JJA still generally has a suppressing trend in this figure, except at the highest LWP range, while winter (December, January and February, or DJF) observes general precipitation enhancement (figure 4.9), as was also seen in the results for the entire dataset. However, the transitional seasons show differing trends with stability, especially at large LWPs, with more stable conditions leading to enhancement of precipitation (appendix figures A3 and A4). The summertime trends of precipitation suppression are relatively consistent with the proposed increase of evaporation of cloud droplets of small cumulus, as discussed in sections 4.1.2.1 and 1.2, since these are common clouds in this season in Oklahoma. However, the buffering mechanism mentioned in sections 4.1.2.1 and 1.5 may also be important in the more transitional or winter time clouds. Nevertheless, there are significantly fewer cases in these seasons and so robust trends are difficult to discern.

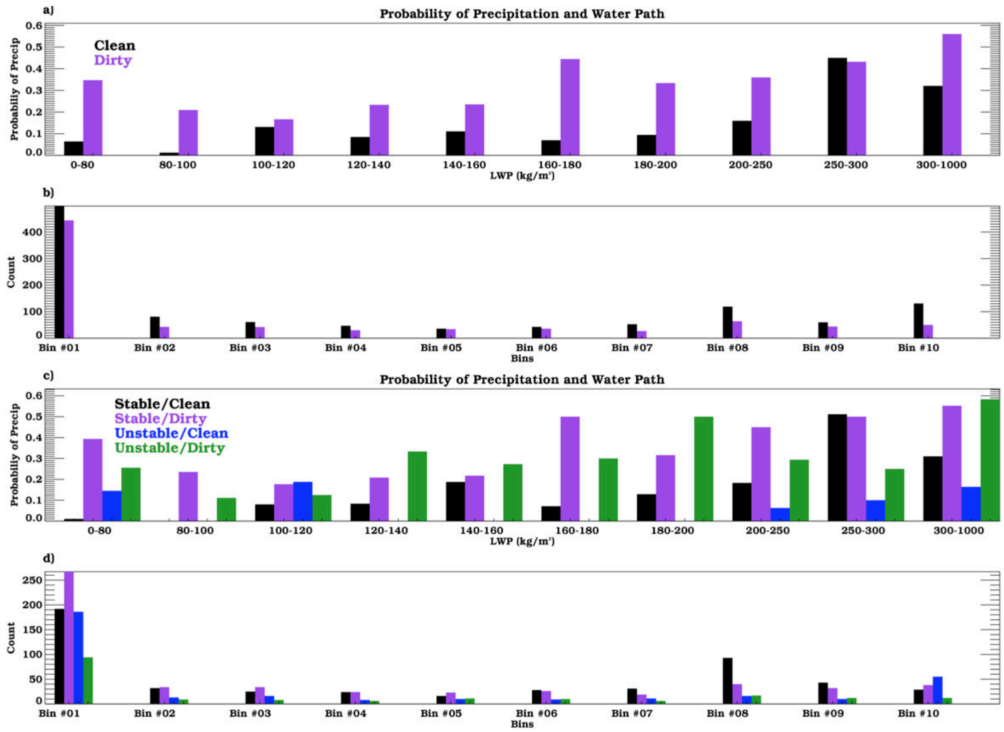

Figure 4.9
Same as figure 4.8, except for winter (DJF).

4.1.2.3 RESULTS BY CLOUD TYPE

When analyzed by cloud type, the results from the SGP site do not behave as hypothesized, even less so than at the Azores site. Figure 4.10 and 4.11 (panels a and b) reveal that the general trend for both cloud types is an enhancement of precipitation, although a few LWP ranges do show suppression, which was not observed in any LWP ranges in figure 4.7. The few bins in which precipitation suppression is observed appear to be simply noise. The precipitation enhancement that occurs throughout the data at the Oklahoma site is thought to be due to the buffering mechanism as discussed in section

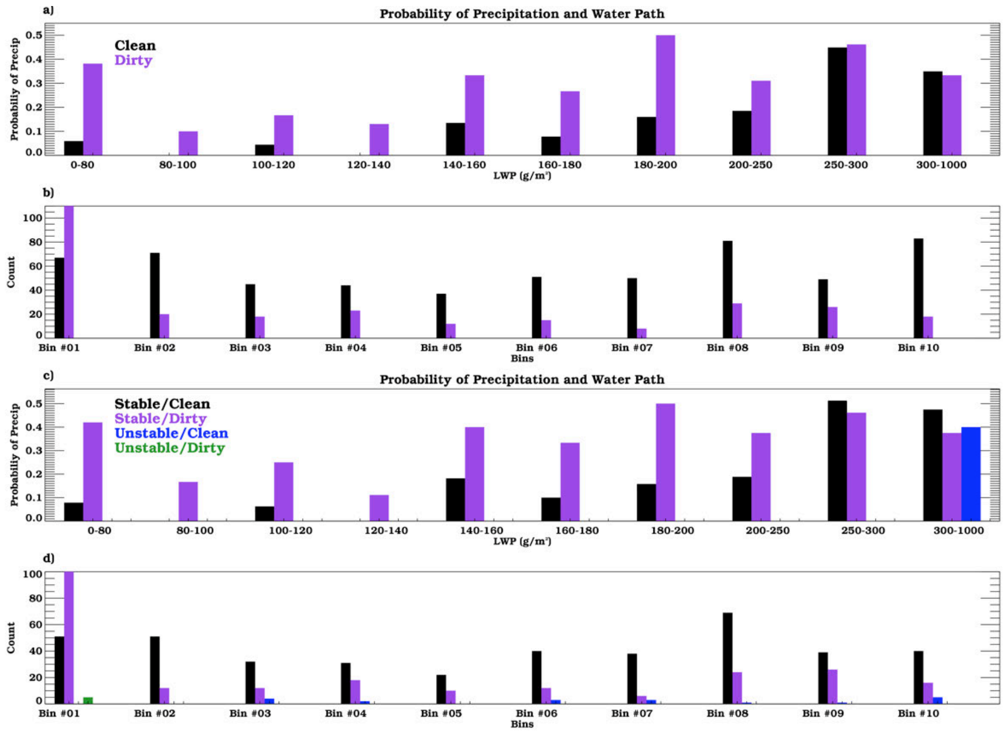

Figure 4.10
Probability of precipitation binned by LWP and histogram of the bin populations at the SGP site for stratus type clouds only. Panels a and b are separated by aerosol, while panels c and d are separated by aerosol and stability.

4.1.1.2 and further in section 1.5. However, this doesn't explain why the stratus type clouds also experienced an enhancement of precipitation, particularly at lower LWPs. The reasoning for this remains unclear.

When these results are separated by stability as well as CN concentration (figures 4.10 and 4.11, panels c and d), there are not really enough cases to determine significant trends, as can be seen in the noisiness of these plots, however, there are a few insights we can gain from examination of the figures. First of all, the algorithm to find stratus and convective type clouds seems to be working fairly well, considering that in figure 4.10, only the stable regime has enough cases to show results in almost every LWP range,

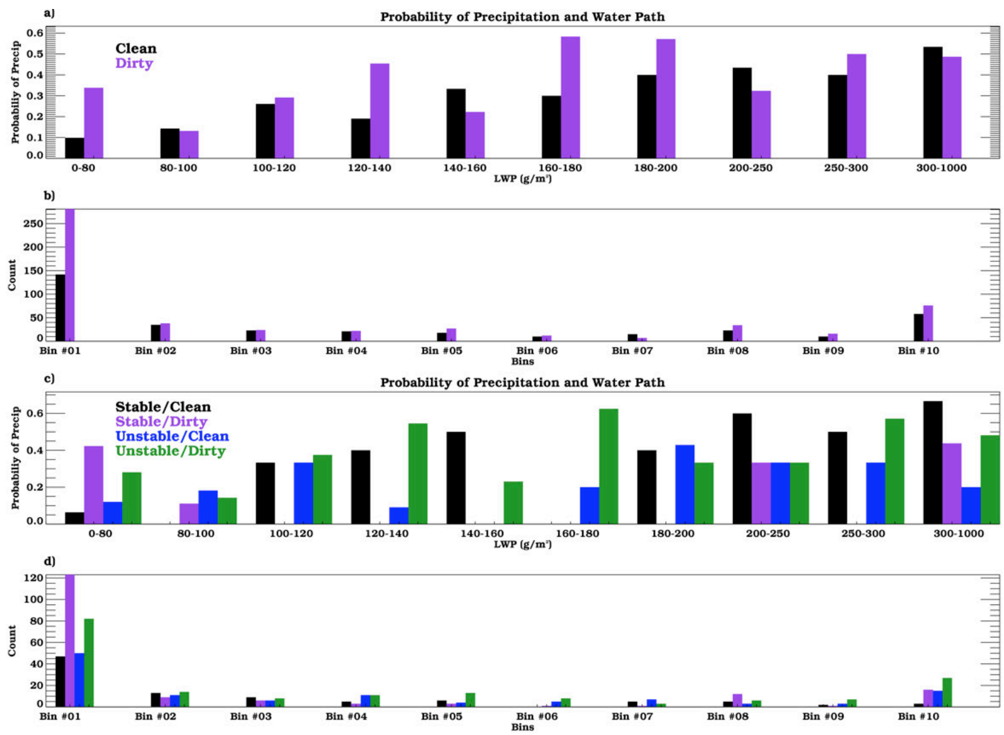

Figure 4.11
Same as figure 4.10, except for convective type clouds.

which captures the stable dynamics of stratus type clouds. Also, in figure 4.11, there does seem to be some semblance of precipitation enhancement for more unstable convective clouds, while the more stable ones generally observe precipitation suppression, at least in the LWP ranges in which one can actually observe such trends (for example, the largest LWP ranges). This is consistent with past theories and the previous results of this study.

4.2 Surface Precipitation

4.2.1 Wind Direction Bias

The average precipitation rate from the rain gauge at each site is included in figures 4.12-4.13. There are slight variations in the rain gauge measurements at each location from certain wind directions, especially at high wind speeds. This could cause a bias in

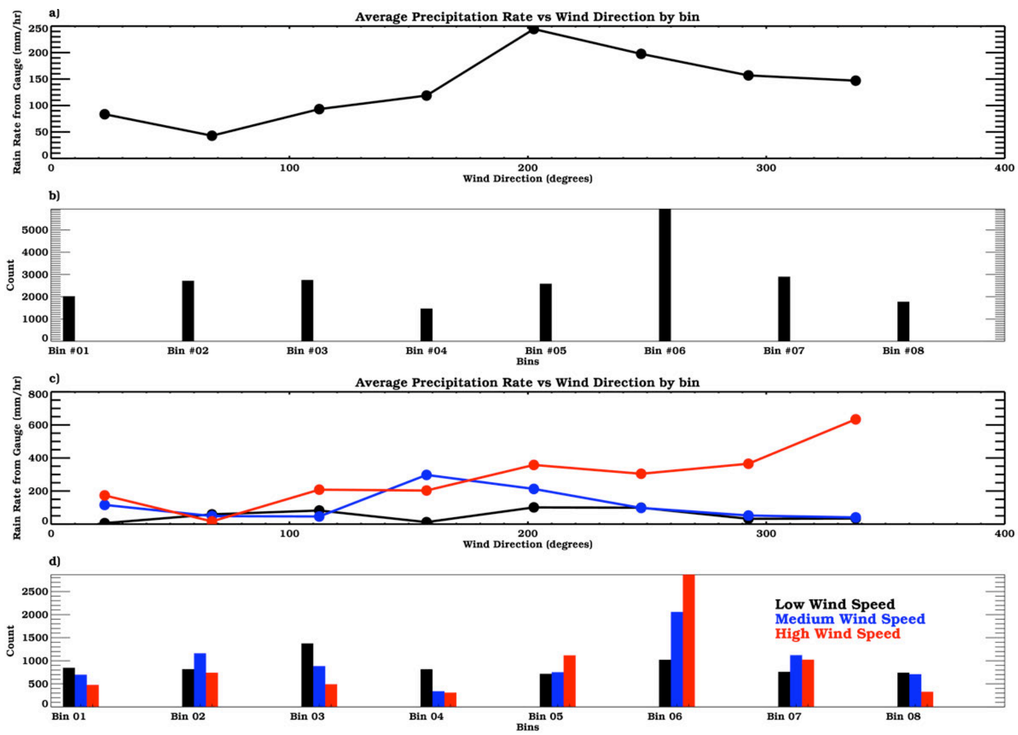

Figure 4.12
Average precipitation rate (in each bin) measured by the rain gauge binned by and plotted against wind direction at the Azores site. Also included are histograms of bin populations. Panels c and d show data separated by wind speed.

results as differences between radar echoes in the cloud and precipitation at the surface likely increase with wind speed. Also, as is evidenced by the histogram counts of figures 4.12-4.13, there were significantly fewer cases of rain reaching the surface and the instrumentation there than were seen by the radar. This is especially true since the radar can pick up drizzle precipitation formation in the cloud, which often evaporates before reaching the ground. The focus of this study was the aerosol effect on the onset of precipitation, since that is arguably one of the times a cloud will be most affected by the hypothesized inhibition of warm rain processes. It is also easier to examine the response of the clouds to aerosols through observations of other cloud variables. Although aerosols

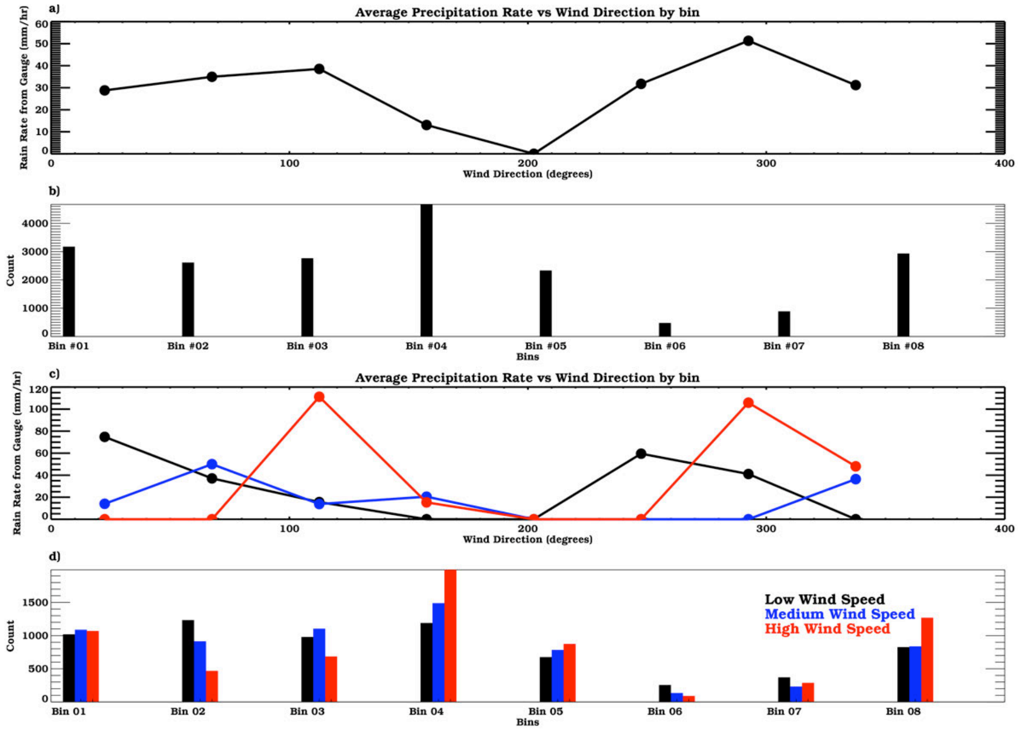

Figure 4.13
Same as figure 4.12, except at the SGP site.

will affect surface precipitation through the response of the cloud and its dynamics, it is more difficult to connect this response to that which happens within the cloud. This section is included as a reminder of why the radar data served this purpose better than the rain gauge data.

4.2.2 Probability of Precipitation

This section presents similar results as those included in section 4.1, however, the probability of precipitation here is calculated using surface precipitation data instead of radar echoes within the cloud. Figure 4.14 shows the probability of precipitation using gauge data at the Azores site as a function of LWP and differentiated by CN concentration; panels c and d are stratified by stability as well as aerosol content. At this

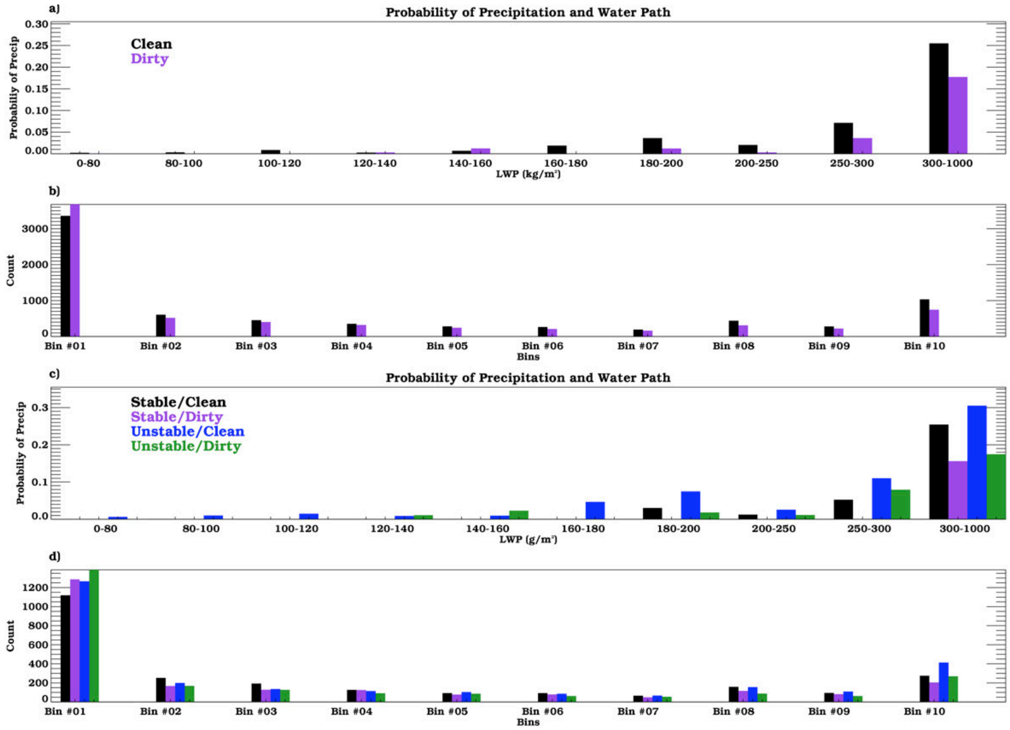

Figure 4.14
Probability of precipitation binned by LWP and histogram of the bin populations at the Azores site. In this figure, however, the probability of precipitation is determined by any rain at the surface as measured by the rain gauge. Panels a and b are separated by aerosol, while panels c and d are separated by aerosol and stability.

site, the rain gauge results indicate that only the very large LWPs lead to precipitation that reaches the ground. The picture is very different at the SGP site, where there is little rainfall reported at all. This is likely due to lower humidities above land. As one can see, only the Azores site has observable trends in large LWPs bins, since these are the only clouds with sufficient water to allow precipitation to reach the surface. The trend observed at the ground is precipitation enhancement, which is consistent with the general results from this location at high LWPs. Since this is the only type of cloud with precipitation reaching the surface, it would appear that precipitation was enhanced at the surface if only the rain gauge results were used. However, as was shown in section 4.1,

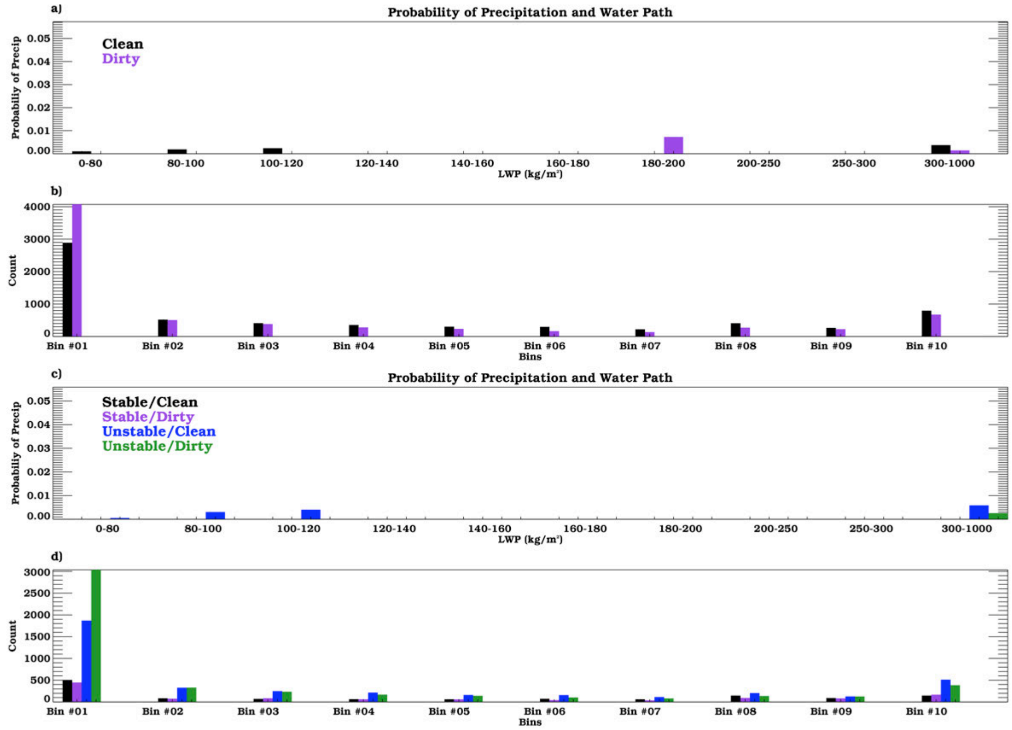

Figure 4.15
Same as figure 4.14, except at the SGP site.

this is not the case at most LWPs and the trend in precipitation within the cloud at this location was suppression with increasing aerosol concentration. As has been mentioned previously, this study focused on the response of the onset of precipitation within the cloud to aerosol, rather than that at the surface, especially considering that precipitation reaching the surface can be altered by the ambient relative humidity, which is not affected by aerosol concentration. This is another reason that the radar data was used instead of the rain gauge.

4.3 Cloud Thickness

4.3.1 General Results

The average cloud thickness is plotted as a function of CN concentration in figure 4.16 (panels a and b show the Azores site, panels c and d contain the SGP data). The results for the cloud thickness are quite similar for both locations, unlike the precipitation results. As one can see, the cloud thickness generally had very little trend with increasing aerosol for all three stability regimes (high, medium and low) when stratified by stability. Included in appendix figure A6, the cloud thickness results were also stratified by cloud base height (CBH). The reason that cloud base height was chosen as an alternative variable to stability is that it can act as a proxy for meteorological conditions, similarly to

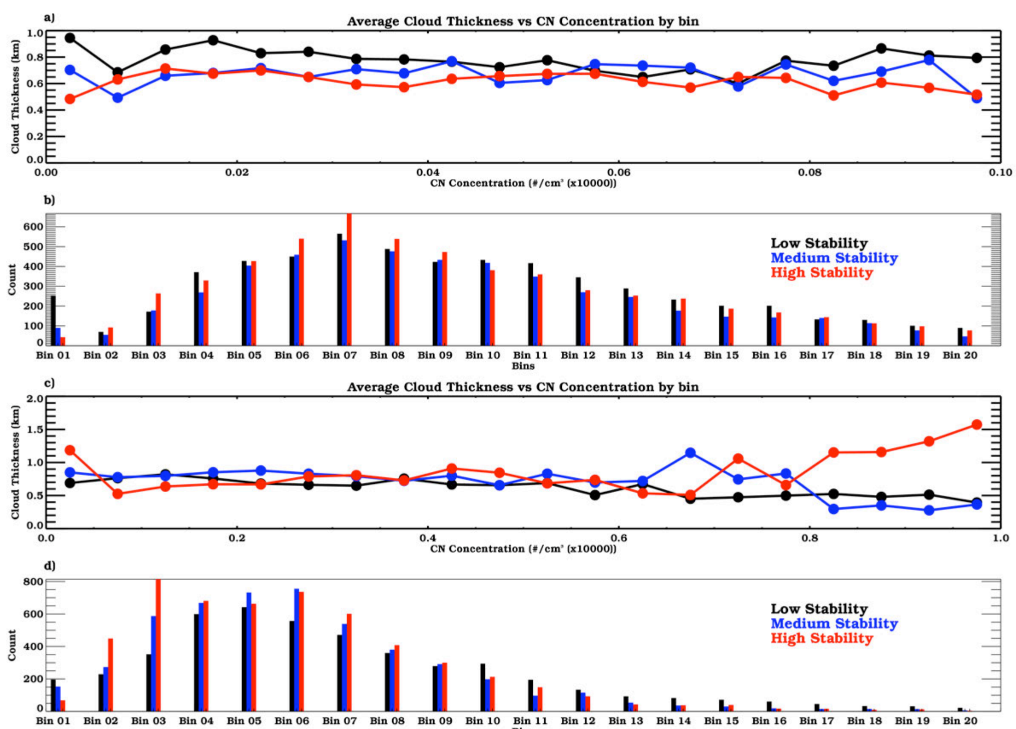

Figure 4.16
Average cloud thickness (in each bin) binned and plotted by CN concentration and separated by stability. Also included are histograms of bin populations. Panels a and b show data from the Azores site, while panels c and d show data from the SGP site.

stability. The lifting condensation level (LCL) is a measure of surface meteorological conditions (temperature and moisture content) and is often similar to cloud base height.

Figure A6 is also included to allow for comparison to the results of Li et al. (2010). The lower panels (c and d) show the Oklahoma site, where Li et al. (2010) performed a similar study. The trends here do not agree with their results, as they found an increase in cloud height with increasing CN concentration, particularly for the lowest two CBH regimes. Figure A6, however, shows similar trends as observed in figure 4.16. The reason that these two studies differ is due to the fact that Li et al. (2010) did not limit their clouds to those beneath the freezing level, but used the CBH as the only limitation for clouds. Therefore, they allowed for vertically developing clouds. In addition, they did not filter out precipitating clouds, which allowed for more vertically developed clouds. Specifically, in deep convection, with more and smaller droplets that are not rained out, more supercooled water can be transported higher in the storm and produces enhanced latent heating and invigoration of convection, most likely producing taller or thicker clouds. This would cause the trend in cloud thickness to increase with aerosol, while the warm clouds themselves do not appear to have this trend. Although the Li et al. (2010) study is more consistent with theory, it appears to be biased by cold cloud processes. It is also in opposition to the results of the current study because if the more vertically developed clouds are undergoing precipitation enhancement through the mechanisms described above, suppression of precipitation should be observable in the initial development of these clouds, yet precipitation enhancement is actually detected in the smaller warm rain clouds of this study. However, the warm rain clouds of this study do not necessarily capture the initial development of more vertically developed clouds. Also, the difference in cloud thickness trend observed in these two studies for shallow and deeper clouds is consistent with van den Heever et al. (2011), which found similarly opposing trends depending on the vertical extent of the cloud.

The trends of cloud height are similar in the two locations. This is noteworthy since the trends of precipitation probability were significantly different. However, this could be due to the competing effects of increased evaporation and suppression of warm rain processes. Suppression of rain should lead to increases in cloud thickness, while increases in evaporation would lead to smaller clouds. However, it could also be that the lack of trends indicates a noisy relationship.

4.3.2 Seasonal Results

In this section, plots of cloud thickness stratified by both stability and cloud base height are shown for each season. This is because during certain seasons, there were not enough cases to show robust trends in all stability regimes, however, in each season and location, the trends seem to be relatively similar when separated by cloud base height or stability. Therefore, showing the trends stratified by both variables allowed for observation of cloud thickness trends for each season and therefore a more detailed analysis of the seasonal results.

When the cloud thickness is broken up by season at the Azores, it tends to have similar trends as is observed in the total data. As can be seen in figures 4.17-4.18 (and A7 and A8), the general trend is relatively constant (or very slight trends) with increasing CN concentration. It is difficult to speculate whether this is pure noise or due to competing or buffering effects within the cloud.

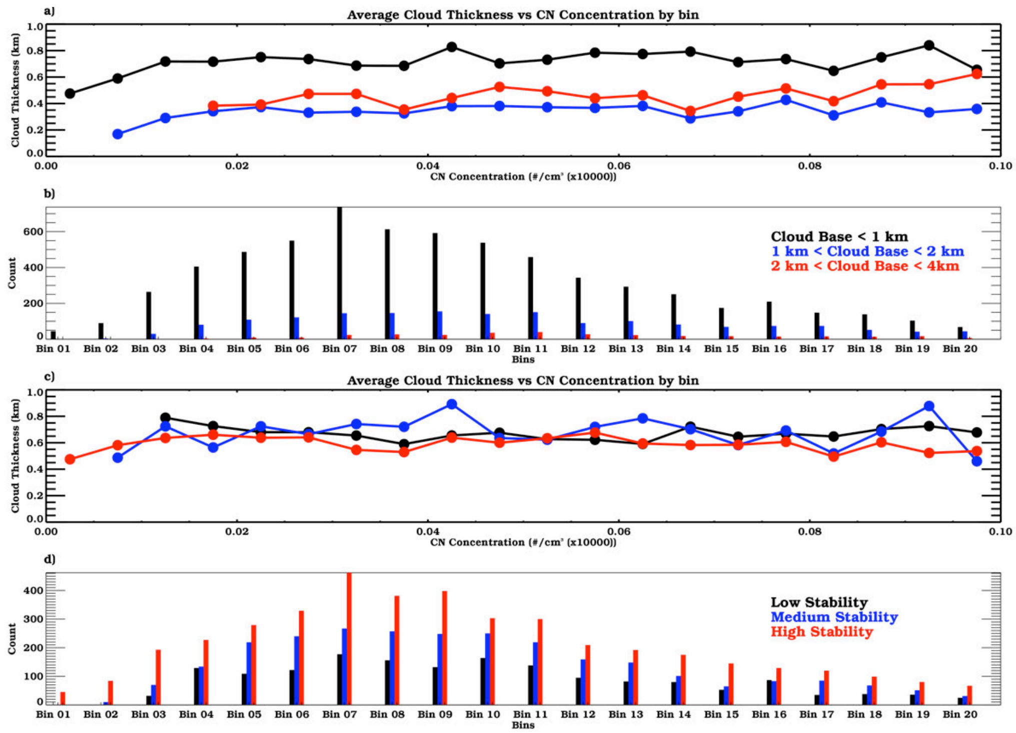

Figure 4.17
Average cloud thickness (in each bin) binned and plotted by CN concentration at the Azores site for summer (JJA) only. Also included are histograms of bin populations. Panels a and b show data separated by cloud base height, while panels c and d show data separated by stability.

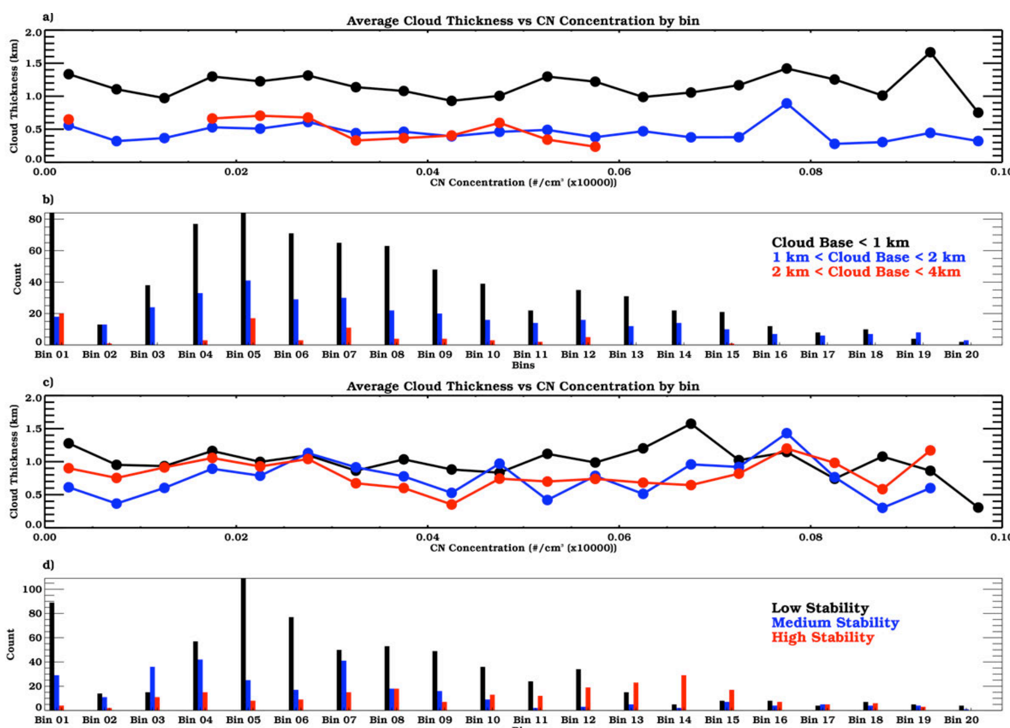

Figure 4.18
Same as figure 4.17, except for winter (DJF).

At Oklahoma, the cloud thickness trends are similar to those observed during the entire year, except during the DJF season. In general, the cloud thickness tended to stay relatively constant or slightly decrease with increasing CN concentration, whether it was stratified by cloud base height or stability (figures 4.19-4.20 and A9-A10). Figure 4.20 shows the DJF season, which exhibits a constant to increasing trend with increasing aerosol. Although this trend is slight at best, it is in opposition to the trends seen in the rest of the year. A possible explanation lies in the fact that Oklahoma winter time tends to have more stratus type clouds associated with large synoptic scale systems than the rest of the year (evidenced by the reduced number of cases with low stability in this season), which should be more susceptible to cloud thickness increases according to theory.

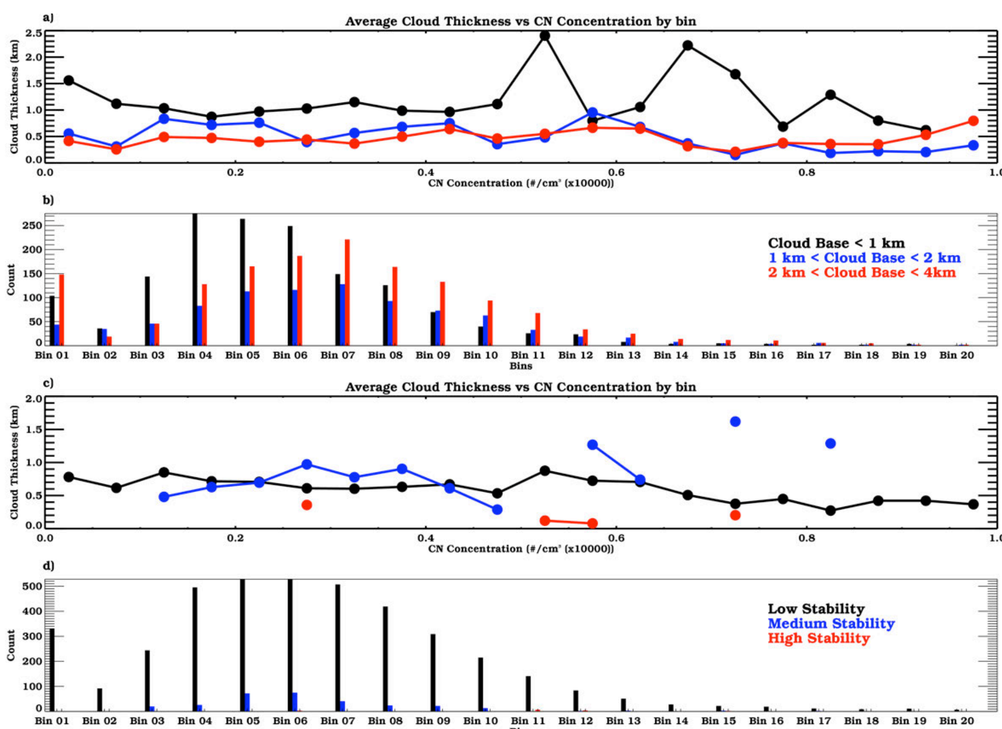

Figure 4.19
Average cloud thickness (in each bin) binned and plotted by CN concentration at the SGP site for summer (JJA) only. Also included are histograms of bin populations. Panels a and b show data separated by cloud base height, while panels c and d show data separated by stability.

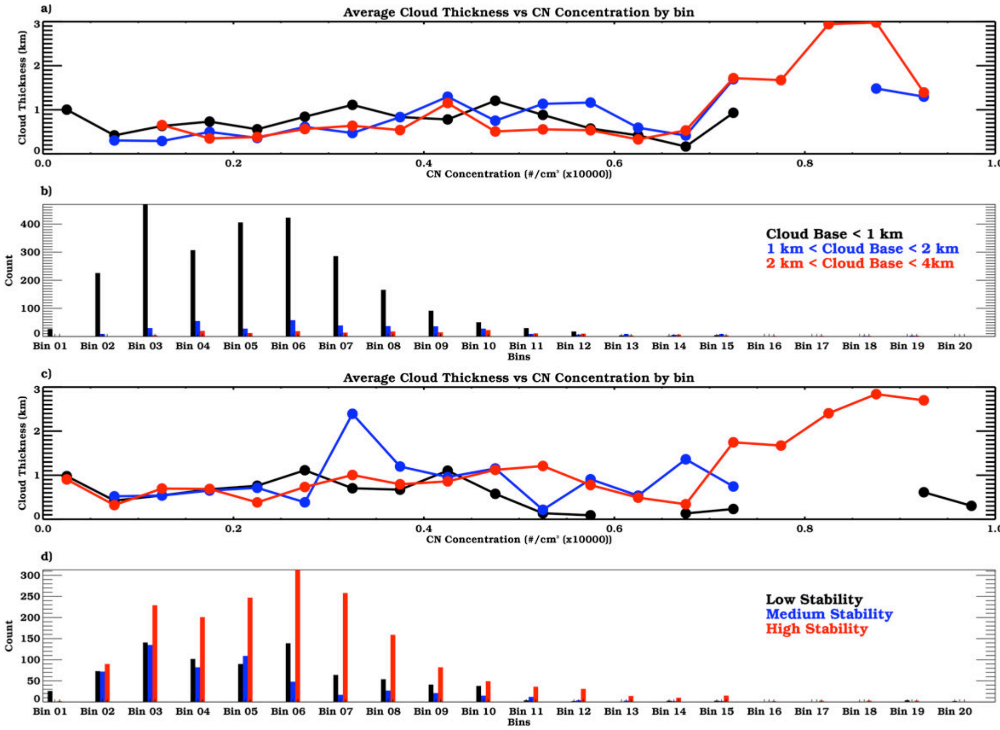

Figure 4.20
Same as figure 4.19, except for winter (DJF).

4.3.3 Results by Cloud Type

Figure 4.21 shows the trend of cloud thickness with increasing CN concentration at the Azores site, divided by stability. Panels a and b show the stratus type clouds, while panels c and d are plotted for the convective type clouds. In this case there are significantly more stable cases in the stratus clouds and more low stability cases in the convective clouds, leading again to the conclusion that these are fairly robust cloud type designations and that the results can be compared to figure 4.16. Examination of figure 4.21 illustrates that neither cloud type shows a distinct trend. Once again, it could be due to a noisy relationship or perhaps even competing aerosol effects as previously mentioned.

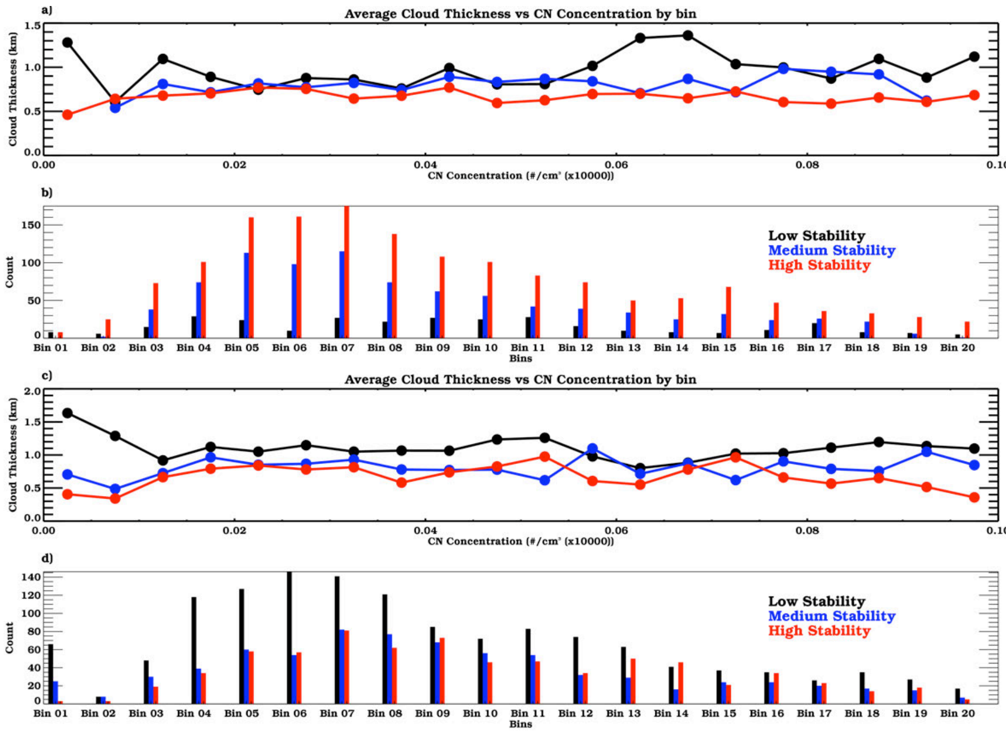

Figure 4.21
Average cloud thickness (in each bin) binned and plotted by CN concentration at the Azores site separated by stability. Also included are histograms of bin populations. Panels a and b show data for stratus type clouds only, while panels c and d show data for convective type clouds only.

At the Oklahoma site, there are not as many cases that fall into the stratus or convective designation, so the trends are even more difficult to distinguish and are most likely less robust. Figure 4.22 shows a relatively constant trend of cloud thickness with increasing CN concentration for both cloud types. This is in opposition to original aerosol theories that suggest that cloud thickness should increase with increasing aerosols due to the suppression of precipitation. Considering that the general trend in precipitation in this site was precipitation enhancement, we might expect an increase in cloud thickness according to the newer buffering mechanism theories. However, these trends are not observed and the cause of this is unkown. We may again speculate on competing effects of aerosols, but it seems impractical considering that no trends are apparent.

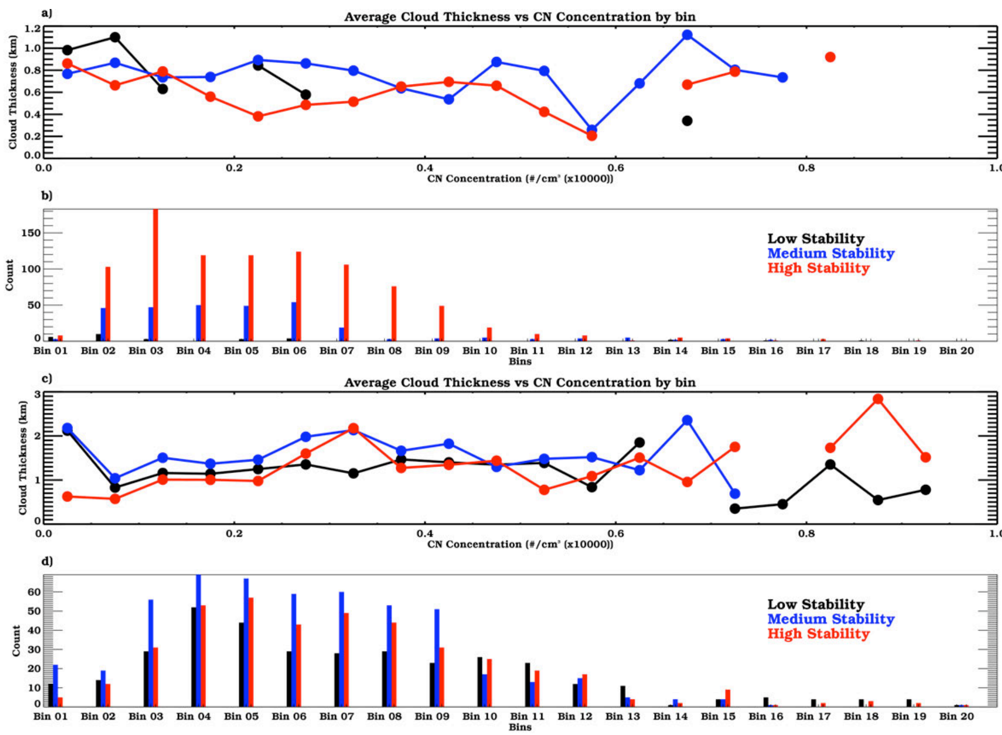

Figure 4.22
Same as figure 4.21, except for the SGP site.

4.4 LWP

4.4.1 General Results

The average LWP as a function of CN concentration is plotted in figure 4.23 for the Azores and figure 4.24 for the SGP site. At both locations, the LWP decreased with increasing CN concentration. This is counter to traditional theories of aerosol effects but is consistent with past studies such as Nakajima et al. (2001) and Han et al. (2002), discussed in section 1.2. This study confirms their results that continental clouds have a stronger LWP susceptibility to aerosols by the fact that at the SGP site, the LWP trend is strong enough to still be observable when stratified by stability (figure 4.24, panels c and d). Also, in the following section it is illustrated that the negative LWP trend holds up for the Azores site in the summer season, which is also consistent with Han et al. (2002).

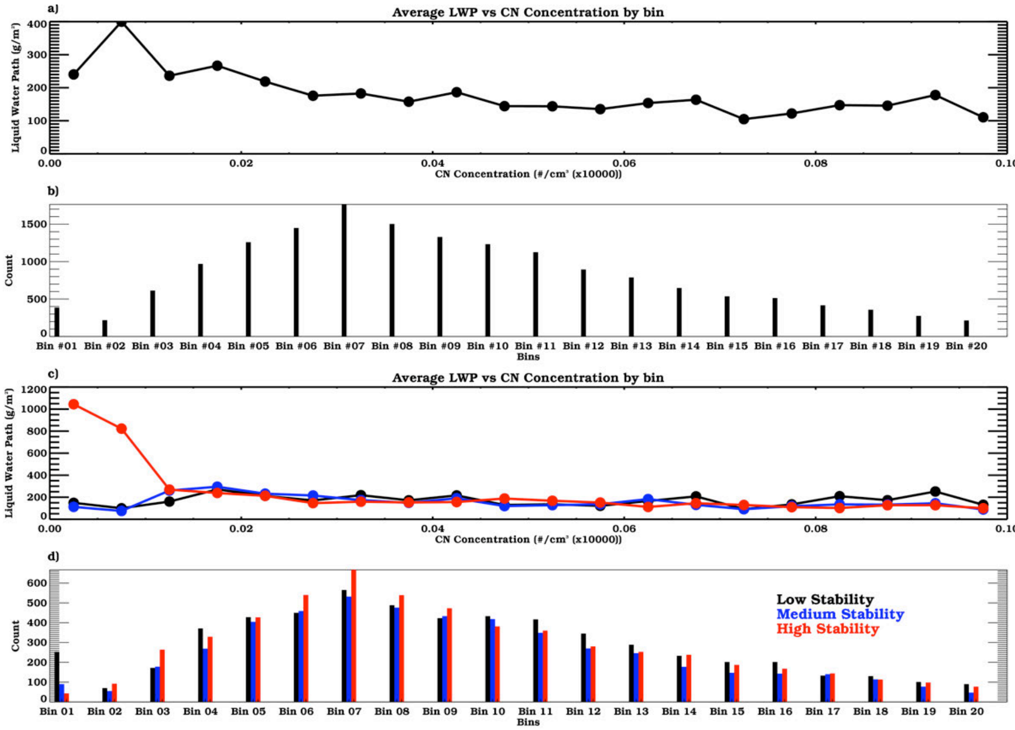

Figure 4.23
Average LWP (in each bin) binned and plotted by CN concentration at the Azores site. Also included are histograms of bin populations. Panels c and d are separated by stability.

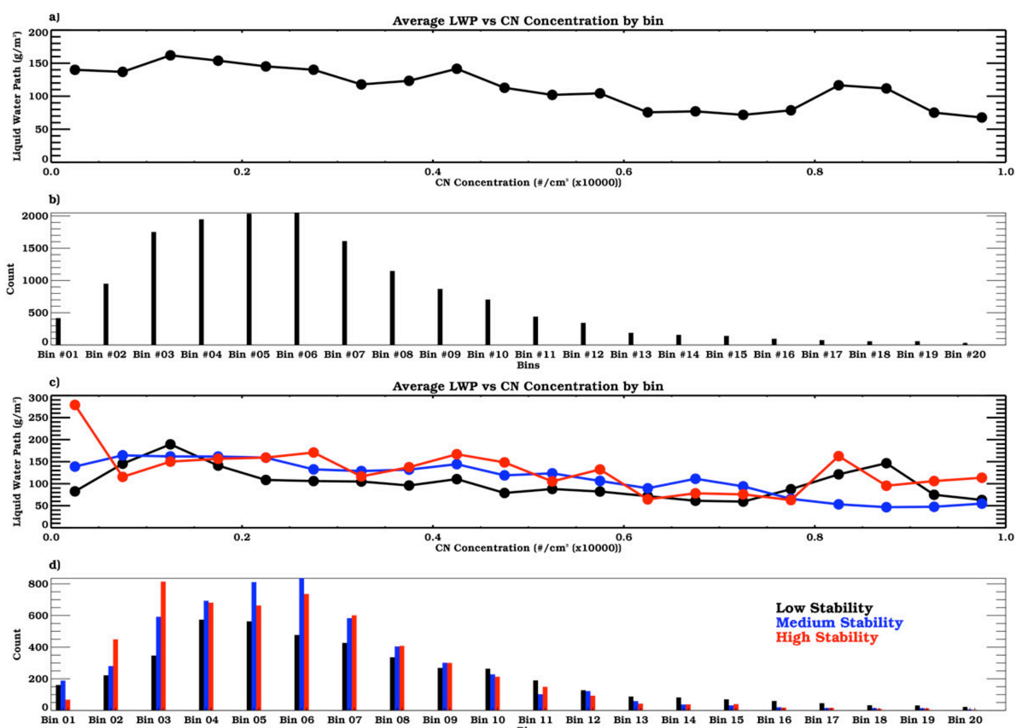

Figure 4.24
Same as figure 4.23, except at the SGP site.

4.4.2 Seasonal Results

At the Azores site, the trend in LWP is much noisier when plotted by season than when the entire data set is included (figures 4.25 and A11). However, one can see a general (if not exactly strong) decreasing trend in the JJA season (figure 4.25, panels a and b), while the other seasons have more constant and noisier trends in LWP with increasing CN concentrations. There are not enough cases in the DJF and SON seasons to observe robust trends, but considering that the MAM season observes a steady trend, we can conjecture that there could be a slight seasonal cycle to the LWP trend. As Han et al. (2002) described, and as is summarized in section 1.2, the warm season in marine environments should have larger LWP susceptibility because of the effect the

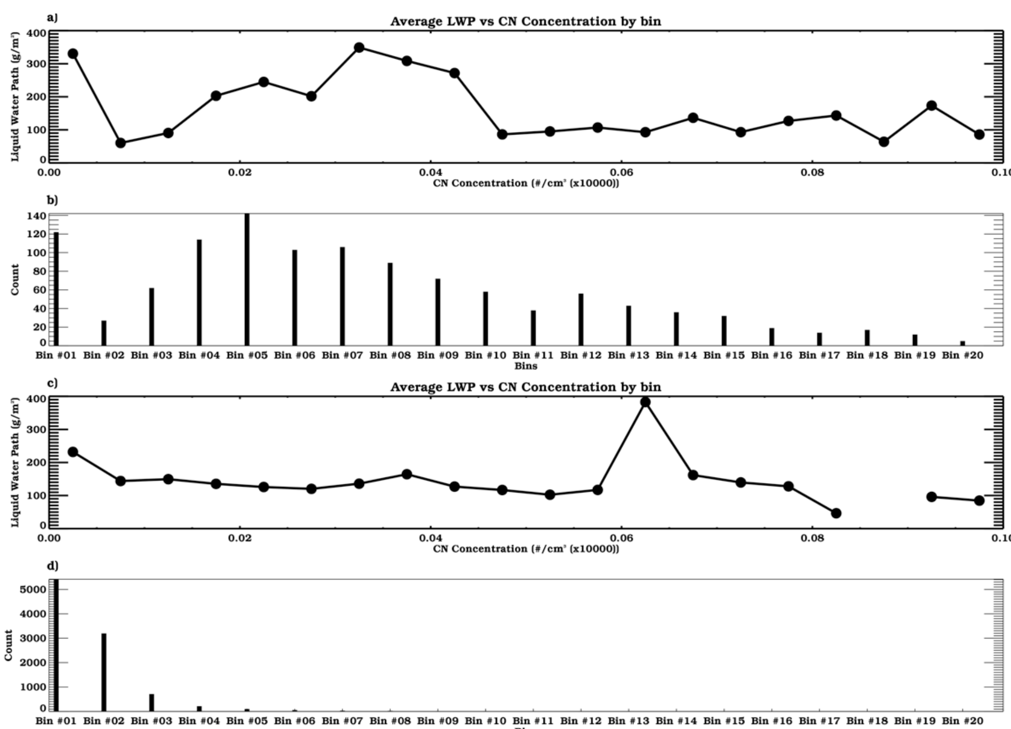

Figure 4.25
Average LWP (in each bin) binned and plotted by CN concentration at the Azores site. Also included are histograms of bin populations. Panels a and b show data from summer (JJA) only, panels c and d show data from winter (DJF) only.

microphysical structure of the these types of clouds has on the boundary layer dynamics in this region and season, as well as the effect sea surface temperature has on these dynamics. The seasonal cycle in LWP trends would be consistent with Han et al., although it is difficult to conclude whether it exists due to the small number of cases in certain seasons.

Although the seasonal plots showing the LWP trend with increasing CN concentration are noisier than the entire data set at SGP, they still generally present a decreasing trend of LWP (figures 4.26 and A12). These decreasing trends, as well as the fact that some of the trends are weak or neutral are consistent with Han, et al. (2002), as presented and described in section 1.2. A seasonal dependence for the LWP susceptibility was not observed in continental clouds in that study and was also not observed in the

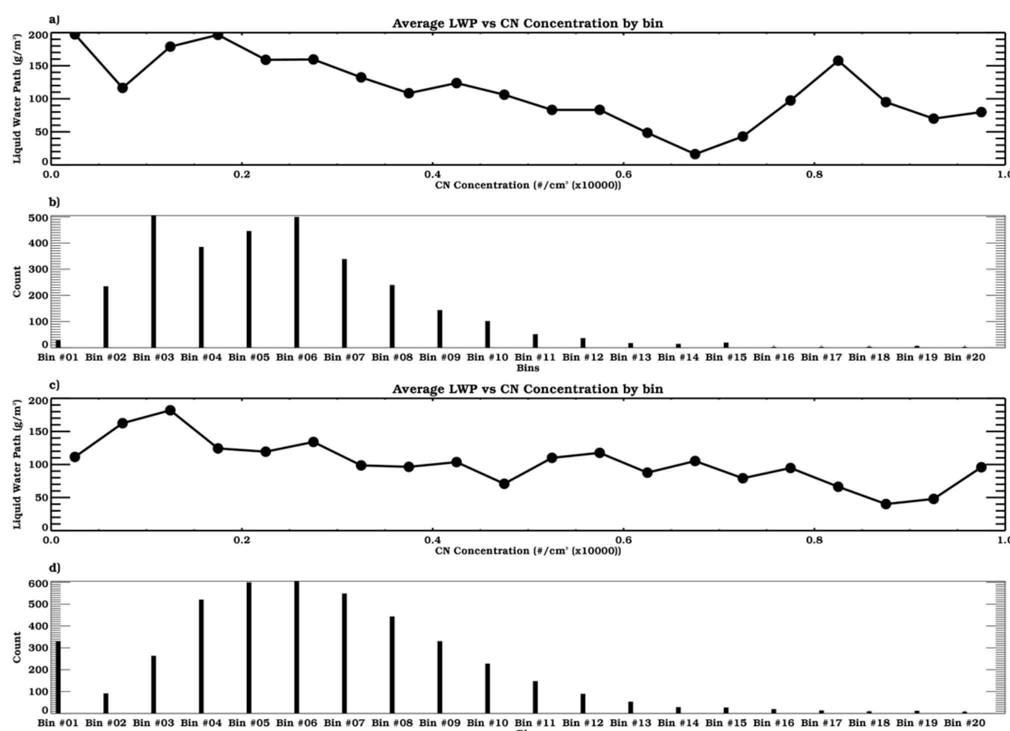

Figure 4.26
Same as figure 4.25, except at the SGP site.

present study. However, that study suggested that marine locales would have more of a seasonal cycle in LWP sensitivity to aerosol.

4.4.3 Results by Cloud Type

The trend of LWP with increasing CN concentration is relatively constant, perhaps very slightly decreasing, for both stratus type and convective type clouds at the Azores site for all stability ranges (figures 4.27 and 4.28). There is a possibility that these trends are noise, or it could be evidence of competing or buffering effects, as have been discussed previously. There is no way to distinguish and therefore, no robust conclusions can be drawn from these plots.

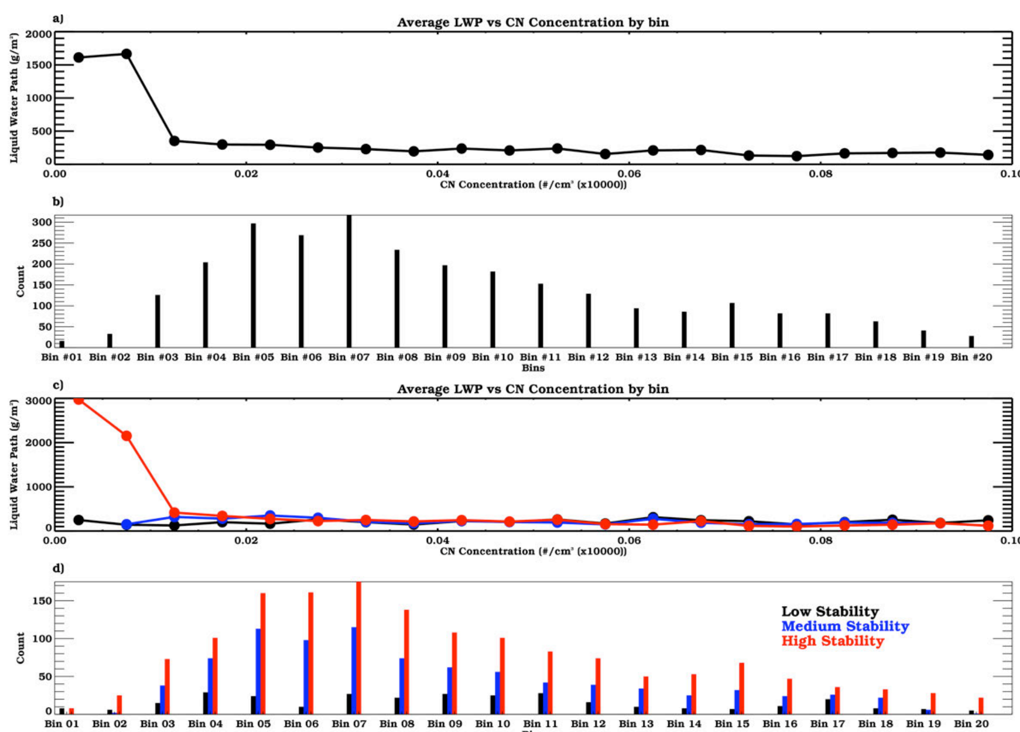

Figure 4.27
Average LWP binned and plotted by CN concentration at the Azores site for stratus type clouds only. Also includes histograms of bin populations. Panels c and d separated by stability.

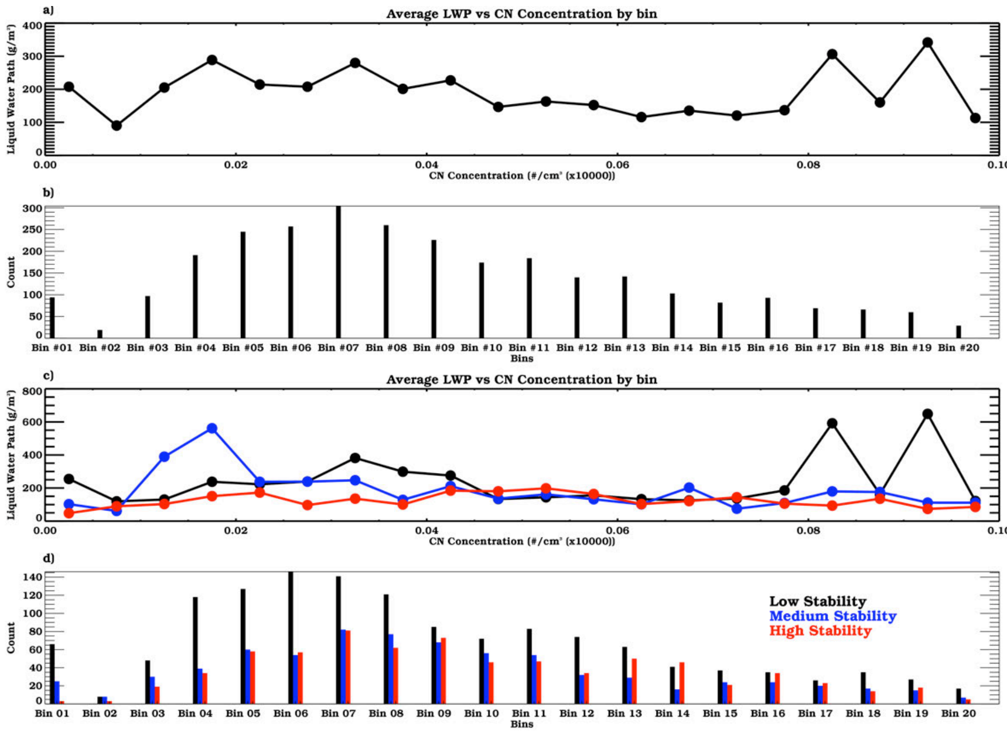

Figure 4.28
Same as figure 4.27, except for convective type clouds only.

The Oklahoma site has some differing results. Here, figure 4.30 shows a decreasing trend in LWP for convective type cases, as would be expected with the increased evaporation of these types of clouds and as is consistent with Han et al. (2002). However, there appears to be little to no trend observable in the stratus type clouds, even when separated by stability (figure 4.29, panels c and d), especially because there are very few cases in certain bins leading to missing data points in the plot. As was the case with many other cloud thickness and LWP trends in this study, there is no explanation for the lack of trends. The absence of these trends are counter to theory, however, the explanation is unknown.

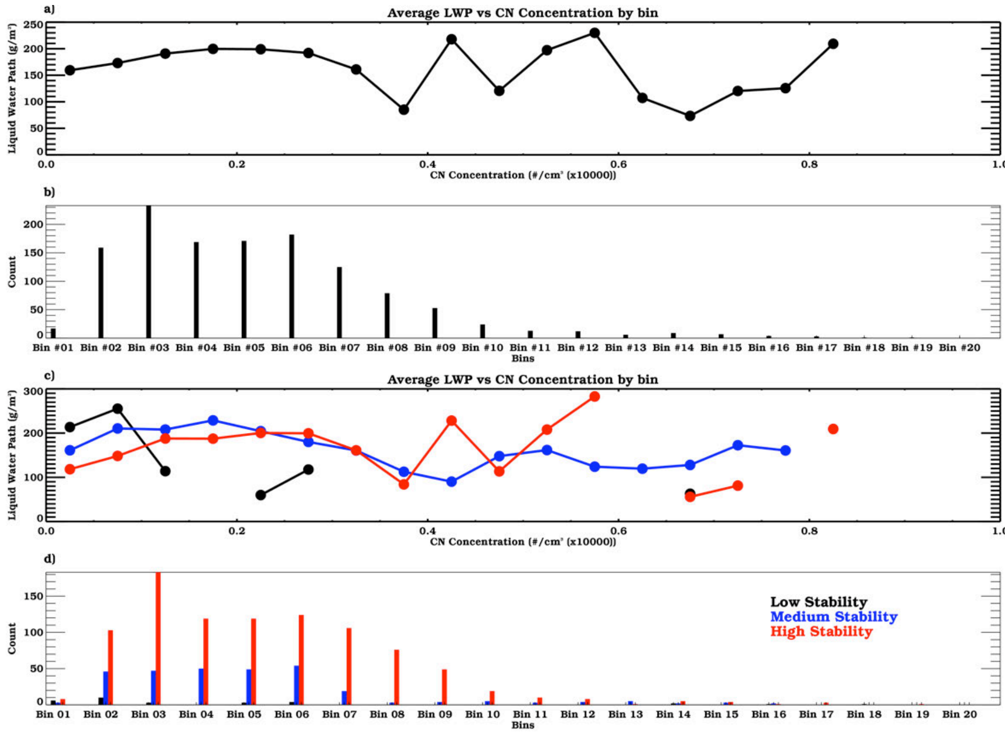

Figure 4.29
Average LWP binned and plotted by CN concentration at the SGP site for stratus type clouds only. Also included are histograms of bin populations. Panels c and d are separated by stability.

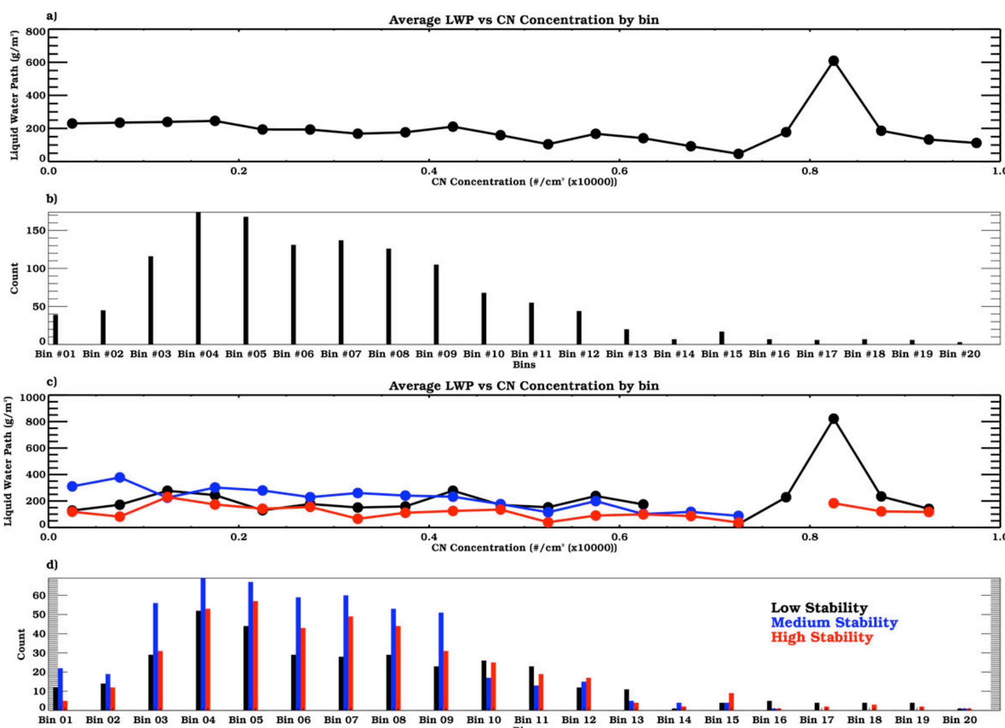

Figure 4.30
Same as figure 4.29, except for convective type clouds only.

4.5 Correlations

This section attempts to present correlations and trends of meteorological conditions with aerosols to see if these may be affecting the response of clouds and precipitation to these aerosols. For example, if a certain air mass brings with it certain meteorological conditions that are not likely to produce precipitating clouds (for example, cold and dry) as well as above average aerosol concentrations, it could appear that there is an aerosol suppression effect when in reality this may not be the case. For this reason, it is necessary to check these relationships, which are examined in this section. Other meteorological conditions were examined, however, stability is the only variable included in these results. This is partly because no noteworthy trends were seen in the other variables and because stability is the most effective at depicting the large-scale meteorological conditions with a single variable. The following figure is plotted for times when clouds are present, so that the analysis is performed on the same dataset as was used in the previous sections.

4.5.1 Stability

As expected, the Azores site shows no trends in stability with CN concentration while the Oklahoma site does have variations in stability when plotted against CN concentration, although there is no distinguishable trend, nor is the correlation significant (figures 4.31 and A13). Considering that stability should be a good measure of meteorological conditions, the lack of relationship between this variable and aerosol is evidence that the results found in this study are not biased by meteorology.

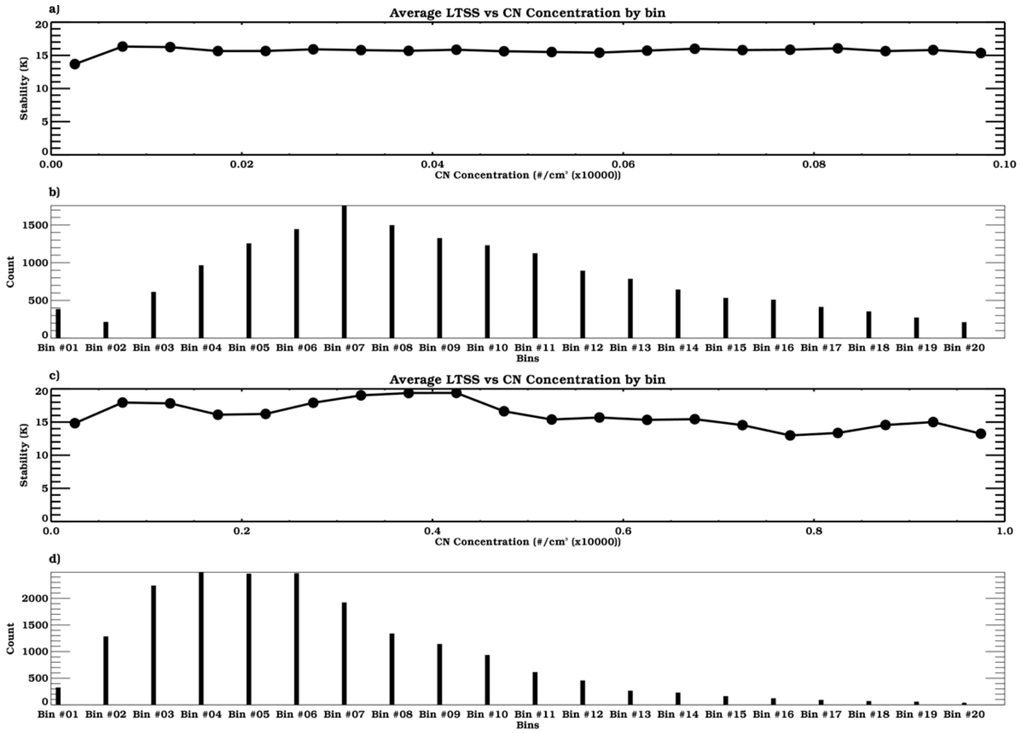

Figure 4.31
Average stability (in each bin) binned and plotted by CN concentration and histogram of bin populations. Panels a and b show data from the Azores site, panels c and d show data from the SGP site.

CHAPTER 5

CONCLUSIONS

In this study, aerosol effects were examined at two different locations in order to understand regional responses of precipitation to increased aerosol particle concentrations. Data was collected by instrumentation at DOE ARM sites in Oklahoma and the Azores, in order to include a continental and marine location, respectively, in the study. The data were also separated by cloud type and season as well as variables related to atmospheric and cloud dynamics in order to determine if these altered the aerosol effect. Meteorological variables were examined to see if these correlated with aerosol concentrations and if they affected the results.

It was generally found that increasing aerosols tended to suppress the onset of precipitation at the Azores and enhance it in Oklahoma. However, these trends did not necessarily hold up for every LWP range or for every season or cloud type. At the Azores, the stratus type clouds and summertime clouds showed suppression of precipitation onset, illustrating that the stratocumulus type clouds common in this region and season are susceptible to the 2nd indirect effect of aerosols. This is also perhaps why the general trend at lower LWPs was precipitation suppression. High LWP ranges did not exhibit precipitation suppression, even in these types of clouds, which was speculated to be due to the enhanced cloud dynamics and warm rain processes that may result from increased cloud water. This is consistent with past studies (e.g. Sorooshian et al. 2009) that suggest that clouds with high LWPs have efficient precipitation processes due to the abundant liquid water present. In these clouds, LWP dominates precipitation formation over the number concentration or size of cloud droplets (Sorooshian et al. 2009), resulting in less susceptibility to aerosol effects. The convective cloud types also appeared to undergo precipitation suppression, but in most LWP bins rather than solely the lower ones. The winter season observed a trend of precipitation enhancement, opposing the other results at this site. This could be evidence of the larger cloud systems passing through the region, which are more likely during this season. These systems have stronger dynamics that may cause them to be less susceptible to aerosol indirect effects. However, it was difficult to prove causes directly from the data. For example, evaporation rates are not measurable in clouds and neither are collision-coalescence rates for the case where high LWP is thought to enhance this process. The results can be compared to modeling studies, and in general, the precipitation at the Azores appears to predictably react to increased aerosols according to theory or hypotheses proposed through these modeling studies.

Situations that resulted in precipitation suppression did not necessarily exhibit all behaviors predicted by the indirect aerosol theories, namely the cloud thickness and LWP did not increase with increasing CN concentration. This was thought to be noise or be due to possible competing effects of the aerosols on the cloud dynamics; suppression of warm rain processes would tend to increase cloud thickness and LWP while increased evaporation of the smaller cloud droplets would decrease these values. It could also be due to one of the macrophysical ‘buffering’ mechanisms discussed in section 1.5. Also, some seasonal dependence of LWP susceptibility to aerosol was observed at this site, consistent with the idea from Han et al. 2002 that seasonal temperature shifts will cause varying affects of aerosols on the boundary layer dynamics of clouds in this region.

The Oklahoma site reacted quite differently to increased amounts of aerosol. The general trend at this site was precipitation enhancement. This is thought to be due to the more convective nature of the clouds in this location, illustrating the possibility of the microphysical buffering mechanism discussed in section 1.5. More water lofted higher in the cloud due to initial suppression is evaporated and causes invigoration, enhancing precipitation processes. However, certain cloud types and seasons (JJA and more stable convective cases) did show some precipitation suppression, which is thought to be related to the cloud dynamical processes. The stable convective cases, which are more common in summertime when convection occurs but deep convection is not as prevalent as the springtime (e.g. fair weather cumulus), most likely have weaker updrafts and thus a less efficient buffering mechanism. This still does not explain why the stratus type clouds in this region also experienced a precipitation enhancement instead of suppression as was expected to occur.

The cloud thickness and LWP at SGP behaved in opposition to original theories and even conflicted with the prediction from the newer microphysical buffering theory that was introduced in this study. However, it is thought that the decreasing trends of LWP are consistent with previous observational studies such as Han et al. (2002). The competing affects of the buffering mechanisms introduced previously may have balanced out to little trend in LWP and no trend in cloud thickness with increasing CN concentration. It is thought that there are competing effects, rather than no aerosol effect on these variables because of the observed effects of increased aerosol concentration on precipitation, however this remains uncertain. Considering that both suppression and enhancement of precipitation were observed at this site for certain cloud types and seasons, it appears that the effects of aerosols are present but not observed in these variables.

Clouds in the Azores clearly behave differently than the continental clouds at SGP. One possible reason could be similar to an idea presented by a few recent studies (e.g. Carrio et al. 2010, Cheng et al. 2009, van den Heever 2007) that there could be a threshold limit at which an increase in aerosols no longer has the same magnitude of effects on the cloud system and precipitation. This hypothesis basically states that if the background aerosol concentrations are relatively high, the effect from increasing aerosols will be weaker in magnitude than if there were low background aerosol concentrations. The site at Oklahoma is very continental in nature, while the Azores is distinctly marine. Hence, the observed aerosol affect may be weaker at Oklahoma because of the already high background concentrations (typically about an order of magnitude higher than at the Azores). Another possibility is that the GCCN concentrations are also enhanced with higher CCN concentrations, leading to more efficient precipitation formation processes (e.g. as in Feingold et al 1999, Cheng et al. 2009, van den Heever and Cotton 2007 and L’Ecuyer et al. 2009). However, this does not explain why the sign of the precipitation response to increased aerosols was different and not just the magnitude.

The most likely reason that the responses in the two locations were so different is because of the diverse environmental conditions and boundary layer dynamics that occur in these two regions. These differing atmospheric conditions also tend to result in varying cloud types between the sites and cloud type can significantly affect results, as was observed in chapter 4. In general, it appeared that both locations had competing aerosol effects occurring. The less stable, more convective nature of the warm clouds in Oklahoma allowed these clouds to overcome the predicted aerosol effects of precipitation suppression, perhaps through the buffering mechanisms (specifically lofting of water and decreasing of stability through evaporative cooling, invigorating precipitation processes) presented throughout this study as well as by Stevens and Feingold (2009). However, the effect of increased aerosols on the boundary layer dynamics of the more stratus or stratocumulus type of cloud observed at the Azores site resulted in more traditional responses. As has been observed in past studies of other oceanic regions (and specifically stratocumulus fields, e.g. Lebsock et al. 2008, Jiang et al. 2002, Lu and Seinfeld 2005, Wang and Feingold 2009, Comstock et al. 2004, etc.), increased concentrations of CN led to precipitation suppression, except at high LWPs, where there is most likely enough cloud water to overcome the decreased efficiencies of precipitation processes. The results for cloud type and season tended to generally agree with these basic ideas, although they were much noisier and more complex due to the fewer number of cases. Also, the fact that the results were not as robust when stratified by stability illustrates the important role that the meteorological conditions and cloud dynamics have.

In summary, regional aerosol effects on precipitation vary widely due to differences in cloud type as well as meteorological conditions. The response of precipitation predicted by traditional theories of aerosol indirect effects are not valid for all cloud regimes, as has also been suggested by other studies such as Khain et al. (2009), van den

Heever et al. (2006), and van den Heever et al. (2011). Further observational studies are required to determine the actual (not solely modeled) aerosol response in continental locations, since the past emphasis has been on marine sites and regional modeling or global studies. Also, it is necessary to determine if the aerosol response in these locations is significant enough to overcome the cloud response to meteorological conditions, considering that the aerosol effect may be secondary to other mechanisms. This study has illustrated that, as predicted by Khain et al. (2009) and Storer et al. (2010), meteorological conditions and regional locations (and hence cloud types) do play an important role in regulating aerosol responses and perhaps even altering the sign of these responses. The response of precipitation to increased aerosols is important for hydrological implications and this study indicates that the response of warm clouds to increased aerosol concentrations may be muted or even in opposition to current theories. This is especially true considering that the marine location, which experienced suppression of the onset of precipitation, witnessed precipitation enhancement at the ground. Therefore, it appears that alterations to current theories and hypotheses may be necessary along with more regional observational studies in order to more fully determine the response of precipitation to increased aerosol concentrations.

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APPENDIX A

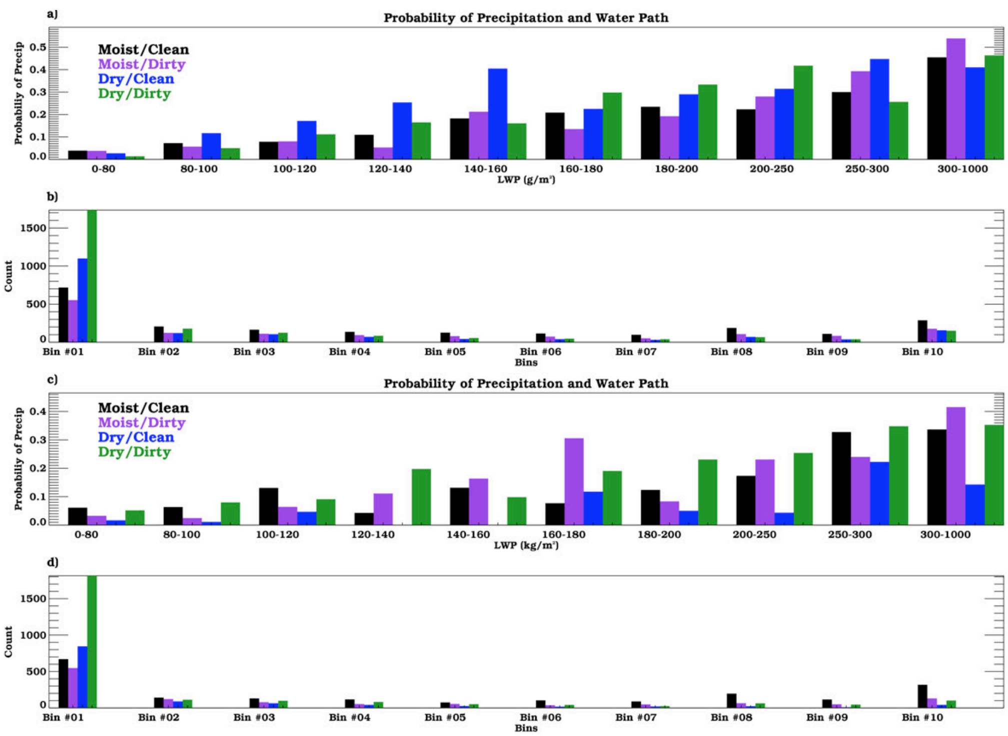

Figure A1
Probability of precipitation binned by LWP and histogram of the bin populations. Both plots are also separated by aerosol (same distinctions as presented in chapter 4 figures) and relative humidity (top third and bottom third of the relative humidity dataset make up the 'moist' and 'dry' distinctions, respectively). Panels a and b show results from the Azores site, panels c and d show results from the SGP site.

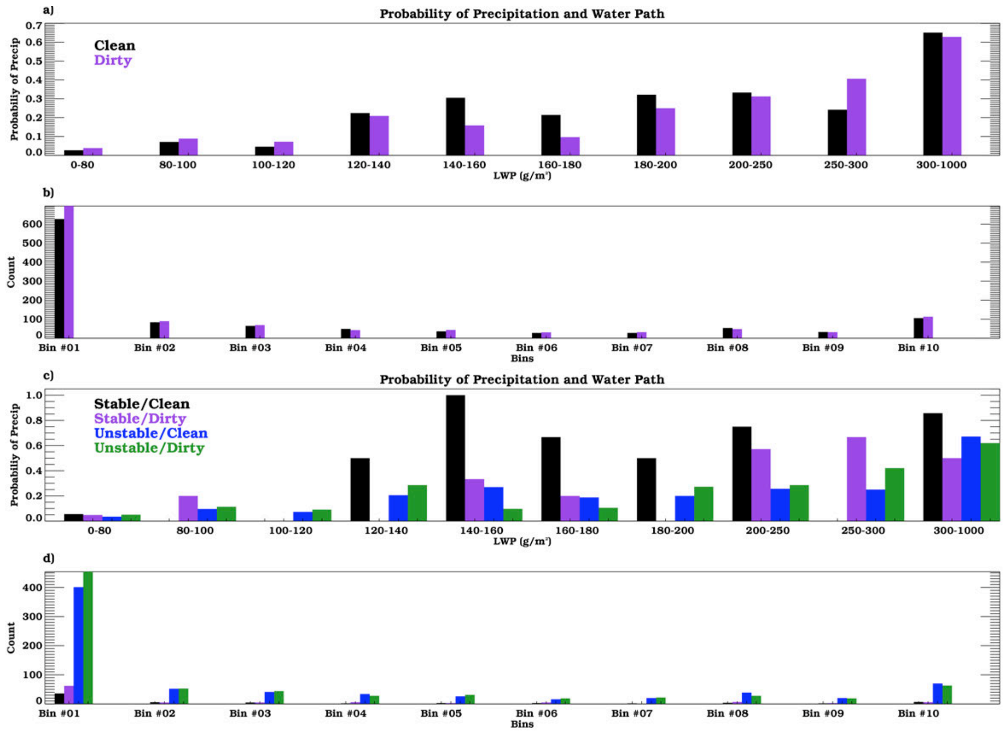

Figure A2
Probability of precipitation binned by LWP and histogram of bin populations at the Azores site for spring (MAM) only. Panels a and b are separated by aerosol, while panels c and d are also separated by stability.

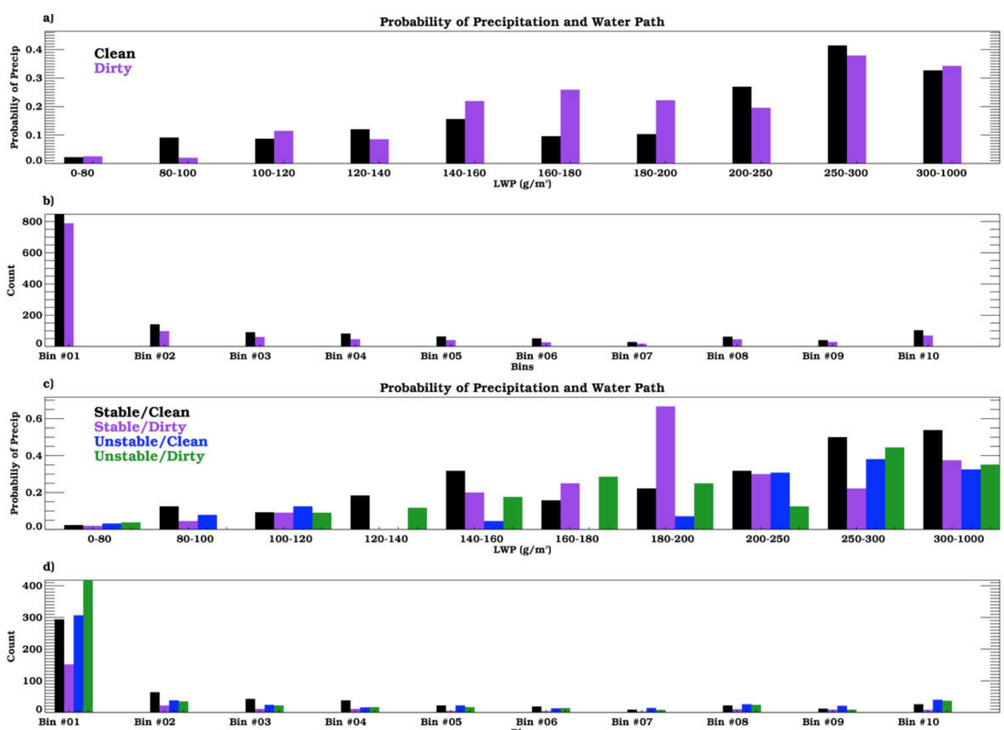

Figure A3
Same as figure A2, but for autumn (SON).

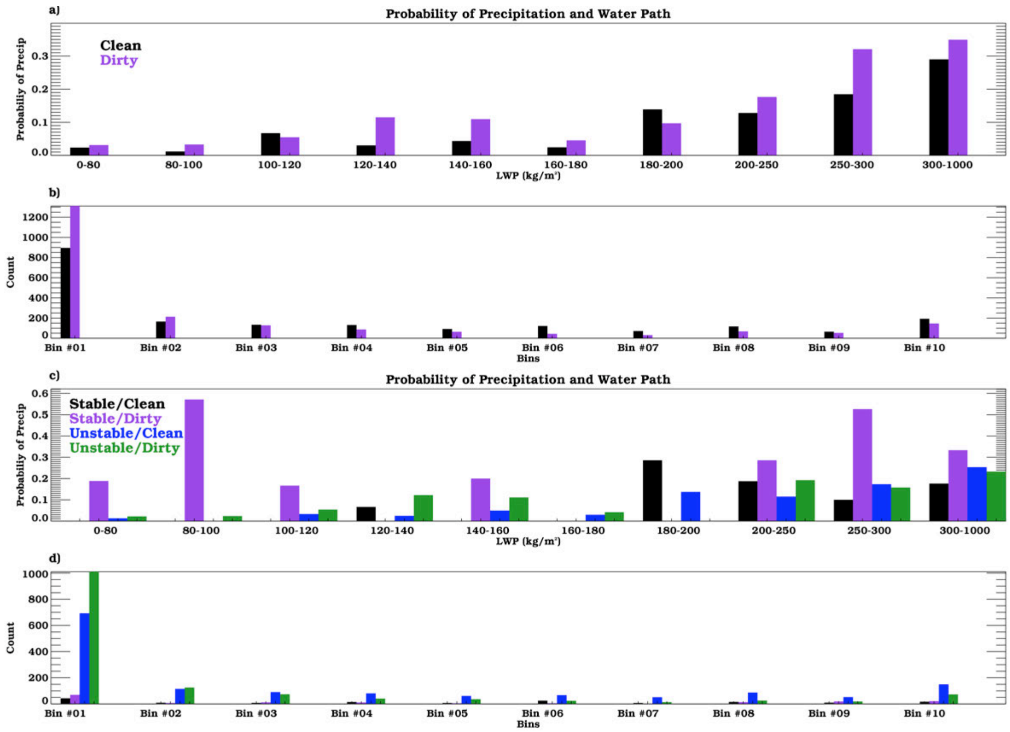

Figure A4
Probability of precipitation binned by LWP and histogram of bin populations at the SGP site for spring (MAM) only. Panels a and b are separated by aerosol, while panels c and d are also separated by stability.

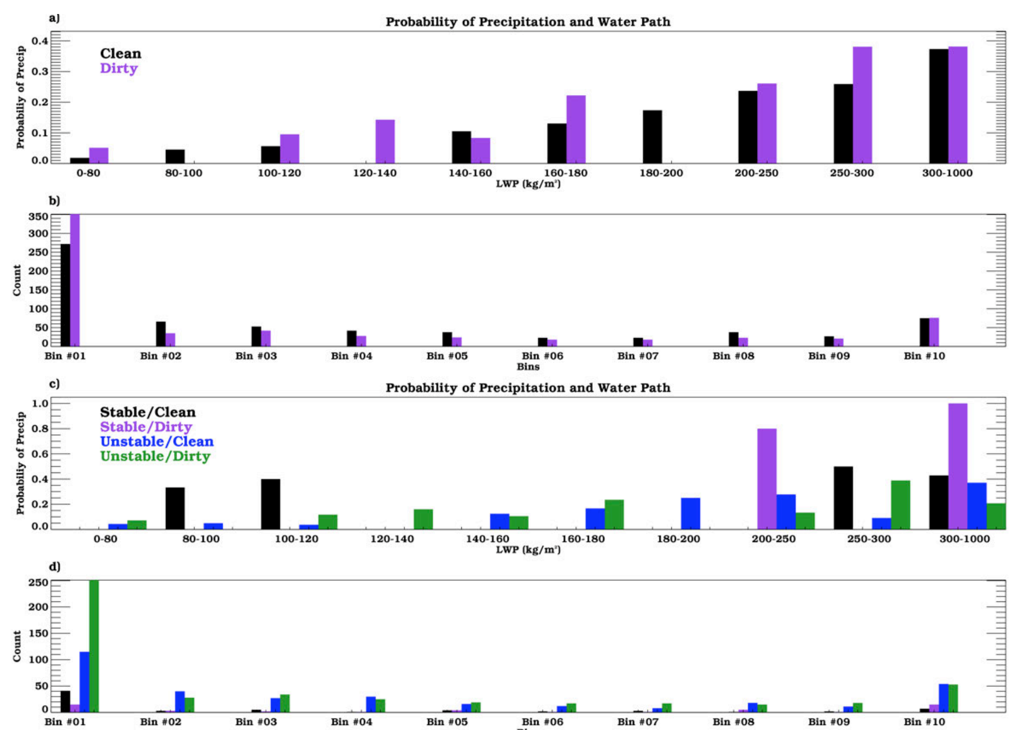

Figure A5
Same as figure A4, but for autumn (SON).

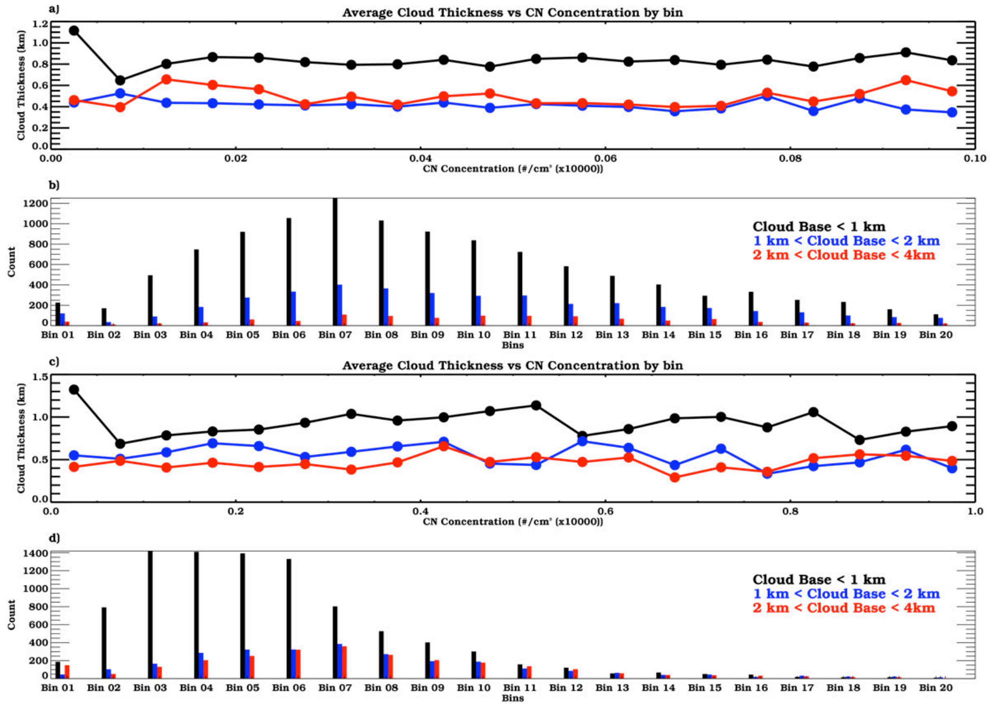

Figure A6
Average cloud thickness binned and plotted by CN concentration and separated by cloud base height. Bin populations are also shown. Panels a and b show data from the Azores, panels c and d show data from SGP.

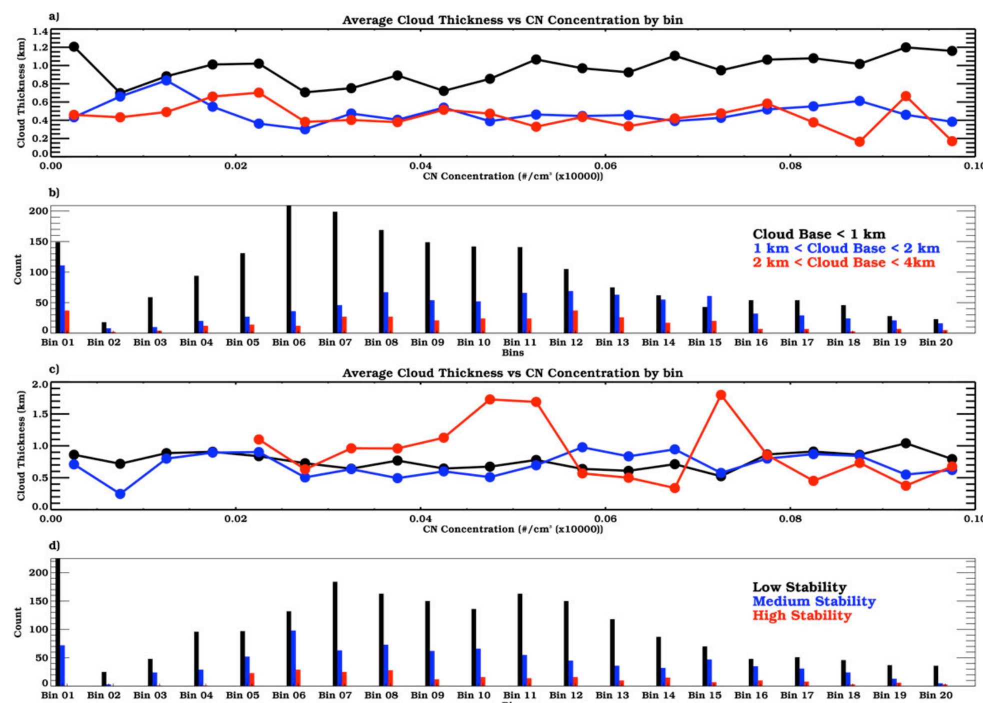

Figure A7
Average cloud thickness binned and plotted by CN concentration at the Azores for spring (MAM). Bin populations are also shown. Panels a and b show data separated by cloud base height, panels c and d show data separated by stability.

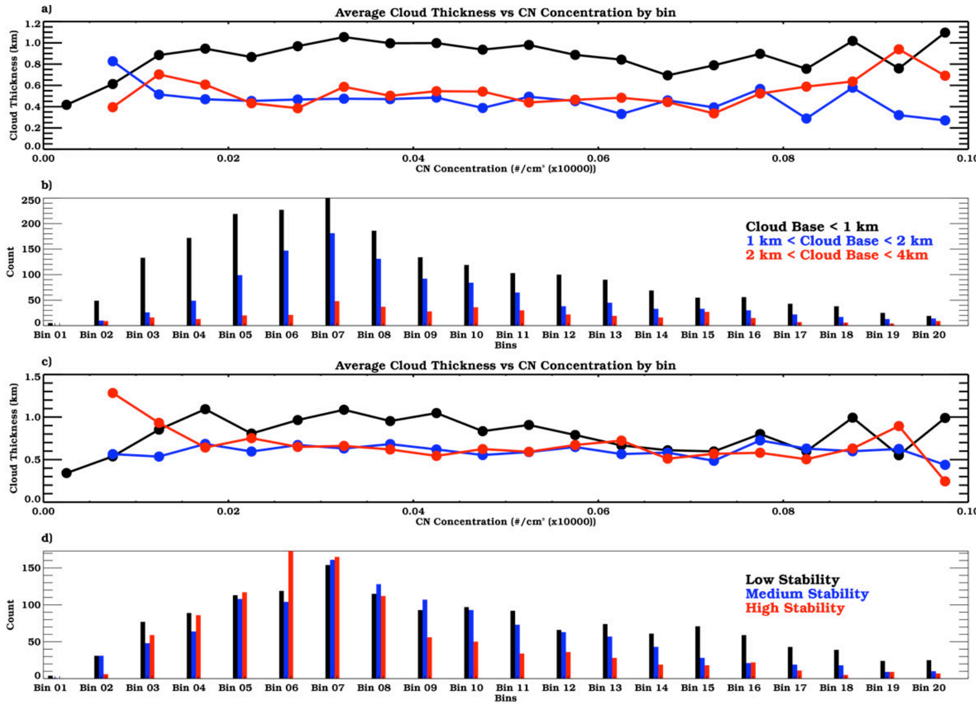

Figure A8
Same as figure A7, except for autumn (SON).

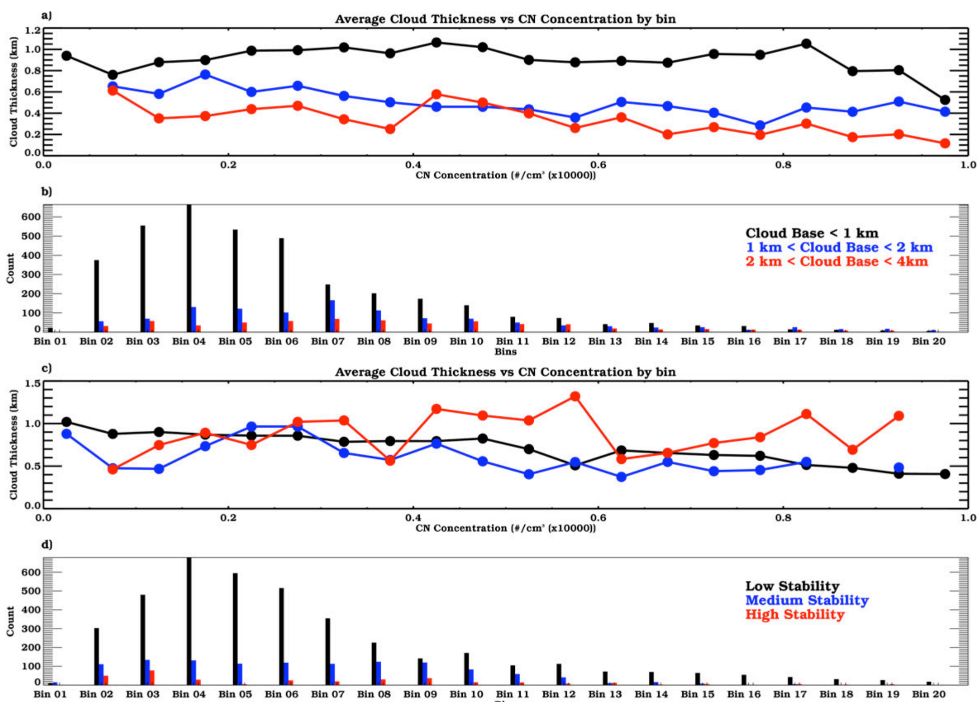

Figure A9
Average cloud thickness binned and plotted by CN concentration at SGP for spring (MAM). Bin populations are also shown. Panels a and b show data separated by cloud base height, panels c and d show data separated by stability.

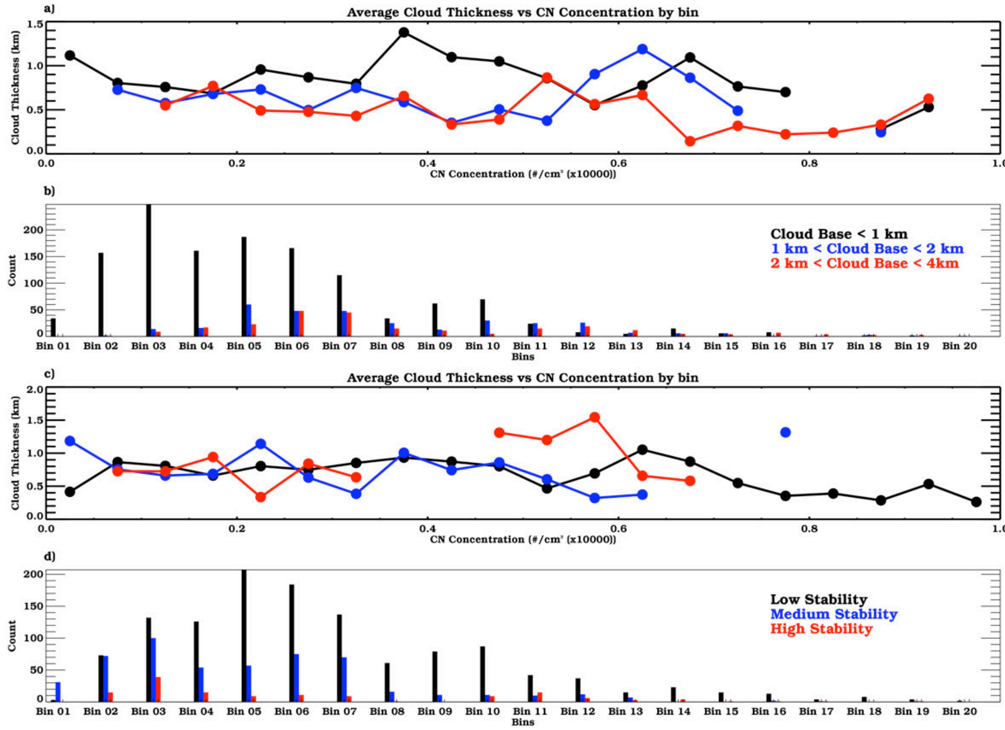

Figure A10
Same as figure A9, except for autumn (SON).

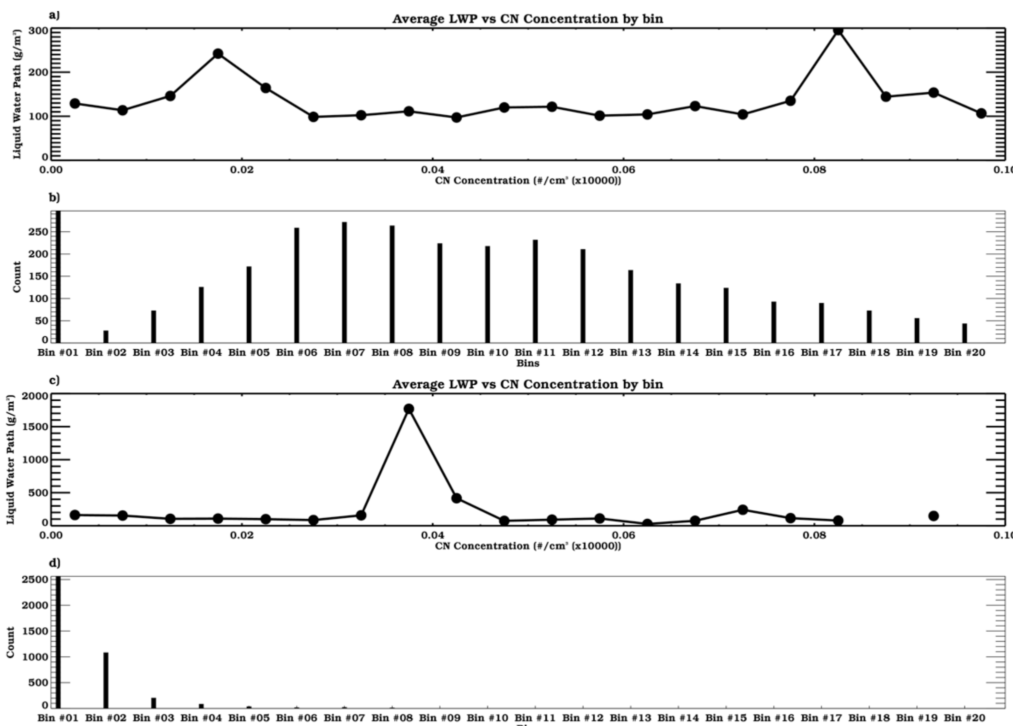

Figure A11
Average LWP binned and plotted by CN concentration at the Azores site. Bin populations are also shown. Panels a and b show data from spring (MAM), lower panels show data from autumn (SON).

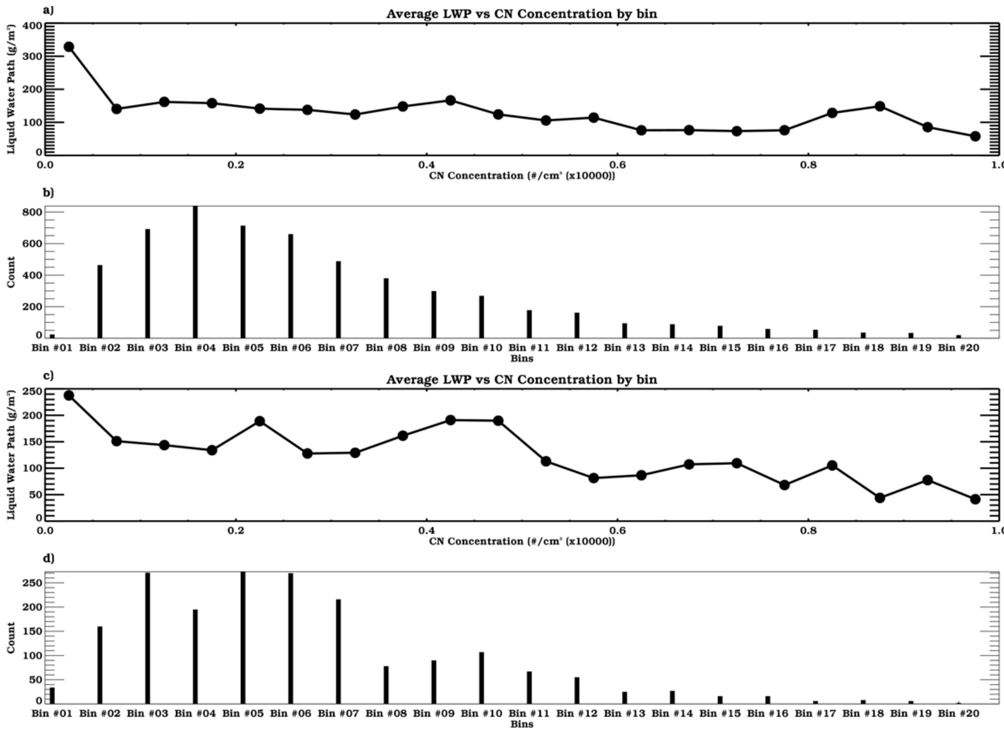

Figure A12
Same as figure A11, except at the SGP site.

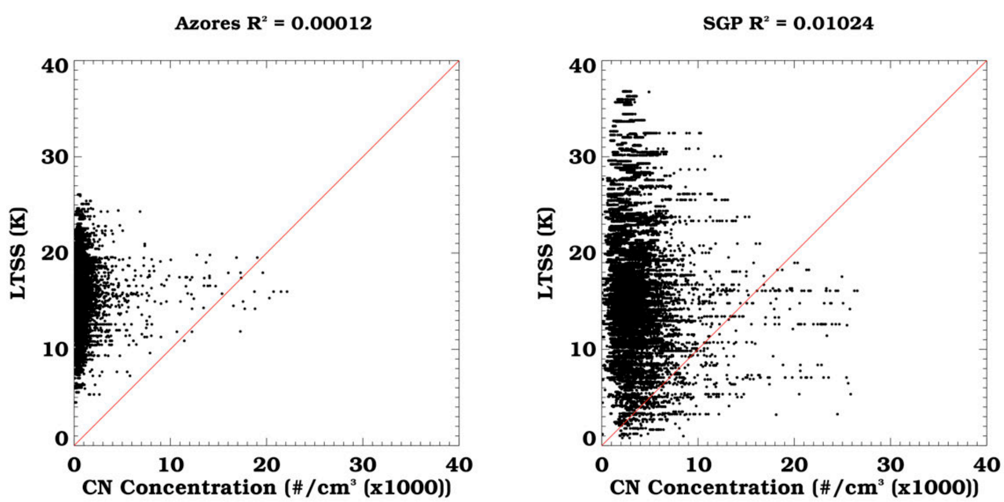

Figure A13
Scatter plots showing stability plotted against CN concentration. Left panel shows data for the Azores site, right panel shows data for the SGP site.