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AEROSOL-CLOUD INTERACTIONS IN TURBULENT CLOUDS: A COMBINED
CLOUD CHAMBER AND THEORETICAL STUDY

By

Kamal Kant Chandrakar

A DISSERTATION

Submitted in partial fulfillment of the requirements for the degree of

DOCTOR OF PHILOSOPHY

In Atmospheric Sciences

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This dissertation has been approved in partial fulfillment of the requirements for the Degree
of DOCTOR OF PHILOSOPHY in Atmospheric Sciences.

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Dedication

To my mentors, family and friends

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Preface

Topics covered in this thesis are derived from the research I have done as a graduate student in the Pi-chamber group at Michigan Tech. In chapter 1, an overview of the topic and some background information is provided. Chapter 2 discussed the moist turbulent Rayleigh-Bénard convection and production of supersaturation fluctuations. It is based on an article under review in *Journal of Fluid Mechanics*. Chapter 3 is about the stochastic condensation theory and *turbulence-induced aerosol effects*, and it is based on an article (Chandrakar et al. [43]) published in *Proceedings of the National Academy of Science*. Chapter 4 is the extension of the work presented in Chandrakar et al. [43] and chapter 3 and is based on a paper (Chandrakar et al. [45]) published in *Journal of the Atmospheric Sciences*. In chapter 5, a transient cloud process, 'cloud cleansing,' and aerosol-cloud feedback during this process with the presence of turbulent flow condition are explored. The work presented in this chapter was published in *Geophysical Research Letters* (Chandrakar et al. [47]). A part of this article was also presented in the master thesis of Sarita Karki (Karki [137]). The chapter 6 is about the laboratory study of the effective radius of cloud droplets, their parameterization, and the dispersion effect. This work was originally published in *Geophysical Research Letters* (Chandrakar et al. [48]). Chapter 7 present a theoretical and laboratory investigation of the functional form of the cloud droplet size distribution in a turbulent cloud condition. It is based on a manuscript under preparation for submission in a peer-reviewed journal. Chapter 8 is the final chapter discussed some implications of studies presented in this thesis and outlined some related scientific questions and topics to explore.

For the research presented in the above articles, experiments, theoretical derivation, data analysis, and writing were done by me with the help of Dr. Raymond A. Shaw and co-authors. We are thankful to all co-authors for their contributions in the listed publications.

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Abstract

The influence of aerosol concentration on the cloud droplet size distribution is investigated in a laboratory chamber that enables turbulent cloud formation through moist convection. In chapter 2, moist Rayleigh-Bénard convection with water saturated boundaries is explored using a one-dimensional-turbulence model. This study provides some background about supersaturation statistics in moist convection. Chapters 3 - 7 discuss the experimental and theoretical investigation of aerosol-cloud interactions and cloud droplet size-distributions in turbulent conditions.

The experiments are performed in a way so that steady-state microphysics are achieved, with aerosol input balanced by cloud droplet growth and fallout. As aerosol concentration is increased the cloud droplet mean diameter decreases as expected, but the width of the size distribution is also observed to decrease sharply. The aerosol input allows for cloud generation in the limiting regimes of fast microphysics ($\tau_c < \tau_t$) for high aerosol concentration, and slow microphysics ($\tau_c > \tau_t$) for low aerosol concentration; here, τ_c is the phase relaxation time and τ_t is the turbulence correlation time. The increase in the width of the droplet size distribution for the low aerosol limit is consistent with the larger variability of supersaturation due to the slow microphysical response. A stochastic theory developed based on the Langevin equation for supersaturation predicts that the standard deviation of the squared droplet radius should increase linearly with a system time scale defined as $\tau_s^{-1} = \tau_c^{-1} + \tau_t^{-1}$, and the measurements are in excellent agreement with this finding. These

experiments are discussed in chapters 3 and 4.

This effect of varying cloud droplet size-distribution width underscores the importance of droplet size dispersion for aerosol indirect effects. An application of this coupling of aerosol and supersaturation fluctuations during the 'cloud-cleansing' process is discussed in chapter 5. Cloud droplet relative dispersion, defined as the standard deviation over the mean cloud droplet size ($d = \sigma_r/\bar{r}$), is of central importance in determining and understanding aerosol indirect effects. The analytical expression of d obtained from the stochastic theory is found to depend on the cloud droplet removal time, which in turn increases with the cloud droplet number density. The results show that relative dispersion decreases monotonically with increasing droplet number density, consistent with some recent atmospheric observations. The albedo susceptibility due to turbulence broadening has the same sign as the Twomey effect and augments it by order 10%. These results, along with the test of a commonly-used effective radius parameterization, are presented in chapter 6.

In chapter 7, theoretical expressions for cloud droplet size-distribution shape are evaluated using measurements from controlled experiments in the II Chamber. Three theoretical distributions obtained from a Langevin drift-diffusion approach to stochastic condensation are tested. Statistical techniques of χ^2 test, sum of squared errors of prediction, and residual analysis are employed to judge relative success or failure of the theoretical distributions to describe the experimental data. In relative comparison, the most favorable comparison to the measurements is the expression for stochastic condensation with size-dependent droplet removal rate.

Chapter 1

Overview

"Clouds come floating into my life, no longer to carry rain or usher storm, but to add color to my sunset sky." - from **Stray Birds** by **Rabindranath Tagore**

Clouds are central to the existence of life from the beginning of Earth's history. Their influence in human life is so conspicuous that their involvement is evident, not just in science, but also in different forms of art.

Clouds are a critical element for a short term weather pattern or a long term climate change. As humanity is progressing technologically, weather prediction plays a significant role in our day to day life from a significant decision making to the planning of a trip. Likewise, long term climate studies help forge policies at government levels. Clouds are one of the wildcards in these weather and climate studies, and its implementation in numerical models appropriately requires a closer understanding of all associated physical processes.

Some of the basic macroscopic processes include influence on radiative fluxes, precipitation formation, interaction with the dynamics of atmospheric boundary-layer, and interaction with pollutants (e.g., aerosols). The small scale microphysics driver drives this macroscopic picture of clouds. In this thesis, some of these microphysical processes are explored using controlled laboratory experiments and theoretical analysis based on the stochastic method. Specifically, aerosol-cloud interaction in a turbulent environment is a central topic here. In the next subsequent sections, an overview of measurement studies of cloud microphysical processes is introduced for a background.

1.1 The complementary roles of field and laboratory measurements

Scientific observation is the firm foundation of reality on which all theories and modeling frameworks are based. The ideal scientific observation is more than a sanity check, it is the reference of reality to which theoretical predictions and numerical models must be compared. What do we mean by an ideal scientific observation? It is a controlled, repeatable measurement of a physical quantity or phenomenon in a well-defined system of interest with known external influences. However, in reality and especially in geophysical systems, an ideal measurement does not exist due to the absence of some of the above criteria. Therefore, these limitations must be identified and carefully considered during analysis, so that they can still serve their intended and indispensable purpose. In the context of this chapter, measurements serve two primary purposes: they provide input and constraints needed for

implementation of known theory (e.g., radiative transfer), and they guide the exploration of poorly-understood physical processes (e.g., ice formation through nucleation and secondary processes).

Broadly, there are two significant categories of observation in atmospheric science: field and laboratory observation. Both complement each other to advance scientific understanding and to provide input for modeling frameworks, and in fact they inform each other: laboratory experiment provides understanding of specific mechanisms for idealized systems needed to interpret the complexity of the field observations, and field observations provide guidance on what ranges of parameter space and what limits are relevant for exploration in the laboratory. Thus both approaches have limitations and strengths, which make them unique in their capabilities. For example, field measurements bring us closest to the actual representation of natural processes, but are complicated due to the presence of multiple simultaneous processes and feedbacks, limited data availability due to the remoteness of systems of interest (such as clouds), limited ability to perform adequate averaging due to the transient nature of many natural processes, and limitations in resources and technical capabilities for the instruments needed for field measurements. Nevertheless, field measurement provide the closest picture of reality and most relevant input data needed for developing and evaluating models. On the other side, laboratory studies are designed to have better control of governing parameters and boundary conditions, and are sufficiently isolated and simplified to provide process level understanding. They can be designed to overcome the sampling limitations, and steady-state conditions are also easier to achieve in some cases. However, lab experiments are not able to provide a complete picture of complex

atmospheric interactions, and therefore must be carefully designed if their results are to be relevant in providing individual pieces of a complex puzzle.

There are significant contributions from field and laboratory measurements in the development of atmospheric science. Undoubtedly, there are an enormous amount of field studies which are constantly providing input over a range of spatial and temporal scales. Laboratory measurements have become somewhat less common, but their contributions have been profound and it is safe to say that a resurgence in laboratory experimentation relevant to ‘fast physics,’ the topic of this book, is occurring. This venue is far too constrained to allow for a thorough review of modern methods and contributions of in-situ and laboratory measurements. Our purpose, therefore, is to provide illustrative examples of measurements that have provided new insight. Our own research makes us most qualified to consider this topic from the perspective of the cloud microphysics and turbulence, so we acknowledge that bias from the outset. Furthermore, simply because of familiarity, and without any intention of priority, we draw some of the examples from our own work.

1.1.1 Illustrative historical examples of field and laboratory measurements

The early cloud chamber experiments of Gunn and Phillips [110] are an instructive example of the insight that can be gained from controlled laboratory measurements. The experiments were performed in an enormous cloud chamber with a diameter of 18.3 meters and a volume of more than 3000 m³ [247]. Findings for two different aerosol conditions are illustrated in

Figure 1.1. The related discussion of Gunn and Phillips [110] is remarkable to us because it encapsulates much of what we consider our current understanding of aerosol indirect effects of cloud microphysics. We can't help but wonder how much sooner such indirect effects could have been thoroughly explored had this cloud chamber facility remained in operation (in passing, we have not been able to determine what factors led to the facility being decommissioned).

"It was found that the abundance of condensation nuclei profoundly influences the character of the rain-initiating processes. Hundreds of clouds artificially produced in the chamber have shown that normal expansions of originally saturated air usually produce a dense clouds of droplets having approximate radii of $7\text{ }\mu\text{m}$ or less. Such clouds are quite stable, persist in the cloud chamber for times exceeding a half hour, and ultimately evaporate as the chamber recovers from the initial cooling of the processed air. It is also observed that if the expansion rate is much less than that typically produced in nature, so that relatively few of the available nuclei are activated, some of the droplets can grow to radii of something like $15\text{ }\mu\text{m}$. In striking contrast to the above results, it is found that the behavior of clouds formed by the expansion of air that has been carefully freed of pollution and of condensation nuclei behaves in a radically different manner. The cloud droplets so formed are notably larger than those formed from ordinary air, and the cloud largely disappears by precipitation rather than evaporation. In fact, we have successfully produced a mist-like rain by expanding well cleaned and saturated air in our 60-ft cloud chamber. This

is impossible when ordinary air is used. These observations are consistent with experimental data reported by Wilson nearly 60 years ago, but their significance and application to meteorological problems were not appreciated.”

1

Figure 1.1: Cloud droplet size distributions measured in a large cloud chamber, illustrating what today is known as the first indirect effect or Twomey effect. The clouds were formed in “uncleaned Gulf air” on the left, and “air electrostatically cleaned” on the right. Figure adapted from Figure 7 of Gunn and Phillips [110]

One of the most influential series of laboratory measurements came from convection tank experiments of Deardorff and colleagues, reported in a series of papers beginning with Deardorff et al. [71] and continuing for more than 15 years. The work was idealized so as to inform the theory of mixed-layer models for the atmospheric boundary layer, such as the the stratocumulus-topped boundary model developed by Lilly [173]. It is not an overstatement that the experiments also eventually led to the first large-eddy simulations [69, 330], which has since become a ubiquitous tool in the atmospheric sciences and beyond. Perhaps symbolically, the experiments were carried out in the basement of the National Center for Atmospheric Research; according to rumor this was so as to avoid attention from administrators unimpressed by such research methods [330]. In spite of that skepticism, the

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measurements continue to inspire and set the standard of comparison for computational work on developing boundary layers [306].

Some of the most influential field measurements are those that were carefully planned in the context of clear theoretical structure and questions. The Kansas experiment that explored boundary layer turbulence was designed with simplifications and measurement strategies guided by such a theoretical framework [327]. One of many outcomes was to confirm the Monin-Obukhov similarity theory for the atmospheric surface layer and mixed-layer models for the region of the boundary layer above the surface layer [135]. Besides the numerous papers that emerged from the project itself, this well-designed experiment has continued to guide theoretical and computational research for decades since [150].

Hobbs and Rangno carried out a set of extensive airborne field studies that have motivated laboratory and computational work in cloud physics for many years since the measurements themselves [118, 246]. These measurements helped inform and strengthen the idea that mechanisms must exist for ice generation beyond simple, primary ice nucleation [17]. A key step in that work was the recognition that aircraft themselves can generate ice when they pass through clouds [245], and the resulting adjustments in sampling strategies led to much greater confidence in the measurements. Subsequently it was determined that ice crystal shattering likely contaminated some of the concentration and size measurements [127, 159]; it is still generally accepted, however, that the discrepancy holds and mechanisms for ice multiplication are needed to account for observed ice concentrations [155].

1.2 What do models need to know about clouds?

Large scale models (e.g., global circulation models), as well as cloud resolving models (or large eddy simulations), are inherently limited in their ability to directly represent phenomena occurring at spatial scales below their grid sizes and temporal scales below their time steps. For example, GCMs are not able to resolve the atmospheric boundary layer or cloud-scale processes; even cloud-resolving models and large eddy simulations require some input for surface fluxes and sub-grid-scale cloud and turbulence processes. Inputs needed for these unresolved processes come in the form of parameterizations from measurements of cloud, boundary-layer, and turbulent flow properties. For example, as a boundary-layer property, information about surface momentum, heat, moisture, and other scalar fluxes is needed. For cloud processes, information related to radiative, precipitation and convective fluxes is needed. In this chapter, we limit the focus to cloud properties which are introduced briefly in the subsequent subsections.

1.2.1 Radiation (radiative fluxes)

Radiative fluxes through a cloud depend on properties like cloud optical depth, single-scattering albedo, and asymmetry parameter. All these properties depend on the size distribution, shape and orientation of hydrometeors, and the partitioning of water phases (liquid versus ice). For a liquid cloud, these dependencies can be further reduced to two microphysical variables, effective radius which is a ratio between the third and second

moment of a droplet size distribution and liquid water path [114, 279, 286]. As an input parameterization, models need the relation between amount of cloud water content and effective radius. This information can be obtained from measurements; for example, Martin et al. [199] showed nearly a linear relation between the cube of mean volume radius and effective radius of cloud droplets for stratocumulus clouds. Therefore, a parameterization of effective radius (r_e) can be expressed as

$$r_e \equiv \frac{\int r^3 n(r) dr}{\int r^2 n(r) dr} \approx \left[\frac{3L}{4\pi\rho_l n_d k} \right]^{1/3}, \quad (1.1)$$

where ρ_l is the density of liquid water, n_d is the droplet number concentration, L is the liquid water content, and k is a constant. The parameter k is a function of relative dispersion and skewness of the droplet size distribution [240]. Consequently, the cloud optical depth can be parameterized with the assumption of a constant liquid water content throughout the cloud depth:

$$\begin{aligned} \tau &\equiv \int_0^h \int_0^\infty Q_{ext} r^2 n(r) dr dz \\ &= 3/4\pi \overline{Q_{ext}} \rho_l^{-1} r_e^{-1} h \\ &\approx \left(\frac{3}{4\pi\rho_l} \right)^{2/3} \overline{Q_{ext}} L^{2/3} n_d^{1/3} k^{1/3} h. \end{aligned} \quad (1.2)$$

Here $\bar{h}$ and $\bar{Q}_{ext}$ are the cloud depth and extinction efficiency (bar indicating average value) respectively.

A lab experiment confirmed the above effective radius parameterization ($r_e^3 = k \overline{r^3}$) in a controlled turbulent environment [48]. However, there are some conditions when this simple parameterization may not be an appropriate assumption, such as in regions dominated by entrainment or when there is a significant amount of drizzle formation. Additionally, these parameterizations also depend on aerosol population [48, 175, 199]. The above picture assumes uniform cloud within a region of interest (for example, within a grid cell of a large scale mode). This assumption might be inadequate if grid cells are large; therefore, numerical models require an additional measure of non-uniformity in a system; indeed, measurements show spatial correlations in cloud liquid water content on kilometer to millimeter scales and this must be represented in the calculation of radiative fluxes [65].

Ice clouds and mixed-phase clouds are still more complicated due to ice nucleation and growth mechanism that are still not fully understood. Moreover, their interaction with shortwave radiation involves additional factors like crystal shape, orientation, and ice fraction (in the case of mixed-phase clouds). These demand substantive field observations and controlled laboratory experiments to understand these clouds so that useful parameterizations can be developed for models. In essence, measurements of cloud properties help guide the development of useful parameterizations and, additionally, aid in placing bounds on their applicability.

1.2.2 Precipitation (water flux)

Similar to the radiative flux through clouds, precipitation flux of water (liquid/ice) depends on the size distribution of hydrometeors, their type, and concentration. An estimate (or parameterization) of the rate of conversion of vapor-grown particles (like cloud droplets or small ice crystals) to precipitating particles requires information about the rate of different growth processes like autoconversion, accretion, riming, and aggregation. In large scale models, these processes are modeled based on the assumed size distribution of hydrometeors. For example, the autoconversion rate of cloud droplets to rain droplets can be estimated if the distribution shape (most commonly assumed as gamma distribution), critical size for conversion, and collision kernel are known [176]:

$$P = \kappa L \int r^6 n(r) dr. \quad (1.3)$$

Here, the Long collision kernel (κ) is used for the derivation. In this example, the autoconversion rate depends on the zeroth (number concentration), sixth and third moments of a droplet size distribution. It can be further simplified in terms of lower moments if a distribution shape is assumed. In case of other precipitation formation processes, additional information like the shape of hydrometeors (ice crystal), fall speed, and density of particles is required. Therefore, observational studies are aimed at determining the size distribution of hydrometeors and other required microphysical details, as discussed above, under

different cloud conditions.

1.2.3 Convection (water flux)

Atmospheric water vapor is one of the crucial factors contributing to climate sensitivity. Convective clouds are one of the vital sources of vertical transport of water vapor and latent heat in the free troposphere [132, 268]. The extent of this transport depends on the entrainment and detrainment flux in these clouds [25, 136, 295]. Moreover, these fluxes affect cloud microphysical process and precipitation formation. Measurement of thermodynamic quantities helps to quantify the profile of these fluxes as well as guide the development of an effective parameterization scheme for large scale models.

In boundary layer clouds, cloud top entrainment influences cloud cover and thermodynamic properties. The layer which governs the entrainment properties are not well resolved in the atmospheric models. Therefore, a parameterization is required to model the cloud top entrainment velocity. This entrainment velocity depends on wind shear, turbulence generated by radiative and evaporative cooling, and coupling to microphysics through drizzle formation and droplet settling [205]. For example, the entrainment velocity due to the droplet settling flux depends on the fifth moment of a droplet size distribution [67].

1.2.4 Processes affecting the droplet size distribution

1.2.4.1 Condensation-growth

Condensation growth is the mechanism for cloud droplet growth at the initial stage after the activation of cloud condensation nuclei. A traditional picture of droplet condensation growth in a closed parcel suggests that the droplet size distribution should get narrower with time, but this is contrary to many observations (e.g., see the size distributions shown in Figure 1.2). This predicted narrowing is a result of the inverse radius dependence of the droplet radius growth rate. In most of the models, a central assumption in the growth calculation is uniform supersaturation within a parcel or model grid cell. If grid cells are large enough, significant fluctuations due to the presence of turbulent flow conditions may be filtered out. In the absence of any sub-grid-scale model or parameterization, the result is a narrow droplet size distribution, spurious liquid water content, and other corresponding feedbacks. How do we know this is the case or how significant are these effects? Answers to these questions lie in exploration of the droplet size distribution primarily via in situ measurements.

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The following basic calculations serve to illustrate: Assume there is an adiabatic cloud parcel experiencing mean updraft velocity ($\overline{w}$) and containing a fixed concentration of activated

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Figure 1.2: Cloud droplet size distribution measured using a FSSP in a) Maritime and b) Continental condition at different altitude level of stratocumulus cloud field. The figure is from Martin et al. [199].

cloud droplets. The starting mean and dispersion of the droplet size distribution are $(\bar{r})$ and (σ_r) . If the growth in droplet size is larger than the assumed starting size, the difference in sizes of two droplets of starting sizes $\bar{r} - \sigma_r$ and $\bar{r} + \sigma_r$, after the parcel has been displaced Δz vertically, would be:

$$\delta r(t) \approx \frac{\sqrt{2}\bar{r}\sigma_r}{\sqrt{\xi s \Delta z \bar{w}^{-1}}}. \quad (1.4)$$

Here, the droplet growth rate due to condensation is approximated as $d_t r^2 = 2\xi s$ with ξ being a thermodynamic constant and s being the supersaturation in the parcel [342]. The vertical displacement of the parcel is simplified as $\Delta z = \bar{w}t$. The equation demonstrates that the width of the droplet size distribution δr decreases with vertical displacement Δz .

The idea that the droplet size distribution gets narrower with time (or with vertical position) under assumed fixed supersaturation is inconsistent with observations (e.g., Martin et al. [199], Miles et al. [210], Pawlowska et al. [229]). Figure 1.2 shows droplet size distributiona measured at different altitude and for different aerosol levels (maritime versus continental) in stratocumulus clouds. The observations confirm the argument that the droplet size

distribution not only does not get narrower with altitude, but instead gets slightly broader. An aspect of this figure related to aerosol concentration will be discussed subsequently in the subsection 1.2.4.3.

There are different proposed mechanism to explain observed broad droplet size distribution: the curvature and solute effects (and associated mechanisms such as spectral ripening and competing activation of cloud condensation nuclei of varying size, solubility, etc.) [134, 156, 322, 339], turbulent fluctuations [46, 60, 225, 256, 277], entrainment and mixing [12, 172, 345], microphysical variability [60, 74], and internal mixing of parcels of different growth history [123, 169]. Liu and Hallett [177] based on the system theory suggest that the width of droplet size distribution also depends on the averaging length scale. If the length scale of averaging is below some minimum threshold, the average spectrum would depend on the length scale [177, 179]. Although, above this threshold, the size distribution would be independent to the length scale and would have a maximum width. The system theory also suggests a Weibull distribution shape for a maximum likelihood distribution with a variable shape factor based on the intensity of turbulence [179].

1.2.4.2 Collision-growth

Condensation growth in a typical adiabatic cloud parcel is very slow and insufficient to produce precipitation hydrometeors within typically observed times required for precipitation formation [18]. Droplet collision-coalescence is the mechanism required to accelerate drop growth for radius approximately $r > 20 \mu\text{m}$. In a quiescent flow, droplet collision

occurs due to the difference in relative settling velocity of large and small droplets with some efficiency of collision due to their hydrodynamic interactions. However, in the case of turbulent flow conditions, the relative velocity of droplets depends on the intensity of turbulence and relative particle response timescale. Additionally, another parameter that influences the rate of collision is the spatial distribution of droplets which also depends on turbulence intensity and particle response timescale, and the radial distribution function (or spatial covariance of droplets) is generally used to characterize the correlations [258]. During a collision event, surface interactions of droplets determine the coalescence efficiency. All of the above microscale interactions are studied extensively using laboratory experiments and high-resolution direct-numerical-simulations [105, 267]. With this information, different collision kernels are proposed for estimating collision rate in atmospheric models where droplets are not resolved explicitly. However, Witte et al. [318] based on estimation from measurements suggests that these collision kernels underestimate the collision rate in large scale models due to the variability of the droplet distribution on scales smaller than their grid cells. Using local-volume measurements the small-scale variability in droplet distribution can be assessed [16, 168].

1.2.4.3 Aerosol effects on cloud droplet number density and moments: first order interest is aerosol concentration

Clouds are significant contributors in Earth’s radiation budget. A change in the boundary-layer cloud coverage can alter the radiative forcing at a similar order as greenhouse effect [280, 321]. Furthermore, atmospheric aerosols can directly alter cloud microphysical properties and, indirectly, cloud macroscopic properties. Cloud microphysical properties are modulated through aerosol indirect effects (first, second, and dispersion effects) [8, 175, 233, 303, 305]. With a decrease in aerosol, the mean size of cloud droplets increases, and the distribution becomes broader. These distribution changes could cause increased precipitation and a decrease in cloud liquid water content (at least in a relatively clean cloud) and, consequently, cause a change in the radiative fluxes. In marine stratocumulus clouds, the precipitation suppression enhances the latent heating and reduces the evaporative cooling. Thus, it alters the dynamics, turbulent kinetic energy, and entrainment rate. Moreover, cloud thickness positively correlates with aerosol concentration [233]. As a result of the above feedbacks, the entrainment rate may be enhanced, effectively forcing the system toward a thinner cloud. Although the cloud thickening increases the liquid water and moisture flux which facilitate buoyant production [321]. In addition, the cloud-top entrainment velocity (w_e) decreases with an increase in aerosol due to a change in droplet settling flux ($w_e \propto -d^5$) [67]. Therefore, there are considerable positive and negative feedbacks governed by aerosol perturbation through microphysics. The overall effect on the cloud system depends on whether these perturbations are amplified or dampened by dynamic forcing

and the aerosol regime (clean versus polluted). Finally, apart from micro-scale properties, the aerosol population can similarly influence mesoscale cloud structure (associated radiative fluxes) and the convection pattern. Indeed, the transformation of a cloud from a closed-cell structure to an open-cell structure is feasible merely due to the reduced aerosol concentration [310].

Most of the aerosol feedbacks were previously considered a result of changes in only the mean droplet size. However, Liu and Daum [175] pointed out that the droplet size relative dispersion changes with aerosol concentration, which influences the effective radius and cloud albedo. Yum and Hudson [344] offers an explanation for this behavior of relative dispersion based on the assumption of an adiabatic parcel, similar to the example presented in section 3.2 (Equation 1.4). For higher aerosol concentration in a cloud parcel, supersaturation decreases due to increased competition for water vapor, which decreases the narrowing effect as a result of inverse radius dependence of droplet radius growth. Consequently, droplet size relative dispersion shows a positive correlation with aerosol concentration. Although, as pointed by Yum and Hudson [344], a model based on this adiabatic parcel produces a lower relative dispersion than typically measured. Moreover, it can not explain the observations of decreasing or nearly a constant trend of relative dispersion with increase in aerosol concentration [10, 187, 189, 193, 194, 210, 294, 348].

In stratocumulus cloud, the mean vertical velocity is relatively low; thus, supersaturation fluctuations and microphysical variability could have a significant effect on aerosol-cloud

feedbacks. Recent lab experiments [46], field observations [276], and some numerical simulations [104, 225, 256] indicate that supersaturation fluctuations can be a crucial part of microphysical interactions. For example, turbulence induced broadening can facilitate precipitation formation by enhancing the droplet collision rate and can influence cloud radiative properties. Furthermore, supersaturation fluctuations might be a key element in the aerosol activation process, specifically, when the mean updraft velocity is low. Similar to the traditional aerosol-indirect-effects due to mean properties, the concentration of aerosols can also modulate these effects of supersaturation fluctuations. Theory and laboratory results suggest that with increase in aerosol concentration, supersaturation fluctuations and associated droplet distribution broadening rate tend to decrease. Therefore, radiative and precipitation fluxes would be influenced as a result of the turbulence-induced aerosol effects [48].

1.2.4.4 Entrainment and turbulence effects on cloud properties

Mixing between a cloud ‘parcel’ and the outside environment results in liquid water contents substantially lower than the quasi-adiabatic ideal. This departure is universally observed near cloud boundaries, such as at the top of stratocumulus clouds and the sides of cumulus clouds. This is particularly relevant for the stability and persistence of stratocumulus clouds that are so radiatively important to determining the climate [195, 205, 287, 307]. Assessing the entrainment rate in stratocumulus clouds remains a primary experimental challenge and major field projects have been developed to address the question [96, 311,

343]. Entrainment of environmental air also is crucial to determining the vertical extent and microphysical properties of cumulus clouds [117, 191]. Cloud microphysical properties respond to the nature of the mixing: the liquid water content is reduced, but because $L \propto n\overline{d^3}$, where n is the cloud droplet number density and d is the cloud droplet diameter, it is evident that a given reduction in L can be achieved with different combinations of n and L . For example, in the limit of homogeneous mixing, a cloud droplets exposed to sub-saturated air evaporate together, resulting in reductions of both n and $\overline{d^3}$; in the limit of extreme inhomogeneous mixing, entrained air is humidified through the complete evaporation of a subset of cloud droplets, with the remaining droplets remaining unaltered, ultimately resulting in a cloud with reduced n but constant $\overline{d^3}$ [38, 188]. Telford [297] offers an alternative hypothesis for a broad droplet size spectra due to entrainment in a turbulent cloud. They suggest that the multiple up and down cycling of entrained 'turbules' (localized eddies of similar properties) from the cloud top to base would lead to a broader droplet spectrum as well as the production of large droplets [296, 297].

The importance of cloud-top entrainment to stratocumulus clouds is illustrated in Figure 1.3. The figure depicts the calculated buoyancy of a mixture of cloud-top air with free-tropospheric air. The buoyancy is plotted versus the mass fraction χ of free-tropospheric air, and the value corresponding to complete evaporation of cloud liquid water and saturation of the resulting mixture is denoted by χ^* . Two different cases corresponding to stratocumulus clouds measured during the POST field campaign are shown to illustrate that in some conditions buoyancy reversal can occur (flight TO6). In other words, the fully-saturated mixture is more dense than either the cloud-top or free-tropospheric air, with the possibility

Figure 1.3: Buoyancy perturbation b of a mixture of air from stratocumulus cloud top with air from the free troposphere. The mass fraction of cloud air is given by $1 - \chi$. The lines plotted are for two flights from the POST field project (flights TO6 and TO14). The vertices denoted χ^* and b^* are the values resulting when all cloud liquid water has evaporated and the resulting mixture has a relative humidity of 100%. The conditions observed in flight TO6 result in negative b^* , indicating the possibility of buoyancy instability. Flight TO14 is an example of conditions under which buoyancy reversal is not expected. Finally, a third flight (TO12) resulted in $b^* = 0$, with the corresponding χ^* denoted by the $\times$. The figure is from Gerber et al. [95].

that this will lead to further entrainment and ultimately to destabilization of the cloud. The relevance of this cloud-top-mixing instability remains a focus of research [205].

3

1.3 A sample of new measurement capabilities and recent results from field studies and laboratory experiments

As discussed in the previous section, the size distribution of hydrometeors is one of the key property determines the interactions of a cloud with incoming solar radiation, precipitation

formation, transport of water vapor, dynamics of boundary-layer, and more. Therefore, investigation of the droplet size distribution (or their moments) using measurement provides some valuable input to the modeling framework. For example, droplet number concentration, second, and third moments of a droplet size distribution are essential for estimating radiative fluxes through a cloud. Similarly, droplet number concentration, third, and fifth moments of a droplet size distribution are important for autoconversion rate and cloud-top entrainment flux.

A next question would be, how do we measure droplet (or ice) size distributions and their moments, as well as other fine-scale or ‘fast’ cloud properties? In the next section we provide illustrative examples of recent advances and findings within the three broad areas of in-situ observation: airborne measurement, ground-based measurement, and laboratory measurements.

1.3.1 Airborne measurements of cloud properties

In situ field observations from airborne platforms such as aircraft, helicopters, drones, and balloons, provide a valuable opportunity to measure cloud microphysical properties in their natural setting. Clouds are typically far from the surface and are often transient in nature, and cloud properties vary widely over different portions of the globe due to thermodynamic and aerosol conditions. Airborne measurements are ideal for reaching cloud systems ranging from marine stratocumulus, to deep convection, to high-altitude cirrus. Instrumentation suitable for airborne deployment has advanced to a very sophisticated and specialized levels

and thorough reviews are available in the literature [313]. Airborne measurements typically provide much more detailed information than ground-based remote sensing, and in fact are necessary in assessing retrieval methods. Through airborne measurements, spatial variation of cloud microphysics, thermodynamic, and turbulent flow properties can be measured in cloud as well as properties of the air above and below cloud. The cloud sampling strategy using airborne instruments depends on the purpose of the study: for example, vertical profiles of cloud properties for cloud optical depth studies may require multiple up and down ‘porpoise dive’ patterns for the aircraft path. On the other hand, for cloud fraction or mesoscale cloud structure, a long, horizontal flight pattern may be required. Ever-present challenges of airborne measurements are the expense and complexity of operating an aircraft, and the high-speed nature of most measurement platforms. The emergence of un-crewed aerial vehicles (drones) for scientific purposes has the potential to significantly alter the type, frequency, duration, and range of in situ measurements available to the community [66].

Airborne instrumentation for measuring hydrometeor size distributions and moments (such as liquid water content) are mainly of the following categories: a) Hotwire b) Forward scattering c) Phase Doppler d) Shadowgraph e) Imaging f) Holographic. More detailed classifications of these instrumentations and descriptions of each type can be found in Baumgardner et al. [14] and Brenguier et al. [33]. Table 1.1 lists some of the latest instruments with their specifications Baumgardner et al. [14].

Here, the first four instruments in the table are only suitable for cloud droplets, not the ice

Airborne Instruments for Cloud Particle Measurement			
Instrument Name	Working Principle	Size Range	Sampling Rate
Hot-Wire Liquid Water Sensor (LWC-300)	Resistance Change	L: 0.05-3 g m ⁻³	4 l s ⁻¹
CAS	Forward Scattering	D: 0.5-50 μ m	25 cm ³ s ⁻¹
Fast-CDP	Forward Scattering	D: 1.5-50 μ m	25 cm ³ s ⁻¹
Fast-FSSP	Forward Scattering	D: 1.5-50 μ m	40 cm ³ s ⁻¹
Small Ice Detector (SID-2)	Scattering	D: 2-140 μ m	100 cm ³ s ⁻¹
Artium Flight PDI	Doppler Effect	D: 0.5-2500 μ m	$\approx$ 100 cm ³ s ⁻¹
2D-C	Shadowgraph	D: 25-800 μ m	5 l s ⁻¹
2D-S	Shadowgraph	D: 10-1260 μ m	8 l s ⁻¹
CIP	Imaging	D: 25-1550 μ m	16 l s ⁻¹
CPI	Imaging	D > 3 μ m	400 cm ³ s ⁻¹
Holodec / Halo-Holo	Holography	D > 6 μ m	Instantaneous sample volume $\approx$ 20 cm ³

Table 1.1

List of airborne instruments for measuring hydrometeor size distribution and moments. The volume sampling rates correspond to a flight speed of 100 m s⁻¹. The table is adapted from Baumgardner et al. [14].

phase. The SID-2, 2D-S, CIP, CPI and holographic instruments can be used for both liquid and ice phases. Hotwire instruments are based on the change in resistance of a hot wire junction due to the evaporation of droplets, and it provides an estimate of the liquid water content and therefore the third moment of the droplet size distribution. The second class of instruments operate by collecting light scattered by spherical droplets in the near-forward direction. They provide individual, drop-by-drop measurement of the diameter within a relatively small instrument cross section that is swept through the cloud the the aircraft. Examples of instruments in this category are the Forward Scattering Spectrometer Probe (FSSP), Cloud Droplet Probe (CDP), and Cloud-Aerosol Spectrometer (CAS). Since the

forward scattering probes are based on the intensity of scattered light, care must be taken to maintain clean optics and regular calibration.

Phase-Doppler-Interferometry (PDI) is based on detection of phase difference rather than intensity, and thereby avoids some of the challenges of forward-scattering instruments. The phase difference between scattered Doppler bursts measured at two angles from a moving droplet is directly proportional to the diameter of that droplet. The coefficient of proportionality is a function of geometrical properties and wavelength of the source light. As with forward-scattering instruments, phase-Doppler instruments have a relatively small sample cross section that is swept through the cloud.

The other class of instruments provides volumetric or at least two-dimensional information about the particle distribution and size. The Shadowgraph system (e.g., 2D Stereo Probe, 2D-S) records the shadow of an illuminated particle in the focal plane of a detector. Similarly, there are other imaging probes which capture two-dimensional images of particles illuminated by light sources (e.g., Cloud Particle Imager (CPI) and Cloud Imaging Probe (CIP)). Holographic instruments have the unique capability of capturing the three-dimensional information about particle position, as well as particle size. These instruments capture the interference pattern created between a reference wave and the light scattered from a dilute population of particles within the measurement volume. Reconstruction of the hologram is performed digitally in modern systems. These instruments provided detailed information on cloud microphysical properties, although, they have a limitation of high computation cost and lower time resolution.

Example: Cloud droplet size distribution moments. As discussed in the previous section, the shape of the cloud droplet size distribution is relevant to radiative properties of clouds. As an example of airborne measurement with a focus on assessing the shape of droplet size distributions in stratocumulus clouds, we refer to the extensive compilation of Miles et al. [210]. They show moments of the droplet size distribution at different altitude and for different aerosol concentrations. Among the other significant aspects, they reported that the cloud droplet distribution gets broader with height contrary to predictions from parcel models.

Microphysical properties of stratocumulus clouds, comparison to satellite-derived properties. Glienke et al. [97], Witte et al. [319] have recently demonstrated the importance of measuring the full droplet size distribution, from cloud droplet to precipitation diameters, for calculating cloud radiative properties. By carefully measuring the full size distribution with phase-Doppler interferometry [319] and digital holography [97], it was shown that effective radius values retrieved from satellite measurements do now show the strong discrepancies previously indicated. Example size distributions from the CSET field project are shown in Figure 1.4. The three panels illustrate how the size distribution responds to different relevant powers of droplet diameter: the top shows a number distribution, the middle shows a surface area (d^2) distribution, and the bottom shows a volume (d^3) distribution. The shaded region shows the range of droplet diameters that are in the ‘gap’ between cloud droplet and drizzle sizes, with the corresponding contributions to total number, surface area, and volume. Some of the most common aircraft instruments for measuring cloud droplet size distributions miss this gap region, and therefore miss significant contributions

Figure 1.4: Cloud droplet size distributions, weighted by number concentration, by surface area, and by volume. HOLODEC data span the gap between the CDP and 2DC measurement ranges. Intriguingly, no gap is observed between cloud droplet and drizzle sizes, as is typically expected from parcel model calculations. The shaded region indicates this ‘autoconversion gap’ and the numbers in that region indicate the number, surface area, and volume contributed by hydrometeors in this ‘drizzlet’ size range. The figure is from Glienke et al. [97].

to radiative properties.

4

Example: Digital holographic measurements of cloud microphysics. The HOLODEC instrument has a novel sampling strategy based on digital in-line holography,

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Figure 1.5: Mixing diagrams generated from HOLODEC data obtained in small, liquid-phase cumulus clouds. Each dot represents the mean-volume diameter and the cloud droplet concentration estimated from a single HOLODEC sample volume. The dashed curve represents homogeneous mixing, and the solid blue line represents extremely inhomogeneous mixing. The figure is from Beals et al. [16].

which provides an estimate of the cloud particle size distribution from a single sample volume. This is in contrast to the typical approach of counting droplets one at a time along a thin line through the cloud. Each instantaneous sample volume is approximately 20 cm^3 , repeated at 3.3 Hz; the time-averaged volume sampling rate is therefore comparable to other instruments (e.g., the CDP), but the local sampling approach means assumptions of spatial homogeneity and ergodicity required when averaging can be relaxed. The ‘local volume’ configuration allows for investigation of the influence of averaging and the variability of local-scale size distributions [16, 74]. The ability to look at three-dimensional locations of particles within the sample volume allows for the investigation of droplet clustering and particle breakup [90, 168].

Data from HOLODEC has helped clarify whether entrainment and mixing leads to uniform evaporation of all droplets (homogeneous mixing) or total evaporation of a subset of droplets (inhomogeneous mixing). This is relevant to determining microphysical properties at cloud boundaries and is therefore important for both drizzle formation and cloud radiative properties. For example, Figure 1.5 shows mixing diagrams acquired with HOLODEC data from liquid-phase cumulus clouds, and the scatter of points along a horizontal line are indicative of extremely inhomogeneous mixing [16]. Each point corresponds to a single hologram, so the spatial averaging that was previously identified as an ambiguity within mixing diagrams [38], was bypassed.

1.3.2 Ground-based field measurements

The third category of In-situ measurements is the ground-based measurements at high altitude sites, for example, Storm Peak Laboratory at Colorado, USA [111], Schneefernerhaus Environmental Research Station at mount Zugspitze, Germany [250], Pico Mountain Observatory in Portugal [56], and the High Altitude Cloud Physics Laboratory at Mahabaleshwar, India [171]. Ground-based stations have advantages of cost-effectiveness, long term statistics, more detailed measurements over the field campaign using aircraft.

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1.3.3 Laboratory methods and experiments

Controlled laboratory experiments are a unique category of experimental cloud investigation, in the sense that specific mechanisms can be explored under idealized conditions. The scale of laboratory experiments ranges from bench-top investigations of single-particle processes, all the way to large-scale facilities for studying full clouds. Some of the currently existed facilities include the Aerosol Interaction and Dynamics in the Atmosphere (AIDA) chamber, the Manchester Ice Cloud Chamber (MICC), the Meteorological Research Institute (MRI) cloud chamber, the Leipzig Aerosol Cloud Interaction Simulator (LACIS), and the Pi Chamber. Two examples of cloud processes that have been investigated in these facilities are described below.

6

Example: Laboratory investigation of turbulence-microphysics interactions.

The Pi Chamber can create conditions for cloud formation using two different principles: turbulent moist convection (Rayleigh-Bénard between differentially-heated, water-saturated surfaces) and adiabatic expansion [50]. The most commonly used mode, turbulent convection, is similar to the convection in atmospheric boundary-layer where the buoyancy different due to heating at the bottom and cooling at the top drives the convective mixing and turbulence. The turbulent kinetic energy dissipation rate generated through this process is

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Figure 1.6: Steady-state moist and cloudy conditions: Measurements of vertical velocity fluctuations (w), kinetic energy dissipation rate (ϵ), temperature (T), water vapor density (ρ_v), supersaturation (ss) during moist convection near the center of the chamber. The vertical black line indicates the point where aerosol injection started, and the last panel shows the formation of cloud liquid water content (LWC) after that point. The figure is from Niedermeier et al. [218].

around $10^{-3} \text{ m}^2 \text{ s}^{-3}$, which is similar to observed values in stratocumulus clouds. However, the Reynolds number is lower, $\text{Re}_\lambda \approx 55$, due to the much smaller length scales. To create a supersaturated environment, the top and bottom boundaries are kept saturated with water at different temperatures. The resultant supersaturation fluctuations are around similar magnitude as some of the reported fluctuation statistics from atmospheric clouds, with standard deviation $\approx 1 \%$, [46, 276]. Moreover, the mean supersaturation for a typical chamber configuration is $< 1\%$. The achievable temperature and pressure range in this facility is -50 to $50 \text{ }^\circ\text{C}$ and 60 hPa to 1000 hPa [50]. Therefore, in this chamber, warm as well as mixed-phase cloud conditions can be created using different cloud condensation and

ice nuclei. Long, steady cloud conditions (timescale of days) can be achieved by maintaining a balance between the steady injection of aerosol particles and the removal of cloud droplets due to settling. It is also possible to perform transient cloud experiments [47].

An example of steady-state moist and cloudy convection in the Pi-chamber is given in Figure 1.6. It shows steady-state thermodynamic properties, turbulent flow, and cloud properties during moist condition, and the transition to cloudy conditions when aerosol injection is started at approximately 9 hours.

1.4 Research Problems:

The laboratory is a powerful venue for scientific discovery because it allows processes to be studied in isolation, to be distilled to their most essential elements so that simple models can be developed. Therefore, we utilized these advantages of laboratory experiments, in combination with a theoretical approach based on the stochastic method, to explore the following problems:

- † Can we characterize the supersaturation fluctuation in turbulent convection?
- † What is the role of turbulence induced supersaturation fluctuations in cloud droplet growth?
- † What are the implications of this turbulence induced droplet growth in precipitation formation and cloud optical properties?

- † How does the presence of aerosol modulate these effects?
- † How is the traditional picture of aerosol indirect effect modified due to the additional influence of supersaturation fluctuation apart from the mean?
- † How are the aerosol population and cloud microphysics coupled during a transient cloud decay (or the cloud-cleansing process)?
- † What is the functional form of the droplet size distribution in non-precipitating turbulent clouds?

Chapter 2

Supersaturation fluctuations in moist turbulent Rayleigh-Bénard convection: a two-scalar transport problem

This chapter is about the numerical study of moist turbulent Rayleigh-Bénard convection and supersaturation fluctuations. It is based on a collaborative research and under review for publication in *Journal of Fluid Mechanics*.

Abstract

Moist Rayleigh-Bénard convection with water saturated boundaries is explored using a one-dimensional-turbulence model. The system involves both temperature T and water-vapor pressure e_v as driving scalars. The emphasis of the work is on a supersaturation s , a nonlinear combination of T and e_v that is crucial to cloud formation. Its mean as well as fluctuation statistics determine cloud droplet growth and therefore precipitation formation and cloud optical properties. To explore the role of relative scalar diffusivities for temperature (D_t) and water vapor (D_v), three different regimes are considered: $D_v > D_t$, $D_v \approx D_t$, and $D_v < D_t$.

Scalar fluxes (Nusselt number: Nu and Sherwood number: Sh) and their scalings with moist Rayleigh number Ra_{moist} are consistent with previous studies of one-component convection. Moreover, variances of the scalars in the bulk region increase with their diffusivities and also reasonably follow derived scaling expressions. Eulerian properties plotted in (T, e_v) coordinates have a different slope compared to an idealized mixing process. Additionally, the scalars are highly correlated, even in the cases of high relative diffusivities (factor of four) D_v and D_t . Based on the above fact and scaling relation of the scalars, the supersaturation variance is found to vary approximately as $Ra_{moist}^{5/3}$, in agreement with numerical results. Finally, the supersaturation profile in the boundary-layer is explored and compares well with scalar boundary-layer models. A sharp peak appears in the boundary-layer-supersaturation profile, not only in the variance but also in the mean profile, due to relative diffusivities of

the scalars.

2.1 Introduction

Buoyancy-driven atmospheric boundary layers capped by strong stable layers can be understood as a large-scale manifestation of turbulent Rayleigh-Bénard convection [37, 68, 329]. The presence of water vapor and its associated phase changes plays a central role in the nature of the convection [34, 35, 207, 226, 227, 288]. The general problem of moist convection, fully coupled with surface fluxes, large-scale dynamics, radiative transfer, and cloud formation remains a grand challenge, with relevance to weather forecasting and climate. In this work, we set as a more modest goal the investigation of how water-vapor supersaturation behaves in traditional Rayleigh-Bénard convection.

2.1.1 Moist convection parameters

The cloud topped convective-boundary-layer is effectively a Rayleigh-Bénard system driven by buoyancy forcing, e.g., radiative forcing from cloud top [173, 205, 212, 288]. In this system, continuous plume eruption from both boundaries transports heat and moisture and produces the mixed layer [85]. The cloud aspect of the problem depends on the presence of water vapor, making this a case of two-scalar Rayleigh-Bénard convection. Similar to a single-component convection, the equivalent Rayleigh-number (or the moist Rayleigh-number for moist convection) determines the marginal stability of the convective system.

For moist convection, it can be approximated as [55, 216]:

$$Ra_{moist} \approx \frac{g\Delta TH^3}{T_0\nu D_t} + \frac{g\epsilon\Delta q_v H^3}{\nu D_t}. \quad (2.1)$$

Here, ΔT , Δq_v and H are the applied temperature difference, the difference in the water vapor mixing ratio, and length scale of the system, respectively. Other constants g , ν , and T_o are the gravitational acceleration, the kinematic viscosity, and the mean temperature (reference temperature). The parameter $\epsilon = M_a/M_w - 1$, where M_a and M_w are the molecular masses of dry air and water, respectively. The above relation is obtained by taking a ratio of buoyancy ($\sim \rho g(\Delta T/T_0 + \epsilon\Delta q_v)$) and viscous force ($\sim \nu \rho w^*/H^2$) scales and assuming that the destruction of density perturbation is larger because of the thermal diffusion than the diffusion of the other scalar (i.e., using $w^* \sim D_t/H$ as a velocity scale in the viscous forcing). Additionally, the velocity scale (w^*) is obtained from the flux balance between advection ($\sim w^* \delta T/H$) and diffusion ($\sim D_t \delta T/H^2$) of scalar perturbation from the base state. The scaling is therefore only valid when the thermal gradient is the largest contributor to buoyancy. In the contrasting case where the second scalar contributes significantly, the diffusivity of the dominant scalar should be used. In the moist convection case, water vapor's contribution to the buoyancy is smaller than the thermal contribution (at least for the temperature range considered here). In the subsequent definition of the Rayleigh number, the underlying assumption is that the thermal gradient is a larger contributor to the buoyancy perturbation than the gradient of the other scalar. However, the contribution of the other scalar in buoyancy is not ignored. Other relevant non-dimensional

variables associated with the fluid properties are the relative diffusivities of water vapor and thermal field, the Schmidt number ($Sc = \nu/D_v$) and Prandtl number ($Pr = \nu/D_t$). These quantify the rate at which the molecular diffusion of scalars destroys the buoyancy forcing. A non-dimensional quantity related to the geometry of a system, the aspect ratio of a convection cell, also influences the flow to some extent.

There is a similarity between double-diffusive convection [142, 262] and the current case of moist convection. In both cases, two scalars involved in the problem which are advecting and diffusing. However, in the traditional double-diffusive convection, scalars usually have opposite contributions to the buoyancy and/or have opposite diffusive fluxes. In the current case of moist convection, both scalars contribute to positive buoyancy and diffusing in the same direction; therefore, it is closer to traditional R-B convection. Indeed, the current case can be expressed as single component R-B convection driven by a difference in virtual temperature. The novel aspect considered here is to explicitly consider the diffusivity difference between the two scalars, as expressed through the Lewis number $Le \equiv D_t/D_v$. The relevance of Lewis number and differential diffusivity is motivated by the water vapor supersaturation, described next.

2.1.2 Supersaturation from two scalar fields

Supersaturation is central to cloud formation, whether it be in earth's atmosphere [27] or in the atmospheres of other planets and stars [40, 161, 198]. Taking water vapor as an example, supersaturation depends upon the two scalar fields temperature and water vapor

Figure 2.1: A thermodynamic vapor-pressure-temperature diagram illustrating the formation of supersaturation through isobaric mixing. An idealized mixing process of two saturated air parcels at different temperature, T_h and T_c (dashed line) and its comparison with the saturation (equilibrium) vapor pressure curve (blue line).

mixing ratio through the water vapor pressure: $s \equiv (e_v/e_{v,s}(T)) - 1$. Here, e_v and $e_{v,s}$ are the water-vapor pressure and saturation (or equilibrium) vapor pressure, respectively. The latter is expressed by the Clausius-Clapeyron equation: $e_{v,s}(T) \propto \exp(-\mathcal{L}/R_v T)$, where $\mathcal{L}$ is the latent heat of vaporization, R_v is the mass-based gas constant for water vapor, and T is the temperature [27, 167].

In turbulent R-B convection, scalar fields are advected and diffused, resulting in their mutual mixing. An idealized view of the mixing of two saturated air parcels is shown in Fig. 2.1, which depicts a thermodynamic space consisting of temperature and water vapor pressure. The equilibrium (Clausius-Clapeyron) curve is shown, and the mixing is assumed to take place between two air parcels with temperatures T_h and T_c with a constant heat capacity.

For isobaric mixing, various mass ratios result in the dashed line [27]. The red dashed lines show a specific example of a fully-mixed parcel with temperature T_{mix} and water vapor pressure $e_{v,mix}$. It can be seen that $e_{v,mix} > e_{v,s}(T)$, resulting in a positive supersaturation in the mixture.

The idealization of Fig. 2.1 can be extended, taking into account the details of how temperature and water vapor fields are mixed in a turbulent flow. The relative humidity ($RH \equiv e_v/e_{v,s}(T)$) problem has been the subject of considerable study in the boundary layer meteorology community, mainly focused on correlations between the two fields [e.g., 282]. The concept also underlies a common approach to representation of sub-grid-scale variability of cloudiness. For example, Sommeria and Deardorff [281] used the joint probability distribution function (PDF) of two moist-adiabatically-conserved scalars to diagnose sub-grid-scale cloud fraction, liquid water content, and buoyancy flux in a large-eddy simulation of trade-wind cumulus clouds. Relative humidity variability is due to joint variability in the two conserved scalars and cloud is assumed to exist when relative humidity $RH \geq 1$. This approach has been extended by several others since then (e.g., Golaz et al. [98], Bogenschutz and Krueger [26] and references therein), in what have come to be known as PDF methods. Further discussion of these approaches is in Section 2.4.

2.1.3 An approach for studying scalar fields in moist Rayleigh-Bénard convection

This work is motivated by the problem of supersaturation in turbulent Rayleigh-Bénard convection within an idealized context, i.e., without the complications of large-scale shear, radiative transfer, and condensate formation that exist in the atmosphere. The work's origins are in questions that arose during the analysis and interpretation of observations taken in the Pi Chamber, a laboratory chamber for studying cloud formation in turbulent R-B convection [216]. In particular, the competing and complementary roles of diffusion versus advection are of interest, as is the relevance of differences in diffusivities for heat and for water vapor. For example, the seemingly negligible difference in diffusivities (Lewis number $Le \equiv D_t/D_v = 0.84$ at 20°C) has been used in other laboratory contexts as the underlying mechanism for precise control of supersaturation [291].

We approach the problem of supersaturation in R-B convection using an idealized, one-dimensional model that faithfully represents the processes of advection and diffusion in turbulent flow. Using this kind of model allows a wide range of parameter space (e.g., Ra) to be explored with computational efficiency and it is well suited to problems that are fundamentally dependent on advection and diffusion of scalars. It represents all relevant scales, so it captures the diffusive boundary layer as well as the larger-scale turbulent mixing in the bulk fluid. We consider this as a first exploration of the phenomena at work, and more detailed approaches, such as using direct numerical simulation, will follow. Finally,

the one-dimensional model is highly scalable, so it can be run at scales comparable to laboratory experiments with turbulent moist convection [216] and then can be scaled up for moist convection in the atmospheric boundary layer. The purpose here is to investigate the mean and fluctuating properties of the supersaturation field in turbulent moist Rayleigh-Bénard convection, in a general sense. Detailed comparison to laboratory experiments and exploration of the atmospheric context are challenges for future work.

The paper is organized as follows: In Section 2.2 the One Dimensional Turbulence (ODT) model is described and the parameters used in this study are defined. We then proceed to use the model to explore the scaling of heat and water-vapor fluxes with relevant dimensionless variables (Sec. 2.3.1); fluctuations of scalars in the bulk fluid (Sec. 2.3.2); fluctuations of supersaturation in the bulk fluid (Sec. 2.3.4); vertical profiles of supersaturation in the boundary layer and the bulk fluid (Sec. 2.3.5); and the sampling within the thermodynamic space of water-vapor pressure versus temperature (Sec. 2.3.3). Finally, we conclude the paper by summarizing and discussing the results in Sec. 2.4.

2.2 Model description and problem setup

The One Dimensional Turbulence (ODT) model was developed to simulate turbulent flows which are statistically homogeneous and isotropic in two dimensions but varying in the third [144]. ODT evolved from the Linear Eddy model [144], which has been previously used to simulate cloud turbulence [163, 164]. ODT has been widely used for turbulence

problems ranging from combustion [84] to LES sub-grid-scale modeling [284]. Subsequent ODT development led to a version of the model specifically designed for problems involving buoyancy, either stably stratified [324] or convective [100, 325]. All versions of ODT rely on the same basic construct. The flow is simulated on a line which is oriented along the direction in which mean flow properties vary, so for buoyant convection ODT represents the vertical coordinate. All fluid properties are functions of location along the line. Turbulent advection occurs through a series of random mappings of the ODT line onto itself. Mappings occur on all relevant length scales, and their statistics reproduce standard scaling relations for turbulence. During the intervals between mapping events, molecular diffusion is modelled using standard partial differential equations. ODT results are typically interpreted statistically e.g., by computing the mean and variance of each flow variable at each position along the line. Due to the restriction to one spatial-dimension, it is computationally feasible to fully resolve the Kolmogorov scale. In the following paragraphs we provide an overview of the ODT model, and readers are referred to the ODT references for details [144, 324, 325].

To simulate moist convection using ODT, a variant of the version employed in Wunsch and Kerstein [324, 325] is employed here. The ODT line is oriented along the z (vertical) axis. The flow variables are the magnitude of the velocity vector (w), temperature (T), and water vapor mixing ratio q_v . During the intervals between mappings, each of these variables (X) obeys

$$(\partial_t - D_X \partial_z^2) X = 0. \quad (2.2)$$

The boundary condition for the current problem is no slip for velocity and water-vapor-saturated bottom and top boundaries at a fixed temperature (hot bottom and colder top). Note that the advection terms are absent from Eq. 2.2, and that the velocity variable (w) do not actually move the fluid elements; in ODT the random mappings do that instead. Here D_X represents molecular diffusivity of variable X . These equations are solved at full resolution (including all scales down to the viscous and/or diffusive scales). Full resolution of all length and time scales in turbulent flow is one of the key advantages of ODT simulations, and it is made computationally affordable by the restriction to one dimension. The fluid density variations are related to temperature and water vapor content by

$$\rho(q_v, T) = \rho_o \frac{T_o(1 + \epsilon q_{v_o})}{T(1 + \epsilon q_v)}. \quad (2.3)$$

Where ρ_o and q_{v_o} are the reference density and water vapor mixing-ratio.

To simulate moist convection in the laboratory Pi Chamber with the ODT model, the velocity boundary conditions replicate no-slip walls at $z = 0$ and $z = H$ and fixed boundary condition for temperature and water vapor are applied. A temperature difference, ΔT is imposed across the convective cell and boundaries are assumed to be saturated with water at the temperature of each boundary. Therefore, there is also an imposed difference for

water vapor (Δq_v) across the convective cell.

The physics of turbulent mixing is implemented in ODT through randomly selected mappings ($M(z)$) of the line onto itself. The mapping is applied to all scalar fields simultaneously. It mimics the motion of a ‘fluid element’ and preserves the properties of each fluid element. The attributes of the mapping function have a strong impact on the model dynamics, so selection of an appropriate function is essential to constructing a realistic representation of turbulent advection within the one-dimensional framework. The mapping used in ODT is measure preserving. This is the one-dimensional equivalent of incompressibility, and implies that all statistical moments of scalar fields are preserved by the mapping. The mapping is also a continuous function, to avoid introducing discontinuities in mapped scalar fields. Finally, the function maps a finite region, to mimic a turbulent ‘eddy’ of defined length. The specific function $M(z)$ used here was introduced by Kerstein [144] and is common to all versions of the ODT model [324]:

$$M(z) \equiv z_o + \begin{cases} 3(z - z_o) & \text{if } z_0 \leq z \leq z_o + L/3 \\ 2L - 3(z - z_o) & \text{if } z_o + L/3 \leq z \leq z_o + 2L/3 \\ 3(z - z_o) - 2L & \text{if } z_o + 2L/3 \leq z \leq z_o + L \\ z - z_o & \text{else.} \end{cases} \quad (2.4)$$

Where z_o is the location of the mapping event on a segment of length L . Essentially, this

function takes the portion of the original scalar profile $T(z)$ between z_o and $z_o + L$, compresses it to $1/3$ of its original size, and places 3 copies of this compressed profile within the original domain. The middle copy is reversed in order to maintain continuity. This mapping function has two additional attributes which are valuable for representing turbulent flows. The compression i.e., mixing of scalar profiles leads to a cascade of turbulent kinetic energy or scalar variance from large to small scales, as illustrated in Kerstein [144] and Wunsch and Kerstein [324]. Also, the mapping results in a net scalar transport along the line when applied to a scalar field with a mean gradient. In the present context, this means that mappings will transport heat and water vapor vertically through the laboratory convection cell.

Since mappings do not change any statistical moment of a scalar field, they conserve kinetic energy. However, if vertical transport of scalars T or q_v results in a change in potential energy, then energy conservation requires potential energy changes to be offset by corresponding changes in kinetic energy (conversion of potential to kinetic energy). The change in kinetic energy requires an additional operation on the velocity scalar, in addition to the applied mapping. The complete set of mapping and energy conserving operations applied to each scalar during a mapping event are given by:

$$X(z) \rightarrow X(M(z)) \tag{2.5a}$$

$$w(z) \rightarrow w(M(z)) + c_w(z - M(z)), \tag{2.5b}$$

where $(z - M(z))$ is the function added to the velocity scalar as part of the mapping process, X represents scalars T and q_v , and c_w is a amplitude selected to enforce energy conservation. Energy conservation determines the constant c_w :

$$c_w = \frac{27}{4L} \left[-w_k \pm \sqrt{w_k^2 - \frac{8gL\rho_k}{27\rho_o}} \right], \quad (2.6)$$

where w_k and ρ_k are defined as:

$$w_k \equiv \frac{4}{9L^2} \int_{z_o}^{z_o+L} w(z)(L - 2(z - z_o))dz \quad (2.7a)$$

$$\rho_k \equiv \frac{-1}{L^2} \int_{z_o}^{z_o+L} (\rho(M(z)) - \rho(z))zdz. \quad (2.7b)$$

The quantity w_k is similar to a wavelet transformation, and w_k can be interpreted as a filtered kinetic energy within the mapped region. Similarly, ρ_k (the mapped density difference) is proportional to the potential energy change. If ρ_k is positive (the mapping increases the potential energy of the fluid), then there must be sufficient filtered kinetic energy in the w_k^2 terms to compensate for this loss of energy; otherwise the amplitudes will be imaginary and the corresponding mapping cannot occur.

The remaining aspect of the turbulence model is to determine the rate at which mappings

occur. Mappings are selected randomly, but the probability of each one is based on standard turbulence scaling relations. The model is therefore not deterministic, and calculations rely on a large ensemble of eddy mappings to produce meaningful average results. Each eddy mapping, defined by its position z_o and length L , is assigned a rate $\lambda(z_o, L)$. Similar to Wunsch and Kerstein [325], the overall rate expression for a mapping of size L at location z_o is

$$\lambda(z_o, L) \equiv \frac{C\nu}{L^4} \sqrt{\left(\frac{w_k L}{\nu}\right)^2 - \frac{8gL^3\rho_k}{27\nu^2\rho_o} - Z}, \quad (2.8)$$

where, C and Z are dimensionless parameters of the turbulence model. Equation 2.8 has been normalized so that filtered kinetic energy term is expressed as an ‘eddy Reynolds number’ ($u_k L/\nu$) and the potential energy term is essentially an ‘eddy Rayleigh number’ ($gL^3\rho_k/\nu^2\rho_o$). The constant Z in Eq. 2.8 represents a small-scale viscous cutoff, or a minimum eddy Reynolds number for a non-zero eddy rate. It determines the smallest eddy mapping that can occur in the model. The constant C sets an overall time scale for all eddy mappings (but has no impact on the relative rates of different mappings). Note that the sign of the potential energy transferred by each eddy can be either positive (convection) or negative (stable stratification).

The eddy rate coefficient (C^2) and viscous cutoff (ZC^2) are set to 1.5×10^3 and 10^5 , respectively, based on the previous study by Wunsch and Kerstein [325]. As in that study, the effect of reasonable changes in the model parameters is tested and results are found to

be consistent.

Equation 2.8 provides the rate for each eddy mapping (defined by its size L and location z_o), given profiles of the velocity components and density (determined from the temperature and water-vapor profiles according to the equation of state). Mappings are randomly selected for implementation, but the selection probabilities are weighted according to the rate λ . The algorithm for doing this in the ODT model is described in detail by Kerstein [144]. Because the rate of mapping events is based on standard physical scalings, the ODT model reproduces the Kolmogorov $-5/3$ power spectrum [144, 324], the viscous ‘law of the wall’ [144], as well as many of the observed properties of Rayleigh-Bénard convection [325]. It is well known that the Nusselt number for a given Rayleigh number also depends to some extent on the aspect ratio of the cell. This effect is neglected in a one-dimensional model. Hence, while the ODT model can be used for parametric studies and scaling relations (as in past studies as well as here), it cannot capture the dependence of Nusselt number on changing geometry. Instead, the model parameters C and Z have been adjusted to produce reasonable quantitative agreement for $Nu(Ra)$ for cylindrical Rayleigh-Bénard cells with aspect ratios of 0.5 to 1.0. Details of the numerical simulations used to determine the model parameter values used here can be found in Wunsch and Kerstein [325]. That work includes tests of the ODT model for Rayleigh-Bénard convection for different parameter values and quantitative assessment of accuracy using comparisons with experimental data. It has been also used and tested carefully for the problem of double-diffusive convection where two scalars involved similar to the current case (e.g. Gonzalez-Juez et al. [100]). Table 2.1 describes several examples of the comparison between the scaling results from

Scaling Law	Experiment	ODT
R-B convection: $Nu(Ra)$	Niemela et al. [220]: $Nu = 0.124 Ra^{0.309}$	Within 5% of experimental values [325]
R-B convection: $\sigma_T \Delta T^{-1}(Ra)$	Niemela et al. [220]: $\sigma_T \Delta T^{-1} = 0.37 Ra^{-0.145}$	Within 10% of experimental values [325]
R-B convection: $Re(Ra)$	Xia et al. [331]: $Re Pr = 3.2 Pr^{0.18} (Ra Pr Nu)^{1/3}$	$Re Pr \sim (Ra Pr (Nu - 1))^{1/3}$ [325]
Double-diffusive convection: $Nu(Ra)$	$Nu \sim Ra^{0.37 \pm 0.1}$ Kelley et al. [142]	$Nu \sim (Ra/R_\rho)^{0.37 \pm 0.03}$ [100]

Table 2.1

Comparison of scaling results from experimental studies and ODT simulations. Results shown are for Nusselt number versus Ra , $\sigma_T/\Delta T$ versus Ra , and Reynolds number versus Ra for single-component convection, and Nusselt number versus Ra for double-diffusive convection. Here Re and R_ρ are the Reynolds number based on velocity fluctuations and buoyancy ratio of two scalar components, respectively.

ODT studies and relevant experimental measurements. More details about the statistical comparison between ODT results and experimental observation for different cases can be explored from the references cited above.

For this study, simulations with ΔT of 6, 8, 12, 14, and 20 K and a mean of 283 K are run. All simulations have saturated bottom and top boundaries. The resulting range of Rayleigh number is $2.05 \times 10^8 \leq Ra \leq 2.75 \times 10^9$. These values are similar to values that have been explored in laboratory experiments [46, 216], and they correspond to Ra sufficiently large that the flows are fully turbulent. Moreover, the Prandtl and Schmidt number ranges used in the current simulations are $0.18 \leq Pr \leq 0.72$ and $0.15 \leq Sc \leq 0.72$.

2.3 Results and discussion

Supersaturation depends on water vapor concentration and temperature fields, so we anticipate that the magnitudes of the diffusivities for the two scalar fields will be relevant to the problem of supersaturation advection and dissipation. In order to understand the relative roles of the two diffusivities, we analyze the following four combinations throughout the rest of the paper.

† Actual D_v and D_t : actual diffusivities for the water vapor and thermal fields, for air at a temperature of 283 K ($D_v/D_t = 1.16$).

† $D_v = D_t$: the water vapor diffusivity is same as the actual thermal diffusivity ($D_v/D_t = 1$).

† $4 \times D_v$: the water vapor diffusivity is four times the actual value, and the thermal diffusivity is same as the actual value ($D_v/D_t = 4.63$).

† $4 \times D_t$: the water vapor diffusivity is same as the actual value, and the thermal diffusivity is four times the actual value ($D_v/D_t = 0.29$).

Figure 2.2(a)-(b) illustrates the vertical-profile for a single scalar field, as simulated by ODT. Specifically, it displays the mean (a) and standard deviation (b) of water vapor mixing-ratio (q_v) versus height. In the plots and rest of the text, the normalized water vapor mixing-ratio and temperature variables are defined as: $q_v^* = (q_v - q_{v_t})/\Delta q_v$ and

Figure 2.2: Water vapor mixing-ratio mean and standard deviation profile: **(a)** vertical profiles of mean water vapor mixing-ratio, **(b)** profiles of mixing-ratio standard deviation for the different diffusivity cases, **(c - d)** semi-logarithmic plot of the scalar mean and standard deviation profiles (for the actual diffusivity case) in the half of the domain towards cold boundary, and the inset figures are the same plots with logarithmic axis and power-law ($\propto (1 - Z^*)^\gamma$) fittings of standard deviation. Here, $Z^* = 0$ corresponds to the hot and 1 corresponds to the cold boundary. The dotted, dashed and dash-dot curves represent power-law expressions obtained from fitting the standard deviation profile outside the boundary-layer at different regions.

$T^* = (T - T_t)/\Delta T$. The subscripts t and b represent properties at the top and bottom boundaries of the domain. These profiles are consistent with results from experimental and fully-resolved computational studies of R-B convection [6, 21, 36, 83, 298]. Specifically, the mean scalar profile shows strong vertical gradients within the boundary layers and a relatively flat bulk profile, and the standard-deviation profile shows peak variability in the boundary layers. When water-vapor diffusivity is increased (the $4 \times D_v$ case), the boundary layers are observed to be thicker and the standard deviation for q_v is increased. These results are expected, and demonstrate the ability of ODT to capture the essential physics of the problem, i.e., the competing roles of advection and diffusion in determining scalar fluxes as a function of height. They are consistent with the findings of Wunsch and Kerstein [325], who first described the ability of ODT to quantitatively represent turbulent R-B convection. Because it is a one-dimensional model, ODT does not capture processes like the formation of a large-scale circulation. In the subsequent sections we will further demonstrate the fidelity of ODT by showing comparison of fluxes with a scaling based on physical arguments.

Additionally, we have analyzed the profiles of scalar mean and standard deviation outside the boundary-layer, similar to Du Puits et al. [83]. From Figure 2.2(c)-(d), it is apparent that the mean and standard deviation profiles of water vapor mixing-ratio (or temperature, not shown here) are nearly logarithmic as observed in Tilgner et al. [298] and Ahlers et al. [6] and contrasting to the observation of power-law in Du Puits et al. [83]. Although, the overall exponent of the power-law fitting for the standard deviation profile (inset of figure 2.2 (d)) is around $\gamma = -0.4$, and it is very close to the value reported by Du Puits et al. [83] for the aspect ratio close to unity. In the current case, this exponent varies from $\gamma = -0.3$

Figure 2.3: Variation of the scalar fluxes of heat (Nu) and water vapor (Sh) with moist Rayleigh number (Ra_{moist}): (a) Nu scaled with Pr according to the scaling relations 2.9 and 2.11 and plotted against Ra_{moist} . (b) Sh scaled with Sc and Pr according to the scaling relations 2.10 and 2.12 and plotted against Ra_{moist} . Inset figures are the compensated plots where Nu and Sh are multiplied with $Ra_{moist}^{-1/3}$ and plotted against Ra_{moist} . Fittings of the scaled Nu and Sh data produce Ra_{moist} exponents around 0.316 ± 0.005 and 0.328 ± 0.006 .

just outside the boundary-layer to $\gamma = -0.62$ near the center of the domain. The aspect-ratio close to unity is a better case for comparison considering the formulation of the ODT model structure. In ODT, the large scale circulation is not explicitly resolved because of its dimensional constraint, although, large mapping events might incorporate the statistical features of it for the aspect ratio of unity [325]. It is intriguing that such a simple model, ODT, can capture such details of the scalar profile in R-B convection.

2.3.1 Flux scaling — Nu and Sh as functions of Ra_{moist} , Pr , and Sc

Can we get the scalar fluxes in moist convection by merely extending the relation of dry convection with additional buoyancy perturbation from another scalar? Does ODT capture the effect of additional buoyancy perturbation correctly? What is the role of scalar diffusivities on the fluxes of both scalars in two-component R-B convection? These questions, along with the appropriateness of the ODT simulation for this case, are the primary motivation of this section. Dimensionless measures of heat and water vapor transfer are given by the Nusselt (Nu) and Sherwood (Sh) numbers, respectively, i.e., the heat and mass transfer for turbulent convection relative to the values calculated for molecular diffusion. Conceptually, heat transfer in turbulent R-B convection is assumed to be limited by conductive heat transfer through the laminar boundary layer (BL), which itself is at a critical thickness [121, 122]. This argument suggests that the Rayleigh number based on the average velocity BL thickness (Ra_{δ_v}) is a constant parameter, such that $Ra_{\delta_v}/Ra = c/Ra = (\delta_v/H)^3$. It is also recognized that the relative thicknesses of the velocity and thermal boundary layers, for $Pr \sim 1$, are $\delta_v/\delta_{th} = \sqrt{\pi\nu t^*}/\sqrt{\pi D_t t^*} = Pr^{1/2}$. Here t^* is a critical timescale for boundary layer development before thermal/plume detaches from boundary [122]. The same argument can be extended to two-component R-B convection by replacing Ra with Ra_{moist} , allowing the following scaling for Nusselt number to be obtained:

$$Nu = \frac{H}{2\delta_{th}} = \frac{1}{2c^{1/3}} Pr^{1/2} Ra_{moist}^{1/3}. \quad (2.9)$$

Similarly, the Sherwood number (Sh) scaling for water vapor can be expressed as

$$Sh = \frac{H}{2\delta_w} = \frac{1}{2c^{1/3}} Sc^{1/2} Ra_{moist}^{1/3}, \quad (2.10)$$

where δ_w is the BL thickness for water vapor. For the case where $Pr \ll 1$, the kinematic BL is much thinner compared to the thermal BL. Therefore, the critical Ra_{δ_v} , which is based on the stability of velocity BL, should contain the friction velocity ($w \sim \nu/\delta_v$) as a velocity scale. This is instead of the velocity scale determined based on advection-diffusion balance of density, since the density change due to scalar diffusion is much smaller in the velocity BL as a result of the larger thermal BL-thickness. Use of the different velocity scale leads to the following relation: $Ra_{\delta_v}/Ra_{moist} = c/Ra_{moist} = Pr^{-1}(\delta_v/H)^3$. Consequently, we obtain the following modified scalings for Nu and Sh :

$$Nu = \frac{H}{2\delta_{th}} = \frac{H}{2\delta_v} \frac{\delta_v}{\delta_{th}} = \frac{1}{2c^{1/3}} Pr^{1/6} Ra_{moist}^{1/3}, \quad (2.11)$$

$$Sh = \frac{H}{2\delta_w} = \frac{H}{2\delta_v} \frac{\delta_v}{\delta_w} = \frac{1}{2c^{1/3}} Sc^{1/2} Pr^{-1/3} Ra_{moist}^{1/3}. \quad (2.12)$$

The above simplified scalings are based on the foundation of Howard [121, 122] and a linear BL approximation for the heat flux estimation. Moreover, any effects of turbulent fluctuations (or eddies) at the edge of BL in the boundary-layer heat transport are ignored.

A more detailed treatment of the scaling relations likely can be obtained by extending the Grossmann and Lohse [108, 109] theory to the two-component RB convection problem. For the current purposes, however, where the goal is to understand the essential features of the supersaturation field within moist convection, we stay with this simplified treatment of the scaling relations.

Figure 2.3 demonstrates the Nusselt (a) and Sherwood (b) number scaling with Ra_{moist} as produced by ODT, and serves as a check for the Nu and Sh scalings described above. It is apparent from the collapse of the data points to a line that the Nu scalings (Eqs. 2.9, 2.11) and Sh scalings (Eqs. 2.10 and 2.12) reasonably describe the heat and water vapor fluxes for the simulated ranges of Pr and Sc . Moreover, fittings of the scaled Nu and Sh produce Ra_{moist} exponents of 0.316 ± 0.005 and 0.328 ± 0.006 for heat and water vapor scalars, respectively. The compensated plots, as shown in inset figures, also display nearly a flat response with Ra_{moist} . However, there is a slight negative slope in the compensated Nu plot, which explains the somewhat lower exponent than $1/3$. These exponents are close to $1/3$ and previous experimental results [89, 219, 220]. Similarly, the Pr scaling presented in numerical studies by Verzicco and Camussi [308] and Breuer et al. [36] for the low Pr case are also roughly consistent with the scaling exponent presented here ($1/6$). These results build confidence in the scaling, as well as in the ability of ODT to reproduce essential statistics of R-B convection.

Figure 2.4: PDFs of (a) temperature and (b) water vapor fluctuations near the domain center for the different diffusivity cases (8K applied temperature difference). The dot-dashed line is a Gaussian curve fit to the scalar PDF for the actual diffusivity case.

Figure 2.5: Normalized standard deviations of (a) temperature and (b) water vapor ($\sigma_T^* \equiv \sigma_T \Delta T^{-1}$ and $\sigma_{q_v}^* \equiv \sigma_{q_v} \Delta q_v^{-1}$) near the domain center versus Ra_{moist} .

Figure 2.6: (a) Normalized covariance of temperature and water vapor pressure at the domain center versus Ra_{moist} . (b) An example of the vertical profile of normalized covariance of temperature and water vapor mixing-ratio across the domain for 20K applied temperature difference (and for the case where water vapor diffusivity is four times the actual value).

2.3.2 Scalar fluctuations in the bulk fluid

The central purpose of this section is first to confirm that the behavior of scalar statistics in the bulk region are in line with expected behavior. Then we extended the analysis to explore the role of differential diffusivity and ultimately, the implications for temperature-water vapor covariance, which is crucial for determining supersaturation statistics.

In the bulk fluid in turbulent R-B convection, scalar fluctuations are determined by turbulent scalar advection from the BL and destruction of scalar gradients due to the molecular diffusion. Therefore, the magnitude of scalar fluctuations in the bulk fluid is not just a function of the applied buoyancy forcing through ΔT , but it also depends on the diffusivity

of the scalar. Figure 2.4 demonstrates this concept, that the scalar diffusivity is important in determining the bulk fluctuations. It displays PDFs of water vapor and temperature fluctuations near the center of the domain. Apparently, the dispersion of the scalar PDF is larger when its diffusivity is higher. The dot-dashed lines in the plots show a Gaussian curve fit of the scalar pdf for the actual diffusivity case. From the comparison with the curve fit, it is clear that far tails of the distributions have a slower decay than the Gaussian, in fact nearly exponential tails, as expected at the center of a turbulent R-B convection cell. Moreover, tails deviate further from Gaussian with an increase in diffusivities of the scalar. Therefore, the dispersion of the scalar pdf increases with increase in respective scalar diffusivities. Moreover, all scalar PDFs from the center of the domain are nearly symmetric as expected and become more skewed closer to the boundaries (not shown here). Can we extend a previously reported scaling approach for scalar variance in one component convection (for example, Castaing et al. [42]) to the moist (or two component) convection case, including an explanation for why σ_T and σ_{q_v} tend to change with Ra_{moist} , Pr , and Sc ? Additionally, do ODT results support these scaling relations? We consider these questions in the remainder of this section.

A simplified relation for the bulk fluctuation with moist-convection parameters (Ra_{moist} , Sc and Pr) can be derived based on scaling arguments similar to Castaing et al. [42]. During steady moist-convection, the bulk convective acceleration ($w \partial w / \partial z$) is mainly supported by the buoyancy forcing in the bulk fluid ($g\beta\delta T_{v,bulk}$), and the viscous force is negligible unless the length-scale of interest is very small. If the buoyancy contribution of water vapor is small, the bulk virtual temperature difference ($\delta T_{v,bulk}$) can be approximated from the

mean temperature profile. The mean virtual temperature drop in the bulk flow can be approximated as: $\kappa \Delta T_v$ based on the assumption of Howard's boundary-layer profile [122], where the constant of proportionality κ depends on Pr and Sc . Consequently, we get the following scaling relation for the normalized vertical-velocity fluctuation ($\sigma_w^* \equiv \sigma_w H / D_t$) in the bulk:

$$\sigma_w^* \sim \kappa^{1/2} Ra_{moist}^{1/2} Pr^{1/2} \left(\frac{z}{H} \right)^{1/2}. \quad (2.13)$$

The heat and water vapor transport in the bulk are due to both the turbulent convection and the diffusion as a result of the mean scalar gradient. In the limit of low Pr or Sc , the diffusion term in the heat or water vapor flux can be neglected. As a result, Nu and Sh can be approximated as $\overline{w'T'^*}$ and $\overline{w'q_v'^*}$, and the order of these terms can be represented as $\sigma_w^* \sigma_T^*$ and $\sigma_w^* \sigma_{q_v}^* Pr^{-1} Sc$ respectively. Here, $\sigma_T^* \equiv \sigma_T \Delta T^{-1}$ and $\sigma_{q_v}^* \equiv \sigma_{q_v} \Delta q_v^{-1}$. By using the Nu and Sh scaling from Eqns. 2.9 and 2.10, the following scalings for the normalized scalar fluctuations can be derived:

$$\sigma_T^* \sim \kappa^{-1/2} Ra_{moist}^{-1/6} \left(\frac{z}{H} \right)^{-1/2} \quad (2.14)$$

and

$$\sigma_{q_v}^* \sim \kappa^{-1/2} Ra_{moist}^{-1/6} Sc^{-1/2} Pr^{1/2} \left(\frac{z}{H} \right)^{-1/2}. \quad (2.15)$$

If $Pr \ll 1$, the above scalings for the temperature fluctuation will not be applicable since

there will be a significant contribution of the heat flux due to the temperature gradient in the bulk, as well as the scaling for Nu will be different. Moreover, the correlation between temperature and vertical velocity will be reduced because of a significantly larger diffusivity of the thermal field. Due to a similar reason as the $Pr \ll 1$ case, for the $Sc \ll 1$ case Eq. 2.15 will also not be applicable. Evidence for a higher mean gradient in the bulk for the $Sc \ll 1$ case is manifested in Fig. 2.2. For the $Pr \ll 1$ case, however, a new scaling of σ_{qv}^* can be obtained, and it is slightly different than Eq. 2.15 due to the different flux scaling in Eq. 2.12:

$$\sigma_{qv}^* \sim \kappa^{-1/2} Ra_{moist}^{-1/6} Sc^{-1/2} Pr^{1/6} \times \left(\frac{z}{H} \right)^{-1/2}. \quad (2.16)$$

Although, as explained above that the scaling for σ_T^* (for $Pr \ll 1$) and σ_{qv}^* (for $Sc \ll 1$) derived based on the Nu/Sh relation may not be completely applicable due to a non negligible mean scalar gradient in the bulk as well as a decrease in correlation efficiency between vertical velocity and scalar fluctuations, we still present these results below for the completeness of the discussion:

$$\sigma_T^* \sim \kappa^{-1/2} Ra_{moist}^{-1/6} Pr^{-1/3} \left(\frac{z}{H} \right)^{-1/2} \quad (2.17)$$

and

$$\sigma_{qv}^* \sim \kappa^{-1/2} Ra_{moist}^{-1/6} Sc^{-1/2} Pr^{1/2} \left(\frac{z}{H} \right)^{-1/2}. \quad (2.18)$$

For these cases, the actual Pr/Sc exponent might be slightly different from above scaling relations for the actual $Pr \sim 1$ and $Sc \sim 1$ ranges.

Figure 2.2 illustrates that the scaling presented above qualitatively represents the shape of the scalar fluctuation profile outside the BL. The normalized q_v standard-deviation decreases nearly in a parabolic profile outside the BL, consistent with scaling. The derived power of Ra_{moist} in the above scalings is close to the experimentally observed $\sigma_T^* - Ra$ scaling for single-component R-B convection [220], as well as to predictions of Grossmann and Lohse [108, 109].

Similarly, as expected from the scaling relations, Fig. 2.5 displays a decreasing trend of the normalized standard deviation of T and q_v with Ra_{moist} . Furthermore, the Ra_{moist} exponent obtained from a data fit is close to the prediction from the scaling law, as can be seen from the dashed reference lines with $(-1/6)$ slope. For the low- Pr case, the σ_T^* data points are slightly offset from the normal trend of the other data set; this is due to the additional Pr dependency because of a higher thermal diffusivity as explained in the earlier paragraphs (Eq. 2.17). Likewise, for the same case, σ_{qv}^* data points are slightly lower, as expected from the scaling relation Eq. 2.16 due to the $Pr^{1/6}$ dependence. Moreover, with a decrease of Sc , i.e., an increase of the water vapor diffusivity, the normalized water vapor fluctuation increases as anticipated by the scaling relation. Although, for the $Sc \ll 1$ case, the Sc dependency in the relation Eq. 2.15 will be slightly different due to a significant mean gradient in the bulk as explained earlier.

The covariance of other scalars with the temperature field in convective flows is an important

quantity for atmospheric applications since it is related to the reaction rate or rate of phase change through the supersaturation. Figure 2.6(a) shows the normalized covariance of water vapor pressure and temperature at the center of the convection cell, as a function of Ra_{moist} . As anticipated, it is close to one for the cases with small diffusivity difference. However, for higher diffusivity differences, it drops down slightly. In these cases, the normalized covariance decreases with an increase in the buoyancy forcing (Ra_{moist}). As depicted in Figure 2.6(b), the covariance magnitude also varies along a vertical profile moving towards boundaries, due to the higher effect of relative diffusion and lower turbulence level closer to the boundaries. In cumulus clouds, the normalized covariance between temperature and water vapor field was estimated between $0.24 - 0.34$ using high-frequency measurements of temperature and water vapor concentration [275]. These values are significantly lower than the current simplistic case. It should be noted, however, that in cloudy convection the presence of water vapor and heat fluxes resulting from entrainment and condensation could decrease this covariance further from the idealized confined convection case.

2.3.3 From two scalars to supersaturation: Mixing diagrams and vertical profiles

In atmospheric R-B convection the relevant scalar for cloud formation is the supersaturation, which as discussed already, is a combination of the two scalars water vapor pressure and temperature: $s \equiv (e_v/e_{v,s}(T)) - 1$. The saturation value $e_{v,s}(T)$ is related exponentially to the temperature through the Clausius-Clapeyron equation. We note that water vapor

Figure 2.7: (a) An example of the mixing diagram for the actual diffusivity case at 8K applied temperature difference with error-bars showing standard deviation. (b) Mixing diagram for different diffusivity cases at a higher applied temperature difference (20K).

pressure and water-vapor mixing ratio can be considered as interchangeable, via the ideal gas law. In Fig. 2.2 and Sec. 2.3.2 we considered the vertical profiles and fluctuations of the two individual scalars, and in this sub-section we extend to supersaturation. In the following two sub-sections we then investigate the bulk and boundary-layer properties of supersaturation in more detail.

Figure 2.7 shows mixing diagrams obtained from ODT grid points near the center of the domain. As discussed in the introduction, in an idealized scenario one would expect the sampled values of e_v and T to lie along a linear mixing line (dashed curve). In the left panel (a), which corresponds to $\Delta T = 8$ K and for the actual diffusivities, this can be seen to be approximately true. In the right panel (b), however, corresponding to $\Delta T = 20$ K, it can be seen that for the two cases with different scalar diffusivities, the points deviate from the idealized mixing line. When the thermal diffusivity is higher than the water vapor,

Figure 2.8: Supersaturation mean and fluctuation profile: **(a)** vertical profiles of the supersaturation mean, **(b)** Profiles of supersaturation fluctuations (standard deviations) for the different diffusivity cases. (Here, $Z^* = 0$ corresponds to hot and 1 corresponds to cold boundary.)

the cloud of points spreads more strongly along the temperature axis and therefore has a shallower slope than the mixing line. This is due to a higher relative transport for the temperature compared to the water vapor due to its higher diffusivity. The exact opposite argument is also valid when considering higher water vapor diffusivity. The spread of points is now greater along the vapor-pressure axis leading to a steeper inclination compared to the mixing line. We can anticipate that this higher spread produces greater fluctuations in supersaturation relative to the idealized mixing scenario with nearly-equal diffusivities (cf. Sec. 2.3.4). The relative supersaturation fluctuation can be visualized by comparing the spread of data points above and below the saturation curve.

Supersaturation mean and fluctuation profiles in the atmospheric boundary-layer are anticipated to be relevant for cloud microphysical processes and dynamics. Figure 2.8 illustrates the profile of supersaturation mean **(a)** and standard deviation **(b)** (σ_s) across the domain.

The mean supersaturation profile is nearly flat in the bulk fluid, just for the individual scalars, when the diffusivity difference is small. For these cases, it is also close to the mean value one calculates from the idealized mixing process. Interestingly, the slope of the mean supersaturation profile flips sign in both halves of the domain when one of the scalar diffusivities is significantly higher than the other. Moreover, a difference in the scalar diffusivities causes a significant peak to appear in the mean supersaturation profile, close to the boundaries. Which diffusivity is larger also determines the sign of the supersaturation (or subsaturation) peak near the two boundaries. If the thermal diffusivity is higher than the water vapor diffusivity, the supersaturation peak is positive at the cold boundary and negative at the hot boundary (and opposite otherwise). A higher diffusivity of a scalar leads to a smaller slope of the mean scalar profile in the boundary layer. Therefore, if the thermal diffusivity is higher than the other scalar, the drop in average temperature from the hot boundary is slower than the case when both are equal. As a result of the above effect as well as the exponential relation of the saturation vapor pressure with temperature, the supersaturation profile produces a negative peak close to the hot boundary and a positive peak near the cold boundary for a higher thermal diffusivity and opposite otherwise. Additionally, for the higher relative diffusivity cases, the mean profile does not cross the mean value and each other precisely at $z^* = 0.5$, due to the nonlinear relation of saturation vapor pressure with temperature (the Clausius-Clapeyron equation). The saturation vapor pressure change is not symmetric at a temperature lower or higher than the mean because of this nonlinearity. Similarly, the multiple peaks in standard deviation profiles near the boundaries could also be a result of this nonlinearity since it does not exist (only one peak)

Figure 2.9: Sample PDFs of supersaturation near the domain center for the different diffusivity cases (8-K applied temperature difference).

in the temperature or water vapor mixing-ratio profiles. The σ_s profile responds analogously to the individual scalar profiles (cf. Fig. 2.2); i.e., it peaks near the boundaries and decreases while approaching the center. This decreasing response with z^* is qualitatively consistent with the parabolic decrease found in Sec. 2.3.4 for scaling of supersaturation in the bulk fluid. Furthermore, in that sub-section we also investigate the observation that σ_s increases significantly for the cases with different scalar diffusivities.

2.3.4 Supersaturation fluctuations in the bulk fluid: contributions from both scalars

In this sub-section we consider the behavior of supersaturation fluctuations in the bulk fluid, away from the BL. Figure 2.9 shows several examples of supersaturation PDFs for different thermal and water vapor diffusivity combinations. Analogous to the temperature and vapor

Figure 2.10: Supersaturation fluctuation statistics at the domain center. (a) Supersaturation standard deviation as a function of Ra_{moist} and its comparison between the different diffusivity cases. (b) Contributions of different scalar statistics (as shown in Eq. 2.19) to the supersaturation variance versus Ra_{moist} .

mixing-ratio PDFs, it responds to a change in the diffusivity of both scalars. However, it produces broader PDFs with an increase in the difference in diffusivity of water vapor and temperature (not the absolute diffusivities of both scalars) due to the nonlinear coupling of both scalars in supersaturation. Moreover, Fig. 2.9 shows that the PDFs are negatively skewed for the lower relative diffusivity cases (left panel (a) of Figure 2.9). This negative skewness of the fluctuation distribution is due to the increasing slope of the saturation curve with temperature (i.e., the exponential temperature dependence).

As described earlier, both scalar fluctuations (normalized by the applied scalar gradients) can be scaled as a function of Ra_{moist} , Pr and Sc . Therefore, we expect the supersaturation fluctuation can also be scaled as a function of these parameters. A detailed exploration of the contributions from both scalars can be made by expressing supersaturation variance as a function of temperature and water-vapor pressure variances [165, 275]:

$$\sigma_s^2 \approx S^2 \left[\left(\frac{\sigma_{e_v}}{\bar{e}_v} \right)^2 + \zeta^2 \left(\frac{\sigma_T}{\bar{T}} \right)^2 - 2\zeta \frac{\overline{T' e_v'}}{\bar{T} \bar{e}_v} \right]. \quad (2.19)$$

Here, $S = e_v/e_{v,s}(\bar{T})$ is the mean saturation ratio, $e_{v,s}(\bar{T})$ is the saturation vapor pressure at temperature the average temperature, and $\zeta = \mathcal{L}/(R_v \bar{T})$. Additionally, primes and bars represent fluctuations and mean quantities, respectively.

With the use of the scaling expressions Eqs. 2.14 and 2.15, a linearized form of the Clausius-Clapeyron equation ($\Delta e_{v,s} \approx \zeta \bar{e}_{v,s} \Delta T / \bar{T}$), and Eq. 2.19, the following scaling relation for supersaturation fluctuations can be obtained:

$$\left(\frac{\sigma_s}{S} \right)^2 \sim \kappa^{-1} \xi^2 \left[1 + \frac{Pr}{S^2 Sc} - \frac{2C_{qv,T}}{S} \left(\frac{Pr}{Sc} \right)^{1/2} \right] Pr^{-2} Ra_{moist}^{5/3} \left(\frac{z}{H} \right)^{-1}. \quad (2.20)$$

Here, $C_{qv,T}$ is the correlation efficiency of water-vapor mixing ratio and temperature and $\xi = \zeta \nu^2 / g H^3$. As can be perceived from Fig. 2.6, the correlation efficiency of water vapor and temperature fluctuations reside mostly in the neighborhood of 100 %, at least for the current range of Ra_{moist} . With this information, Eq. 2.20 can be simplified to:

$$\left(\frac{\sigma_s}{S} \right)^2 \sim \kappa^{-1} \xi^2 \left[1 - \frac{1}{S} \left(\frac{Pr}{Sc} \right)^{1/2} \right]^2 Pr^{-2} Ra_{moist}^{5/3} \left(\frac{z}{H} \right)^{-1}. \quad (2.21)$$

The left panel of Fig. 2.10 displays the variation of supersaturation fluctuations with Ra_{moist} as a result of fluctuations in both scalars. There is an increasing trend of the supersaturation

fluctuations with Ra_{moist} as anticipated from the scaling relation Eq. 2.20, and as indicated by the dashed line with the predicted scaling slope for Ra_{moist} . The magnitude of σ_s are larger for a higher difference between the water vapor and thermal diffusivity. This effect of diffusivity difference on σ_s is also predicted by the scaling relation Eq. 2.21. Contributions of the different terms of Eq. 2.19 to the magnitude of the supersaturation fluctuations are shown in the right panel of Fig. 2.10. It can be seen that all terms are equally important, although because of the strong covariance, Eq. 2.19 can be further simplified by replacing the covariance term with the standard deviations of the two scalars. Moreover, the scalar with a higher diffusivity dominates the contribution, and the covariance term always has the largest magnitude.

2.3.5 Supersaturation profiles in the boundary layer

In this sub-section we move from the bulk to the boundary layer (BL). The mean and instantaneous BL profiles in R-B convection have been observed to be close to the BL theory of Prandtl-Blasius-Pohlhausen [350, 351]. Therefore, to explore the supersaturation profile within the boundary layer, we have used the Prandtl- Blasius-Pohlhausen profiles for the water vapor and temperature fields. As mentioned above, some studies claim the Prandtl-Blasius-Pohlhausen profile reasonably represents the scalar profile in the boundary layer of single-component Rayleigh-Bénard convection. In the current case, even though we have two driving scalars and the air is supersaturated with water vapor, both scalars contribute to positive buoyancy, avoiding the complexities of double-diffusive convection.

Figure 2.11: BL mean supersaturation profile at the hot boundary: **(a)** an example of the s profiles for the actual diffusivity case, **(b)** the s profile for the case where four times higher (than actual) diffusivity of water-vapor is used. Here, the black-dash curve is the simulation output, the solid red-curve shows an estimation using the simplified Blasius-BL solution, and the solid cyan-curve represent an estimation using Howard's model [122].

Figure 2.12: Supersaturation mean and peak standard deviation inside the BL. **(a)** Variation of the mean supersaturation peak with ΔT and its comparison with the Howard model. **(b)** Maximum supersaturation standard deviation inside the BL versus applied temperature difference.

In fact, the moist convection case can be expressed as single-component R-B convection with the virtual temperature difference as the driving scalar. The only complication arises due to the different diffusivities of both scalars, which changes the relative slopes of the two profiles. However, the concept of the Prandtl-Blasius-Pohlhausen profile due to the large scale circulation can also be extended for this case since two scalars do not have contrasting buoyancy contributions, unlike in double-diffusive convection. Its applicability may be questioned even in case of the single-component R-B convection due to the presence of the plume structure near boundaries. Therefore, we also have compared this profile with Howard's model [122] to explore the supersaturation profile.

With the assumption of low Pr/Sc (i.e., the thermal and water vapor boundary layer completely encapsulates the velocity boundary layer), we can obtain a simplified solution of the water vapor and thermal boundary layer. This assumption ($\delta_v < \delta_{th}$ and $\delta_v < \delta_w$) might be a reasonable approximation for the Pr/Sc range considered in this study. It implies that the velocity variation in the thermal or water vapor boundary-layer is neglected. Since the Blasius boundary layer is expected to be self-similar, using a similarity transform with similarity variables $\eta = z/\delta(x)$, $\Theta = 2(T_b - T)/\Delta T$ and $\psi = U\delta(x)f(\eta)$ (or $f'(\eta) = u(z)/U$), a simplified solution can be obtained [20, 260]:

$$\Theta(\eta) = \operatorname{erf}\left(\frac{\eta\sqrt{Pr}}{2}\right). \quad (2.22)$$

Here ψ , δ , T_b and U are the velocity stream-function, BL thickness, temperature at the

bottom boundary and free stream velocity, respectively. This solution is also applicable for the water-vapor field with corresponding changes in variables (e.g., Pr replaced with Sc and δ_{th} with the BL thickness for water vapor, δ_w). The streamwise velocity scale U determines the timescale for the boundary layer growth, i.e., the kinematic boundary layer thickness at location x from the leading edge scales as $\sqrt{\nu\tau}$, where $\tau = x/U$. For Rayleigh-Bénard convection, the streamwise velocity is thought to be a result of the large scale circulation, which is still questionable due to the presence of the intermittent plume structure near boundaries. The plume structure of the boundary layer suggests that the intermittent boundary layer develops locally until a plume detaches from the boundary. In ODT simulations the large scale circulation is not represented explicitly due to its one-dimensional structure; however, the effect of a large scale circulation is represented stochastically by the large-scale mappings [325]. Therefore, for the completeness of the discussion, we have included the Prandtl-Blasius-Pohlhausen solution of the thermal and water vapor profiles. The thermal BL thickness can be estimated from $\delta_{th} = H/(2 Nu)$ together with the $Nu-Ra$ relation. We use the above solution in conjunction with the empirical relation of Nu and Sh from fitting (as explored in section 2.3.1) and the Clausius-Clapeyron equation to get the BL profile of supersaturation.

Figure 2.11 shows the mean supersaturation profile (near the hot boundary) obtained from the ODT simulation and its comparison with the BL solution as discussed above. Both the simulation and the estimate produce a peak of supersaturation in the BL, and the estimated magnitude of the peak is roughly consistent with the simulation. Considering the approximations used in the BL solution, it has reasonable predictive ability. As an

alternative, we also adapt the model proposed by Howard [122] to predict the BL profile of supersaturation profile. As seen in Fig. 2.11, it provides a significantly improved match to the ODT. The Howard model is based on the concept of continuous eruptions of ‘thermals (plume)’ from the BL: it assumes that at some place on the hot boundary, a thermal develops during the conduction stage and because of the convective instability, it becomes detached from the boundary instantly once it reaches some critical size. A mean critical time to which a thermal develops, until which the corresponding diffusion-equation solution can be applied near the plate, is represented here as t^* . In turbulent convection these thermals are continuously developing and breaking off at different times and places. Therefore, the time-dependent solution of the diffusion equation needs to be averaged from 0 to t^* and the resulted expression is [122]

$$\bar{T} = T_m + \Delta T/2 \left[(1 + 2 \xi^2) \operatorname{erfc}(\xi) - \frac{2 \xi e^{-\xi^2}}{\sqrt{\pi}} \right]. \quad (2.23)$$

Here, $\xi = z/(2\sqrt{D_t t^*})$, $t^* = H^2(Ra_{\delta_v}/Ra)^{2/3}/(\pi\nu)$, and Ra_δ is the Rayleigh number based on the critical boundary layer thickness ($\delta = \sqrt{\pi\nu t^*}$). The expression for t^* is obtained from the definition of Rayleigh number ($Ra/Ra_\delta = (H/\delta)^3$) and the critical boundary layer thickness ($\delta = \sqrt{\pi\nu t^*}$). Howard [122] reported $Ra_\delta = 2800$ obtained from fitting measurements made in air by Townsend [300]. We use the above relation (and a similar relation for water vapor), along with $Ra_\delta = 2800$ for obtaining the supersaturation profile shown in Fig. 2.11. The Howard model better predicts the magnitude and z -position of the peak in mean supersaturation and σ_s .

Figure 2.12 displays the supersaturation mean **(a)** and σ_s **(b)** peaks near the hot boundary at different ΔT and for the different diffusivity cases. Both increase monotonically with ΔT . The prediction of the peak mean- s from the Howard model is also shown in the left panel **(a)**, and it is able to capture the trend and the magnitude remarkably well for all cases. The peak magnitude of the mean supersaturation is positive and higher for the higher water vapor diffusivity case and negative for the higher thermal diffusivity case. The fluctuations are influenced by the absolute diffusivity difference between water vapor and temperature, and is greater if one of the diffusivity is significantly larger than the other. Moreover, the peak position of supersaturation is nearly insensitive to the temperature difference in the current range of Ra_{moist} (not shown here).

2.4 Summary and Discussion

The problem of moist Rayleigh-Bénard convection involves buoyancy components from two driving scalars, temperature and water vapor, in a flow field, which makes it more intriguing and complex compared to its dry (single-component) convection counterpart. There is a rich variety of multi-component convection problems, ranging from double-diffusive convection [125] to particle-settling-driven convection [39]. In this work, we consider the problem from the perspective of supersaturation, which is a nonlinear coupling of water-vapor concentration and temperature fields, and is the fundamental driver of phase transformations in clouds. Cloud formation itself leads to profound changes in flow structure through latent heating, radiative transfer, and particle settling, but in this work we focus solely on the

supersaturation field. This view can be considered complementary to the boundary-layer perspective of humidity flux budgets ([328], [207], and reference therein), where covariances of temperature and absolute humidity are considered. One aspect of the problem we explicitly account for here is the difference in diffusivities of both scalars; although it is seemingly slight, we consider the extent to which it influences scalar statistics. The intent in varying the two diffusivities has been primarily to elucidate and clarify the role of differential diffusivity in determining supersaturation profiles and statistics. Beyond its purpose in exploring the dependence on differential diffusivity, there may be applications (e.g., semiconvection or cloud formation in exoplanet atmospheres) in which varying Lewis numbers play a role [198, 347].

Supersaturation statistics, not simply mean quantities, but properties of the fluctuations, are very important for cloud microphysical processes [239]. The supersaturation statistics in the atmospheric boundary-layer may be dependent on surface and radiative fluxes, and other complex dynamical features of the atmospheric convection. In this article, we have investigated the supersaturation statistics in an idealized scenario of the moist R-B convection in order to gain insight into the fundamental roles of temperature and water-vapor covariance, differential scalar diffusivity, and diffusion versus advection. This R-B convection perspective is also motivated by experiments performed in a turbulent moist-convection chamber, in which supersaturation variability has been proposed as contributing to important cloud properties [46]. Eventually, we aim to compare these results to experimental observations, but this will require careful development of methods capable of accurate measurement of temperature and water vapor, highly-resolved in both space and time.

Central results and findings from this study may be summarized as follows:

† We have investigated moist R-B convection using a stochastic model (ODT), covering a range of Rayleigh number $2.05 \times 10^8 \leq Ra \leq 2.75 \times 10^9$. ODT is a numerically-efficient method of exploring advection and diffusion within turbulent convection [325]. To explore the role of relative diffusivity, four different combinations of scalar diffusivities for temperature and water vapor have been used. As a primary check, profiles of mean and standard deviation of both scalars are consistent with profiles one would expect in R-B convection.

† A simplistic scaling for Nu and Sh is developed, based on the approaches of Malkus [196] and Howard [121]. The scalings and simulation results are generally self-consistent, and are in line with previous findings.

† In the bulk fluid, σ_T^* and σ_{qv}^* both follow a $Ra_{moist}^{-1/6}$ scaling relation. Moreover, the magnitude of scalar fluctuations increases with an increase in the respective scalar diffusivity, as predicted from the scaling relations. An increase in thermal diffusivity decreases water vapor fluctuations, which is also consistent with derived scaling. Derivation of the scaling relations are based on the argument presented in Castaing et al. [42] for the single-component convection case.

† The correlation of both scalars is very close to 1 for a small difference in scalar diffusivities, and it decreases slightly with an increase in relative diffusivity and Ra_{moist} . It also decreases further towards boundaries.

† In a mixing diagram ($e_v - T$ coordinate), data points obtained from the center of the

domain shows deviations from the idealized mixing scenario for cases with relatively different water vapor and thermal diffusivities. The scatter of data points within the space $(e_v - T)$ is greater for the scalar with higher diffusivity. This inclination of data points corresponding to the mixing line signifies higher supersaturation fluctuations compared to the idealized mixing case.

† Vertical profiles of supersaturation mean and standard deviation both are nearly flat in the bulk fluid for cases with low diffusivity difference. Additionally, the mean value is close to the prediction from the ideal mixing-diagram. In contrast, for cases with higher diffusivity difference, the mean profile develops a more pronounced slope and its sign flips depending on $D_v > D_t$ or $D_v < D_t$. The σ_s profile resembles the shape of both scalars and its magnitude scales with relative diffusivity difference.

† The PDF of supersaturation become broader with an increase in absolute value of the diffusivity difference. Also, the PDF is slightly negatively skewed for cases with a low diffusivity difference, unlike the T and q_v PDFs. Moreover, we have developed a scaling relation for supersaturation variance in the bulk fluid. It suggests an increase in supersaturation variance as $Ra_{moist}^{5/3}$, likewise, a relationship with Pr and Sc . The supersaturation sampled from the center of the domain approximately agrees with the scaling. Furthermore, the analysis of numerical output shows contributions to the supersaturation variance from both scalar variance and covariance. It indicates that all terms are equally important, although the covariance contribution can be simplified as a function of individual scalar variances since the correlation efficiency is nearly 100%. However, this correlation efficiency might be significantly lower in atmospheric

clouds (e.g., $0.24 - 0.34$ in cumulus cloud [275]) due to additional relative fluxes of water vapor and heat as a result of entrainment and condensation. This certainly is a topic for further investigation.

† Finally, the supersaturation profile within the boundary layer has been investigated. It develops opposite-sign peaks near the two boundaries for a nonzero diffusivity difference. These peaks flip sign depending on the condition $D_v > D_t$ or $D_v < D_t$. Additionally, supersaturation profiles in the BL obtained using scalar profiles from Prandtl-Blasius-Pohlhausen solution (for a low Pr/Sc case) and from Howard’s model, both resemble the shape reasonably well. Howard’s model predicts the shape, as well as the peak magnitude and location extremely well.

The insight into the behavior of supersaturation fluctuations in the idealized scenario considered in this study could have implications for sub-grid-scale representation of fluctuations relevant to cloud droplet growth [e.g., 46]. Already, the two-scalar concept has been used in representation of cloudiness in computational models of the atmosphere by combining assumed probability density functions (PDFs) with higher-order turbulence closure (HOC). HOC uses the equations that govern selected moments of the SGS variables. For example, Sommeria and Deardorff [281] and Mellor [208] independently connected the concept of assumed probability density functions with HOC. They proposed that within the grid cells of a large-eddy simulation (LES), the liquid water potential temperature θ_l and total water mixing ratio (vapor plus cloud droplets) q_t , could be assumed to have a joint Gaussian distribution. They further showed how this assumption can be used to diagnose the fractional cloudiness from the means, variances, and covariance of θ_l and q_t . Their idea was that these

moments could be determined by HOC, then used to determine the parameters of the assumed joint Gaussian for each grid cell of a model. The joint distribution could then be used to diagnose the fractional cloudiness, which in turn affects the buoyancy flux, microphysical process rates, and radiative fluxes. Cloud is assumed to exist when q_t exceeds the saturation mixing ratio of water vapor, q_v^* and the cloud water mixing ratio is diagnosed as $q_t - q_v^*$ integrated over the joint PDF. The approach has been further generalized [98, 99, 244] and has been widely adopted. The work presented here provides additional insight, by illustrating the role of scalar diffusivity, and by providing scaling relations for boundary layer and bulk supersaturation profiles and fluctuations. A next step is to make direct comparison with laboratory measurements, as well as extending to atmospherically-relevant scales.

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Chapter 3

Aerosol indirect effect from turbulence-induced broadening of cloud-droplet size distributions

This chapter is about the laboratory study of microphysical interactions of aerosol and cloud in the presence of turbulent fluctuation. It is based on a collaborative research published in the Proceedings of the National Academy of Sciences of the United States of America [43]¹.

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3.1 Abstract

The influence of aerosol concentration on the cloud droplet size distribution is investigated in a laboratory chamber that enables turbulent cloud formation through moist convection. The experiments allow steady-state microphysics to be achieved, with aerosol input balanced by cloud droplet growth and fallout. As aerosol concentration is increased the cloud droplet mean diameter decreases as expected, but the width of the size distribution also decreases sharply. The aerosol input allows for cloud generation in the limiting regimes of fast microphysics ($\tau_c < \tau_t$) for high aerosol concentration, and slow microphysics ($\tau_c > \tau_t$) for low aerosol concentration; here, τ_c is the phase relaxation time and τ_t is the turbulence correlation time. The increase in the width of the droplet size distribution for the low aerosol limit is consistent with larger variability of supersaturation due to the slow microphysical response. A stochastic differential equation for supersaturation predicts that the standard deviation of the squared droplet radius should increase linearly with a system time scale defined as $\tau_s^{-1} = \tau_c^{-1} + \tau_t^{-1}$, and the measurements are in excellent agreement with this finding. The result underscores the importance of droplet size dispersion for aerosol indirect effects: increasing aerosol concentration changes the albedo and suppresses precipitation formation not only through reduction of the mean droplet diameter, but also by narrowing of the droplet size distribution due to reduced supersaturation fluctuations. Supersaturation fluctuations in the low aerosol / slow microphysics limit are likely of leading importance for precipitation formation.

3.2 Introduction

The optical properties of warm clouds depend on the droplet size distribution and its moments such as number density and effective radius, which in turn are influenced by the aerosol particles that act as nuclei for the formation of cloud droplets [86, 175]. Thus, aerosol indirect effects are considered among the largest uncertainties in climate response to changes in radiative forcing [266]. This work addresses how the aerosol number concentration affects the cloud droplet size distribution in a turbulent environment, which is relevant to both the aerosol first and second indirect effects (albedo and lifetime effects). The lifetime effect links the development of precipitation and thus cloud lifetime, to aerosol number concentration. The logic is that a higher aerosol concentration leads to smaller cloud droplets and narrower size distributions, and therefore suppression of the collision and coalescence of droplets, thereby increasing cloud lifetime and maintaining higher cloud liquid water content [8, 193, 229, 233]. The microphysical details of the transition from condensation growth to collision growth are not fully understood, however, and it is fair to say that the underlying mechanism of the second indirect effect is still a matter of active research [86, 184]. Initiation of precipitation in warm clouds is favored when the average droplet size is increased, but perhaps even more crucial is the sensitivity to an increase in the width of the droplet size distribution [181, 202, 320]. For decades the cloud physics community has grappled with the theoretical finding that cloud droplet growth in a closed (adiabatic) parcel leads to a size distribution that becomes narrower with height above cloud base, tending to suppress the onset of coalescence through differential gravitational

sedimentation. Observations, in contrast, often reveal that cloud droplet size distributions are relatively broad, and this is usually interpreted as a consequence of entrainment and mixing and secondary activation [24, 261]. Supersaturation fluctuations due to entrainment and droplet recycling have been postulated as a mechanism for producing droplets larger than are able to be generated in an adiabatic parcel [61, 169, 338?]. These approaches have close ties to the theory of stochastic condensation developed decades ago and still being refined [79, 158, 235]. In this paper we seek insight into the mechanisms underlying the lifetime effect by asking the question, How do changes in aerosol concentration influence the moments of the cloud droplet size distribution that are relevant to precipitation formation?

Cloud droplet size dispersion is also directly relevant to the albedo effect: for example, cloud albedo is enhanced not only when the droplet number density n increases (for constant liquid water content), but also when size dispersion decreases with increasing n [86, 240]. Various contributions to this dispersion effect have been identified, such as condensational narrowing and collisional broadening, which respectively tend to decrease and increase albedo susceptibility [87, 175, 193]. What other processes influence the dispersion? Here we consider the concept that cloud droplet growth occurs in a turbulent environment, and that the microphysical response to turbulent fluctuations depends on relative magnitudes of the characteristic time for droplets to adjust to varying humidity (τ_c) and the correlation time of the turbulence (τ_t). The cloud response time τ_c is inversely proportional to droplet number density and the mean droplet diameter, so it stands to reason that aerosol properties can push a cloud into contrasting regimes with $\tau_c < \tau_t$ or $\tau_c > \tau_t$. For example, it has been suggested [86], based on the conceptual picture of competing time scales for turbulence and

cloud response [12, 172], that varying aerosol number density can shift a cloud into regimes where the nature of the cloud response to mixing and entrainment would be distinct. And recent field evidence seems to support the concept that limiting τ_c -regimes influence the supersaturation variability resulting from turbulence [271]. We investigate these concepts using a cloud chamber capable of generating turbulent clouds with controlled aerosol input.

3.3 Experimental Approach

The concept of the experiment is outlined in Fig. 3.1, showing a turbulent cloud generated through moist Rayleigh-Bénard convection and the resulting isobaric mixing of warm and cool water-saturated air [50]. Steady state cloud properties are achieved as a constant rate of aerosol injection eventually is balanced by cloud droplet growth in the turbulent environment and subsequent removal by gravitational settling. The resulting steady-state cloud can be sampled for long times, allowing for high statistical significance, and the changes in cloud droplet size distribution to varying aerosol concentrations can be observed in detail. A crucial aspect of the experiment is that the clouds are generated with constant forcing: the usual confounding feedbacks that are always present in the atmosphere are absent because the thermodynamic and turbulent forcing of the cloud is externally controlled within the convection chamber. The two vertical images shown in the figure illustrate conditions achieved under two different aerosol injection rates.

Experiments are performed in the Pi Chamber [50] ($\approx 5 \text{ m}^2$ horizontal cross-section and 1 m

high), with an unstable temperature gradient (26 °C bottom and 7 °C top boundary) applied between top and bottom surface to create the turbulent environment. All the experiments started with moist Rayleigh-Bénard convection containing no aerosols (no cloud droplets), and are allowed to run for $t > 10$ hrs to ensure steady-state condition. Then aerosol injection started at a constant rate and after a relatively brief transient (of order 10 min), cloud properties also reach steady-state conditions. Aerosol injection then continued for the next 10 – 12 hours to ensure sufficiently long cloud and turbulent samples to minimize random sampling uncertainties. Aerosol inlet concentrations and background aerosol concentration inside the chamber during the experiment are measured with a TSI Scanning Mobility Particle Sizer (SMPS). The aerosol injection occurs at a point and then aerosols are rapidly mixed by the turbulent flow; measurements at several points in the chamber confirm that n_a varies by less than 10%. Cloud properties (n and d) are measured by a Dantec Dynamics phase Doppler interferometer at a point close to the supersaturation measurement. In steady state, the cloud droplet number density and mean diameter are observed to be stationary in time to within the sampling uncertainty. Other properties such as temperature, water vapor concentration, turbulence intensity and dissipation rate, and interstitial aerosol concentration also are observed to be steady throughout the experiment due to the constant forcing.

3.4 Results

3.4.1 Steady-state cloud properties for different aerosol concentrations

Figure 3.2 shows the probability density of droplet diameter for five different aerosol input rates. The input rates $\dot{n}_a$, measured cloud droplet number density n and mean diameter $\bar{d}$ are summarized in Table 3.1. The standard deviation σ_d and relative dispersion $\sigma_d/\bar{d}$ for droplet diameter are also included. With the decrease in the aerosol input and corresponding decrease in the cloud droplet number concentration, mean (and mode) droplet diameter is observed to increase. Intriguingly, the size distribution also is observed to become broader and positively skewed, consistent with the dispersion effect observed in naturally occurring clouds [193]. The change in the mode diameter and the distribution width with aerosol injection rate is very clear and reproducible, and it is especially intriguing that the pronounced large-droplet tail approaches the drizzle range for the cleanest cases.

Table 3.1

Time averaged microphysical properties for the five steady state clouds:
Chamber-averaged aerosol injection rate $\dot{n}_a$, interstitial aerosol concentration $n_{a,int}$, cloud droplet number density n , mean cloud droplet diameter $\bar{d}$, standard deviation of cloud droplet diameter σ_d , calculated phase relaxation time τ_c , and calculated Damköhler number Da . The turbulence correlation time $\tau_t = 2$ s is obtained from the measured velocity fluctuations.

$\dot{n}_a$ (cm ⁻³ min ⁻¹)	$n_{a,int}$ (cm ⁻³)	n (cm ⁻³)	$\bar{d}$ (μm)	σ_d (μm)	$\sigma_d/\bar{d}$	τ_c (s)	Da
1	2	21.3	16.6	6.7	0.40	58.1	0.03
2	10	76.9	15.1	5.7	0.38	17.6	0.11
4	36	201.2	12.7	4.5	0.35	8.0	0.25
12	372	564.6	8.6	2.4	0.28	4.2	0.5
1515	22000	1944.3	7.6	2.1	0.28	1.4	1.5

Figure 3.1: Overview of the experimental approach, showing two photographs of the turbulent cloud illuminated by a laser light sheet for two different aerosol injection rates. Panels a) and b) show low and high aerosol injection rates, respectively. Panel a) also schematically shows the microphysical steady state achieved by steady aerosol injection rate, followed by aerosol activation, droplet growth in a turbulent environment, and eventual removal by droplet sedimentation. The fluctuations in droplet number density are especially visible in panel b). Turbulent convection transports energy and water vapor from the lower (warm) surface to the upper (cool) surface, both of which are maintained at water saturation. The turbulent mixing of air from the two surfaces leads to a nearly uniform vertical profile of supersaturation. Note that the light sheet is seen from a side perspective and is approximately 2 m in depth. The red dots at the bottom are from the crossing laser beams from the phase Doppler interferometer.

Figure 3.2: Probability density functions (PDFs) for cloud droplet diameter observed under steady-state conditions for five aerosol injection rates. PDFs are shown instead of size distributions in order to emphasize the change in shape rather than the change in number density. The aerosol injection rates shown in the legend correspond to the aerosol source concentrations multiplied by the injection flow rate and divided by the chamber volume so as to obtain volumetric sources within the Pi Chamber. Droplet size distributions are measured with a Dantec phase Doppler interferometer, which measures in the size range of approximately $d = 3$ to $220 \mu\text{m}$ with a resolution of 1-2%.

3.4.2 Origin of droplet size distribution broadening

The appearance of a large-droplet tail with reduced aerosol injection rate is striking, and begs for further interpretation. We consider here two hypotheses for the increasing breadth of the size distribution with decreasing aerosol concentration: cloud droplet collisions and

supersaturation fluctuations.

One possible explanation is that as the aerosol injection rate is reduced, the mean cloud droplet diameter increases and thereby leads to a higher droplet collision rate. As collisions become more frequent a large-droplet, ‘drizzle tail’ appears. We assess this hypothesis by estimating the mean collision time based on the observed size distribution, by using a parcel model, and finally through large eddy simulation of the convection chamber, including water vapor and bin microphysics. The estimated collision times (see Supporting Information) are of order 1 hour or greater. A typical droplet residence time from the chamber τ_{res} is estimated by observing the decay of n in the chamber when the aerosol injection rate is turned off (for the lowest aerosol case, which has an interstitial concentration of nearly zero). We find an exponential decay with time constant $\tau_{res} \approx 9.7$ min, much less than the estimated collision times. Detailed parcel model and LES calculations (Supporting Information) confirm this conclusion that the large droplet tail does not result from droplet collisions.

3.4.2.1 Supersaturation fluctuations

Supersaturation s is derived from two scalar quantities, temperature T and water vapor mixing ratio q_v , which themselves are advected and diffused within the turbulent atmosphere. Thus, in the absence of processes that deplete the supersaturation, such as phase transitions, s will exhibit fluctuations that are the hallmark of all turbulent flows [270].

Indeed, under cloud-free conditions in the Pi Chamber (i.e., no aerosol injection) the supersaturation can be calculated from high resolution, collocated measurements of q_v and T , and the resulting PDF of fluctuations s' shown in Fig. 3.3 is observed to be nearly Gaussian. We now explore whether these supersaturation fluctuations can account for the change in shape of the droplet diameter PDF. From prior work [86, 271?], we anticipate that a cloud can exist in limiting cases of fast and slow microphysical response compared to turbulent mixing. They can be expressed as high and low Damköhler number, $Da = \tau_t/\tau_c$, where τ_t is the turbulence correlation time and τ_c is the microphysical response time. In our experiments the cloud can be pushed toward the fast or slow limits by changing the steady aerosol injection rate. The microphysical response time is the phase relaxation time, which is related to cloud droplet mean diameter and number density as $\tau_c \propto (n\bar{d})^{-1}$. If liquid water content $w \propto n\bar{d}^3$ is constrained to be constant (e.g., due to constant thermodynamic forcing in the convection chamber) then it follows that $\tau_c \propto n^{-2/3}$, i.e., increasing with decreasing cloud droplet concentration and therefore except for highly over-seeded conditions, increasing with decreasing aerosol injection rate. Values for the five experiments are shown in Table 3.1, and this trend is indeed observed to be valid. We can speculate that the supersaturation quickly adjusts to the quasi-steady value for fast microphysics, but that it spends more time away from the quasi-steady value for sluggish microphysics, thereby leading to greater variability in the local droplet condensation growth rates and a broader droplet size distribution. To quantitatively evaluate this hypothesis for existence of the significantly enhanced droplet size variability at low aerosol injection rate we consider analytical solutions to a stochastic differential equation for supersaturation, and we perform large eddy simulations of the turbulent moist convection with cloud represented using bin

Figure 3.3: Supersaturation fluctuation distribution observed with no aerosol injection and therefore no cloud droplets in the chamber. The fluctuations are derived from measurements of water vapor mixing ratio using a LI-COR LI-7500A H₂O analyzer and temperature using a resistance temperature detector. The absolute magnitude of s is susceptible to biases, but the fluctuations s' are significant compared to instrument resolutions. The best fit Gaussian has a standard deviation of 0.014.

microphysics (discussed later).

We extend the stochastic differential equation approach of Sardina et al. [256] to our experimental scenario. Droplet growth due to diffusion can be written as $dr^2/dt = 2\xi s$, from which an expression for the variability in r^2 can be obtained:

$$\frac{d\sigma_{r^2}^2}{dt} = 4\xi \overline{s' r^2}, \quad (3.1)$$

where r , σ_{r^2} , and ξ are the radius of a droplet, RMS r^2 fluctuation, and condensation growth parameter, respectively. Over-bar and prime indicate the mean and fluctuations. Thus the challenge becomes finding an expression for the covariance of droplet size and supersaturation, $\overline{s'r^{2'}}$. To that end we write a stochastic differential equation for the supersaturation

$$ds(t) = s(t + dt) - s(t) = \left[\frac{s_{eq} - s}{\tau_t} - \frac{s}{\tau_c} \right] dt + \left(\frac{2\sigma_{s_0}^2 dt}{\tau_t} \right)^{1/2} \eta(t). \quad (3.2)$$

Here supersaturation $s(t)$ and Gaussian white noise $\eta(t)$ are both continuous random variables but are not differentiable [241]. The first term on the right side of Eq. 4.4 represents a source term due to moist convection that always tends to drive the system toward an equilibrium value of supersaturation s_{eq} . The second term represents the sink due to cloud droplet growth, and the last term is a random fluctuation term (Wiener process) representing variability due to turbulence ($\sigma_{s_0}^2$ and $\eta(t)$ are the supersaturation variance without cloud (cf., Fig. 3.3), and a Gaussian white noise term with mean of 0 and standard deviation of 1, respectively). From this starting point it is possible to obtain differential equations for supersaturation variance and for $\overline{s'r^{2'}}$: $d\overline{s'(t)^2}/dt = -2\overline{s'(t)^2}/\tau_s + 2\sigma_{s_0}^2/\tau_t$, where $\tau_s = \tau_c\tau_t/(\tau_c + \tau_t)$ is the combined time scale of the system, and $d\overline{s'r^{2'}}/dt = 2\xi\overline{s'^2} - \overline{s'r^{2'}}/\tau_s$.

Under steady-state conditions, expressions for $\overline{s'^2}$ and $\overline{s'r^{2'}}$ can then be obtained from the differential equations:

$$\overline{s'^2} = \frac{\sigma_{s_0}^2 \tau_s}{\tau_t} \quad (3.3)$$

and

$$\overline{s' r'^2} = 2\xi \overline{s'^2} \tau_s = \frac{2\xi \sigma_{s_0}^2 \tau_s^2}{\tau_t}. \quad (3.4)$$

Combination of equation 4.3 and 4.14 and integration with time gives the expression for the width of a droplet size distribution,

$$\sigma_{r^2}^2 = \frac{8\xi^2 \sigma_{s_0}^2 \tau_s^2}{\tau_t} \tau_{res}. \quad (3.5)$$

Here τ_{res} is the time available for growth, which for these experiments is the typical droplet residence time in the cloud chamber. Droplets settle out of the chamber with a time scale of approximately 10 min, and at least initially we may assume that the lifetime is independent of the aerosol injection rate. Writing the global time scale as a function of Damköhler number, $\tau_s = \tau_t/(1 + Da)$, we obtain $\sigma_{r^2} \propto \sigma_{s_0}/(1 + Da)$. Thus, the width of the cloud droplet size distribution depends on the relative magnitudes of the turbulent mixing time and the phase relaxation time. The limits $Da \gg 1$ and $Da \ll 1$ therefore result in very different behavior, with the former ‘fast microphysics’ scenario resulting in relatively narrow droplet size distributions and the latter ‘slow microphysics’ scenario leading to relatively wide droplet size distributions. This is qualitatively consistent with the trends observed in

Figure 3.4: Width of the r^2 distribution versus the system time scale $\tau_s = (\tau_c^{-1} + \tau_t^{-1})^{-1}$. The theory predicts a linear dependence between σ_{r^2} and τ_s , and this is observed for all runs except that with much higher aerosol injection rate. A linear fit to the four data points yields slope $88 \text{ m}^2 \text{ s}^{-1}$. Uncertainties in the steady-state cloud properties calculated using the typical Poisson statistic are very small because of the long data samples. Instead, uncertainty is dominated by the turbulent fluctuations. Therefore, the uncertainty is estimated by dividing a steady-state data sample into smaller subsets and, for each subset, all the physical quantities are calculated. Error bars in the figure represent the maximum symmetric deviation of the individual quantity from the mean of all data subsets.

Fig. 3.2 and Table 3.1.

To test the linearity prediction we plot the calculated σ_{r^2} versus $\tau_s = \tau_t/(1 + Da)$ in Fig. 3.4. Except for the point corresponding to the highest aerosol concentration, the linear dependence predicted by Eq. 4.18 is excellent. In contrast, σ_{r^2} is plotted versus $\tau_c \propto Da^{-1}$ in the inset, and the lack of collapse confirms that a simple linear dependence on Da does

not describe the system behavior.

The outlier point corresponds to the experiment with the highest aerosol injection rate and therefore the largest droplet number density n . The aerosol injection rate for that experiment is two orders of magnitude larger than in the other experiments (cf., Table 3.1) and it therefore provides insight into how the simple linear prediction from Eq. 4.18 breaks down. First, it is possible that instrumental artifacts are influencing this data point: due to the very high droplet concentration, n may be underestimated due to coincidence effects; and due to the very small mean droplet diameter, the width of the size distribution is getting closer to the inherent size resolution of the measurement method. Second, the linear prediction $\sigma_{r^2} \propto \tau_s$ depends on the assumption of constant droplet residence time τ_{res} . When aerosol concentration and n are very large, however, we may anticipate that the mean supersaturation in the chamber is suppressed due to strong competition between droplets, therefore resulting in a slow average growth rate and an increase in the residence time. This is corroborated by the transient data, which results in $\tau_{res} \approx 10$ min for all cases except the highest aerosol case, for which τ_{res} is approximately 4 times larger.

The predicted slope for the linear dependence of σ_{r^2} on τ_s is $\sqrt{8}\xi\sigma_{s_0}(\tau_{res}/\tau_t)^{1/2}$. The parameter $\xi = 109 \mu\text{m}^2 \text{s}^{-1}$ is calculated from the thermodynamic properties in the chamber and $\tau_t = 2$ s is obtained from the turbulence measurements. As discussed already, the droplet lifetime $\tau_{res} \approx 9.7$ min. A measurement of σ_{s_0} is also needed, and from Eq. 3.3 it is evident that this is obtained under cloud-free conditions: for no cloud, $\tau_s = \tau_t$ such that

$\overline{s'^2} = \sigma_{s_0}^2$. As shown in Fig. 3.3, the PDF of s fluctuations is consistent with the Gaussian-distributed noise assumption in Eq. 4.4, and has an observed width of $\sigma_{s_0} = 1.4\%$. Using these values from directly measured properties in the cloud chamber, the slope is estimated to be $74 \mu\text{m}^2 \text{ s}^{-1}$. This prediction is compared to the fitted slope in Fig. 3.4 of $88 \mu\text{m}^2 \text{ s}^{-1}$. These agree to within about 20%, which is an encouraging result given the uncertain estimate of τ_{res} and the challenge of measuring water vapor supersaturation necessary for estimation of σ_{s_0} .

3.5 Atmospheric implications

The experiments described here provide strong evidence for aerosol-mediated broadening of cloud droplet size distributions in turbulent environments: clean clouds can reside in a slow microphysics regime in which supersaturation fluctuations are large and lead to broad size distributions, whereas polluted clouds reside in a fast microphysics regime with small supersaturation fluctuations and narrow droplet size distributions. But how general are the results, what are the limiting assumptions, and how relevant is this idealized laboratory result to naturally occurring clouds in the atmosphere? As mentioned already, strong fluctuations of scalars within turbulence is a ubiquitous and general problem [270]. Indeed, it is known that supersaturation is a fluctuating quantity in the turbulent atmosphere [81, 158, 165, 239]. For example, supersaturation variability in the atmosphere, without the influence of cloud droplet activation and growth, can be estimated for one source, the RMS vertical velocity: $\sigma_s \approx \tau_t(ds/dz)\sigma_w$. For warm clouds with $ds/dz \approx 5 \times 10^{-4} \text{ m}^{-1}$

[251], turbulence measurements suggest the ranges $\sigma_w \sim 0.1$ to 1 m s^{-1} and $\tau_t \sim 10$ to 100 s [267], resulting in $\sigma_s \sim 10^{-4}$ to 10^{-2} . Other sources of supersaturation fluctuations include entrainment and isobaric mixing of cloud parcels with different temperatures. These estimates and similar ones in the literature [158, 165, 239, 271] confirm that the Pi Chamber experiment produces fluctuations through isobaric mixing, with a σ_s within the reasonable range for the atmosphere. Unlike the atmosphere, however, in the Pi Chamber σ_{s_0} is fixed, allowing the response of droplets to varying aerosol conditions to be studied in isolation. In the atmosphere, feedbacks between the microphysical evolution and σ_s may exist, e.g., due to varying radiative flux divergence. In this context, the results presented here certainly provide support for similar stochastic differential equation approaches for cloud microphysics [79, 202, 256] and perhaps even for macroscopic cloud properties [285]. It will be intriguing to investigate exactly how the current turbulence-induced aerosol effect relates to that observed in closed parcel studies that consider wide ranges of aerosol concentrations [52, 249].

A key assumption in the derivation leading to the linearity prediction underlying Fig. 3.4 is that τ_c is a constant for a given aerosol injection rate. In reality, fluctuations of n and $\bar{d}$ (where the bar here would represent a local average) lead to a τ_c that varies in time and space [?]. Thus the theoretical approach given here only allows for a $\overline{s'^2}$ that varies between the σ_{s^2} and zero, whereas fluctuations in the integral radius can lead to variance of s greater than σ_{s^2} . The variance of supersaturation is relevant not only to the droplet size dispersion, but also to the number of activated aerosol particles [81]. Therefore the mean (quasi-steady) supersaturation and $\overline{s'^2}$ will together influence both the droplet concentration and the shape

of the size distribution. How significant these effects are, their relative importance, and their relevance for atmospheric clouds, is a topic for further research.

The most central result is the influence of aerosols on broadening of the droplet size distribution, known as the dispersion effect. Our experiments explore the Da range 0.03 to 1.5 (from lowest to highest aerosol injection rate), corresponding to a transition from the very slow microphysics limit to the edge of the fast microphysics regime. What range of Da exists in the atmosphere? For the most typical conditions $\tau_c \sim 1$ to 10 s [158?] and $\tau_t \sim 10$ to 100 s, suggesting that Da can vary from order 100 to 1, thereby residing primarily in the cloud-droplet-dominated, fast microphysics regime. Under very clean conditions, however, with droplet concentrations of order $n \sim 10 \text{ cm}^{-3}$ and a corresponding $\tau_c \sim 100$ s and $Da \sim 0.1$, clouds can reside in the turbulence-dominated, slow microphysics regime. Our results suggest that precipitation formation would be strongly enhanced due to the increase in droplet size distribution width in such clean clouds [181]. Even under relatively polluted conditions, the turbulence broadening mechanism can operate as cloud-processed air is entrained into clouds, thereby reducing the droplet number density without fully evaporating the cloud droplets themselves [261]. Perhaps most compelling is the speculation that the emergence of high aerosol loading resulting from industrialization has fundamentally altered the microphysics of clouds and suppressed their ability to form precipitation.

Finally, we suggest that the results reported here can serve as the basis for physical-based representations of aerosol indirect effects, including the dispersion effect [175, 193, 229], in coarse-resolution models. Added aerosols tend to reduce the phase relaxation time relative

to the turbulence correlation time, and thereby suppress the broadening of the size distribution making development of precipitation difficult. The notion of Da together with the stochastic differential equation for supersaturation allows expressions for the relative dispersion $\sigma_d/\bar{d}$ to be obtained, and this in turn can be coupled with expressions for onset of precipitation that depend on the relative dispersion [87, 181, 203]. The results in Table 3.1 confirm that not only does σ_d increase, but $\sigma_d/\bar{d}$ does as well with reduction of aerosol concentration. Following similar logic as used in obtaining Eq. 4.18, the result $\sigma_{r^2}/\bar{r^2} = \sqrt{2\tau_t/\tau_{res}}\sigma_{s_0}/s_{eq}$ can be obtained. The result is compelling because it is independent of Da , and indeed the relative dispersion (cf., Table 3.1) is observed to be nearly, but not exactly constant (the slight decrease is consistent with an increase in droplet lifetime with increasing aerosol concentration). In the atmosphere an increase or decrease in relative dispersion would depend on how the particle lifetime varies, which is related to precipitation development itself, and therefore becomes a more complex scenario. Nevertheless, this work provides an pathway for quantifying the central result: the width of the droplet size distribution increases with decreasing cloud droplet number density; this in turn can be expected to enhance the cloud albedo susceptibility directly (first indirect effect) and indirectly by accelerating precipitation formation (second indirect effect).

Chapter 4

Influence of Turbulent Fluctuations on Cloud Droplet Size Dispersion and Aerosol Indirect Effects

This chapter is an extension of the work presented in chapter 3 about the aerosol and cloud interactions in a turbulent environment. It is based on a collaborative research published in the Journal of the Atmospheric Sciences [49] ¹.

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4.1 Abstract

Cloud droplet relative dispersion, defined as the standard deviation over the mean cloud droplet size, is of central importance in determining and understanding aerosol indirect effects. In recent work it was found that cloud droplet size distributions become broader as a result of supersaturation variability, and that the sensitivity of this effect is inversely related to cloud droplet number density. The subject is investigated in further detail using an extensive dataset from a laboratory cloud chamber capable of producing steady-state turbulence. An extended stochastic theory is found to successfully describe properties of the droplet size distribution, including an analytical expression for the relative dispersion. The latter is found to depend on the cloud droplet removal time, which in turn increases with the cloud droplet number density. The results show that relative dispersion decreases monotonically with increasing droplet number density, consistent with some recent atmospheric observations. Experiments spanning from fast to slow microphysics regimes are reported. The observed dispersion is used to estimate time scales for autoconversion, demonstrating the important role of the turbulence-induced broadening effect on precipitation development. An initial effort is made to extend the stochastic theory to an atmospheric context with steady updraft, for which autoconversion time is the controlling factor for droplet lifetime. As in the cloud chamber, relative dispersion is found to increase with decreasing cloud droplet number density.

4.2 Introduction

Aerosols play a crucial role in cloud microphysical processes. For example, the number concentration and size distribution of aerosol particles affect the corresponding cloud droplet properties, which in turn affect properties like cloud reflectivity and rate of precipitation formation. Taken together, these factors are known as the first and second indirect effects [8, 305]. The essence of these ideas is that a higher aerosol concentration leads to a greater number of cloud droplets with smaller sizes. These more numerous, smaller droplets reflect more solar radiation and suppress precipitation formation. Aerosol indirect effects are significant elements in the uncertainties of Earth's radiation budget [41, 266].

The aerosol effect on precipitation was first explored experimentally by Gunn and Phillips [110], who found that a clean cloud precipitated faster compared to a polluted one. Albrecht [8] arrived at a similar conclusion and connected the precipitation suppression in a polluted cloud to a decrease in the collision rate. Moreover, he made a connection between the effects of precipitation suppression on cloud albedo and ultimately on climate change. Previous interpretations of precipitation suppression by aerosols have been based primarily on a reduction in the mean droplet size. However, recent studies show clear evidence of a change in relative dispersion with an increase in aerosol loading [86, 175, 187, 193, 199, 346]. Since the droplet collision rate is also a function of relative dispersion, the aerosol effect on relative dispersion will contribute in the cloud lifetime, or second indirect, effect.

Beyond the influence of size dispersion on the precipitation rate, it also directly influences

cloud optical properties [5, 86, 175, 240]. The relative dispersion of the cloud droplet size distribution is defined as $d \equiv \sigma/\bar{r}$, where σ is the standard deviation of the drop size distribution and $\bar{r}$ is the mean droplet radius. It is expected that an increase in the relative dispersion has a warming effect, opposite to the cooling effect induced by an increase in the cloud droplet number [86, 175]. The actual effect of aerosol loading on cloud droplet relative dispersion are contradictory, however; some studies suggest an increase in the relative dispersion with a decrease in the aerosol loading (e.g. [187, 189, 193, 209]) while other observations show the opposite trend (e.g. [175, 199, 223, 229]). Similarly, a theoretical study based on growth by condensation in a cloud parcel also suggested an increase in the relative dispersion with aerosol loading [182]. Chen et al. [52] proposed a regime dependent dispersion effect behavior to reconcile these two contradictory observations.

Recent laboratory experiments [43] have demonstrated a mechanism by which aerosol concentration influences the relative dispersion of cloud droplet size distributions: for high cloud condensation nucleus (CCN) concentrations clouds tend to reside in a fast microphysics regime, where all droplets are exposed to similar thermodynamic conditions and therefore experience similar condensation growth histories. For low CCN concentrations clouds can enter a slow microphysics regime in which thermodynamic fluctuations become large and diverse droplet condensation growth histories lead to broad size distributions. The results are supported by field observations suggesting that large fluctuations in supersaturation even in thin, recently formed clouds can lead to very wide size distributions [275]. Similar effects have also been observed with direct numerical simulation of droplet growth in turbulence [277].

The thermodynamic driving force for cloud droplet growth, water vapor supersaturation, fluctuates within a turbulent cloud environment [239?]. The fluctuating supersaturation field can result from fluctuations in the vertical velocity, entrainment, and isobaric mixing. Apart from the inherent fluctuations, large-scale turbulent structures also cause entrainment of the surrounding air at cloud top and edges. Entrainment and mixing of dry air are associated with a decrease in mean supersaturation and a further increase in fluctuations. In either case, two characteristic time scales can be defined – the turbulent and phase relaxation (or evaporation) time scales [172]. The turbulent mixing time scale is typically the Lagrangian correlation time associated with fluctuations in scalar quantities (τ_t), and the phase relaxation time scale ($\tau_c = (4\pi D_v \bar{r} n_d)^{-1}$) is associated with the exponential relaxation by a group of cloud droplets to a change in the supersaturation field [160]. The relative magnitude of the two is given by the Damköhler number ($Da = \tau_t/\tau_c$) [172].

In this article, we extend our previous work [43] to explore the two aerosol-mediated regimes for turbulence induced broadening of the drop size distribution, and macroscopic cloud properties. In that work it was found that the two regimes are quantified by the dimensionless number Da : The range $Da > 1$ defines relatively fast microphysics, resulting in suppressed supersaturation variability and narrow droplet size distributions; whereas the range $Da < 1$ defines relatively slow microphysics, large supersaturation variability, and broad droplet size distributions. Specifically, in this paper we significantly expand the range of conditions explored, the transition between the two Da regimes, and we consider the implications of relative dispersion for precipitation formation via the autoconversion rate. In order to understand the aerosol effects on the autoconversion rate and optical properties, in section

4.4 we explore the droplet size dispersion in relation to the droplet number concentration and supersaturation forcing by expanding the analytical approach presented in Chandrakar et al. [43]. Moreover, in section 4.5.3 this theoretical framework is tested with experimental results, and some limitations of the theory are confirmed with an expanded range of the data set. To understand the process in an atmospheric context, in section 4.7 we present a simplified theoretical framework for a rising air parcel. This basic approach can be further enriched to incorporate the complexity of atmospheric clouds and can serve as a basis for parameterization of cloud properties. Finally, aerosol effects on autoconversion rate are explored (also in section 4.7) using the experimentally obtained steady-state droplet size distribution and cloud properties estimated from it.

4.3 Experimental Approach

Similar to the work described in Chandrakar et al. [43], the turbulence-induced aerosol effects on clouds are studied by creating a mixing cloud in the Π -Chamber [50]. A buoyancy driven convective flow (Rayleigh-Bénard convection) is created by applying an unstable temperature gradient between the top and bottom surfaces of the chamber. In this study, two different temperature differences ($\Delta T = 16$ and 19 K) are used to obtain two different turbulence intensities, mean supersaturations (in the absence of aerosols), and liquid water contents. The presence of water-saturated top and bottom boundaries leads to a supersaturated environment through isobaric mixing within the the turbulent convective flow. In this steady, supersaturated environment, we inject salt aerosol at a steady rate (~ 1 to

$1000 \text{ cm}^{-3} \text{ min}^{-1}$); a balance between aerosol activation and droplet settling enables steady cloud properties to be generated [43, 50].

The presence of long-duration, steady, and controlled cloud conditions in the II-Chamber makes it ideal for studying cloud microphysics and aerosol interaction. During these conditions, the cloud droplet size distribution is measured using a Phase Doppler Interferometer (DanTech, Accuracy: 1-2 % , reliable size range: 3-220 μm). The collocated measurement of water vapor concentration and temperature are done using a hygrometer (LI-COR, Accuracy: 1%) and an RTD (Minco, Accuracy: 0.1 K). To get a reliable measure of the droplet size distribution and its moments inside the chamber, a long time measurement (~ 200 min) of the steady-state cloud droplet size distribution obtained using the Phase Doppler is made. In a separate study we investigated the influence of spatial and temporal variability of the droplet size distribution on the stochastic condensation process using data from a holographic imaging system [73]. Justified by that work, here, as in Chandrakar et al. [43], we consider the role of the average phase relaxation time.

We extend the work of [43] by significantly increasing the number of experiments at distinct steady-state conditions. In the prior work the range of Da was relatively limited, and the theory developed at that point assumed constant droplet lifetime and therefore diverged from the measurements under polluted conditions. Later in this paper we extend the theoretical analysis to relax that condition, and in order to make a thorough comparison the transition from low to high Da needs to be resolved. It is worth noting that the experiments reported here amount to 200 to 300 minutes of data collection per run (with approximately

24 hours needed for preparation for each run), implying ≈ 5000 minutes of observational data. The region of Da space explored in the experiments is, for a temperature difference of 16 K: $Da = 0.21, 0.27, 0.65, 1.06, 1.29, 1.30, 1.31, 3.89, 4.65$; For a temperature difference of 19 K: $Da = 0.44, 0.63, 2.11, 2.13, 4.71, 8.71, 9.53, 16.60, 24.91, 25.61, 40.23$.

Droplet size distributions that result from different aerosol injection rates for $\Delta T = 16$ K are shown in Figure 4.1. The observations confirm our previous finding [43], now with a more complete data set and over a wider range of conditions; Specifically, Figure 4.1 shows that the droplet size distribution mode and width increase with a decrease in the aerosol loading, or more directly, with the resulting droplet number concentration. The new results further suggest that the width of the droplet size distribution saturates (*i.e.* it does not respond to a increases in the droplet number concentration above some level). Once that occurs, only the mode diameter decreases, which is an indication of the supersaturation limited regime, where asymmetry of the droplet growth and evaporation limits the response of the droplet size distribution width.

4.4 Analytical Approach for Condensational Growth in a Turbulent Environment

In a quest to understand our experimental observations of turbulence induced broadening of the drop size distribution, we have devised a theoretical framework based on previous studies [43, 88, 129, 224, 256?]. The analysis is based on stochastic differential equations,

Figure 4.1: Steady-state droplet size distributions: each of the distributions represents a different steady-state droplet number concentration for a temperature difference in the chamber of 16 K. The corresponding Damk ohler numbers are 0.21, 0.27, 0.65, 1.06, 1.29, 1.30, 1.31, 3.89, 4.65 (in order of increasing n_d).

which are used to model scalar fields in a turbulent environment [241], where scalars are randomly fluctuating on a wide range of scales (dissipation to integral). Stochastic growth of cloud droplets by condensation in a turbulent environment is analogous to a random walk, but in r^2 space [92, 128]. The derivation was only outlined by [43], so we provide more complete details here, as well as extending it to atmospheric conditions.

Cloud droplet growth due to condensation can be expressed as [251]:

$$\frac{dr^2}{dt} = 2\xi s. \quad (4.1)$$

For an ensemble of cloud droplets, expressions for the mean and fluctuation in droplet size

can be written as:

$$\frac{d\overline{r^2}}{dt} = 2\xi\overline{s} \quad (4.2)$$

$$\frac{d\sigma_{r^2}^2}{dt} = 4\xi\overline{s'r'^2} \quad (4.3)$$

where r , σ_{r^2} , ξ , and s are the radius of a droplet, the RMS fluctuation in r^2 , the growth parameter, and supersaturation respectively. Over-bar and prime indicate mean and fluctuating quantities. In the above equations, the growth parameter ξ is assumed to be a function only of the mean temperature; its dependences on temperature and water vapor fluctuations are weak and thus those terms are neglected. Equation 4.3 indicates that the growth of the fluctuations in droplet size depend upon the correlation between fluctuations in s and r^2 . Subsequent steps are dedicated to finding a simplified relation for this correlation term and to estimate the evolution of the mean and fluctuations in the supersaturation.

4.4.1 Stochastic differential equation for a turbulent supersaturation field

In a turbulent environment, cloud droplet growth after activation is nearly independent of the small scale fluctuations. This is true if the Lagrangian turbulent correlation time, τ_t , is short compared to the droplet condensation growth time scale, $\overline{r^2}/2\xi\overline{s}$ [277]. In this case, the largest scale of turbulent fluctuations (integral scale) is the dominant scale for fluctuations

in the scalar fields (e.g., supersaturation) to interact with droplet condensational growth. It is analogous to the small scale, inter-particle interactions in Brownian motion; in that case, if the molecular interaction time scale is much shorter compared to the process of interest, the molecular process can be modeled as random noise [92, 128].

For most of the time after droplet activation, the droplet condensation growth time scale is large compared to the correlation time scale of turbulence due to the small value of the mean supersaturation in clouds. Consequently, we modeled the supersaturation equation using a stochastic differential equation with the integral time scale as the supersaturation correlation time scale; all smaller scale fluctuations are considered as Gaussian random noise. The justification for using Gaussian distributed supersaturation fluctuations is the behavior of scalar fluctuations in the turbulent field. Scalar fluctuations in a turbulent environment are nearly Gaussian distributed with slight deviations near tails due to intermittency [241, 270]. Indeed, a nearly Gaussian supersaturation field has been reported in recent work [43, 275]. We take supersaturation forcing as a Gaussian distribution as a first approximation for droplet growth. The overall approach is evaluated by comparison with the laboratory measurements later in the paper.

The source of the supersaturation in the Π -chamber is due to turbulent mixing, and the sink is conversion of vapor to liquid water (*i.e.* droplet growth). Fluctuations in the vapor and/or temperature field perturb s from its equilibrium value. It then tends to relax back exponentially with time scale, τ_t . Depletion of s as droplets grow also has an exponential time scale, τ_c . With this framework in mind, the governing equation for supersaturation in

a turbulent environment for cloudy conditions can be modeled as [92, 241]:

$$ds(t) = \left[\frac{s_o - s}{\tau_t} - \frac{s}{\tau_c} \right] dt + \left(\frac{2\sigma_{s_o}^2 dt}{\tau_t} \right)^{1/2} \eta(t). \quad (4.4)$$

Here supersaturation $s(t)$ and Gaussian white noise $\eta(t)$ are both continuous random variables, but are not differentiable. Therefore, the tools of the Ito calculus are used in the derivation that follows. A list of all symbols with a brief explanation of their meaning is in Appendix A.

As discussed above, the first term on the right side of Eq. 4.4 is the drift or source term due to turbulent mixing and the second represents cloud droplet growth. The last term is a Wiener process (the random fluctuation term). τ_t , s_o , $\sigma_{s_o}^2$ and $\eta(t)$ are the turbulent correlation time scale (turbulent mixing time), equilibrium supersaturation without a cloud, the variance in supersaturation without a cloud, and Gaussian white noise (with a mean of 0 and standard deviation of 1) respectively. In this equation, we assume homogeneity in the turbulence and supersaturation fields. As discussed above, τ_c is the phase relaxation time and $\bar{r}$, n_d , and D_v are the mean radius, total droplet number concentration, and a modified water vapor diffusion coefficient (accounting for the latent heat release by droplet during condensation) respectively. Fluctuations of D_v due to fluctuations in the temperature and water vapor field are neglected. Also, we assumed a uniform phase relaxation time throughout the volume; a homogeneous cloud droplet sink for water vapor is assumed. The impact of assuming constant τ_c in the droplet growth is the topic in another, parallel study [73].

Equations for the mean supersaturation and its fluctuations can be derived from Eq. 4.4:

$$d\bar{s}(t) = \left[\frac{s_o - \bar{s}}{\tau_t} - \frac{\bar{s}}{\tau_c} \right] dt \quad (4.5)$$

and

$$ds'(t) = \left[-\frac{s'}{\tau_t} - \frac{s'}{\tau_c} \right] dt + \left(\frac{2\sigma_{s_o}^2 dt}{\tau_t} \right)^{1/2} \eta(t). \quad (4.6)$$

We now wish to obtain terms related to the supersaturation variance $\overline{s'^2}$. Ito calculus gives [92, 128]

$$d(s'^2) = 2s'ds' + (ds')^2 \quad (4.7)$$

We substitute the value of ds' from equation 4.6 into Eq. 4.7 and then average:

$$\begin{aligned} d(\overline{s'^2}) = & - \left[\frac{2\overline{s'^2}}{\tau_t} + \frac{2\overline{s'^2}}{\tau_c} \right] dt + 2 \left(\frac{2\sigma_{s_o}^2 dt}{\tau_t} \right)^{1/2} \overline{s'\eta(t)} + \left(\frac{2\sigma_{s_o}^2 dt}{\tau_t} \right) \overline{\eta(t)^2} \\ & - 2 \left[\frac{\overline{s'\eta(t)}}{\tau_t} + \frac{\overline{s'\eta(t)}}{\tau_c} \right] \left(\frac{2\sigma_{s_o}^2 dt}{\tau_t} \right)^{1/2} dt + \mathcal{O}(dt^2). \end{aligned}$$

Since $s'(t)$ and $\eta(t)$ are independent at any time, t , (*i.e.* the increment in s' is independent of its past and current value), and $\eta(t)$ is δ -correlated with itself, all the $s'(t)$ and $\eta(t)$ correlation terms will be zero and $\overline{\eta(t)^2} = 1$. Furthermore, all the higher order dt terms can be neglected in the limit of $dt \rightarrow 0$. The final, averaged differential equation is then:

$$\frac{d\overline{s'^2}}{dt} = -2 \left[\frac{\overline{s'^2}}{\tau_t} + \frac{\overline{s'^2}}{\tau_c} \right] + \left(\frac{2\sigma_{s_o}^2}{\tau_t} \right). \quad (4.8)$$

The three terms on the right side of the equation describe how the rate of change of supersaturation variance is related to turbulent relaxation to the mean, cloud droplet growth, and the turbulent fluctuation source term, respectively. We can see that with no cloud droplets and in steady state, $\overline{s'^2} = \sigma_{s_o}^2$, as expected.

In order to obtain the width of the cloud droplet size distribution, Eq. 4.3 makes it clear that we need to obtain the correlation of s' and $r^{2'}$. This is written as

$$\begin{aligned} d(s'r^{2'}) &= s'(t+dt)r^{2'}(t+dt) - s'(t)r^{2'}(t) \\ &= (s'(t) + ds'(t)) (r^{2'}(t) + dr^{2'}(t)) - s'(t)r^{2'}(t). \end{aligned} \quad (4.9)$$

Substituting the expression for $ds'(t)$ and $dr^{2'}(t)$ from Eq. 4.6 and Eq. 4.1 in the preceding equation results in:

$$\begin{aligned} d(s'r^{2'}) &= \left(s'(t) + \left[-\frac{s'}{\tau_t} - \frac{s'}{\tau_c} \right] dt + \left(\frac{2\sigma_{s_o}^2 dt}{\tau_t} \right)^{1/2} \eta(t) \right) \\ &\quad \left(r^{2'}(t) + 2\xi s'(t)dt \right) - s'(t)r^{2'}(t). \end{aligned} \quad (4.10)$$

Similar to the procedure used for Eq. 4.8, we average and eliminate all the cross-correlation terms with $\eta(t)$. Now, if we divide both sides by dt and take the limit $dt \rightarrow 0$, the $(dt)^2$ term will be zero, and the final differential equation for the average is

$$\frac{d\overline{s'r^{2'}}}{dt} = 2\xi\overline{s'^2} - \frac{\overline{s'r^{2'}}}{\tau_t} - \frac{\overline{s'r^{2'}}}{\tau_c}. \quad (4.11)$$

Physically, this equation describes the rate of change of the cross-correlation between supersaturation and droplet size fluctuations, resulting from supersaturation variability and droplet growth in a turbulent cloud.

4.4.2 Quasi steady-state solution

In quasi-steady-state (assuming the temporal variation of the phase relaxation time is slow), expressions for $\overline{s}$, $\overline{s'^2}$ and $\overline{s'r^{2'}}$ can be written from Eqs. 4.5, 4.8, and 4.11 as:

$$\overline{s} = \frac{s_o\tau_s}{\tau_t} \quad (4.12)$$

$$\sigma_s^2 = \frac{\sigma_{s_o}^2\tau_s}{\tau_t} \quad (4.13)$$

and

$$\overline{s'r^{2'}} = 2\xi\overline{s'^2}\tau_s = \frac{2\xi\sigma_{s_0}^2\tau_s^2}{\tau_t}, \quad (4.14)$$

where $\tau_s = \tau_c\tau_t/(\tau_c + \tau_t)$ is the global system time scale. By substituting the above values of the mean supersaturation and fluctuation correlations into Eqs. 4.2 and 4.3, we get the final, simplified expressions for droplet growth in a turbulent environment [43]:

$$\frac{d\overline{r^2}}{dt} = \frac{2\xi s_o \tau_s}{\tau_t}, \quad (4.15)$$

and

$$\frac{d\sigma_{r^2}^2}{dt} = \frac{8\xi^2\sigma_{s_o}^2\tau_s^2}{\tau_t}. \quad (4.16)$$

Assuming a short growth time or a quasi-steady supersaturation field, the variation of the system time scale τ_s will be negligible, and Eqs. 4.15 and 4.16 can be integrated:

$$\overline{r^2} = \frac{2\xi s_o \tau_s}{\tau_t} t \quad (4.17)$$

and

$$\sigma_{r^2}^2 = \frac{8\xi^2\sigma_{s_o}^2\tau_s^2}{\tau_t} t. \quad (4.18)$$

These equations predict that the mean and the width of r^2 increase with time. To make a quantitative comparison with our measurements, we first consider the lifetime of a droplet

in the chamber, which can be estimated as:

$$\tau_l = \left(\frac{H}{a\xi s_o} \right)^{1/2} \left(\frac{\tau_t}{\tau_s} \right)^{1/2}. \quad (4.19)$$

(See A.2 for the details of the derivation and complete explanation of symbols in the equation.)

Note that Eq. 4.19 is the estimate for the lifetime of a single droplet while Eqs. 4.17 and 4.18 are expressions for the distribution of droplets in the chamber. It is the distribution of lifetimes that is needed for the comparison of theory and measurements. As a first-order approximation, we use an average lifetime $\tau_l/2$. Thus the average value of r^2 to compare with our measurements becomes

$$\overline{r^2} = \frac{\xi s_o \tau_s \tau_l}{\tau_t}. \quad (4.20)$$

While consideration of the average droplet lifetime in the chamber is sufficient for a comparison of the measurements of $\overline{r^2}$ to the theory, a further refinement is needed for the second (or higher) moment. Specifically, the theory treats droplet radius square as a Gaussian random variable and therefore the radius is not inherently positive definite, while of course in the physical system droplet radius is bounded by zero. Examination of the equations for the supersaturation and r^2 show that both quantities can be negative. Although the chamber can be subsaturated, droplets must have a size greater than or equal to zero. Therefore,

negative trajectories (in the theory) for r^2 lead to unrealistically high values for the width of the distribution.

Figure 4.2: An example of the measured r^2 distribution and its comparison with the Gaussian fit (with mode $\mu = 0$). Some of the data point at the lower range are excluded for a better fit since they have higher measurement uncertainties.

Both the variation in droplet lifetime and the constraint of a positive droplet size tend to produce a measured distribution which peaks close to zero. (Droplets close to the beginning of their lifetime and droplets experiencing subsaturation along their local trajectory will have a size close to zero.) Therefore, as a first approximation, one can calculate the second order moment of the r^2 distribution using a truncated Gaussian distribution. Because the mean supersaturation is typically small in our experiments, the peak of the size distribution is close to zero and we can use a half Gaussian. Figure 4.2 shows a measured r^2 distribution with a corresponding Gaussian distribution fit with mode $\mu = 0$. The measured distribution fits the half Gaussian distribution reasonably well, and reinforces this approach for obtaining σ_{r^2} . Therefore, a final corrected form of the σ_{r^2} equation, beyond Eq. 4.18 is:

$$\sigma_{r^2}^2 = \frac{4\xi^2\sigma_{s_o}^2\tau_s^2\tau_l}{\tau_t} \left(1 - \frac{2}{\pi}\right). \quad (4.21)$$

Here, the correction factor for σ_{r^2} is obtained by calculating the ratio between the variance of full Gaussian distribution and same distribution with the lower limit of 0 instead of $-\infty$.

4.5 Comparison of Stochastic Theory and Experiments

Comparison of the stochastic theory to experiments will allow its validity to be assessed or, more likely, will help reveal the conditions under which the assumptions built into the analysis are reasonable, and when those assumptions begin to fail. As part of the effort to evaluate why the theory may deviate, we use a numerical simulation of the stochastic equations that allows more detail to be included regarding droplet removal by sedimentation, and to impose the positive-definite radius constraint. The model is described first, and then measurements are compared in the two following sub-sections.

4.5.1 Numerical simulation of the stochastic model

Incorporating physical constraints such as the distribution of droplet lifetimes and positive values of droplet radius in the mathematical model requires some approximations that should be checked. In addition to analytical solution, another approach to address these problems is to use a numerical model based on set of stochastic differential equations. In

a numerical approach, physical constraints like variation in droplet growth history due to settling and new droplet activation, and positive droplet size, can be directly implemented. The numerical model will allow us to evaluate when conditions are reached under which the analytical solutions break down, and can serve as a bridge between the theory and the measurements. Therefore, we have developed a computational stochastic model based on the Lagrangian supersaturation equation 4.4 for each particle trajectory. In this model, similar to the II-chamber experiments, a steady droplet number is forced, by allowing droplets to activate and settle (details of the activation process are ignored, i.e., droplets are simply generated at a rate). All droplets are assumed to fall with the Stokes terminal settling speed and are removed once they reach the bottom boundary; the effect of the turbulent environment in droplet settling is ignored. Moreover, the spatial distribution of particles in the vertical dimension is assumed random at the start, and whenever there is a new activation, the droplet's vertical position is assigned randomly. Each droplet is allowed to grow or evaporate based on the current supersaturation value determined by the Lagrangian supersaturation Eq. 4.4. The input parameters τ_t , τ_c , s_o and σ_{s_o} are kept constant during individual runs, as is expected to be the case for the steady-state experimental conditions. The stochastic equivalent of Euler's method (also called the 'EulerMaruyama method') is used for the numerical simulation [128].

In order to compare the stochastic simulation with steady experimental data, the model is run with same τ_c value as in the experiment, until a steady state is reached. Then the model output is processed similar to the experiments to allow direct comparison. At a fixed instance, during the steady period, the droplet size distribution is obtained from the

model output. All droplets below $5\mu m$ are removed from both the measured data (close to the instrument lower cutoff, which can vary slightly due to condensation on windows, etc.) and from the model output. The r^2 moments measured as a function of τ_c are fitted with model output using the grid search method [124]. In this method, model is run for a range of turbulent and thermodynamic parameters (i.e. τ_t , s_o and σ_{s_o}) in the neighborhood of the physically expected value; a best-fit curve and corresponding optimal parameters are obtained by searching for the minimum residual.

Figure 4.3: Variation of mean droplet size ($r_m^2 / \overline{r^2}$) with the phase relaxation time scale (τ_c) for 19K (**left**) and 16K (**right**) cases. Measurements are shown as red circles. The analytical expression from the stochastic theory is shown as a dashed line, and the more detailed computational solution, processed as the measurements, is shown as black circles.

4.5.2 Experiment and theory: $\overline{r^2}$ and σ_{r^2}

In this part, results from measurement, theory, and model for the mean and second moment of the size distribution are compared. Before comparison, it should be kept in mind that

Figure 4.4: Dependence of the droplet size distribution width ($\sigma_{r,2}$) on the phase relaxation time scale (τ_c) for 19K (**left**) and 16K (**right**) cases. Measurements are shown as red circles. The analytical expression from the stochastic theory is shown as a blue dashed line, and the modified analytical expression (described in the text) as a black dashed line. The more detailed computational solution, processed as the measurements, is shown as black circles.

both experimental measurement and model only contain droplets with $D \geq 5 \mu\text{m}$. However, the theoretical results have not been so constrained; therefore, the τ_c and moments of droplet size distribution will be expected to be different on this basis alone, in addition to other limitations. Figures 4.3 and 4.4 display the dependence of the moments of the droplets size distribution on τ_c . Alternatively, the dependence on τ_s could be plotted, but we have chosen τ_c because it is more directly tied to the microphysical measurements. The plots show that both $\overline{r^2}$ and $\sigma_{r,2}$ increase with τ_c due to the decrease in competition for water vapor as expected from the theoretical prediction. Larger values of τ_c represent a cleaner cloud (resulting from the lower droplet concentration, assuming fixed liquid water content), hence mean and fluctuations in supersaturation are expected to be higher, and that, in turn, drives a larger mean and fluctuation in droplet size. It can also be observed that the measurement, model, and theory all exhibit similar trends, and that the model result indeed

matches the measurements better than the theory. The model points fit the measurements quite well for the mean values $\overline{r^2}$, but deviate somewhat for the second moments $\sigma_{r,2}$. The optimal value of the model parameters τ_t , s_o and $\sigma_{s,o}$ for these fits are $37.8 - 40$ s, $1.96 - 2.5$ % and $1.50 - 1.82$ %, respectively, for the 19-K experiment (48.2 s, $0.1-0.2$ % and $0.55-0.63$ % for 16-K experiment). These parameter ranges are obtained by fitting the $\overline{r^2}$ and $\sigma_{r,2}$ data separately. In some cases, the fits give a similar value of the parameter, so it is listed as a single value instead of range. These values are obtained using the grid search method for optimal fitting as explained in the previous subsection, and the values of the parameters are all within the physically-reasonable range. Uncertainties in the measurement are obtained by dividing steady-state time series into smaller time bins (but still much greater than the large eddy timescale) and calculating the corresponding standard errors. Deviations between measurements and model, specifically at the lower size range, are due to larger uncertainties in determining accurate values of measured droplet number concentration. At the lower size range, estimation of the droplet number concentration using the PDA (Phase Doppler Anemometry) is influenced by the lower signal strength and detection limit of PDA.

The parameters obtained through the fitting of the model are also used for obtaining the theory results, and those are plotted as lines together with the experimental and model results. For $\overline{r^2}$ we plot Eq. 4.20. For $\sigma_{r,2}$ we plot the Eq. 4.18 for the unmodified theory, i.e., that allowing for trajectories with ‘negative radius,’ with Eq. 4.19 used for the lifetime; and Eq. 4.21 for the approximate, ‘half-Gaussian’ modification of the theory that allows only trajectories with positive-radius. There are some differences in magnitude between

theoretical results and measurements, specifically for $\sigma_{r,2}$, due to the previously-discussed limitations of negative droplet trajectories and different droplet growth history. The correction for these effects on $\sigma_{r,2}$ (eq. 4.21) bring the theory closer to the measurement, however, further improvement will be needed. We expect that the deviations from the observations, even for the model that accounts for the limitations in the PDA size range, may come from several sources. First, as discussed earlier, the approximation of Gaussian distributed supersaturation fluctuations needs to be further investigated, considering that the theory and model depend on Lagrangian rather than Eulerian sampling for the distribution. Second, and related to the first point, the spatial distribution of supersaturation needs to be considered in more detail. For example, the influence of wall effects in the chamber and their influence on supersaturation variability likely cause deviations from the Gaussian model. Third, the droplet lifetime, both mean and distribution, needs to be investigated because it now rests on an idealized analytical model. Such an investigation will be challenging from an experimental perspective because it requires Lagrangian tracking of cloud droplets over long times. Fourth, the time scale τ_t needs to be more thoroughly understood. In prior work we have assumed it is approximately equivalent to the Eulerian large-eddy correlation time, although the theory requires a Lagrangian correlation time. From a system-level perspective of the governing equation (Eq. 4.4), the first term suggests that τ_t is a relaxation time and therefore could be related to the large-scale circulation in the chamber rather than the local turbulence properties. Indeed, the τ_t values obtained above are consistent with that interpretation: specifically, a path around the outermost part of the chamber has length of order 5 m and the observed flow speeds are of order 10 cm s⁻¹, resulting in a circulation time of 50 s. Investigations into the nature of the large-scale circulation are ongoing [216].

The reasonable agreement between theory and measurement implies that the basic approach is sound, and that the implications of the theory for clean versus polluted cloud regimes are valid. These regimes are achieved in the limits of $Da \ll 1$: faster turbulent mixing compared to microphysics, and uniform cloud properties, versus $Da \gg 1$: slower turbulent mixing compared to microphysics and non-uniform supersaturation and cloud properties. The role of Da can be appreciated through its contribution to the global system time scale $\tau_s = \tau_t / (1 + Da)$, where again $Da = \tau_t / \tau_c$. In the current study, based on the stochastic model fitting, we expect $\tau_t \approx 40$ s (for 19K temperature difference) and 48 s (for 16K temperature difference) as explained earlier. Therefore, it suggests that there is a crossover between the clean ($Da < 1$) to polluted ($Da > 1$) cloud regime around $\tau_c \approx 40$ s in Figs. 4.3 and 4.4. However, the exact value of this transition point is uncertain because of the instrument limitations and uncertainties associated with the τ_t estimation. Nevertheless, there is a clear distinction between the relatively clean and polluted cloud regime. In the clean cloud regime, both, $\overline{r^2}$ and $\sigma_{r,2}$ are relatively high compared to the polluted regime. We expect that these physical regimes will exist in the atmosphere as well. In the atmosphere, unlike in a comparatively small chamber however, clouds are not restricted to a small domain and therefore the temporal evolution of the system time scale will be determined by other processes. These are considered in more detail in Sec. 4.7.

4.5.3 Aerosol effect on relative dispersion

Cloud radiative properties and droplet collision rates are both functions of relative dispersion. Therefore, understanding the aerosol effects on droplet size dispersion is important for the atmospheric radiation budget and understanding precipitation formation process. The relative dispersion for droplet area (d_a) can be obtained directly from Eqs. 4.21 and 4.20, with the resulting form:

$$d_a = \frac{\sigma_{r^2}}{r^2} = \sqrt{\frac{2\tau_t}{t} \frac{\sigma_{s_0}}{s_0}} \sqrt{1 - \frac{2}{\pi}}. \quad (4.22)$$

It is worth noting that the relative dispersion for droplet area is approximately twice the relative dispersion for droplet radius (d) for a Gamma radius distribution. (It can be theoretically shown for a Gamma distribution with $d < 1$). In addition, our measured droplet distributions are close to a Gamma distribution. Therefore, $d_a \sim 2d$ is a reasonable approximation and can be verified from the measurements. Eq. 4.22 suggests that the relative dispersion is independent of the phase relaxation time (and Damköhler number), but it does depend on the droplet growth time, which is a function of τ_s in the case of the Π chamber.

A simplified expression for the relative dispersion can be obtained from Eq. 4.22 after substituting the expression for droplet lifetime t from Eq. A.2:

$$d_a = \sqrt{4\tau_t \left(\frac{a\xi s_o}{H} \right)^{1/2} \frac{\sigma_{s_o}}{s_o} \left(\frac{\tau_s}{\tau_t} \right)^{1/4} \sqrt{1 - \frac{2}{\pi}}}. \quad (4.23)$$

The dependence of relative dispersion on lifetime results in an implicit dependence on the mean supersaturation due to the role of condensation growth and sedimentation in removing droplets from the II chamber. Since $\bar{s}$ is a function of τ_s and τ_t (Eq. 4.12), the lifetime of a cloud droplet will also depend on τ_s and τ_t . In this case, τ_t is fixed hence the relative dispersion increases with $\tau_c \propto n_d^{-1}$ (and decreases with n_d). For a relatively clean cloud (larger τ_c), Eq. 6.4 suggests that the relative dispersion decreases approximately linearly with the droplet number concentration. Similarly, in the atmosphere, the lifetime of a droplet, τ_l , is constrained by different loss mechanisms like collisions with other droplets, sedimentation, and evaporation. The onset of these processes also depends on the supersaturation experienced by a cloud droplet; therefore, the lifetime of a cloud droplet in an atmospheric cloud will also be a function of τ_s . Consequently, droplet size dispersion will be a function of τ_s . Moreover, continuous growth of a cloud droplet size distribution will result in a time dependent phase relaxation time. As a result, the final expression for the droplet size dispersion will contain τ_s dependent terms apart from the lifetime.

Figure 4.5 shows the variation of relative dispersion with droplet number concentration. Here, we have plotted d versus n_d instead of τ_c in order to be consistent with and to allow direct comparison with previous literature. Moreover, τ_c depends on both $\bar{r}$ and n_d ,

Figure 4.5: Relation between relative dispersion of the droplet radius distribution and the droplet number concentration: steady-state values of the relative dispersion at different n_d (corresponding to different aerosol loading) for 19 K (**left**) and 16 K temperature differences (**right**).

but measurements show that the change in n_d dominates changes in the magnitude of τ_c . Therefore, a smaller n_d represents the relatively clean cloud condition. In this plot it is clearly observed that relative dispersion decreases with increasing droplet concentration (and corresponding aerosol loading). A decrease in the relative dispersion with droplet number concentration is consistent with the stochastic model coupled with the expression for droplet lifetime, developed earlier.

Figure 4.5 summarizes the dispersion effect as observed in the turbulent-cloud experiments, with d decreasing with increasing n_d . It is consistent with some atmospheric observations [187, 189, 193, 209], but it should be emphasized that the behavior observed in the cloud chamber experiments is based on condensation growth alone, with no observable influence of coalescence growth. In that respect, it is of interest that the behavior contrasts with

results of studies based on adiabatic condensation growth where supersaturation fluctuations are not included [52, 182, 344]. In an adiabatic cloud parcel (without turbulence), a higher CCN concentration leads to a reduced narrowing of a droplet size distribution (relative to the low CCN case) due to decrease in the mean, quasi-steady supersaturation and the corresponding droplet growth rate. This is a consequence of the inverse radius dependence of the droplet radius growth rate ($dr/dt = \xi s/r$). In essence, in the absence of fluctuations, droplet size dispersion decreases with an increase in the mean supersaturation. For example, Yum and Hudson [344] show more broadening for low supersaturation environments. The rate of change of size dispersion due to stochastic condensation, as expressed by Eq. 4.16, in contrast, has no dependence on mean supersaturation. Rather, the size dispersion increases in proportion with variability in supersaturation σ_{s_0} . The result is that stochastic condensation can produce much larger magnitudes of relative dispersion compared to adiabatic condensation alone. Indeed, when compared with observations, the latter are always significantly higher than the calculations for adiabatic growth [344].

From the perspective of relative size dispersion, this ‘condensation narrowing effect’ for a closed parcel is compounded because the numerator σ_r decreases with time (as discussed in the previous paragraph) and the denominator $\bar{r}$ increases with time. A starting relative dispersion determined by the CCN spectrum will rapidly decrease due to growth in a uniform supersaturation field. In a turbulent environment with supersaturation variability, each droplet experiences a different Lagrangian supersaturation history. As a result, droplet growth is analogous to a random walk in r^2 space and, like any Brownian process, the dispersion increases with time. The behavior of the relative dispersion for stochastic

condensation, as for adiabatic condensation, also depends on mean supersaturation and corresponding mean droplet growth. This is apparent in Eq. 6.4, which shows that the relative dispersion is inversely proportional to the mean supersaturation.

4.6 Autoconversion Rate: Impact of Aerosol and Turbulence on Precipitation

Increased CCN concentration can affect cloud lifetime by suppressing precipitation formation through the warm rain process [86]. An increase in aerosol loading suppresses the mean supersaturation, which results in a decrease in the mean droplet size, which in turn reduces collisions among droplets. However, recent work [43, 44, 73, 275] and the analysis presented above suggest that a turbulent environment induces fluctuations in the supersaturation, and that τ_c determines how the width of the droplet size distribution responds to the fluctuations. Because τ_c depends on the number density of cloud droplets, it is therefore influenced by the CCN concentration. (Note that the aerosol activation rate is also a function of the mean supersaturation and fluctuations [44].) In this section we explore this effect on cloud lifetime by estimating the autoconversion rate of cloud water content ($P_L = dL/dt_{\text{cloud}}$) as a function of aerosol concentration. Though we cannot directly investigate collision-coalescence in the clouds in the Π Chamber, the autoconversion rate can nevertheless be calculated. It gives us an estimate of the rate at which cloud water content would be expected to transform to a category of rain water content due to droplet collisions, if sufficient vertical extent were available.

The droplet size distributions obtained from the experiments with variable aerosol concentrations are used to estimate the autoconversion rates based on a gamma size distribution fit, using an expression for a gamma size distribution and the Long collision kernel given by Liu et al. [183] and Xie and Liu [332]. In this estimate, effects of the turbulent flow field on the droplet collision process are ignored. The critical radius for starting the autoconversion process is assumed to be a constant; it is taken as the average value for a marine cloud, $r_{cr} = 10.3\mu m$ [180]. With those assumptions,

$$P_L = \left(\frac{3}{4\pi\rho_w} \right)^2 k_l \frac{\Gamma(\alpha_d)\Gamma(\alpha_d + 3, x_{cq})\Gamma(\alpha_d + 6, x_{cq})}{\Gamma^3(\alpha_d + 3)} \frac{L^3}{n_d}, \quad (4.24)$$

where, $x_{cq} = r_{cr}/\beta_d$, $k_l = 1.9 \times 10^{11} \text{ cm}^{-3} \text{ s}^{-1}$ is the coefficient for the Long collision kernel, α_d and β_d are shape and scale parameters of the gamma distribution, and Γ is the gamma function. We define the autoconversion timescale for cloud liquid water content as $\tau_a \equiv L/P_L$.

We fit the measured cloud droplet radius distributions to obtain the corresponding gamma parameters. These parameters are then used in Eq. 4.24 to obtain the autoconversion rate and corresponding time scale. Figure 4.6 shows the dependence of autoconversion time scale on the steady-state droplet number concentration for clouds in the chamber. For relatively clean cloud cases (the lower droplet number concentration range), the autoconversion time scale is on the order of 10 hours. This time scale is much larger than the typical cloud droplet lifetime, consistent with the prior conclusion that collision-coalescence can be neglected for

Figure 4.6: Autoconversion timescale ($\tau_a = L/P_L$) as a function of droplet number concentration for steady-state cloud droplet size distributions: 19 K (**left**) and 16 K (**right**) temperature differences.

similar conditions in the cloud chamber [43]. However, as n_d transitions to the polluted cloud regime, the autoconversion timescale increases sharply to a value on the order of 10^{13} hours. The specific values are not as significant as the relative change: increase in cloud droplet concentration by a factor of 10 to 100 leads to an increase by many orders of magnitude in the expected time for collision-coalescence to convert the cloud droplets to precipitation.

This behavior of the autoconversion rate is consistent with the relative dispersion trend. We showed in the previous section that the mean and width of the droplet size distribution increase with the system time scale (decrease with droplet number concentration); therefore, the observed change in the autoconversion rate with number concentration is due to changes in both the mean droplet size and fluctuations. It is also noticeable that the autoconversion

rate is higher, leading to smaller τ_a , for the larger temperature gradient (higher supersaturation forcing). This follows from the fact that the mean droplet size and fluctuations are proportional to the supersaturation forcing. The large magnitudes of autoconversion time scale for both clean and polluted regimes shown in the figure are due to the limited growth of the cloud droplet distribution and lower liquid water content in the constrained chamber environment. However, the main purpose of this study is to understand the effect of aerosol loading in the autoconversion process which is clear from the overall trend. In clouds in the atmosphere, the functional dependence on fast and slow microphysics regimes should be similar, but the autoconversion times lower because of the unconstrained growth and higher liquid water contents.

4.7 Analytical Approach Extended for Atmospheric Applications

The analysis presented thus far has been for clouds formed through isobaric mixing, the primary cloud formation process in the II Chamber. Atmospheric clouds, of course, can form through isobaric mixing, but cloud formation through adiabatic ascent is also common. Here, we develop a simple model of stochastic condensation for such conditions. Our aim is not to capture the full complexity of clouds in the atmosphere, but to present the basic physical mechanisms behind cloud droplet growth in a turbulent, ascending parcel, and to consider the implications for relative dispersion, precipitation formation and cloud optical properties. Similar to the approach discussed above for the chamber, a stochastic differential

equation for supersaturation in a rising cloud parcel (of a mean vertical velocity $\bar{w}$) can be written as:

$$ds(t) = \left[\alpha \bar{w} + \frac{\bar{s} - s}{\tau_t} - \frac{s}{\tau_c} \right] dt + \left(\frac{2\sigma_{s_0}^2 dt}{\tau_t} \right)^{1/2} \eta(t) \quad (4.25)$$

where α is a thermodynamic constant (with a temperature dependence) [251] and other symbols have been previously defined. The first term on the right hand side is the mean supersaturation forcing because of the mean vertical velocity of a cloud parcel, the second term represents the turbulent mixing of supersaturation to a mean value, and the last term is the sink due to droplet growth. Here, the driving force for supersaturation fluctuations (the last term) is considered as a turbulence-correlation-dependent random forcing which could be due to a combination of vertical velocity fluctuations, entrainment, and isobaric mixing. We have neglected the aerosol activation part of the droplet growth process.

Equations for the supersaturation fluctuations and variance will be similar to Eqs. 4.6 and 4.8 respectively. Therefore, the derivation of $\overline{s'r^{2'}}$ will follow a similar development as for the II- Chamber. However, the equation for the mean supersaturation in a cloud parcel will be [275]

$$\frac{d\bar{s}}{dt} = \alpha \bar{w} - \frac{\bar{s}}{\tau_c}. \quad (4.26)$$

4.7.1 Quasi steady-state solution

At quasi steady-state, the expressions for the mean supersaturation and variance are

$$\bar{s} = \alpha \bar{w} \tau_c \quad (4.27)$$

and

$$\sigma_s^2 = \frac{\sigma_{s0}^2}{1 + Da} \quad (4.28)$$

After substituting the value of the mean supersaturation from Eq. 4.27 and $\tau_c \approx (4\pi D_v n_d (\bar{r}^2)^{1/2})^{-1}$ into Eq. 4.2 and then solving it for a mean droplet size, the final expression is

$$(\bar{r}^2)^{3/2} = 3 \int_0^t \frac{\xi \alpha \bar{w}}{4\pi D_v n_d} dt. \quad (4.29)$$

The assumption of $\bar{r} \approx \bar{r}^2^{1/2}$ is necessary to obtain a closed-form solution, and is shown later to result in a small underestimation of the mean and fluctuations for droplet radius. For a steady droplet number concentration in the parcel, this expression can be simplified to

$$(\bar{r}^2)^{3/2} = \frac{3\xi\alpha\bar{w}t}{4\pi D_v n_d}$$

$$\bar{r}^2 \approx 3\xi\alpha\bar{w}\tau_c t \approx 3\xi\bar{s}(t)t \quad (4.30)$$

where we neglected the temperature dependence of the thermodynamics constants ξ , α , and D_v since they are weak functions of the air temperature.

Integration of Eq. 4.16 after substituting the expression for $\bar{r}^2$ from Eq. 4.30 in an approximate form of $\tau_c \approx (4\pi D_v n_d (\bar{r}^2)^{1/2})^{-1}$, gives a final expression for a droplet size distribution width:

$$\sigma_{r^2}^2 = 8\sigma_{s_0}^2 \tau_t \int_0^t \frac{\xi^2}{(1 + b(T)\tau_t n_d^{2/3} \bar{w}^{1/3} t^{1/3})^2} dt \quad (4.31)$$

where $b(T) = 4\pi D_v \left(\frac{3\xi\alpha}{4\pi D_v} \right)^{1/3}$. For a constant droplet number concentration and mean velocity, the solution is:

$$\sigma_{r^2}^2 = \frac{3a}{B^3} \left[\frac{B\sqrt[3]{t}(B\sqrt[3]{t} + 2)}{(B\sqrt[3]{t} + 1)} - 2\log(B\sqrt[3]{t} + 1) \right] \quad (4.32)$$

where $B = b(T)\tau_t n_d^{2/3} \bar{w}^{1/3}$ and $a = 8\sigma_{s_0}^2 \tau_t \xi^2$.

Figure 4.7: An example of the cloud droplet mean size $r_m^2 / \bar{r}^2$ (black curve) and fluctuation σ_{r^2} (red curve) growth with time for a rising cloud parcel of fixed droplet number concentration 100 cm^{-3} . It also shows the comparison of direct calculation (solid line) from the set of differential equations to the analytical solution based on the quasi-steady-state assumption (dotted line). The atmospheric conditions used for this stochastic condensation growth calculation are: cloud base temperature=278 K, $\tau_t = 20 \text{ s}$, $\bar{w}=0.1 \text{ m s}^{-1}$ and $\sigma_{s,0} = 0.01$.

Figure 4.7 presents an example of the numerical solution of droplet growth in a rising cloud parcel using Eq. 4.2, 4.3, 4.14, 4.26, and 4.8 without any approximation, as a quasi-steady analytical solution. In this figure, the analytical solution (Eqs. 4.30 and 4.32) is also compared with the numerical output of the governing set of equations. It closely follows the numerical results with slight underestimation due to the quasi-steady state assumption and approximation of $\tau_c \approx (4\pi D_v n_d (\bar{r}^2)^{1/2})^{-1}$. It can also be observed that, for a reasonable value of mean velocity and starting turbulent fluctuations, droplet size dispersion increases significantly with time. Although, there is no feedback, entrainment, and droplet loss or activation included in the theory, it can be thought of as an elementary framework for illustrating the basic physics. The model can be further developed in the future to incorporate complexities of an atmospheric cloud parcel.

4.7.2 Relative dispersion and autoconversion rate for a rising cloud parcel

In atmospheric clouds, $Da > 1$ is more frequently observed in the early growth stage before entrainment and mixing of surrounding dry air start dominating. In this case, a simplified solution of Eq. 4.31 can be expressed as

$$\sigma_{r^2}^2 = \frac{3a}{B^2} \sqrt[3]{t} \quad (4.33)$$

and the relative dispersion ($\sigma_{r^2}/\overline{r^2}$) for this case (from Eq.4.30 and 4.33) will be

$$d_a = \sqrt{\frac{8}{3\tau_t} \frac{\sigma_{s_0}}{\alpha} \bar{w}^{-1} t^{-1/2}}. \quad (4.34)$$

These equations suggest that for the high Da case, the relative dispersion of droplet size is independent of the droplet number concentration, and is inversely proportional to the mean vertical velocity and droplet lifetime. The only dependence of the relative dispersion on droplet number concentration will be due to the dependence of the droplet lifetime on number concentration.

The major loss mechanism of cloud droplets in the atmosphere is expected to be droplet collisions and subsequent precipitation formation. A time scale associated with the cloud droplet loss due to autoconversion process can be obtained from the autoconversion rate

of cloud droplet number concentration for a gamma radius distribution [183, 332]) $\tau_{l,c} = (n_d/d_t n_d)$:

$$\tau_{l,c} = \left(\frac{4\pi\rho_w}{3} \right)^2 \frac{\Gamma^2(\alpha_d + 3) n_d}{k_l \Gamma(\alpha_d) \Gamma(\alpha_d + 6, x_{cq}) L^2}. \quad (4.35)$$

We note the similarity to Eq. 4.24; here we consider rate of change of number, whereas before we considered rate of change of mass. Similar to the droplet settling lifetime, Eq. A.2, this equation shows that the cloud droplet lifetime based on the autoconversion rate is an increasing function of the droplet number concentration for a constant liquid water content.

Equations 4.34 and 4.35 suggest that the relative dispersion for a droplet distribution in a cloud parcel is a non-linear function of the droplet number concentration (without considering a time-dependent droplet number concentration). It can also be inferred that the relative dispersion is a non-linear function of the mean vertical velocity. These points are consistent with observations by Lu et al. [187], who showed a power law dependence of relative dispersion with n_d and $\bar{w}$.

As noted at the beginning of the section, we consider a simplified scenario for this analysis. In the atmosphere, complicating factors include a vertical aerosol profile and supersaturation (mean+fluctuation) dependent CCN concentrations [249, 301]. Here, we have assumed that the droplet number concentration is constant and independent of the mean vertical velocity. Of course, in an atmospheric cloud, the droplet number density is a function of the mean

vertical velocity and available aerosol, as well as the fluctuations in supersaturation [44].

Figure 4.8: Autoconversion rate and autoconversion time scale as a function of time in a rising cloud parcel, obtained from the theoretical expression based on the gamma droplet radius distribution. Five different cloud droplet number densities are assumed, in order to understand the role of the turbulence-broadening effect. The left panel displays the ratio of the autoconversion rate as predicted by the theory, to the autoconversion rate assuming fixed size distribution width. The corresponding fixed dispersion cases are arbitrarily set with dispersion ($\sigma_{r,2}$) growth stopped after 500 s. The right panel displays the time dependence of the autoconversion time scale for different cloud droplet number densities. The time varying gamma distribution parameters for a rising cloud parcel are obtained from the stochastic theory of droplet growth presented in this section. Atmospheric conditions used for the stochastic condensation growth calculation are: cloud base temperature of 278 K, $\tau_t = 20$ s, $\bar{w}=0.1 \text{ m s}^{-1}$, and $\sigma_{s,0} = 0.01$. Each of the profiles is for a fixed droplet number concentration (50; 100; 250; 500; 1000 cm^{-3}). For the $n_d=50 \text{ cm}^{-3}$ case, τ_a is also shown for a constant dispersion after $t=500$ s to demonstrate the effect of stochastic dispersion growth in the autoconversion process (i.e., compare the dashed and solid purple curves).

With these simplifications kept in mind, Figure 4.8 shows (left panel) the autoconversion rate compared to a cloud with fixed dispersion and (right panel) the autoconversion time for a rising cloud parcel. Each of the lines in the figure represents a cloud parcel with a fixed droplet number density, ranging from 50 to 1000 cm^{-3} . The autoconversion rate is calculated based on the equation described above (Eq. 4.24); the gamma parameters for a rising cloud parcel are obtained from Eqs. 4.30 and 4.32 (by approximating the droplet

radius distributions as gamma distributions). The relationship between the droplet area relative dispersion and the radius relative dispersion for a gamma radius distribution is used to estimate the gamma parameters from $\overline{r^2}$ and $\sigma_{r,2}$ values, and the cloud liquid water content is estimated from gamma parameters.

In the left panel, the autoconversion rate for a rising cloud parcel with the full turbulence-broadening effect is normalized by the autoconversion rate for an identical rising parcel but with constant dispersion after 500 s. This is done to illustrate the influence of supersaturation variability and the resulting change in size distribution width with time. As can be seen in the figure, the autoconversion rate is enhanced by factors of 2 to 5 by the turbulence-induced broadening mechanism.

Furthermore, as expected and as illustrated in the right panel, an increase in the droplet number concentration decreases the autoconversion rate and increases the autoconversion time scale due to decreases in the mean droplet size and size distribution width. There is a significant change in the autoconversion time scale as the cloud distribution grows with time. The autoconversion time scale at the end of the simulation varies from a few minutes to order of 10 hours for very clean and polluted conditions, respectively. To appreciate how much of this effect is due to changes in mean properties compared to increases in size distribution width resulting from supersaturation fluctuations, we compare to a case with constant dispersion $\sigma_{r,2}$. It is clear that a decrease in the autoconversion rate with an increase in the aerosol concentration is not just because of the depression in the mean size but also due to a decrease in the droplet size fluctuations.

4.8 Discussion and Concluding Remarks

We have extended and refined the observations first shown in Chandrakar et al. [43]. Measurements in controlled, laboratory conditions show that the droplet size distribution broadens with a decrease in aerosol loading. This broadening process is associated with fluctuations in the supersaturation in a turbulent environment. As expected, higher aerosol loading suppresses the mean supersaturation, leading to a smaller mean droplet size. The higher aerosol loading also suppresses fluctuations in supersaturation, which results in a narrower droplet size distribution.

The relevant time scale of the turbulent supersaturation fluctuations for droplet growth by condensation is the Lagrangian correlation time of the turbulence, while the time scale associated with droplet response to the supersaturation field is the phase relaxation time. The relative magnitude of these time scales (quantified by system time scale) determines the supersaturation fluctuations and corresponding droplet size dispersion.

In our experiments, the phase relaxation time is controlled by the aerosol input and the turbulence time scale by the applied temperature gradient. With a decrease in the aerosol input, cloud droplet number concentration decreases and the phase relaxation time scale increases. Consequently, for a fixed turbulence time scale, droplet size dispersion increases with the system time scale due to a decrease in the aerosol loading. Moreover, similar response is also observed in the supersaturation fluctuations and mean droplet size as described by the theory of stochastic condensation.

Novel results and contributions resulting from this work can be concisely summarized as follows:

† The results of [43] were an initial exploration of the dependence of microphysical properties on the Damköhler number. In this work we have performed experiments under a wider range of conditions to more carefully resolve the n_d dependence of microphysical properties in a turbulent environment as it makes the transition below and above $Da \approx 1$. For the two sets of thermodynamic conditions that have been used, Damköhler numbers in the range $0.2 \leq Da \leq 4.6$ (nine cases at $\Delta T = 16$ K) and $0.4 \leq Da \leq 40.2$ (eleven cases at $\Delta T = 19$ K) have been achieved.

† The work of [43] left a question about what led the theory, as formulated there, to break down at very large Da . In that work we assumed constant cloud droplet lifetime, and there was one data point for which the predicted linear dependence on τ_s was clearly violated. That work also allowed unphysical negative-radius trajectories. In this work we have extended the theory to account for droplet lifetime and have implemented an approximate positive-radius condition, with the result that the theory produces reasonable agreement with observations of mean and dispersion of cloud droplet size.

† We report an analytical result for relative dispersion and find that it is predicted to be constant except for the implicit dependence of droplet lifetime on n_d . We explicitly show the dependence of dispersion on n_d and compare the theory to observations with favorable results.

† We calculate an autoconversion time scale for the observed droplet size distributions and show that it has an explicit dependence on n_d . The time scale changes by many orders of magnitude just from the relatively modest microphysical changes existing in the cloud chamber.

† In order to learn more about the relevance of the results in an atmospheric context, we extended the stochastic theory to a rising-parcel viewpoint, and illustrate the results by estimating how the autoconversion time changes for the theory compared to a constant dispersion case. Implications of the onset of coalescence for droplet lifetime and therefore the dependence of relative dispersion on n_d is briefly explored.

The theory for a rising, quasi-adiabatic cloud parcel merits further discussion. In this idealized model, the cloud parcel is assumed to have a constant mean updraft velocity, and the cloud droplet number concentration is also assumed to be steady. Moreover, any feedback to turbulent convection due to the release of latent heat from growing cloud droplets is ignored. A relation for relative dispersion of droplet size is extracted from this theory and is simplified for the $Da > 1$ case, which is the more frequent condition in atmospheric clouds. It suggests that the relative dispersion of cloud droplet size is inversely proportional to the mean updraft velocity and proportional to the supersaturation fluctuations. It also varies with droplet lifetime ($\propto t^{-1/2}$), which is proportional to the droplet number concentration for a fixed liquid water content due to the autoconversion process. Therefore, the relative dispersion of droplet size is predicted to decrease with an increase in droplet number concentration ($\propto n_d^{-1/2}$). This prediction of power law dependence of relative dispersion with droplet number concentration and mean updraft velocity is consistent with

some observations of cumulus clouds [187].

Like clouds in the atmosphere, the lifetime of droplets in the lab experiments is constrained. However, the mechanism of droplet loss is different for the two cases; in the lab experiments, it is governed by the droplet settling due to the finite vertical dimension of the chamber. In the atmosphere, droplet loss is primarily by collision-coalescence or by entrainment, and the former has been considered here. Despite the differences in loss mechanisms, the dependence of the cloud droplet lifetime on droplet number concentration is similar; in both cases, droplet lifetime is proportional to the droplet number concentration. For the chamber experiments, theory predicts a decrease in the relative dispersion with droplet number concentration and this prediction is nicely validated with the steady-state experimental results.

Finally, with this analysis of the turbulence-induced aerosol effect on the droplet size distribution, we explored the aerosol effect on the autoconversion process. The theoretical model of adiabatic stochastic condensation growth for a rising cloud parcel is used together with the expression given by Liu et al. [183] and Xie and Liu [332] for the autoconversion rate. The result shows an increase in the autoconversion rate with a decrease in the droplet concentration. However, this increase is not just due to increase in the mean size, but also a result of an increase in the dispersion of droplet size; it is clearly evident from the estimation for a fixed dispersion. Similarly, the autoconversion time scale is estimated for the experimentally observed droplet size distributions in the cloud chamber, corresponding

to different aerosol loading. Those results also show a strong dependence of the autoconversion time scale for clean versus polluted cloud conditions; it is much smaller for clean cloud conditions, which have larger mean droplet radius and size dispersion. This work demonstrates the relevance of stochastic condensation to the autoconversion process. It adds to the growing evidence that turbulence contributes significantly to autoconversion both through the condensation and coalescence processes [54, 103, 105, 221, 254].

It is reasonable to ask how the cloud chamber observations reported here compare to atmospheric observations. Because not all information is available to determine Da for the observations reported in the literature, we consider the dependence of relative dispersion on droplet number density. The observed trends are consistent with some reported atmospheric observations [187, 189, 193, 209] and are opposite to some others [175, 199, 209, 223, 229, 230]. There are some observations suggesting nearly constant relative dispersion with a change in the aerosol concentrations [194] and some that have unclear trends [209, 349]. In [209], averages of all marine and continental cloud data suggest that marine clouds have higher relative dispersion than continental clouds; but a comparison based on different mean size range produces an unclear trend. However, the droplet size dispersion σ_r for marine clouds increases with a decrease in the aerosol concentration (or corresponding droplet concentration) and is consistent with current laboratory observations and accompanying theory. For continental clouds, σ_r is shown to be nearly constant for different droplet number concentrations. Lu et al. [193] suggest that an average over multiple cloud samples produces dispersion broadening with increase in aerosol concentration, contrasting to the observations from single clouds, for which relative dispersion decreases

with increase in aerosol concentration. They concluded it is an effect of difference in cloud conditions, such as different cloud top entrainment, updraft velocity, or evaporation at cloud base. Similarly, Lu et al. [187] shows an unambiguous observation of the relative dispersion decrease with aerosol loading. Not surprisingly, the atmospheric observations are scattered, especially when data from different cloud types or even different realizations of one cloud type are mixed. This suggests that other factors come into play, such as dynamical feedbacks and the role of entrainment, neither of which have been treated here. Nevertheless, it appears that much of the data, when carefully conditioned on cloud type, are consistent with the conceptual framework that has been treated here. Having such a framework in mind may help in evaluating aerosol effects on relative dispersion in future work.

As a final remark, we consider these results in the context of aerosol indirect effects, which are thought to be significant for global radiative energy balances [41, 266]. In their most elementary form, these effects are a result of the influence of aerosol concentration on mean droplet size. For example, an increase in aerosol concentration reduces mean droplet size due to enhanced competition for water vapor, ultimately increasing the cloud reflectivity (first indirect effect) and the cloud lifetime by reducing the precipitation efficiency (second indirect effect) [8, 305]. However, some recent studies suggest that aerosol concentration also influences the cloud droplet size dispersion, which in turn enhances or suppresses aerosol indirect effects depending on the response of relative dispersion with aerosol concentration [86, 175, 193]. The work presented here addresses this dispersion aerosol indirect effect. As explained in the previous paragraph, the exact nature of the aerosol-dispersion response is unclear through atmospheric observations, likely because multiple processes are interacting.

The current experimental study suggests that the process of stochastic condensation, by itself, leads to a decrease in relative size dispersion with increasing aerosol loading. This would therefore be expected to enhance the aerosol indirect effect.

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Chapter 5

Aerosol removal and cloud collapse accelerated by supersaturation fluctuations in turbulence

This chapter is about the laboratory study of the aerosol-cloud feedback during the 'cloud-cleansing' process (or during the transient decay of clouds). It is based on a collaborative research published in the Geophysical Research Letters [44] ¹.

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5.1 Abstract

Prior observations have documented the process of cloud cleansing, through which cloudy, polluted air from a continent is slowly transformed into cloudy, clean air typical of a maritime environment. During that process, cloud albedo changes gradually, followed by a sudden reduction in cloud fraction and albedo as drizzle forms and convection changes from closed to open cellular. Experiments in a cloud chamber that generates a turbulent environment show a similar cloud cleansing process followed by rapid cloud collapse. Observations of 1) cloud droplet size distribution, 2) interstitial aerosol size distribution, 3) cloud droplet residual size distribution, and 4) water vapor supersaturation are all consistent with the hypothesis that turbulent fluctuations of supersaturation accelerate the cloud cleansing process and eventual cloud collapse. Decay of the interstitial aerosol concentration occurs slowly at first then more rapidly. The accelerated cleansing occurs when the cloud phase relaxation time exceeds the turbulence correlation time.

5.2 Introduction

Microphysical and radiative properties of stratocumulus clouds are strongly influenced by the number of aerosol particles in the boundary layer [31, 248, 264, 304]. Aerosol properties are not static, however, but are coupled to cloud droplet formation and the presence of drizzle [11, 141]. For example, even for modest drizzle rates, aerosol populations can

be strongly modified by collection [232, 320]. Furthermore, Ackerman et al. [3] proposed that under conditions with weak aerosol production, stratocumulus cloud lifetime may be limited by droplet coalescence and eventual decay of radiatively driven convection. A particularly striking example was documented by Goren and Rosenfeld [102], using satellite data to perform a Lagrangian study demonstrating that stratocumulus clouds containing aerosols from a relatively polluted environment undergo a cleansing process over several days. The cleansing process, initially taking place in closed cell clouds, was followed by a rapid transition to drizzling open cells. They referred to this process as “maritimization,” suggesting that the clouds evolve from a relatively polluted, continental state to a relatively clean, maritime state. Indeed, it has been demonstrated that closed versus open cellular stratocumulus convection patterns can be induced just by varying aerosol concentration [253, 310].

We focus here on obtaining a deeper understanding of the aerosol-cloud interactions that lead to cloud cleansing and perhaps even to cloud collapse. Specifically, we investigate the transient response of clouds in a laboratory environment that allows for aerosol-cloud interaction in the presence of constant thermodynamic forcing and turbulence properties [50]. This greatly simplifies the problem relative to atmospheric observations, in which there is full coupling of microphysics with radiation and large-scale dynamics, with the resulting nonlinearity introduced by feedbacks.

An idealized statement of transient response of cloud droplet number density n_d can be written as a balance between the rate of activation to form new droplets and removal

with characteristic time scale τ_{removal} : $dn_d/dt = \dot{n}_{d,\text{activation}} - n_d/\tau_{\text{removal}}$. Removal in the atmosphere is expected to be dominated by collision-coalescence, but this is efficient only after sufficient numbers of large cloud droplets have been generated; in other words, the condensation growth stage is the bottleneck, so a study focused on that part of the process is likely to yield insight. When aerosol are depleted, exponential decay with time scale τ_{removal} is suggested. But τ_{removal} itself is expected to be a function of the supersaturation: if the number of cloud droplets is large, the quasi-steady supersaturation [251] will be small and droplet growth will be slow, resulting in a long residence time. If the number of cloud droplets is small, however, the quasi-steady supersaturation can be expected to be larger, leading to small τ_{removal} . We can anticipate, therefore, that the cloud microphysical properties and resulting supersaturation will lead to slow initial decay followed by a rapid decay stage, qualitatively similar to the cloud cleansing observations of Goren and Rosenfeld [102].

Conceptually, the experiments consist of injecting aerosols at a constant rate into a turbulent cloud chamber (the Pi-chamber [50]) such that steady-state cloud microphysical and interstitial aerosol properties are achieved: the rate of aerosol injection is balanced by activation and subsequent loss of cloud droplets as they grow by condensation and settle out of the chamber. Then suddenly the aerosol injection is switched off and we watch as the cloud and aerosol populations evolve. Based on prior experiments in the Pi-chamber, which suggest that reduction in steady-state aerosol concentration leads not only to higher mean supersaturation but also results in stronger supersaturation fluctuations [43], our working hypothesis is that the mean and fluctuations of supersaturation are suppressed until the

aerosol activation rate is sufficiently low, and that is followed by rapid cloud cleansing and droplet growth.

5.3 Observations of cloud decay

The Pi-chamber [50] is used to create the turbulent mixing conditions that lead to cloud formation by applying an unstable temperature gradient between the saturated top and bottom boundaries. Cloud formation is initiated by injecting NaCl aerosol. In these experiments, we employ 19-K (26 °C bottom boundary and 7 °C top boundary) and 8-K (14 °C and 6 °C) temperature differences to drive the turbulent convection. Once the cloud properties reach steady-state, the aerosol source is turned off to initiate the cloud cleansing process.

Water vapor concentrations are measured at 20 Hz with a LI-COR hygrometer (7500A), positioned near the center of the chamber. Temperature is measured at 1 Hz at eight places in the chamber using resistance thermometers (RTDs, Minco), recorded with a Lakeshore 218. Size distributions of the interstitial aerosols are acquired continuously using a Scanning Mobility Particle Sizer (SMPS, TSI). The total aerosol number concentration reported here is derived by integrating the size distributions. Cloud droplet size distributions are measured with a Phase Doppler Anemometer (Dantec). Further details of the experiment can be found in the Supplementary Material.

Figure 5.1 is an overview of the temporal evolution of cloud and aerosol properties as the

Figure 5.1: Variation of microphysical properties during the transient decay of a cloud initially in steady-state for $\Delta T = 19$ K in the chamber. Aerosol injection ceases at $t = 0$ min. **Top panel:** Droplet number concentration (light blue circles) and its running mean (blue line) are shown on the left axis. The mean droplet diameter (black line) is shown on the right axis; **second panel:** standard deviation of the droplet radius r ; **third panel:** interstitial aerosol number concentration; **bottom panel:** phase relaxation time, τ_c , (light gray squares) as calculated from the measured droplet size distributions and its running mean (black line). The horizontal red line shows the turbulent mixing time scale, τ_t , which is constant.

system decays after the aerosol source is turned off. The figure illustrates the essential coupling between the cloud droplet and aerosol concentrations. First, the droplet number concentration very slowly decays and the mean diameter increases slightly. During that time, the loss of droplets to sedimentation is nearly balanced by activation of interstitial aerosol particles. The loss of interstitial aerosol is seen in the third panel.

In a turbulent environment, supersaturation is a fluctuating quantity [270], which cloud microphysical properties adjust to with a characteristic time given by the phase relaxation time, $\tau_c = (2\pi D_v \bar{d} \bar{n}_d)^{-1}$, where d is the cloud droplet diameter and D_v is the diffusion coefficient for water vapor in air adjusted for thermal effects [251]. The measured τ_c is nearly constant to this point and it is lower than the turbulence correlation time scale τ_t (bottom panel), suggesting that the cloud is in the polluted, fast-microphysics state [43]. (See Supplementary Material for details of the time scales.) As interstitial aerosol are activated and removed, decreasing the reservoir of potential cloud droplets, the cloud eventually crosses from a fast microphysics regime ($\tau_c < \tau_t$) to a slow one, ($\tau_c > \tau_t$). Those two cases are characterized by the Damköhler number, $Da \equiv \tau_t/\tau_c$. For $Da > 1$, corresponding to small τ_c , the supersaturation is expected to be low and fluctuations in s are diminished, as a large number of small droplets quickly smooth out potential spikes (or deficits) in the vapor field [43, 271]. Conversely, we can expect that once the cleansing process has reduced the droplet number and $Da < 1$, the reduced competition among a smaller number of cloud droplets results in an increase in the mean supersaturation as well as an increase in the fluctuations about the mean.

At the 100th minute, the phase relaxation time crosses the value of turbulent mixing time scale (*i.e.* the cloud crosses from fast to slow microphysics). Near $Da \approx 1$ the droplet number concentration decreases more sharply and the mean diameter increases at a faster rate. The interstitial aerosol concentration also decays more rapidly once $Da < 1$. Finally, the width of cloud droplet size distribution, shown in the second panel, increases as the phase relaxation time crosses the $Da \approx 1$ level. Therefore, both the mean droplet size and

the width of a droplet size distribution increase as the cloud gets cleaner, consistent with the findings from steady-state experiments [43?].

5.4 Removal of Interstitial Aerosol

The cloud’s mean properties do not change appreciably in the first hour after the aerosol source is turned off. Why? The answer is revealed in the third panel of Figure 5.1, which shows the concentration of interstitial (*i.e.*, unactivated) aerosol particles as a function of time. When the aerosol source is turned off, there is already a substantial reservoir of interstitial aerosol in the chamber ($> 10^4 \text{ cm}^{-3}$). As droplets in the chamber grow and settle out, there are ample particles which can be activated to replace them, and indeed, the concentration of interstitials begins to decrease almost immediately.

To further quantify the losses of aerosol particles in the chamber, we consider each size bin in the aerosol number distribution, as measured by the SMPS, as an independent time series. By fitting that to an equation of the form $n = n_0 \exp(-t/\tau_a)$, we obtain characteristic aerosol decay times, τ_a , as a function of size. Those decay times are shown in the top panel of Figure 5.2. (Note that for this portion of the analysis we fit a single exponential to the data. As subsequent analysis shows (see below), the decay times actually change with time. The average decay time, however, does reveal important differences in removal rates for different sizes of particles.) The pronounced variation with dry diameter indicates different removal mechanisms. The data in the figure suggest three size ranges: small ($D_p < 40 \text{ nm}$),

Figure 5.2: Interstitial aerosol decay. **Top panel:** Aerosol decay timescales obtained from exponential fits of number concentration as a function of time for different aerosol sizes during cloud cleansing; the variation in characteristic decay times as a function of size shows that there are different removal mechanisms in the chamber. The primary removal process for aerosol diameters greater than about 40 nm is activation (see text for details). **Bottom panel:** Plot of $\ln(dN_{\text{bin}})$ as a function of time. The red squares correspond to medium size aerosol particles (dry diameter: $D_p = 45\text{--}62$ nm, critical supersaturation $s_c = 0.4\text{--}0.25\%$). The curvature in the line indicates that the characteristic decay time decreases with time. As the cloud becomes cleaner, the removal of these particles is more efficient. The black circles correspond to larger particles ($D_p = 188\text{--}260$ nm, $s_c = 0.05\text{--}0.03\%$). The straight line indicates that there is a single characteristic decay time. The transition from slow to fast aerosol decay for a medium size range aerosol particles roughly coincides with the transition from $Da > 1$ to $Da < 1$.

intermediate ($40 < D_p < 150$ nm), and large ($D_p > 150$ nm).

Settling is negligible for all of the aerosol sizes considered here. Measurements with aerosol

particles in the chamber at 100% relative humidity show that losses to the walls can be neglected for all but the smallest particles, as can diffusive losses to cloud droplets. (See Supplementary Material for details.) Characteristic decay times are 200 minutes or more. Activation is the dominant removal mechanism, as once particles become cloud droplets, they grow and settle out of the chamber.

The exception to this is small particles, with dry diameters less than approximately 40 nm. As the top panel in Figure 5.2 shows, they are removed from the chamber relatively quickly. In these cases, the particles' critical supersaturation, $s_c \approx 0.5\%$, is large enough that not even strong fluctuations exceed it in steady state conditions. For these smaller particles, measurements and calculations show that the most important removal mechanisms are diffusive loss to the walls and to cloud droplets [243, Sec. 17.4.2.1].

For all other particles, the primary removal mechanism is activation. The time scales for removal by other mechanisms greatly exceed the measured times shown in the figure. For the largest particles ($D_p \approx 150$ nm; $s_c < 0.06\%$), even when the supersaturation in the chamber is at the cloudy, low, quasi-steady state value, they are activated and removed as they encounter regions where fluctuations take the local supersaturation above the value needed for activation. As a consequence of this, characteristic decay times are 20 minutes or less for these large aerosol particles.

For aerosol particles with dry diameters between about 40 and 150 nm, the principal removal mechanism is also activation, but it proceeds in a two stage process. The lower panel in Figure 5.2 illustrates this. For this plot, we have averaged 10 adjacent aerosol size bins to

improve the signal to noise ratio. The two curves shown are for large aerosol (black circles), which exhibit a single exponential decay, and intermediate size aerosol (red squares), which show a slow, then fast decay. The larger aerosol, with a mean diameter of 222 nm, have low critical supersaturations, so fluctuations within the chamber are always enough to activate them. In contrast, the smaller particles, with a mean diameter of 53 nm, are removed slowly at first, as rare fluctuations of supersaturation exceed their critical supersaturation, activating and removing them. As more and more aerosol particles are removed from the chamber, the cloud droplet number starts to decay with a concomitant increase in the mean cloud droplet diameter. As the cloud crosses from the polluted ($Da > 1$) to the clean ($Da < 1$) regime, the aerosol removal rate increases suddenly. The lower panel of Figure 5.2 shows that the removal rate of the smaller aerosol particles increases dramatically at about 90 minutes, which corresponds to the time at which the phase relaxation time starts to increase rapidly, as shown in the bottom panel of Figure 5.1.

5.5 Supersaturation

Cloud and aerosol properties are coupled through the supersaturation, and in a turbulent environment that coupling extends throughout the lifetime of the cloud. To estimate the mean supersaturation in the chamber, we compare the probability distributions of cloud droplet residuals (*i.e.*, the aerosol particles upon which the cloud droplets formed) and interstitial aerosol. Cloud droplet residuals are sampled for steady-state cloud conditions, using a pumped counter-flow virtual impactor (CVI), which separates larger from smaller

particles [29]. In this way, we are able to sample cloud droplets, isolated from the interstitials. The droplets are dried and the residuals counted and sized with an SMPS. The range of supersaturation in the chamber can be determined from the distribution of cloud droplet residuals by computing their critical supersaturation.

Figure 5.3: The PDF of interstitial aerosols (blue) and residuals of cloud droplets (red): These PDFs are for steady-state cloud conditions in the chamber ($\Delta T = 19$ K and aerosol injection rate $5.0 \times 10^5 \text{ cm}^{-3}$ at 2 lpm). The cutpoint of the CVI was set to $4.5 \text{ }\mu\text{m}$. A comparison of the PDFs of the cloud droplet residuals and the interstitials indicates that, in steady-state cloud conditions, the mean supersaturation is low and that the fluctuations about that value rarely (if ever) exceed $\approx 0.5\%$. The apparent difference in area under the PDFs is a result of linear size binning displayed in a semilog plot.

As Figure 5.3 shows, the distribution of residuals is shifted relative to the distribution of interstitial particles from the chamber. While there are plenty of particles with diameters smaller than $\approx 40 \text{ nm}$ present in the interstitial distribution, those particles are not found in the cloud droplet residuals, which is compelling evidence that, under those conditions, the supersaturation did not exceed $\approx 0.5\%$, the critical supersaturation for those particles.

Figure 5.3 also indicates that there is a distribution of supersaturations within the chamber, as is expected in a turbulent environment [80]. While the peak of the distribution of residuals is at ≈ 110 nm ($s_c = 0.1\%$), the tail extends for another 50 nm in dry diameter, up to at least 0.3% supersaturation.

Long sampling times are needed for measurement of the cloud droplet residuals, so we cannot simply repeat the measurement shown in Figure 5.3 to derive the evolution of the supersaturation in the chamber as the cloud cleansing process proceeds. The time variation of supersaturation can be estimated, however, from co-located measurements of the water vapor concentration and temperature (using a hygrometer and an RTD). These measurements are not accurate enough to provide a reliable estimate of the mean supersaturation, but relative changes can be determined [271]. (The hygrometer is also susceptible to a long term (tens of minutes) drift in the mean in environments where water condenses on the instrument, as is the case for the $\Delta T = 19$ K experiments.)

Figure 5.4 shows the change in supersaturation, both measured and estimated from theory, for a case similar to that discussed in section 5.3 but for weaker turbulence and lower s_0 . Here, s_0 is the supersaturation in the chamber in the absence of a cloud as a result of forcing due to the temperature gradient. (A case for $\Delta T = 8$ K is used to avoid interference from condensation on the hygrometer, which occurs for higher ΔT , and thus higher, s_0 .) A comparison of the two panels shows that both the mean and fluctuations of s increase sharply at the same time that n_d and d change. This occurs at the transition near $Da \approx 1$,

Figure 5.4: Transient cloud properties for an 8K temperature gradient. **Top panel:** droplet number concentration (light blue circles) and its running mean (blue line) are shown on the left axis and the mean cloud droplet diameter (black line) is shown on the right axis; **bottom panel:** The mean supersaturation and its fluctuations increase as the cloud becomes cleaner. The measured standard deviation of the supersaturation (*i.e.* fluctuations) are shown on the left axis (black line). The increase in the saturation ratio as the cloud progresses from polluted to clean conditions, is shown on the right axis. The values derived from measured water vapor concentrations and temperature (red line) and an estimate using Eq. 5.1 from the cloud measurements (red squares) both show an increase at $t \simeq 150$ min.

and is consistent with the theory of Chandrakar et al. [43]. They showed that the quasi-steady value of s and its fluctuations, σ_s^2 , can be expressed as:

$$s = \frac{s_0}{1 + Da} \quad (5.1)$$

and

$$\sigma_s^2 = \frac{\sigma_{s_0}^2}{1 + Da} \quad (5.2)$$

where $\sigma_{s_0}^2$ is the variance in s_0 . As these two equations show, s and σ_s^2 both increase as Da decreases, resulting in an increase in the activation rate of interstitial aerosols due to a higher probability of experiencing supersaturation that exceeds their critical value. (We note that Equations 5.1 and 5.2 depend on the assumption of constant τ_c . Turbulence and settling will cause some local variability [?] and this is a topic of current investigation.)

5.6 Relevance for the atmosphere and concluding remarks

These experiments show that in polluted cloud conditions, like those present at time $t = 0$ in Figure 5.1, the mean supersaturation is low and fluctuations are suppressed. Cloud conditions (*i.e.* concentration and mean diameter) are slowly varying in this state as shown in both Figures 5.1 and 5.4. The cloud gradually becomes cleaner as cloud droplets settle out of the volume and interstitial aerosol are activated. Given time, enough droplets and interstitials are removed such that the cloud crosses into the limit of slow microphysics,

$Da < 1$. Then, s begins to increase to the value that would exist in the absence of a cloud, and fluctuations about the mean are enhanced. These increases in s and σ_s result in an accelerated rate of activation of interstitials (cf. bottom panel of Figure 5.2). The process continues to deplete the aerosol reservoir until the cloud collapses.

While these results are applicable for the conditions achieved in the Pi-Chamber, are they relevant for the atmosphere? Accelerated aerosol removal and cloud collapse in the chamber depends on removal of cloud droplets through settling. (In the atmosphere, significant removal by settling normally requires onset of coalescence.) The settling time scale in the one meter depth of the chamber is of order minutes, while, in the atmosphere, loss by sedimentation for small droplets is negligible because of the much larger cloud depths and the presence of mean vertical velocities. To answer the question, we consider coupled differential equations for droplet number density, n_d , and aerosol concentration, n_a . In general, n_d increases as a result of aerosol activation and diminishes as a result of settling and collisions between cloud droplets.

$$\frac{dn_d}{dt} = \dot{n}_{a,\text{activation}} - \dot{n}_{d,\text{settling}} - \dot{n}_{d,\text{collision}} \quad (5.3)$$

whereas n_a increases due to aerosol formation or evaporation of cloud droplets (injection for the case of the chamber) and decreases as a result of losses from activation, coagulation, and diffusion:

$$\frac{dn_a}{dt} = \dot{n}_{a,\text{injection}} - \dot{n}_{a,\text{activation}} - \dot{n}_{a,\text{coagulation}} - \dot{n}_{a,\text{diffusion}} \quad (5.4)$$

In the chamber, the time scale for collision between drops is large in comparison to settling (see Supplementary Material for details). In the atmosphere, the situation is reversed. For small droplets, such as those present in a polluted cloud regime, settling can be neglected in comparison to collision-coalescence (and subsequent removal of precipitation). The key, however, is that in both cases droplets are removed. The difference in the time scales implies that, in the atmosphere, the cloud cleansing process that we have outlined would take days, not hours as it does in the chamber.

Aerosol processes are similar. While coagulation and diffusion can be neglected in comparison to other processes in the chamber (see Supplementary Material for detailed calculations of timescales), those processes will remove interstitial aerosol in the atmosphere, which reduces the reservoir of potential cloud droplets. Activation is a sink for interstitial aerosol, both in the chamber and in the atmosphere. Again, the combination of slow aerosol removal through diffusion, coagulation, and activation, which is governed by a slow reduction in the number of cloud droplets, results in a cloud cleansing process that proceeds over the course of days in the atmosphere, not hours as it does in the chamber.

In either case (chamber or atmosphere), if the number of cloud droplets is large, the quasi-steady supersaturation and σ_s will be small and droplet growth will be slow, resulting in a long residence time. In the chamber, the long residence time is a consequence of a smaller settling velocity while in the atmosphere, the longer residence time follows from the reduced probability of collisions between small drops. In contrast, if the concentration of cloud droplets is small, the quasi-steady supersaturation and σ_s are both larger [43, 271], with a

resulting increase in both the mean and width of the droplet size distribution. This leads to smaller residence times through an increased settling velocity and a greater probability for collision and coalescence. As the cloud droplet number is further reduced, the mean and fluctuations of supersaturation are enhanced in a positive feedback loop, which eventually results in cloud collapse. Though the detailed removal mechanisms differ, we anticipate that feedbacks between aerosol, cloud microphysical properties, and supersaturation, will be parallel in the chamber and in the atmosphere. This runaway polluted-to-clean-cloud transition results in slow initial decay of n_a and n_d , followed by a rapid collapse. The cloud cleansing process demonstrated in the experiments described here, therefore, represents a simplified version of that occurring during the evolution of polluted clouds to a maritime state [e.g., 102]. Indeed, field observations support the notion that both aerosol activation and droplet growth take place in the presence of a broad distribution of supersaturations [80, 271].

In summary, feedbacks among aerosol particles, cloud properties, and a turbulent supersaturation field result in a two-stage cloud cleansing process. In the first stage, when $Da > 1$, cloud properties are relatively constant and aerosol are removed slowly. In the second stage, as the cloud crosses into the slow microphysics regime ($\tau_c > \tau_t$ and $Da < 1$), the removal of aerosol is more efficient as the mean and variance of supersaturation rise. Those changes result in broadening of the cloud droplet size distribution, thereby opening the bottleneck from condensational growth to runaway collision-coalescence and accelerating the cloud cleansing process.

Chapter 6

Dispersion Aerosol Indirect Effect in Turbulent Clouds: Laboratory Measurements of Effective Radius

This chapter is about the laboratory study of the dispersion aerosol effect and effective radius parameterizations in a turbulent cloud. It is based on a collaborative research published in the Proceedings of the Geophysical Research Letters [48] ¹.

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6.1 Abstract

Cloud optical properties are determined not only by the number density n_d and mean radius $\bar{r}$ of cloud droplets, but also by the shape of the droplet size distribution. The change in cloud optical depth with changing n_d , due to the change in distribution shape is known as the dispersion effect. Droplet relative dispersion is defined as $d = \sigma_r/\bar{r}$. For the first time, a commonly-used effective radius parameterization is tested in a controlled laboratory environment by creating a turbulent cloud. Stochastic condensation growth suggests d independent of n_d for a non-precipitating cloud, hence nearly zero albedo susceptibility due to the dispersion effect. However, for size-dependent removal, such as in a laboratory cloud or highly clean atmospheric conditions, stochastic condensation produces a weak dispersion effect. The albedo susceptibility due to turbulence broadening has the same sign as the Twomey effect and augments it by order 10%.

6.2 Introduction

In a significant way, aerosols affect cloud albedo and lifetime by altering the droplet size distribution. A higher cloud condensation nucleus (CCN) concentration tends to cause a higher concentration of smaller cloud droplets; that, in turn, enhances the cloud reflectivity and also increases the cloud lifetime by suppressing precipitation formation [8, 233, 303, 305].

Collectively, these are referred to as the first and second aerosol indirect effects. Semi-empirical parameterization of aerosol and cloud microphysical properties and their influence on cloud optics have been implemented in large-scale models and used to estimate aerosol effects on the radiative balances at a global scale [32, 86, 199, 266, 280].

The optical properties of cloud can be expressed through optical depth, single-scattering albedo, and asymmetry parameter. These are functions of two microphysical variables, effective radius of the droplet size distribution $r_e = \overline{r^3}/\overline{r^2}$ and liquid water path [32, 114, 279, 286]. Coarse-scale atmospheric models and remote sensing retrieval algorithms often depend upon assumptions on how r_e is related to the mean volume radius $r_v = (\overline{r^3})^{1/3}$ and cloud droplet number density n_d [32, 53, 113, 238, 292]. For example, it has been empirically observed that the two are approximately linearly related: $r_v^3 = k r_e^3$ [199]. This empirical observation is then utilized to parameterize r_e as a function of n_d and other microphysical variables.

The ratio k can be expressed as a function of the relative dispersion of the drop size distribution $d = \sigma_r/\bar{r}$ and its skewness $\mathcal{S}$ [174, 199, 240]:

$$k = \frac{(1 + d^2)^3}{(1 + 3d^2 + \mathcal{S}d^3)^2}, \quad (6.1)$$

where σ_r is the standard deviation of the droplet size distribution and $\bar{r}$ is the mean droplet radius. The effective radius of a droplet size distribution can then be expressed as [193, 199, 240]:

$$r_e \equiv \frac{\int r^3 n(r) dr}{\int r^2 n(r) dr} \approx \left[\frac{3L}{4\pi\rho_l n_d k} \right]^{1/3}, \quad (6.2)$$

where ρ_l is the density of liquid water, n_d is the droplet number concentration, and L is the liquid water content. Eq. 6.2 suggests $r_e \propto n_d^{-1/3}$ as a parameterization [30, 240, 280]. However, this assumes constant k and liquid water content, L , neither of which need be true.

With Eq. 6.2 in mind, we consider how an increase in the aerosol concentration not only increases n_d , but also may change the shape of the droplet size distribution. Equation 6.2 suggests that the cloud optical properties are not only affected by n_d (*i.e.* the first indirect effect) and liquid water content L , but also by the relative dispersion d through k . For example, an increase in n_d enhances cloud albedo directly, but also indirectly by decreasing precipitation efficiency and therefore tending to decrease the size dispersion (assuming constant L) [86].

Atmospheric observations suggest that the dependence of relative dispersion d on n_d has wide scatter, and therefore that the first aerosol-cloud indirect effect (Twomey effect) can be enhanced or suppressed due to changes in the shape of the cloud droplet size distribution [4, 87, 175, 187, 192, 193, 230, 293]. This can be expressed through the ‘albedo susceptibility’ obtained by from the expression of cloud optical depth, $\tau \propto L^{2/3} n_d^{1/3} k^{1/3} h$, where h is the cloud depth [4, 87, 192, 304]:

$$\frac{d \ln \tau}{d \ln n_d} = \frac{1}{3} + \frac{2}{3} \frac{d \ln L}{d \ln n_d} + \frac{1}{3} \frac{d \ln k}{d \ln n_d} + \frac{d \ln h}{d \ln n_d}. \quad (6.3)$$

Here, the first term on the right side is the classical effect of droplet number concentration on τ (Twomey effect). The third term $(1/3)(d \ln k / d \ln n_d)$ is known as the dispersion effect, and is the focus of this paper. Equation 6.3 implies that the dispersion (and liquid water content and cloud thickness effects) can enhance or offset the overall aerosol indirect effect beyond the Twomey effect. And yet even the sign of the dispersion effect remains uncertain, with field observations suggesting that it likely depends on CCN spectral width, entrainment, and precipitation development [87, 182, 344]. Here we consider just the dispersion effect resulting from activation and condensation growth in a turbulent environment (i.e., assuming no collision-growth).

Previous studies have generally suggested that condensation growth of cloud droplets leads to narrowing of the droplet size distribution and associated reduction in the relative dispersion, whereas collision-coalescence leads to broadening and increase in d . In recent experimental work, we identified a turbulence-induced size distribution broadening effect that also depends on the CCN concentration [43]. The results show that as CCN concentration is reduced, the droplet size dispersion σ_r increases dramatically even though droplets only grow by condensation. The droplet size dispersion is thought to be a result of modification of supersaturation fluctuations by cloud droplet growth in a turbulent environment, and a stochastic theory predicts that $d = \sigma_r / \bar{r}$ should be a constant [43] or slightly increasing [49] depending on assumptions made about droplet removal rate.

In this letter, we explore the dependence of r_e , k and d on CCN concentration using cloud chamber experiments. The experiments are designed such that measurements are made in an environment with steady-state turbulence and microphysical properties: droplet growth occurs by condensation and other processes such as entrainment or growth by collision-coalescence can be neglected. These idealizations allow the behavior of r_e , k and d to be explored in the context of droplet activation, condensation-growth, and sedimentation within a turbulent environment. The experiments therefore provide a way to avoid the complexity that has led to widely varying estimates of the dispersion effect from atmospheric measurements. Furthermore, results can be compared to theoretical predictions for stochastic condensational growth. Our aim is to provide physical insight into aerosol effects on cloud optical properties.

6.3 Measurements of size dispersion and the effective radius parameter k

Experiments are performed in the Michigan Tech II-chamber by creating a turbulent mixing cloud [50]. An unstable temperature difference is applied between water saturated top and bottom boundaries of the chamber to create a turbulent supersaturated environment (moist Rayleigh-Bénard convection). In this supersaturated environment, salt aerosols are injected at a constant rate to create cloud droplets. By changing the aerosol injection rates, a range of steady cloud conditions ranging from highly polluted to clean is achieved; i.e., in an individual experiment, steady state is achieved through balance between constant

aerosol injection and droplet settling. The cloud droplet size distribution and relevant thermodynamic variables are measured for each set of steady state cloud properties. The droplet size measurements were truncated at diameter $\approx 5 \mu\text{m}$ due to lower detectability at smaller sizes. This truncation will influence estimates of n_d and moments of the size distribution, most notably for high aerosol concentrations [49]. Quantities such as r_e that depend on high moments such as $\overline{r^2}$ and $\overline{r^3}$ are not strongly influenced because of their dependence on the large-size tail of the distribution. Fitting of the measurements with a gamma distribution and computing changes in r_e and k confirms that variation with n_d only change slightly. The details of the experiments and the data set that are used in this paper, as well as an overview of the stochastic condensation growth theory are provided by Chandrakar et al. [49]. Here we focus on analysis of those results in the context of r_e , k , and implications for the dispersion effect via Eqs. 6.1, 6.2, and 6.3.

Steady-state experiments in the cloud chamber with different input values of CCN result in observed dependence of r_v and r_e on n_d as shown in Figure 6.1. The left panel shows the decrease in r_v with n_d in response to an increase in the aerosol injection rate, along with contours of constant liquid water content L . It varies from 0.06 to 0.77 gm^{-3} with increasing aerosol injection rate, which is a result of the decreased droplet settling speed and associated increase in droplet lifetime. The r_e corresponding to these cases is shown in the right panel of the figure, along with curves obtained from Equation 6.2 assuming constant L and k . Deviation of the data points from the curves implies departure from the simple $r_e \propto n_d^{1/3}$ parameterization [30, 240, 280]. Moreover, the solid and dashed lines in the right panel of the figure demonstrate that the assumption of a constant parameter

Figure 6.1: Left, variation of the mean volume radius for different aerosol injection rates: measurements of mean volume radius (black dots) for different steady-state cloud droplet concentrations are shown, and its variation is compared with constant liquid water content contours (solid curves). The constant liquid-water-content L contours ($r_v = (3L/4\pi n_d)^{1/3}$) were drawn for the lowest, highest, and intermediate L observed in the measured data set. Right, variation of the effective radius with the droplet number concentration: steady-state values of r_e (black dots) are shown for different n_d (corresponding to different aerosol injection rates). The solid center line is for $L = 0.3 \text{ g m}^{-3}$, $k = 0.84$ or $L = 0.22 \text{ g m}^{-3}$, $k = 0.62$, and upper and lower dash lines are for $L = 0.3 \text{ g m}^{-3}$, $k = 0.62$ and $L = 0.22 \text{ g m}^{-3}$, $k = 0.84$ respectively. These values of k are obtained from the fitting in Fig. 6.2. The measurements almost, but do not quite, have a $n_d^{-1/3}$ dependence. Red circles are the theoretical estimates using Eqs. 6.2, 6.1 and 6.4 (please refer to Section-3 for detailed discussion).

k does not fully describe the data. As can be seen, different data points follow different constant- k lines, and the L and k combination for a data point is not unique.

These results suggest that insight can be obtained into the linearity assumption $r_v^3 = kr_e^3$ using steady-state cloud data from the II-chamber. The left panel of Fig. 6.2 shows r_v^3 and r_e^3 for a range of different aerosol injection rates, with constant turbulence forcing in the cloud chamber. Each of the data points in the figure corresponds to a steady-state droplet size distribution (averaged over a time scale $\sim \mathcal{O}(200 \text{ min})$) for different aerosol injection rates. The figure confirms an approximately linear relationship between the cubic mean

Figure 6.2: Left, the nearly (not exactly) linear relationship between r_v^3 and r_e^3 : experimental results (red dots) are fitted with a linear curve (black line), resulting in $k = 0.66$. Inset figures reveal that the nearly linear behavior of r_v^3 vs r_e^3 is not precisely true, rather the slope k is different for the clean (lower right) and polluted (upper left) regimes. Right, variation of the parameter $k = r_v^3/r_e^3$ with the relative dispersion (black square) and its comparison with the estimates (open red square) from Eq. 6.1. The solid red line shows Eq. 6.1 for zero skewness. Dashed lines are for three skewness values, showing that $S \approx 0.5$ to 1.5 . For comparison, Martin et al. [199] observed $k = 0.67$ and $k = 0.80$ for continental and maritime stratocumulus cloud cases, respectively.

volume radius (r_v^3) and cubic effective radius (r_e^3). The slope of the best fit line to the data is $k = 0.66$, which is close to the range of k (0.67–0.8) observed in the atmosphere [199]. However, closer examination reveals that k does vary, *e.g.* with aerosol loading: the two insets show linear fits for just the polluted cloud (upper left) and clean cloud (lower right), resulting in k varying from 0.62 to 0.84. The results are numerically similar but opposite from the in situ observations of Martin et al. [199], which resulted in $k = 0.67 \pm 0.07$ for continental stratocumulus clouds and $k = 0.80 \pm 0.07$ for maritime stratocumulus clouds.

In the right panel of Fig. 6.2, the parameter k is calculated directly from each steady-state cloud droplet size distribution using Eq. 6.1. The measurements (black squares) show a

monotonic dependence of k on relative dispersion d , decreasing with increasing d as expected from Eq. 6.1. The experimental data points lie away from the theoretical curve with $S = 0$ (red line), showing the importance of the skewness of the distributions as expressed in Eq. 6.1. Skewness is observed to range from $S \approx 0.5$ to 1.5. The primary contribution to the observed variation in k is from d , with skewness making a smaller contribution. Martin et al. [199] and Liu and Daum [174] showed that the effect of skewness in r_e parameterization is significant when d is large. In the current study, the effect of skewness is explored without assuming any distribution shape. But taking a gamma distribution as an example, the relative dispersion d and the skewness S are both proportional to $a^{-1/2}$, where a is the unitless shape parameter, and are independent of the scale parameter. Therefore, a gamma distribution is only able to describe the current observations if a is taken to be a function of d , and therefore of n_d . Finally, as a consistency check, calculations of k from Eq. 6.1 are made (red squares). The results adequately follow the values from a direct calculation, confirming that higher moments do not contribute and Eq. 6.1 is a reasonable representation of the dependence of k on d .

The change in d observed in the measurements is a direct consequence of varying the CCN concentration and the resulting cloud droplet concentration. In Fig. 6.3 we show the dependence of k on n_d for each observed, steady-state cloud droplet size distribution (black squares). The measurements show that k is a weak function of droplet number concentration n_d , consistent with the original findings of Martin et al. [199]. The parameter k is observed to increase nearly linearly with increasing n_d in relatively clean clouds, similar to the observations from marine stratocumulus clouds by Lu et al. [193]. This is in contrast

Figure 6.3: Variation of the parameter k with droplet number concentration n_d : steady-state values of k (black squares) obtained for the n_d that occur for different aerosol injection rates. Inset figure: An estimate of the dispersion effect, as defined in Eq. 6.3. The albedo susceptibility due to droplet size dispersion is obtained using a power-law fit of the k versus n_d measurements for the range $n_d < 10^3 \text{ cm}^{-3}$ (since k is nearly constant after that point). The magnitude of the dispersion effect in the inset is to be compared to the Twomey effect $1/3$ defined in Eq. 6.3. Calculated k values using Eqs. 6.1 and 6.4 are shown as red squares.

to Martin et al. [199], who observed larger k for maritime compared to continental stratocumulus clouds, and therefore an opposite sign of the dispersion effect. Interestingly, the data seem to suggest two regimes: a nearly linear increase in k with increasing n_d under low- n_d conditions, and a nearly constant k for relatively large n_d . However, the variation of k with n_d is modest, changing by only 22% for a factor of nearly 200 increase in the droplet concentration. The inset in Fig. 6.3 shows an estimate of the magnitude of the dispersion effect $(1/3)d \ln k / d \ln n_d$ for the low- n_d regime, using a power-law fit to the data. The dispersion effect is observed to be of order 10% of the Twomey-effect value $1/3$.

6.4 Dispersion effect and atmospheric implications

We now attempt to interpret the dependence of k on n_d using the stochastic condensation approach outlined by Chandrakar et al. [43]. The approach can be qualitatively summarized as follows: supersaturation is assumed to vary randomly due to mixing, entrainment, fluctuations in vertical velocity, or variations in cloud microphysical properties [72]. Given s_o as the expected value of the mean supersaturation without any cloud droplet formation (i.e., it is the expected value of the supersaturation that would exist for the same forcing if there were no water vapor loss due to cloud formation), the mean supersaturation in the presence of cloud, $\bar{s}$, is given by $\bar{s} = s_o / (1 + \tau_t / \tau_c)$. Here τ_t / τ_c , is the ratio of the turbulence correlation time scale, τ_t , and the phase relaxation time, $\tau_c = (4\pi D'_v \bar{r} n_d)^{-1}$, where D'_v is the water vapor diffusion coefficient modified to account for heat conduction [166, 251]. τ_c corresponds to the characteristic time for adjustment of supersaturation resulting from droplet growth by condensation. Chandrakar et al. [43] showed that the width of a cloud droplet size distribution becomes much broader for small τ_t / τ_c , corresponding to relatively low CCN and n_d , in what can be termed the slow microphysics / fast turbulence regime. The data set used in this study spans a τ_t / τ_c range of 0.44 - 40.23, with the $\tau_t / \tau_c = 1$ transition occurring between the $n_d = 75.9$ and 198.4 cm^{-3} data points. Thus, the observed saturation of k with increasing n_d in Fig. 6.3 corresponds to clouds in the $\tau_t / \tau_c > 1$ regime, i.e., to fast microphysics and slow turbulence.

Eq. 6.1 suggests that the parameter k depends mainly on the relative dispersion. As just

discussed, however, the relative dispersion resulting from stochastic condensation depends on the aerosol loading and corresponding cloud droplet concentration. Using the central result of Chandrakar et al. [43] for σ_{r^2} and the fact that $\overline{r^2} \propto \bar{s}t$ [251], the relative dispersion for r^2 can be written as:

$$d_a = \frac{\sigma_{r^2}}{\overline{r^2}} \propto \left(\frac{\sigma_{s_0}}{s_0} \right) \left(\frac{\tau_t}{t} \right)^{1/2}. \quad (6.4)$$

We can take t as the average droplet lifetime τ_l . This is an interesting result because it suggests that d_a has no explicit dependence on n_d . For the II chamber results shown in Figs. 6.1 and 6.2, however, there is at least a modest change in d_a as aerosol injection rate and n_d are increased. It is indeed reasonable to expect that the droplet lifetime depends on the rate of droplet growth and therefore the mean supersaturation. Following Yang et al. [337], that dependence is $\tau_l \propto \bar{s}^{-1/2}$, and as discussed earlier, mean supersaturation has a dependence on the phase relaxation time $\bar{s} = s_0/(1 + \tau_t/\tau_c)$. Consequently, $\tau_l \propto (s_0/(4\pi D'_v n_d \bar{r} \tau_t))^{-1/2}$, where the expression for the phase relaxation time τ_c has been used. The implication is that d_a should have a dependence on the product $n_d \bar{r}$ since other factors such as τ_t are held constant during the experiments. Taken together, these results lead to the following scaling for relative dispersion in the II chamber: $d_a \propto (\sigma_{s_0}/s_0) (1 + 4\pi D'_v \tau_t \bar{r} n_d)^{-1/4}$. A more complete derivation is given by Chandrakar et al. [49], but this simpler approach is adequate for our purposes because all other factors in the final expression are constant in the experiments except $\bar{r}$ and n_d .

Figure 6.1 (right panel) and Fig. 6.3 show estimates of r_e and k using Eqs. 6.1 and 6.4. To compare, we make the approximation $d_a \approx 2d$. The individual points are based on the measured values of $\bar{r}$ and n_d , and use the same, constant proportionality factor for Eq. 6.4, obtained through best fit of the measured droplet size statistics. Eq. 6.4 with the functional dependence of droplet lifetime reasonably captures the behavior of the experimental data points. Thus, we conclude that the weak observed dispersion effect in the cloud chamber experiments is fundamentally a result of the droplet-size dependence of the removal rate. The stochastic condensation process itself suggests constant k , assuming constant environmental conditions leading to supersaturation variability, and therefore zero dispersion effect; the positive and small dispersion effect results from larger droplets being removed more rapidly than small droplets.

Using the expression for droplet relative dispersion, Eq. 6.4 and the definition of k , Eq. 6.1, the albedo susceptibility due to the dispersion effect can be obtained. It is relevant to droplet condensation growth in the presence of turbulent fluctuations of water vapor concentration and temperature, and in the absence of collision-coalescence. It also is based on the assumption that droplet removal is due to gravitational sedimentation, as occurs in the cloud chamber. This scenario has atmospheric relevance as well, for clouds in extremely clean (low CCN concentration) environments where cloud droplets growth solely by condensation are removed as precipitation [200]. The dependence of sedimentation rate on droplet radius, along with the dependence of mean radius on n_d , leads to a dependence of d and k on n_d . The resulting albedo susceptibility due to the dispersion effect has a positive sign. We obtain a quantitative estimate of the dispersion effect by fitting a smooth function to

the observations of k in Fig. 6.3, and then taking its derivative. The result is shown in the inset to Fig. 6.3 for the range $n_d < 10^3 \text{ cm}^{-3}$. As expected from the n_d dependence of droplet lifetime in Eq. 6.4, it increases with an increase in n_d . Its magnitude is small in comparison to the Twomey effect, with a maximum value around 15%. That is a direct result of the relative dispersion having a weak dependence on droplet concentration, i.e., d_a changes by a factor of 2 with a $\approx 200\times$ increase in n_d .

As can be inferred from Eq. 6.4, the relative dispersion of droplet size distribution is independent of n_d if droplet removal is not size dependent. It just depends on thermodynamic and turbulence properties, and the cloud droplet lifetime. If there is no size-dependent droplet removal (like, sedimentation in case of the cloud chamber) and activation, i.e., a closed parcel, the stochastic theory suggests dependence of the size-distribution width on aerosol concentration. However, the relative dispersion at a fixed growth time t turns out to be independent of aerosol number. This statement is based purely on stochastic condensation without any influence of other factors, such as the activation-based dispersion effect for different aerosol-cloud interaction regimes [52, 182, 344]. Consequently, it implies that the albedo susceptibility due to dispersion effect based on stochastic condensation alone will be zero for a cloud with no size-dependent removal. This condition is nearly true for non-drizzling stratocumulus clouds where size-dependent removal processes (such as gravitational removal and droplet collision-coalescence) are not active. However, more subtle effects are possible: for example, a change in the aerosol population might indirectly affect the dispersion effect by suppressing the onset of droplet collision process and enhancing the cloud droplet lifetime. Observations in the atmosphere indeed show differing

relative dispersion trends with aerosol concentration: some suggest an increase in relative dispersion (negative dispersion effect) [175, 199, 209, 229, 230], some a decreasing relative dispersion (positive dispersion effect) [187, 189, 193, 209], and some a relatively constant or unclear trend [194, 209, 294, 348?] with aerosol concentration. These observation of non-constant behavior of relative dispersion might result from a variety of processes, like onset of collision-coalescence, dynamic effects (entrainment and mixing), or predominance of an early growth stage in which the initial aerosol distribution influences the size dispersion [87, 189, 231, 293, 344]. Consequently, one needs to be cautious in comparing the dispersion effect from clouds having different thermodynamic conditions, different stages of development, or the different regions or stages where entrainment and mixing dominate the overall growth process [261].

6.5 Discussion and conclusions

Physically-based representations (i.e., parameterizations) of aerosol-cloud microphysical processes are needed for remote sensing retrieval methods and for coarse-resolution atmospheric models, including those used for climate studies. Much effort has been made to develop empirical microphysical parameterizations using field measurements of clouds. However, variability in the atmospheric cloud conditions and complex feedbacks make it attractive to test some of the parameterizations in a controlled laboratory environment. Therefore, we have generated turbulent cloud conditions in an isolated laboratory chamber and have tested some commonly-used expressions using the resulting statistically-steady

cloud microphysical dataset. This approach allows us to have well-characterized thermodynamic, turbulence, and microphysical conditions, and long enough, steady data sets for critical scrutiny of these parameterizations. Furthermore, the experiments allow certain physical processes, namely condensation growth of cloud droplets in a turbulent environment, to be isolated from other processes, including dry-air entrainment and collision-coalescence. We address the influence of condensation growth in turbulence, modulated by aerosol concentration, on calculated cloud radiative properties; specifically, this study is focused on the effective radius and the dispersion aerosol-indirect effect. Observations are interpreted in the context of a theoretical description of cloud droplet growth in a turbulent environment. A positive, weak albedo susceptibility is observed for the cloud chamber conditions, resulting from the droplet-size-dependence of the removal rate. It is hoped that this work will help in establishing a physical foundation for further development of microphysical parameterization for atmospheric clouds.

A commonly used parameterization of the effective radius ($r_v^3 = kr_e^3$) developed based on measurements of the stratocumulus clouds is observed to also hold for turbulent clouds generated in the II-Chamber. The measurements of slope parameter k are consistent with the range of values from stratocumulus cloud measurements [193, 199]. The validity of this linear fit over a large range of n_d suggests that the parameter k is weakly changing over this range. Pioneering work has shown that the parameter k is a function of the relative dispersion and skewness of the cloud droplet size distribution [174, 199, 240]. Given the dependence of k on the width and skewness of the droplet size distribution, we expect from prior work there will be a link to aerosol (CCN) concentration when droplet growth occurs

in a turbulent environment [43, 49, 72], and indeed a positive, albeit weak dispersion effect is observed.

Martin et al. [199] suggested that measurement data points from stratocumulus and cumulus clouds sometimes deviate strongly from a linear $r_v^3 = kr_e^3$ relation. They interpreted this deviation as a result of entrainment of cloud-free air from cloud top and edges. This indicates the importance of turbulent flow conditions and demands further investigation of the influence of turbulence and entrainment. The results presented here also suggest an influence of turbulent fluctuations on the microphysics and related cloud optical parameters. A particularly intriguing possibility arises if the main droplet removal mechanism is entrainment and subsequent evaporation. If the dilution and removal of droplets through evaporation takes place via homogeneous mixing, then there is a preference to remove smaller droplets earlier, which would tend to result in a negative dispersion effect. In contrast, if the removal of droplets through evaporation takes place via inhomogeneous mixing, then all droplets have equal probability of removal, resulting in zero dispersion effect.

Chapter 7

Functional form of droplet size-distributions in turbulent clouds: experimental evaluation of theoretical distributions

Abstract

Theoretical expressions for cloud droplet size distribution shape are evaluated using measurements from controlled experiments in the Π Chamber. Three theoretical distributions obtained from a Langevin drift-diffusion approach to stochastic condensation are tested: (i)

stochastic condensation with a constant removal timescale; (ii) stochastic condensation with a size-dependent removal timescale; (iii) droplet growth in a fixed supersaturation condition and removal due to the gravitational settling. In addition, a similar Weibull distribution that can be obtained from the drift-diffusion approach, as well as from mechanism-independent probabilistic arguments (e.g., maximum entropy), is tested as a fourth hypothesis. Statistical techniques of χ^2 test, sum of squared errors of prediction, and residual analysis are employed to judge relative success or failure of the theoretical distributions to describe the experimental data. The experiments are carried out with steady-state cloud microphysical properties, with different aerosol injection rates, and corresponding cloud droplet concentrations. Five different aerosol injection rates are run both for narrow-distribution, size-selected aerosol particles, and six aerosol injection rates are run for broad-distribution, polydisperse aerosol particles. In relative comparison, the most favorable comparison to the measurements is the expression for stochastic condensation with size-dependent droplet removal rate. However, even this optimal distribution breaks down for broad aerosol size distributions, primarily due to deviations from the measured large-droplet tail. A possible explanation for the deviation is the Ostwald ripening effect coupled with deactivation/activation in polluted cloud conditions.

7.1 Introduction

The shape of the droplet size distribution (DSD) is central to understanding atmospheric clouds, with applications ranging from remote sensing retrieval to parameterization of precipitation and radiative transfer in climate models. However, the uniqueness of a functional shape for the cloud droplet size distribution is questionable due to the existence of different physical mechanisms for droplet formation, growth and removal, and their significance in different regions of cloud or stages of cloud growth. Physical processes affecting the cloud droplet size distribution include the curvature and solute effects (and associated mechanisms such as spectral ripening and competing activation of cloud condensation nuclei of varying size, solubility, etc.) [134, 156, 322, 339], turbulent fluctuations [46, 60, 225, 256, 277], entrainment and mixing [12, 172, 345], microphysical variability [60, 74], internal mixing of parcels of different growth history [123, 169], and droplet collision-coalescence [97, 242, 318]. Cloud droplet growth prior to onset of collision-coalescence occurs through water vapor condensation, and therefore the central variable affecting the droplet size distribution is the water vapor supersaturation, including both its mean value and its variability [81, 276].

Often in applications of cloud physics, such as for climate and numerical weather prediction models and for remote sensing, empirical distribution shapes are used for obtaining relevant moments. Empirical functional forms commonly used for distribution shape are the gamma (Khrgian and Mazin distribution), log-normal, and exponential [63, 178, 210, 242]. Undoubtedly it would be a significant advance to identify theoretical distribution shapes from

fundamental physical principles, even if restricted to some limiting atmospheric conditions. Several different theoretical models have been proposed to characterize the shape of cloud droplet size distributions formed through the condensation process. Two approaches that have received considerable recent attention are systems-theory derivations based on the principle of maximum entropy [177, 179, 323, 341] and derivations based on a Langevin equation representation of stochastic condensation [2, 46, 203, 225, 255, 277]. By and large, however, these theoretical models have not been subjected to systematic experimental evaluation to validate the proposed functional forms for the droplet size distribution. This is primarily because the sparsity of measurements from field experiments and the natural variability of atmospheric clouds makes this evaluation process ambiguous: large uncertainties in field measurements do not effectively constrain distribution shape, especially in the important but often under-sampled distribution tails. In this article we describe well-controlled and characterized laboratory experiments to assess several predicted distribution shapes from a theoretical model of stochastic condensation. We use statistical techniques for goodness-of-fit analysis to judge the relative success or failure to describe the experimental data over a range of aerosol conditions, i.e., from relatively clean to polluted conditions. Furthermore, in addition to varying the aerosol concentration, we consider size-selected aerosols as well as a broad distribution of aerosol sizes. The experiments are conducted under conditions that are statistically steady in time and without the complications of collision-coalescence. This allows us to evaluate the theoretical models under conditions as close as possible in replicating the assumed simplifications in their derivation.

In the next section we describe the Langevin model for cloud droplet growth and the analytical solutions to the resulting Fokker-Planck equation that correspond to different assumptions regarding turbulence and droplet removal. The three distributions correspond to assumptions of: (i) stochastic condensation growth with size-independent removal rate, (ii) stochastic condensation growth with size-dependent (Stokes settling) removal rate, (iii) condensation growth due to just mean supersaturated environment (no fluctuations) with droplet removal due to Stokes settling. For completeness, we also include a distribution resulting from the maximum-entropy (systems theory) approach, based on the constraint of conservation of liquid water. In Section 7.4 we outline the design of the experiment and the methodology for fitting analytical distributions to the observations, as well as the statistical tools used for evaluating suitability (via χ^2 test) and goodness of fit of the distributions. In Section 7.5 we present the data along with the fitted distributions for results from two sets of experiments: one with size-selected aerosol particles at several different aerosol concentrations, and one with a broad size range of aerosol particles, also at several different aerosol concentrations. We conclude with a discussion of the results and their implications for our understanding of atmospheric clouds.

7.2 Solutions to the Fokker-Planck equation for the cloud droplet size distribution

Atmospheric clouds are inherently turbulent. This implies that if we could measure the supersaturation experienced by a population of droplets at a given instant or alternatively, the

supersaturation experienced by a droplet over time, it would be a highly variable quantity. The droplet condensation process is therefore stochastic, and at least approximately, we can draw an analogy between random ‘diffusion’ of cloud droplet radius within size space, and Brownian molecular diffusion. The turbulence statistics, including the distribution of supersaturation and the Lagrangian correlation timescale of scalar fluctuations, drive this stochastic condensation process. Effectiveness of stochastic methods in modeling processes such as Brownian diffusion and turbulent combustion [?] has motivated the application of this mathematical technique to droplet condensation as well. The condensation process can be approximately represented as Brownian diffusion in the space of droplet surface area. Therefore, a Langevin equation for Brownian diffusion of droplet surface area (condensational growth of cloud droplets) with a drift due to mean supersaturation ($\bar{s}$) experience by droplets and diffusion due to the supersaturation fluctuations (standard-deviation σ_s) can be represented as [91, 128]:

$$dx = a \, dt + \sqrt{\mathcal{D}} dw \quad (7.1)$$

Where, $a = 2\xi\bar{s}$, ξ is the droplet growth factor and depends on thermodynamic quantities [252], $\mathcal{D} = \partial_t \sigma_x^2 \propto \sigma_s^2$, $x \equiv r^2$, and r is the droplet radius. In Eq. 7.1, the first term is the drift due to the mean supersaturation experienced by a group of cloud droplets; it represents mean r^2 growth, $\partial_t \overline{r^2} = 2 \, \xi \bar{s}$ [252]. The last term is the diffusion in r^2 space ($\partial_t \sigma_x^2$) due to supersaturation fluctuations with diffusivity $\mathcal{D}$, and dw is the differential of the Wiener process. A model for r^2 diffusivity can be obtained from the expression of droplet size dispersion growth presented in Chandrakar et al. [46], where $\partial_t \sigma_x^2 \propto \sigma_s^2 \tau_s$. The

time scale governing droplet growth in the fluctuating system is τ_s , the harmonic mean of the Lagrangian correlation time for supersaturation in the turbulent flow and the cloud phase relaxation time.

A Fokker-Plank equation describing the probability density function for droplet size can be obtained from the Langevin equation 7.1 with additional source and sink term due to droplet activation and removal [91, 128, 203, 255]:

$$\frac{\partial p}{\partial t} = -a \frac{\partial p}{\partial x} + \frac{\mathcal{D}}{2} \frac{\partial^2 p}{\partial x^2} + \dot{N}_I \delta(x - x_{cr}) - \frac{p}{\tau_r}. \quad (7.2)$$

Here, the first term on the right side represents drift in r^2 -space (mean growth) due to the mean supersaturation, the second term signifies diffusion of cloud droplet size due to turbulence (with diffusivity $\mathcal{D}$), the third term is probability gain due to activation of particle with critical size x_{cr} (and the activation rate $\dot{N}_I$), and the last term is the droplet loss or removal rate with timescale τ_r . We now proceed to describe three solutions for $p(r)$ from Equation 7.2 corresponding to different assumptions that could plausibly represent the conditions existing in a laboratory turbulent-cloud chamber, or in a natural cloud.

7.2.1 Size distribution for mean and fluctuating supersaturation with size-independent removal

Saito et al. [255] considered the simplifying assumptions of fixed critical size, equivalent to

mono-disperse aerosol particles of the same composition, and a constant, size-independent rate of droplet removal, which is equivalent to a constant droplet lifetime (i.e., $\tau_r = \tau_\ell = \text{constant}$). Under those conditions, Equation 7.2 leads to the following steady-state solution for $x \geq x_{cr}$:

$$p(x) = A \exp(-A x). \quad (7.3)$$

In this equation the coefficient A is

$$A = \frac{\sqrt{x_m^2 + 4\sigma_{x,m}^2} - x_m}{2\sigma_{x,m}^2}, \quad (7.4)$$

where, $x_m = 2 \xi \bar{s} \tau_\ell$, and $\sigma_{x,m}^2 = \mathcal{D} \tau_\ell / 2$. Alternately, in terms of droplet radius the size PDF can be expressed as:

$$p(r) = 2 A r \exp(-A r^2). \quad (7.5)$$

It is important to note here that the effect of curvature and solute terms on the equilibrium supersaturation are ignored. Ostwald ripening is therefore not accounted for in the derivation. Furthermore, because of the assumption of fixed activation size, cloud formation with

a broad spectrum of aerosol sizes could lead to deviation from the measured (physical) distribution. Similarly, the assumption of constant removal (or growth) timescale (τ_ℓ) could be violated if the primary removal mechanism is gravitational settling. Interestingly, a similar functional form of the steady-state solution as Equation 7.3, i.e., a Weibull distribution with a shape factor of two in a radius space, can also be achieved by considering a balance between the mean drift and the diffusion terms [203].

7.2.2 Size distribution for mean and fluctuating supersaturation with size-dependent removal

The constant-removal-rate assumption can be replaced with a combination of a size-independent removal timescale (e.g., due to turbulent transport) and a size-dependent removal rate (e.g., due to gravitational settling). For a removal timescale of the form $\tau_r^{-1} = \tau_\ell^{-1} + \kappa x$, it is possible to derive an analytical solution of the corresponding Fokker-Plank equation. Here a linear form of the removal timescale with droplet size $x \equiv r^2$ corresponds to gravitational settling with Stokes drag. It is also possible to write a similar form of cloud droplet removal due to collision-coalescence process. An analytical solution of Eq. 7.2 with this size dependent removal timescale (for $x \geq x_{cr}$) is:

$$p(x) = C \exp(x_m \sigma_{x,m}^{-2} x/2) Ai \left(\frac{x_m^2 + 4\sigma_{x,m}^2 (\tau_\ell \tau_{st}^{-1} x_m^{-1} x + 1)}{4 \sigma_{x,m}^{8/3} x_m^{2/3} (\tau_\ell \tau_{st}^{-1})^{2/3}} \right). \quad (7.6)$$

Here x_m and $\sigma_{x,m}$ are defined as in the preceding subsection, $\tau_{st} \equiv (\kappa x_m)^{-1}$ is the average

timescale associated with the Stokes settling, and Ai is the Airy function of the first kind.

The normalization constant is given by

$$C = \left[\int_{x_{cr}}^{\infty} \exp(x_m \sigma_{x,m}^{-2} \zeta/2) Ai \left(\frac{x_m^2 + 4\sigma_{x,m}^2 (\tau_\ell \tau_{st}^{-1} x_m^{-1} \zeta + 1)}{4 \sigma_{x,m}^{8/3} x_m^{2/3} (\tau_\ell \tau_{st}^{-1})^{2/3}} \right) d\zeta \right]^{-1}. \quad (7.7)$$

Fitting this function to data is cumbersome, and requires that the full size distribution is measured. We seek a simpler form that is valid for the large-droplet tail of the distribution, for which measurements are more reliable. An asymptotic form of Eq. 7.6 can be obtained by Taylor series expansion of $Ai(\alpha^*x + \beta^*)$ (or by the steepest descent method) with the limit $\alpha^*x + \beta^* \gg 1$. Here α^* and β^* are constants and can be interpreted from Eq. 7.6. We also assume zero mean supersaturation (i.e., $a = 0$) and just Stokes settling (i.e., $\tau_r = (\kappa x)^{-1}$ and $\tau_\ell = 0$) for simplicity. The resulting distribution is

$$p(x) \approx \frac{C}{2\sqrt{\pi}} \beta^{-1/12} x^{-1/4} \exp \left(-2/3 \sqrt{\beta} x^{3/2} \right), \quad (7.8)$$

where $\beta = 2\kappa/\mathcal{D}$. Again it should be emphasized that this expression is valid for the right tail of the distribution. In terms of radius this can be expressed as

$$p(r) \approx \frac{C}{\sqrt{\pi}} \beta^{-1/12} \sqrt{r} \exp \left(-2/3 \sqrt{\beta} r^3 \right). \quad (7.9)$$

7.2.3 Size distribution for fixed supersaturation with size-dependent removal

If we ignore supersaturation fluctuations in Eq. 7.2 by setting $\mathcal{D} = 0$, we obtain an equation analogous to that familiar in the cloud physics literature (cf., Equation 7.31 of Rogers and Yau [252]). The additional term describing removal of particles by gravitational settling is preserved (again assuming Stokes drag with $\tau_r = (\kappa x)^{-1}$. Krueger [162] showed that the resulting solution describes a droplet size distribution growing in a non-turbulent parcel with gravitational settling as the only removal mechanism:

$$p(x) = \sqrt{\frac{2\kappa}{\pi a}} \exp\left(-(\kappa/2a) x^2\right). \quad (7.10)$$

In terms of droplet radius it is

$$p(r) = 2 r \sqrt{\frac{2\kappa}{\pi a}} \exp\left(-(\kappa/2a) r^4\right). \quad (7.11)$$

This solution for the droplet size distribution in a fixed-supersaturation environment can reasonably be interpreted as the limiting solution when $\bar{s} \gg \sigma_s$.

Equations 7.8 and 7.10 provide two convenient extremes for comparison with observations; in the first case, only the supersaturation fluctuations determine the shape of the size

distribution and in the second case only the mean supersaturation contributes. The Stokes-settling removal process is common in both cases. Equation 7.5 describes the simplest case in which turbulence is dominant, and for which droplet removal does not depend on particle size. We see already that the exponent in the distribution, which determines the shape of the large-droplet tail of the distribution, is different for the three derived distributions: r^2 versus r^3 versus r^4 . Moreover, the slope of the tail is inversely proportional to σ_s or $\bar{s}$ depending on the case.

7.3 Size distributions from the principle of maximum entropy

Liu and Hallett [177] proposed a functional form of the cloud droplet size distribution based on Shannons maximum entropy principle for a system of droplets of fixed mass and number concentration. This distribution does not have any information about the physical processes responsible for the cloud droplet growth. Instead, a maximum-likelihood size distribution is derived by maximizing the system entropy and with a constraint of conservation of liquid water content. The result in terms of droplet radius is

$$p(r) = \frac{4\pi\rho n_t^2}{L} r^2 \exp\left(\frac{-4\pi\rho n_t}{3L} r^3\right), \quad (7.12)$$

where n_t is the total droplet concentration and L is the liquid water content. This is a

Weibull distribution with a shape factor of 3. In droplet-volume coordinates ($x \equiv r^3$), the distribution is transformed to a simple exponential function of x , i.e., $p(x) \propto \exp(-x)$. Interestingly, a similar form of the DSD was obtained by Williams and Wojtowicz [317] also using probabilistic arguments and the constraint of fixed cloud droplet concentration and liquid water content. Both derivations rest on the assumption that all droplet sizes allowed by the constraints have equal probability, and then consider the implications using the machinery of Boltzmann statistics.

Liu et al. [179] proposed a generalized system theory which produces a Weibull distribution with a variable shape factor. The shape factor of the maximum likelihood distribution depends on the conserved variable, for example in the above case, it is the liquid water content. Whether liquid water content, or some other variable like droplet surface area is conserved, then becomes the central physical question. It was suggested that this scale factor may depend on the scale of averaging and the level of turbulent fluctuations Liu et al. [179]. If the averaging is below some threshold, the shape factor of the distribution should depend on the length scale of averaging. Moreover, with decrease in turbulent fluctuations, the shape factor increases, and the distribution approaches towards a delta function corresponding to the zero fluctuation case. Connecting these general, probabilistic approaches to the diffusion-drift (Fokker-Planck) approach will be worth exploring.

7.4 Experiments and procedure for data analysis

7.4.1 Experimental Description

Experiments are performed in the controlled environment of the Π cloud chamber. Here the thermodynamics and turbulent flow properties are regulated by maintaining the boundary conditions for temperature and water vapor concentration. Turbulent moist convection is driven by an imposed vertical temperature gradient, and the water-saturated top and bottom boundaries create supersaturated conditions via isobaric mixing. A steady-state cloud is produced through the continuous injection of aerosol particles (cloud condensation nuclei) into the chamber, balanced by the loss of cloud droplets by sedimentation. Technical details about the Π chamber, associated instrumentation, and typical experimental configurations are described in more detail by Chang et al. [50].

Conditions used in the current study are the same as the experiments reported in Chandrakar et al. [46, 47]. The analysis of interstitial (nonactivated aerosol particles present in a cloud) and residual (aerosol samples obtained from collecting cloud droplets) aerosol distributions presented in Chandrakar et al. [47, see their Figure 3] indicates the maximum supersaturation fluctuations in the chamber rarely exceed 0.5 %. Moreover, the magnitude of supersaturation fluctuations (standard deviation) for these conditions is reported to be approximately 1.4 % [46]. These measurements indicate that the mean supersaturation for

the data set used for the current study is very close to zero. Direct measurement of the absolute value of supersaturation is challenging with the currently available instrumentation.

A well-characterized aerosol distribution for injection (nearly a log-normal distribution with a mode diameter $50 - 70$ nm and range $10 - 270$ nm) is generated by atomizing a solution of NaCl and passing it through a diffusion dryer. The size distribution and concentration of aerosols in the injection system and the cloud interstitials are measured using a scanning mobility particle sizer (TSI). For the experiments with size-selected aerosols, the full distribution of aerosols from the atomizer is passed through a differential mobility analyzer (TSI) set to select 200-nm aerosol particles. The primary part of the distribution is in the approximate range $145 - 235$ nm. There was a secondary mode at the larger size ($\approx 235 - 350$) associated with doubly charge particles, but their contribution to the overall concentration of aerosols were smaller compared to the primary mode.

The data used for this study were obtained when the microphysical conditions in the Π chamber were in steady state, corresponding to a given aerosol injection rate. In other words, we establish the fixed thermodynamic boundary conditions and wait for transients to decay, then begin injecting aerosol particles and wait approximately an hour for microphysical properties to stabilize. For size-selected aerosols, five aerosol injection rates were used, i.e., five separate steady-state experiments carried out over a period of approximately one week (each sample is from $6 - 8$ hrs of steady-state data segments, and number of droplets used for the analysis were between $2000 - 19200$). For the full-size-distribution aerosols, another six aerosol injection rates were used (each sample is from $6 - 20$ hrs of steady-state data

segments, and number of droplets used for the analysis were between 25800 – 862690).

Cloud droplet diameters are measured using a Phase-Doppler Interferometer (Dantec), and by averaging over time a size distribution can be estimated, subject to sampling uncertainty. For this study, droplet size bins below $7.5\text{ }\mu\text{m}$ are excluded during data fitting since there are indications that the Phase-Doppler system underestimates the droplet concentration at smaller sizes due to reduced signal-to-noise ratio. The minimum detectability of the instrument based on the manufacturer specifications for a configuration of transmitter and receiver optics used here is approximately $2.7\text{ }\mu\text{m}$. However, this lower limit is for optimal conditions, whereas in the II-chamber experiments the transmitted and received signals must pass through windows, which often have some condensation present. As a conservative approach, we use this slightly higher cutoff; it is consistent with the emphasis on the large-droplet tail during the analysis, described next.

7.4.2 Data analysis

The purpose of the analysis is to test whether the previously introduced functions are able to describe the measurements, to within acceptable statistical bounds. To that end, and with the capabilities of the available instrumentation in mind, the analysis is guided by the following principles:

1. Coordinates are defined so that the function is linear; i.e., as an exponential distribution on a semi-log plot.

Summary of Fitting Functions and Coordinates				
Case Label & Equation	Assumptions	Functional Form of Fit	Binning Method	Plotting Coordinates
D^2 distribution; Eq. 7.3	Size-independent removal	$p_1(x) = A \exp(-Ax);$ $x \equiv D^2$ (7.13)	Logarithmic bins in D^2	D^2 versus $\log(p(x))$
D^3 distribution; Eq. 7.10	Zero mean supersaturation and size-dependent removal	$p_2(x) = b_1 \sqrt{x} \exp(-b_2 x^3);$ $x \equiv D$ (7.14)	Logarithmic bins in D	D^3 versus $\log(x^{-1/2}p(x))$
D^3 distribution (Weibull); Eq. 7.12	System theory with the conservation of liquid water content	$p_3(x) = c_1 \exp(-c_2 x);$ $x \equiv D^3$ (7.15)	Logarithmic bins in D^3	D^3 versus $\log(p(x))$
D^4 distribution; Eq. 7.8	Zero supersaturation fluctuation and removal due to Stokes settling	$p_4(x) = d_1 \exp(-d_2 x^2);$ $x \equiv D^2$ (7.16)	Logarithmic bins in D^2	D^4 versus $\log(p(x))$

Table 7.1

Summary of the functions that are fitted to the data for the purpose of goodness-of-fit and residual analysis. The first column gives a label to be used in figure legends and the equation number from the text; the second column briefly summarizes the assumptions underlying the expression; the third column shows the function to be fitted to the data; the fourth column states the method used for binning the data; and the fifth column states the coordinates used in plotting the data. In all cases, the binning method and plotting coordinates are intended to provide a linear shape of the large-droplet tail of the distribution.

2. Data are binned logarithmically by droplet size so that reasonably uniform counts occur within all bins.

Both of these principles serve to make the comparison to theory as direct as possible, and to place emphasis on the behavior of the large-droplet tail of the size distribution. That is important because of the limitations in robustly measuring concentrations of small droplets. The approach also is intended to be as neutral as possible, allowing all functions to be compared to data in a way that is naturally suited to the distribution shape. The fitting functions, coordinates, and binning methods are summarized in Table 7.1. We discuss several more aspects of the two analysis principles in the following two paragraphs.

In order to express the four functions as simple exponential distributions, D , D^2 , or D^3 coordinates are used. The coordinate transformation employed to simplify the theoretical distributions is

$$\hat{p}(x_2) = p(x_1) \left| \frac{dx_1}{dx_2} \right|. \quad (7.17)$$

Using this approach has a distinct advantage since the resulting distributions can be plotted as linear functions in semilog- y coordinates, and therefore simplifies the fitting and evaluation processes. Significantly, the linear distributions also allow comparison of measurements with theoretical distributions even though we are missing the smaller droplets range ($D < 7.5 \mu\text{m}$) in the measurements.

Droplet measurements are used to generate size distributions with the logarithmic binning described in Table 7.1. The logarithmic bins lead to a more uniform distribution of the

data across bins, with the aim of maintaining relatively uniform sampling uncertainty for all data points [13]. Finally, a minimum threshold of > 5 droplet counts per bin is applied for the distribution calculations.

In order to perform a goodness-of-fit test, it is necessary to have a reliable estimate of the measurement uncertainties. The uncertainties are estimated by breaking the time series of droplet size into smaller time segments and calculating the standard deviation per size bin. This method is used for estimating the uncertainties because simple counting (sampling) uncertainties under-represent the actual variability in the data. In the turbulent flow generated in the Π chamber, uncertainties in the measurements due to counting and instrumental errors are dominated by the physical variability in the system. The main driving factor for the physical variability is the turbulent convection and the associated large-scale circulation [216]. The timescale associated with the large-scale circulation is the longest timescale in the convective system. Therefore, the data sample is divided into segments of several circulation timescales to estimate uncertainties ($\sim 45 - 60$ times the large-scale circulation timescale).

7.4.3 Weighted Least Square Fitting, χ^2 Test, and Goodness of Fit Evaluation

The functions described by Eqs. 7.13, 7.14, 7.15, and 7.16 are fitted to the data using weighted least-squares (WLS) regression. Again, suitable coordinates are used such that the functions are linear when plotted, as summarized in Table 7.1. The inverse of the

variances of counts for each bin are used as the weighting factors ($w_i = \sigma_i^{-2}$) for the least-squares fitting [13].

The primary statistical tool we use for determining goodness of fit by the theoretical distributions to the measurements is the χ^2 test. In this test, $\chi^2 \equiv \sum w_i(Y_i - \hat{Y}_i)^2$, i.e., the sum of squared error (SSE) and its probability is estimated for a fit. Here Y_i represents a measurement value, $\hat{Y}_i$ is the corresponding value from the fit, and again, w_i is the weighting factor. The expectation value (probability = 0.5) of the χ^2 distribution is equal to the degrees of freedom (DOF) of a fit, where the DOF is the number of data points minus the number of free parameters of the fit [13]. If a fit produces $\chi^2 \gg \text{DOF}$ or the χ^2 probability ($p|_{\chi^2}^\infty$) is significantly lower than the desirable cutoff, the fit is considered as failing to describe the measurement. The SSE can be used for hypothesis testing (i.e., pass or fail), but it can also be considered as a fitting statistic signifying how close the fitted function values are to the data set. In this work, we use it as a goodness-of-fit parameter.

We also apply residual analysis to obtain additional information about the goodness of a fit, and how it varies between different candidate fitting-functions. Unlike hypothesis testing or evaluations based on a single goodness-of-fit parameter (e.g., χ^2), residual analysis accounts for whether the measurement points are above or below the fitted function. In our application, therefore, the fit residuals are plotted versus the droplet size bin. Based on the trend of the residual plot, relative appropriateness of a fit can be judged [124]. For example, in an ideal case, i.e., for a good fit, the fit residuals should be distributed randomly, being equally likely to be positive or negative, and their magnitudes should be small. However, if

they have a monotonic (increasing or decreasing) trend, it means that there is a consistent deviation between the measurements and the fitted function. In our case, because the functions are linear and are fitted to the data, only a nonlinearity of the data would result in a consistent pattern (a ‘run’) of residuals with droplet size. A consistent change in the sign of residuals is easily interpreted as indicative of a poor fit. In this study, all three methods are used to evaluate the theoretical functions described in Table 7.1: the χ^2 -test, the relative comparison of the goodness of fit parameter (SSE), and the residual analysis.

7.5 Results

As indicated earlier, two different sets of experiments are compared to the four distribution functions: experiments with input of monodisperse, size-selected aerosol and experiments with a broad, polydisperse distribution of aerosol. The droplet size distributions from both of the experiment sets, the weighted least-squares fitting results, and the statistical analysis as described in the previous section are presented next.

7.5.1 Experiments with monodisperse aerosols

We begin with the analysis of cloud droplet size distributions formed on size-selected aerosols (injection of a narrow, monodisperse distribution of aerosols with a mode diameter of around 200 nm). Five different aerosol concentration settings are used to achieve progressively increasing cloud droplet concentrations, corresponding to extremely clean to moderately

Figure 7.1: Weighted Least-square fitting of the D^2 -distribution Eq. 7.13 to the measured cloud droplet size distributions, for size-selected aerosol. The five panels correspond to, from top to bottom and left to right, increasing aerosol injection rate and cloud droplet concentration. The binning method and coordinates are defined in Table 7.1.

Figure 7.2: Weighted Least-square fitting of the D^3 -distribution Eq. 7.14 to the measured cloud droplet size distributions, for size-selected aerosol. The five panels correspond to, from top to bottom and left to right, increasing aerosol injection rate and cloud droplet concentration. The binning method and coordinates are defined in Table 7.1.

Figure 7.3: Weighted Least-square fitting of the D^3 -Weibull-distribution Eq. 7.15 to the measured cloud droplet size distributions, for size-selected aerosol. The five panels correspond to, from top to bottom and left to right, increasing aerosol injection rate and cloud droplet concentration. The binning method and coordinates are defined in Table 7.1.

Figure 7.4: Weighted Least-square fitting of the D^4 -distribution Eq. 7.16 to the measured cloud droplet size distributions, for size-selected aerosol. The five panels correspond to, from top to bottom and left to right, increasing aerosol injection rate and cloud droplet concentration. The binning method and coordinates are defined in Table 7.1.

Figure 7.5: Ratio of the sum of squared error (SSE) and the degrees of freedom (DOF) for WLSQ fittings of measured distributions, versus steady-state cloud droplet concentration n_d for the five size-selected aerosol cases.

clean cloud conditions (from about 11 to 77 cm^{-3}). The droplet size distributions (blue circles with measured uncertainties) and fitted functions (red lines) are presented in Figures 7.1, 7.2, 7.3, and 7.4. Underneath each size-distribution plot is a plot of the corresponding fit residues. The blue circles are the residuals (difference between the measurement and the fitted value for each size bin), with dashed lines to aid in detecting trends, and the red line indicates a zero residual, perfect fit to the data. The panels are arranged from top-left to bottom-right in order of increasing droplet number concentration.

Figure 7.1 shows the fitting of the D^2 function, Eq. 7.13. This solution is for stochastic condensation with a constant (size-independent) droplet removal rate. The fits all appear to be reasonable, as judged by eye. The residual plots, however, show a modest but consistent trend with a concave-down shape, indicating that the theoretical function falls off somewhat

too weakly in the large-droplet tail compared to the measurements.

Figure 7.2 shows the fitting of the D^3 function, Eq. 7.14, corresponding to stochastic condensation with size-dependent droplet removal. Again, the fits appear quite reasonable by eye. The residual plots are also encouraging, showing no obvious trends, i.e., with residuals randomly distributed about the zero-line, and also with smaller magnitudes (mostly between ± 0.1) compared to the previous case.

Figure 7.3 shows the fitting of the D^3 -Weibull distribution, Eq. 7.15, which is obtained from probabilistic approaches based on assumption of fixed concentration and total volume. The fits are okay, given the uncertainties, as confirmed by the χ^2 test presented later, but in this case it is evident even to the eye that in the plotting coordinates that result in a linear theoretical function, the measurement points have a concave-up shape. This is readily confirmed in the residual plots, also showing a distinct, concave-up shape consistent for all cloud droplet concentrations. It should also be noted that the range of the residuals is considerably larger than in the previous two cases.

Figure 7.4 shows the fitting of the D^4 distribution, Eq. 7.16, corresponding to growth of cloud droplets with fixed supersaturation and size-dependent removal. The situation here is similar to the D^3 -Weibull distribution just discussed, with clearly identified nonlinear shape of the measurements in the D^4 coordinates. The theoretical function falls off too rapidly with increasing size compared to the measurements. As in the previous case, the residual plots confirm this, all showing a distinct, concave-up pattern. The range of residuals is largest in this case.

It is worth noting that there is no clear trend in the relative distribution shape between different aerosol injection rates, apart from the change in the distribution width. Based on the visual inspection and residual analysis made thus far, the D^2 (Eq. 7.13) and D^3 (Eq. 7.14) distributions appear to be most appropriate. That they can capture the size distribution shape over an order of magnitude change in droplet number concentration seems quite remarkable to us.

In order to make an additional, quantitative comparison between the suitability of the four functions for describing the measurements, we use the χ^2 test. Figure 7.5 shows the sum of squared errors of prediction (SSE or χ^2) for all functional fits and for the five different cloud droplet concentrations. In most of the cases (except, the fourth case where only the D^2 distribution and D^3 distribution for size-dependent removal pass), all distribution fits pass the χ^2 test since the χ^2 values are close to the DOF. By pass, we mean that SSE is less than DOF. Here it is worth recalling that SSE greater than DOF implies a poor fit, and low likelihood of the function describing the measurements. The opposite extreme, however, with SSE much less than DOF implies that the fit is too good to be true, given the estimated uncertainties; this is usually an indication that the uncertainties of the data are overestimated. In this case, in four out of the five cases, all theoretical models technically pass the fit test. However, when we compare the relative SSE values as a goodness-of-fit parameter, the D^3 distribution is consistently the lowest and therefore the optimal fit. In four of the five cases, this is followed by the D^2 distribution. These quantitative conclusions are consistent with those reached from the residual analysis earlier in the section.

Furthermore, the residual plots suggest a Weibull distribution with a shape factor between 2 and 3 would fit the large-droplet tail of the distribution well. The residuals in Fig. 7.1, corresponding to Weibull with shape factor 2, were concave down, and the residuals in Fig. 7.3, corresponding to Weibull with shape factor 3, were concave up. The exponential in the D^4 distribution certainly decays too sharply. The D^3 distribution function corresponding to stochastic condensation with size-dependent removal apparently is the best fit: it has an additional $\sqrt{D}$ multiplied with the D^3 -exponential, resulting in a tail that decays at a slightly slower rate than the D^3 Weibull distribution, but sharper than the D^2 Weibull distribution.

7.5.2 Experiments with polydisperse aerosols

Experiments with a broad, polydisperse size distribution of aerosol input were carried out for three reasons: first, to allow larger cloud droplet concentrations to be achieved, thereby allowing more polluted cloud conditions to be explored, second, to obtain lower counting uncertainties as a result of the higher concentrations, thereby placing stronger constraints on the theoretical fits, and third, to explore any possible dependence of the cloud droplet size distribution on the shape of aerosol size distribution. In the six steady-state experiments, cloud droplet concentration is varied over two orders of magnitude, from very clean to very polluted (from about 21 to 1790 cm^{-3}) conditions. The observed droplet size distributions and the four functional fits are summarized in Figures 7.6, 7.7, 7.8, and 7.9. As before,

Figure 7.6: Weighted Least-square fitting of the D^2 -distribution Eq. 7.13 to the measured cloud droplet size distributions, for polydisperse aerosol. The six panels correspond to, from top to bottom and left to right, increasing aerosol injection rate and cloud droplet concentration. The binning method and coordinates are defined in Table 7.1.

Figure 7.7: Weighted Least-square fitting of the D^3 -distribution Eq. 7.14 to the measured cloud droplet size distributions, for polydisperse aerosol. The six panels correspond to, from top to bottom and left to right, increasing aerosol injection rate and cloud droplet concentration. The binning method and coordinates are defined in Table 7.1.

Figure 7.8: Weighted Least-square fitting of the D^3 -Weibull-distribution Eq. 7.15 to the measured cloud droplet size distributions, for polydisperse aerosol. The six panels correspond to, from top to bottom and left to right, increasing aerosol injection rate and cloud droplet concentration. The binning method and coordinates are defined in Table 7.1.

Figure 7.9: Weighted Least-square fitting of the D^4 -distribution Eq. 7.16 to the measured cloud droplet size distributions, for polydisperse aerosol. The six panels correspond to, from top to bottom and left to right, increasing aerosol injection rate and cloud droplet concentration. The binning method and coordinates are defined in Table 7.1.

Figure 7.10: Ratio of the sum of squared errors (SSE) and the degrees of freedom (DOF) for WLSQ fittings of measured distributions, versus steady-state cloud droplet concentration n_d for the six full-distribution, polydisperse aerosol cases.

panels are organized by increasing cloud droplet concentration, from upper-left to lower-right.

Figure 7.6 shows the fitting of the D^2 function, Eq. 7.13. This solution is for stochastic condensation with a constant (size-independent) droplet removal rate. The fits at the lowest concentration (upper left panel) and at the fourth level of concentration (lower left panel) appear reasonable. Intriguingly, however, the two moderate-concentration cases in the upper middle and right panels show distinctly weaker tails in the measurements compared to the theory, whereas the high-concentration cases in the lower middle and right panels show much stronger tails. The residual plots show that even the two fits that seemed reasonable do, in fact, display strong biases, with concave-down shape similar to the two moderate-concentration cases. The two high-concentration cases show a flip in the shape

of the residual curve and display very high magnitudes indicative of a poor fit.

Figure 7.7 shows the fitting of the D^3 function, Eq. 7.14, corresponding to stochastic condensation with size-dependent droplet removal. The first four panels show reasonable fits. The goodness-of-fit in the upper-left panel, corresponding to the lowest droplet concentration, is further borne out in the residual plot with small magnitudes. The next three droplet concentration levels (upper middle and right plots, and lower left plot) show deviations in the tails indicated by the distinct, curved shape of the residual patterns. The residual patterns flip from concave-down to concave-up for these three panels, but the magnitudes of the residuals are reasonably small compared to other fits to the polydisperse-aerosol cases. The two highest droplet concentration cases (lower middle and right panels) are clearly poor fits, with the measurements displaying pronounced large-droplet tails that are not captured by the theoretical distributions.

Figure 7.8 shows the fitting of the D^3 -Weibull distribution, Eq. 7.15, which is obtained from probabilistic approaches based on assumption of fixed concentration and total volume. Most of the fits can be seen to deviate from the measurements. The first four panels, corresponding to the lowest droplet concentrations, show obvious, concave-up residual patterns, but the magnitudes are generally below 1. Once again, the two cases with highest droplet concentration show obvious deviations in the tails, with the measurements displaying enhanced large-droplet concentrations.

Figure 7.9 shows the fitting of the D^4 distribution, Eq. 7.16, corresponding to growth of cloud droplets with fixed supersaturation and size-dependent removal. The situation here is

again similar to the D^3 -Weibull distribution just discussed, with clearly identified nonlinear shape of the measurements in the D^4 coordinates. And once again, the deviations are more pronounced with increasing cloud droplet concentration: the theoretical function falls off too rapidly with increasing size compared to the measurements. As in the previous case, the residual plots confirm this, all showing a distinct, concave-up pattern. The range of residuals is largest in this case.

Figure 7.10 shows the SSE, normalized by the DOF, for all the fittings of the distributions from the different aerosol cases. In a relative sense, the results from the polydisperse aerosol cases are similar to the monodisperse aerosol experiments: in these cases as well, the D^3 and D^2 distributions are the most successful in describing the measurements. In the four cases with lowest cloud droplet concentration the D^3 distribution has the lowest SSE/DOF, whereas in the two most polluted cases the D^2 distribution is the best fit. Except for the last two cases, the D^3 distribution passes (or nearly passes) the χ^2 -test. The D^2 distribution passes in the lowest-concentration case. The D^3 -Weibull and D^4 distributions are reasonably close to passing the χ^2 for the four lowest-concentration cases. However, it should be recalled that for most of these borderline χ^2 test results, it is indeed true that there were significant deviations in the tail of the distribution for the measurements compared to the fit.

For the last two, polluted cloud conditions the χ^2 test clearly rejects all distributions, with SSE/DOF values much greater than unity. This is consistent with what already was evident from visual inspection of the fits: all four theoretical distributions fail to faithfully capture

the shape of the measured distribution in very polluted conditions. Unlike the size-selected, monodisperse aerosol cases, there is an evident trend in the deviation of the distribution shapes relative to the measurements as aerosol concentration is increased under polydisperse conditions.

7.6 Discussion

The purpose of this study was to evaluate a drift-diffusion framework for stochastic condensation that leads to a Fokker-Planck equation for the cloud droplet size distribution (Eq. 7.2). It can be solved to obtain analytical size distribution shapes under certain simplifying assumptions. Three solutions are explored: stochastic condensation with a constant cloud droplet removal rate [255], stochastic condensation with a removal rate proportional to the square of the particle diameter (relevant for settling with Stokes drag), and droplet growth in a fixed supersaturation field, with a similar diameter-squared removal rate [162]. In addition, a Weibull distribution with shape factor of 3 can be obtained from the Fokker-Planck equation with suitable assumptions for the drift or diffusion terms, and also results from mechanism-free probabilistic arguments [179, 317]. The functions are summarized in Table 7.1.

The II chamber provides an ideal environment in which to test idealized distributions because the number of physical processes contributing to cloud droplet evolution is relatively small compared to the atmosphere: activation on CCN, growth due to condensation in a

fluctuating environment, and eventual removal due to settling or turbulent transport. The II Chamber configuration used here allows the boundary and aerosol conditions to be well characterized and steady in time. In addition, because the experiments are carried out in steady state, microphysics can be measured over long durations, allowing large-droplet tails to be well resolved.

Comparison of the SSE/DOF values along with analysis of fitting residuals suggests that the D^3 distribution corresponding to stochastic condensation with size-dependent droplet removal most successfully fits the observations. For these conditions, we can confidently state that the drift-diffusion approach and resulting Fokker-Planck equation are able to capture the behavior of the cloud droplet size distribution over the parameter range explored. Application of the χ^2 test by itself suggests that all four theoretical distributions can plausibly describe the observations under conditions of monodisperse aerosol injection. However, a careful comparison of the goodness of fit parameter (SSE/DOF) and residual analysis favors the D^3 distribution for the size-dependent removal followed by the D^2 Weibull distribution.

In the experiments with polydisperse aerosol injection the situation is more complex. Again, the D^3 distribution is most successful in fitting the measurements, except for the two experiments with highest aerosol injection rate, for which none of the functions are deemed acceptable by the χ^2 test. In those cases the large-droplet tail of the measured size distributions far exceeds what can be described by any of the distribution functions. This suggests that there is missing physics in the governing equation. We can speculate on two causes. The first is the possibility that activation and deactivation of aerosol particles of different

size with different efficiency may be the cause of the pronounced growth of the large-droplet tail. It stands to reason that this would be most effective when cloud droplet concentrations are high, with plentiful interstitial aerosol, all residing in a low mean supersaturated environment with turbulent fluctuations. Overall, this type of broadening is related to the Ostwald ripening effect [9, 131, 156, 322, 339]. During polluted cloud conditions, the average supersaturation is expected to be nearly zero, but the presence of turbulent flow condition allows the supersaturation to fluctuate above and below the mean value. Therefore, there is a possibility of multiple activation-deactivation cycles. Due to the irreversibility of the droplet growth with the curvature and solute effects, size distribution broadening results. Obtaining analytical solutions when including the curvature and solute effects in the Fokker-Planck equation will remain a challenge for future work. The second possible explanation for the unaccounted broadening of the droplet size-distribution in the most polluted cloud condition would be the effect of a highly-skewed supersaturation distribution. In polluted cloud conditions the mean supersaturation in the chamber should be very close to zero. Although the phase relaxation time is expected to be very short compared to mixing times, there could be localized growth and activation at boundaries before complete mixing of the scalar fields. The possibility of strongly non-Gaussian behavior in the supersaturation statistics is not accounted for in the stochastic condensation growth model.

Rejecting or accepting theoretical expressions rests heavily on the quality and uncertainty of the measured large-droplet tails. Higher statistical confidence than obtained here will require further reducing statistical uncertainties, mainly by measuring farther into the large-droplet tail. For a given cloud droplet concentration, that implies measuring for a longer

time (or using an instrument with a larger sample volume). Longer measurement time places more severe constraints on the ability to maintain steady thermodynamic conditions in the Pi Chamber; most notably, drifts in the water-vapor boundary conditions over the period of days have been noted. Relying on rare droplet counts also places high demands on the ability of the droplet measurement to reject noise and ensure high probability of detection. Finally, reliable measurement ability in the small-droplet part of the distribution is sure to help substantially.

Lastly, we briefly consider implications for clouds in the atmosphere. The laboratory measurements favor the D^3 and D^2 distribution resulting from stochastic condensation, either with size-dependent removal or constant removal rate. When polydisperse aerosols were used, however, there was some deviation near the tail of the distribution possibly due to the significant effect of Ostwald ripening and associated deactivation and activation. In non-precipitating atmospheric clouds, the removal due to the gravitational settling is usually considered to be negligible. Therefore, the D^2 Weibull distribution would be a more appropriate form of DSD in such clouds unless there are other dominant and complicating processes like droplet collision-coalescence or entrainment effect near the cloud top. The stratocumulus cloud core before the start of significant drizzle formation would be an ideal location for applicability of this model. Exploring under what conditions the mean versus fluctuating supersaturation will be of primary importance, and when other factors such as Ostwald ripening will become significant, will be of great interest for future exploration.

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Chapter 8

Concluding Remark and Future Outlook

Aerosol-cloud interactions are crucial for the climatological impact of pollution due to their influence on global radiative forcing and precipitation formation process. An essential aspect of this problem is the connection with the cloud microphysical processes. Turbulence is ubiquitous in atmospheric clouds and crucial for microphysical interactions and cloud dynamics. With this motivation, the problems presented in this thesis are focused on the investigation of aerosol-cloud interactions in a controlled manner. We investigated the following central questions in the previous chapters: How are the turbulent fluctuations in clouds affect the traditional picture of aerosol indirect effects and aerosol-cloud feedback? Can we quantify these effects? How do the supersaturation fluctuations affect droplet size-distribution growth? Is there any general shape of droplet size distribution in turbulent

clouds, and what is its form?

These problems are studied experimentally by creating steady-state turbulent clouds in a controlled laboratory setting, Michigan Techs Pi-chamber. It provides unique advantages of longtime statistics of cloud, aerosol, and turbulent flow properties in relatively know conditions which are critical for this study. A theoretical analysis based on the stochastic method is also presented here to explore the cloud droplet growth in a turbulent environment. The theoretical framework also provides some useful relation to investigating the effect of aerosol on the cloud microphysics and supersaturation field.

A central result of all analysis presented in this thesis is that the supersaturation fluctuations in a turbulent cloud are a crucial element of all microphysical interactions and aerosol feedbacks. Key variables associated with microphysical interaction in turbulent clouds (before the start of significant collision-coalescence) are

1. Supersaturation statistics (mean and fluctuations)
2. Timescales associated with the problem: the Lagrangian scalar-correlation timescale in a turbulent flow (τ_t), the phase relaxation timescale (τ_c), and the droplet removal timescale (τ_r)
3. Aerosol properties: chemical composition, concentration, and size.

Turbulence-induced aerosol effects: In chapter 3 and 4, we showed the droplet size dispersion $\sigma_{r^2} \propto \sigma_s \tau_s \tau_r^{1/2}$. Analogous to a traditional first aerosol-indirect-effect or Twomey

effect, cloud droplet concentration increases with an increase in aerosol concentration in turbulent clouds. In the classical picture of the first indirect-effect, increase in droplet concentration with aerosol concentration is considered as a result of only mean supersaturation field. Although, in turbulent clouds, effects of supersaturation fluctuations in droplet activation process might be significant [2, 9, 81, 165]. Therefore, as a combined effect of supersaturation mean and fluctuations, cloud droplet concentration rises in response to increases in aerosol population, which decreases the phase relaxation timescale of cloud droplets and the system timescale (a harmonic mean of the turbulent correlation and phase relaxation timescales). Consequently, the mean, as well as the fluctuations of supersaturation, decreases with the aerosol population, and so as the droplet size mean and dispersion. It is because the dispersion growth of droplet size-distribution depends on the cross-correlation of droplet size and supersaturation fluctuations.

This turbulence-induced aerosol effects on cloud droplet size-distribution have implications for the cloud optical properties and precipitation formation. The narrowing of droplet size distribution, with an increase in aerosol concentration, reduces the effective radius of scattering; hence, the cloud optical-depth increases. This change in the cloud optical-depth, due to change in droplet size-distribution width, is also known as the dispersion effect. Likewise, a change in the dispersion of a droplet size-distribution, with aerosol population, also influences the precipitation formation rate. Therefore, apart from the first and second aerosol indirect-effects due to the mean supersaturation field, the presence of turbulent fluctuations causes an additional contribution to these processes.

Turbulence-induced cloud-cleansing: In a transient cloud system, aerosol population, cloud droplets, and supersaturation field are coupled in a positive feedback loop. It is a scenario where a polluted cloud system with limited aerosol sources undergoes a cleansing process (for example, the transition of a closed-cell stratocumulus cloud system to an open-cell structure) [102, 310]. With a decrease in the aerosol population, supersaturation mean and fluctuations increases, and so as the droplet size mean and dispersion. It increases the removal rate of cloud droplets for a size-dependent removal process (as well as the activation loss of aerosols). Therefore initially, aerosol and cloud droplets decay slowly as a result of suppressed supersaturation mean and fluctuations. Once the phase relaxation timescale reaches a value close to the turbulence timescale, the supersaturation mean and fluctuations start increasing and so as the decay rates of aerosol and cloud droplets. At the end of this initially slow and then faster decay, cloud collapse entirely.

Implications for boundary-layer clouds: In stratocumulus clouds, the mean vertical velocity is relatively low; thus, supersaturation fluctuations could have a significant effect on aerosol-cloud feedbacks. In marine stratocumulus clouds, the precipitation suppression enhances the latent heating and reduces the evaporative cooling. Thus, it alters the dynamics, turbulent kinetic energy, and entrainment rate. Moreover, cloud thickness positively correlates with aerosol concentration [233]. As a result of the above feedbacks, the entrainment rate may be enhanced, effectively forcing the system toward a thinner cloud. Although the cloud thickening increases the liquid water and moisture flux which facilitate buoyant production [321]. In addition, the cloud-top entrainment velocity (w_e) decreases with an increase in aerosol due to a change in droplet settling flux ($w_e \propto -d^5$) [67]. Therefore,

there are considerable positive and negative feedbacks governed by aerosol perturbation through microphysics. The overall effect on the cloud system depends on whether these perturbations are amplified or dampened by dynamic forcing and the aerosol regime (clean versus polluted). Apart from micro-scale properties, the aerosol population can similarly influence mesoscale cloud structure (associated radiative fluxes) and the convection pattern. Indeed, the transformation of a cloud from a closed-cell structure to an open-cell structure is feasible merely due to the reduced aerosol concentration [310].

Implications for climate studies and sub-grid-scale models: Liu and Daum [175] and subsequent studies pointed out the change in the relative-dispersion of cloud droplet size distribution with aerosol concentration, which influences the effective radius and cloud albedo. Therefore, it could have a significant influence on the overall radiative forcing. Studies presented in this thesis would help to understand some aspects of this dispersion effect, which might not be clear from field observations. For example, we explored the effect of aerosol concentration in droplet size relative-dispersion in turbulent cloud conditions using controlled laboratory experiments and stochastic theory. This study could provide some physical basis for their parameterization in climate models. The current study explored an additional aspect of this problem, the effect of turbulence and supersaturation fluctuations, and it was not explicitly considered before. In climate models, the effects of turbulence or supersaturation variability are unresolved and far from implementation. Considering that, studies like current would help to develop some simplistic models for incorporating such effects. For example, the relation of relative dispersion ($d = \sigma_r/\bar{r}$) presented in chapter 5: $d \propto \sigma_s/\bar{s} t^{-1/2}$. Here, t is an average droplet growth timescale or lifetime.

In climate models, and even in most of the cloud-resolving models, the sub-grid-scale microphysical interactions with the turbulent fluctuations are not resolved or modeled (only the mean effects are incorporated). There are some recent developments in the Large-eddy-simulation where these effects of fluctuations are modeled by applying random vertical velocity fluctuations [104] or using linear-eddy-model for sub-grid-scale mixing [119] with a Lagrangian microphysics model. Although, these models are computationally expensive as well as do not resolve the fluctuations and the Lagrangian correlation completely. Besides, for large-scale models, only the bulk microphysics schemes are computationally feasible. Therefore, the relationships presented in chapter 3 and 4 about the effect of supersaturation fluctuations on droplet size growth (example, $\partial_t \sigma_{r_2}^2 \propto \sigma_s^2 \tau_s$) can be used as a sub-grid-scale bulk parameterization.

Similarly, in bin microphysics schemes, the physical diffusion process due to sub-grid-scale turbulent fluctuations is not included. Using the stochastic approach presented here, this diffusion flux of a droplet size distribution can be modeled as [255]:

$$F_{diff} = \mathcal{D} \frac{\partial^2 n_d(x)}{\partial x^2}, \quad (8.1)$$

where $\mathcal{D} = \partial_t \sigma_{r_2}^2 \propto \sigma_s^2 \tau_s$, and $x \equiv r^2$.

8.1 Future Directions

8.1.1 Future work related to the Pi-chamber:

Droplet removal timescale in turbulent Rayleigh-Bénard convection: The droplet removal timescale in the chamber is very central to the growth of a droplet size distribution. It was coarsely estimated in [43] based on the decay rate of a clean cloud (a low interstitial aerosol concentration case). In a turbulent moist Rayleigh-Bénard convection when droplets are activating, growing, and at the same time settling due to gravity, an estimate of the removal timescale is not straightforward since the gravitational settling velocities of droplets are much smaller than the turbulent fluctuations. Moreover, there are other dynamical features of the flow like, large-scale circulation, boundary-layer structure, and high supersaturation near boundaries, which are central to the droplets dynamics. It would be interesting to explore this problem using a direct numerical simulation or by tracking droplet trajectories near the bottom of the chamber using the holographic instrument. It might also be worth exploring this problem by analyzing the budgets of aerosol and cloud droplet concentration in the chamber.

Turbulence-induced cloud droplet activation: Can we get a relation between aerosol activation and supersaturation statistics in a turbulent environment? A commonly used CCN activation parameterization in models is given by Twomey [301] : $N_{CCN} = C S^k$,

where N_{CCN} is number of activable CCN, and C and k are parameterization constant. This relation assumes an adiabatically rising parcel with no turbulence. Therefore, it is not suitable for a turbulent cloud field where supersaturation is significantly fluctuating. For atmospheric cloud models, a relationship which accounts for a supersaturation variability at sub-grid-scale is required. The activation/deactivation flux would depend on supersaturation PDF (or its statistics like mean, variance, and skewness), aerosol size-distribution, and turbulence correlation timescale. The droplet activation process involves the Lagrangian interaction of supersaturation field and aerosol particles. Consequently, both timescales, activation timescale based on aerosols properties and turbulence timescale, are critical [2, 9].

Activation-deactivation flux and the Ostwald ripening effect: As shown in chapter 7, the droplet size distribution for a polluted condition (high aerosol concentration) diverges from a theoretical distribution based on the stochastic condensation approach. The measured distributions have a slightly broader tail. In polluted conditions, mean supersaturation is expected to be very low (near zero), and there could be a significant amount of subsaturated region. As a result, multiple activation-deactivation cycles of cloud droplets might lead to broadening of size-distribution as observed. The irreversibility of the activation-deactivation process, due to the curvature and solute effect, causes the broadening of a droplet size distribution, also known as the Ostwald ripening effect [9, 131, 156, 322, 339]. It would be worth exploring this topic further with the help of Pi-chamber experiments and stochastic modeling tools to determine the condition for this process. For example, the following questions might be of interest: Is the timescale of supersaturation fluctuations in the chamber is sufficient to produce this effect? What is the fraction of supersaturation PDF

which needs to be subsaturated to produced observable effect? Is there any conceivable condition in atmospheric clouds (e.g., regions dominated by entrainment-mixing) sufficient for this process?

Characterization of inhomogeneity in droplet distribution: The stochastic condensation theory presented in chapter 3 and 4 assumes a uniform distribution of cloud droplets in a volume of interest. We extended this stochastic model to account for additional effects of spatial nonuniformity of droplet number concentration (or integral radius) in the growth of cloud droplets. Additionally, we tested this theoretical formulation with measurements performed in the Pi-chamber using digital-inline-holography in collaboration with Neel Desai and others [74]. The holographic measurements provide volumetric information about droplet sizes and their location. This study indicates significant variability in droplet concentration in turbulent moist-convection (at least, in the case explored with a low aerosol injection rate). Although, the cases with high aerosol concentration require further investigation.

An important question arises from these measurements, what is the source of variability in droplet concentration (or integral radius)? The measured droplet sizes are small enough for any inertial effects (the Stokes number, defined as the ratio of the particle response timescale and the characteristic timescale of fluid flow, is much less than one). Another possible hypothesis would be the skewness in supersaturation PDF due to boundary effects or the presence of the plume structure and their organization, which can lead to the nonuniform

activation of droplets. It would be worth exploring this topic further by doing some experiments with different aerosol injection rates to cover the clean and polluted cloud regimes. It might be possible that the nonuniformity would be more pronounced in polluted conditions due to lower mean supersaturation. However, these experiments will require improvement in the Holographic setup to measure smaller cloud droplets. Atmospheric implications of this study include the aerosol-induced inhomogeneity in the region dominated by entrainment in clouds, where supersaturation distribution expected to have a high skewness [207]. For example, Dodson and Small Griswold [82] reported some measurements indicating that the cloud droplet inhomogeneity is correlated with aerosol concentration.

Stochastic condensation with non-Gaussian distribution of supersaturation fluctuations: The stochastic condensation theory assumes a Gaussian shape of supersaturation PDF. This assumption might not be appropriate in the regions dominated by the entrainment and mixing process [207]. Therefore, the theoretical framework needs to account for this non-Gaussian behavior to appropriately capture the effect of entrainment and mixing in droplet activation and growth.

Mixed-Phase Clouds: We have also explored the mixed-phase clouds in the Pi-chamber. A study presented in Desai et al. [75] explored this topic by creating a steady-state mixed-phase cloud through continuous injection of cloud condensation nuclei and ice-nucleating particles at a controlled thermodynamic and turbulent condition. By controlling the relative

fraction of ice nucleating particles, we achieve different degrees of the *Wegener-Bergeron-Findeisen* process, and the findings are consistent with the theoretical criteria of glaciation (Korolev [153]) within experimental uncertainties. Measurements of cloud particles with digital-inline-holography also show significant variability in the phase partitioning (segregation of ice particles and liquid droplets). This observation motivates us to think about the effect of phase partitioning in the liquid and ice phase microphysics. Can we develop a theoretical model to incorporate the effect of variability in phase-partitioning in the supersaturation field and cloud particle size-distribution? What would be its implications for the activation of cloud droplets and the nucleation of new ice particles? Moreover, the Pi-chamber would be the best place to explore the optical properties of mixed-phase clouds in a controlled manner. There are many uncertainties and open questions related to the light propagation through ice or mixed-phase clouds which can be investigated in the Pi-chamber.

8.1.2 Future work related to the atmospheric modeling:

Supersaturation variability in the stratocumulus clouds: Boundary-layer clouds are strongly linked to climate sensitivity and are susceptible to aerosol perturbations[269]. Turbulence is ubiquitous in atmospheric processes, including convection in the atmospheric-boundary-layer and cloud processes. Moreover, the turbulence-microphysical interactions in boundary-layer clouds would depend on the magnitude of supersaturation fluctuations. Therefore, it would be worth to analyze the supersaturation variance and their scaling

in boundary-layer clouds. Cloud models and large-scale models (e.g., General circulation models) are typically do not resolve the sub-grid-scale microphysical interactions with turbulence; hence, the study of supersaturation variance would help develop some useful parameterizations for these interactions.

Supersaturation is nonlinearly coupled with two scalars, water vapor mixing ratio q_v and temperature T . Therefore, the supersaturation variance depends on the variance of water vapor and temperature field and their covariance [276]. In turbulent convection, a scalar variance is directly proportional to the net scalar flux. Significant scalar fluxes in boundary-layer clouds include surface flux due to turbulent transport, convective flux, and entrainment flux from the free troposphere. These fluxes contribute to overall scalar variance in the mixed-layer. However, some of these fluxes might have significant local influence. For example, entrainment flux dominates near the interface-layer (between the capping-inversion and the cloud-layer). Therefore, scalar variances and their scaling might be different in the mixed-layer and the entrainment-zone. Previous studies [70, 207, 213] focused on the moisture variance at the entrainment zone and its parameterization. They showed that the moisture variance peaks in the entrainment zone. Moreover, Wyngaard et al. [328] showed that the covariance of water-vapor and temperature varies with height and become negative at the top part of atmospheric boundary-layer.

As discussed above, this problem involves multiples aspect, which demands a detailed investigation using numerical tools and field observations. There are some recent development

in measurement technique (e.g., helicopter-based method: ACTOS [276]), which would allow high-frequency measurements of scalar fields in boundary-layer clouds. The problem of supersaturation variance can be explored numerically by implementing the scalar variance and covariance prognosis in atmospheric Large-eddy-simulations (none of the cloud models have such sub-grid-scale variance and covariance prognosis). It would also be interesting to investigate the supersaturation variance profile in boundary-layer with and without the presence of cloud forcing.

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Appendix A

A.1 Symbols used

Definitions of the symbols used in the dissertation are provided in Table A.1.

Table A.1
List of Symbols

Symbols	Description
d	Relative Dispersion of Droplet Radius
d_a	Droplet Area Relative Dispersion
Da	Damköhlerr number
D_v	Molecular Diffusivity of Water Vapor in Air (Corrected for Latent Heat Release During Phase Change)
H	Average Droplet Vertical Displacement Inside the Chamber
k_l	Coefficient of Long Collision Kernel
L	Liquid Water Content of Cloud
n_d	Droplet Number Concentration of Cloud
P_L	Cloud Liquid Water Autoconversion Rate
r	Radius of Droplet
r_{cr}	Critical Radius for Autoconversion
s	Supersaturation
s_o	Mean Equilibrium Supersaturation in the Absence of Cloud
v_t	Stokes velocity
$\bar{w}$	Mean Vertical Velocity of a Cloud Parcel
x_{cq}	Critical Mass Ratio
σ	RMS Fluctuations in Droplet Radius
σ_{r^2}	RMS Fluctuations in r^2
σ_s	RMS Supersaturation Fluctuations
σ_{s_o}	RMS Supersaturation Fluctuations in the Absence of Cloud
τ_a	Autoconversion Time Scale for Cloud Liquid Water Content
τ_c	Phase Relaxation Time
τ_l	Droplet Lifetime
$\tau_{l,c}$	Cloud Droplet Lifetime Based on Autoconversion of Cloud Droplet Number Concentration
τ_s	Global System Time Scale
τ_t	Turbulence Correlation Time
$\eta(t)$	RMS Supersaturation Fluctuations
ξ	Condensation Growth Parameter
ρ_w	Density of Water
α_d	Shape Parameter for a Gamma Droplet Radius Distribution
β_d	Scale Parameter for a Gamma Droplet Radius Distribution
Γ	Gamma Function

A.2 Lifetime of a droplet in the chamber

To estimate the lifetime of a single droplet in the chamber, τ_l , the vertical velocity of a droplet is approximated as the Stokes velocity: $v_t = ar^2$, with the constant $a = 1.2 \times 10^8 \text{ m}^{-1} \text{ s}^{-1}$. Droplets are assumed to be in the Stokes regime and the effect of turbulent flow on the settling speed is not significant. Specifically, in the current case, this effect is expected to be very small because of the Stokes number range under consideration (10^{-4} to 10^{-5}) [126]. The vertical displacement of a droplet (z) is then

$$z = \int_0^{\tau_l} v_t dt = \int_0^{\tau_l} ar^2 dt. \quad (\text{A.1})$$

By neglecting the initial radius, the size of a cloud droplet at any time t can be expressed as $\overline{r^2} = 2\xi\langle s \rangle_t t$ where $\langle s \rangle_t$ is the average supersaturation that a single droplet experiences along its trajectory in the chamber. This expression with Eqs. A.1 and 4.12 in the main text, results in a relationship between displacement of a single droplet and its lifetime [335]:

$$z = \int_0^{\tau_l} 2a\xi\langle s \rangle_t t dt = a\xi\langle s \rangle_t \tau_l^2.$$

Considering the average for all cloud droplets, which are allowed to fall over distance H while experiencing mean supersaturation $\bar{s}$, we obtain

$$\langle z \rangle = H \approx a\xi\bar{s}\bar{\tau}_l^2.$$

Finally, we solve for the mean droplet lifetime:

$$\tau_l = \left(\frac{H}{a\xi\bar{s}} \right)^{1/2} = \left(\frac{H}{a\xi s_o} \right)^{1/2} \left(\frac{\tau_t}{\tau_s} \right)^{1/2}, \quad (\text{A.2})$$

where, H is the average distance that a droplet falls and can be approximated as half of the chamber height. In the above expression and subsequent derivation, the average droplet lifetime ($\bar{\tau}_l$) is represented as τ_l .

Appendix B

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