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INVESTIGATION OF MICROPHYSICAL PROPERTIES OF LABORATORY
AND ATMOSPHERIC CLOUDS USING DIGITAL IN-LINE HOLOGRAPHY

By

Neel Desai

A DISSERTATION

Submitted in partial fulfillment of the requirements for the degree of

DOCTOR OF PHILOSOPHY

In Atmospheric Sciences

MICHIGAN TECHNOLOGICAL UNIVERSITY

2018

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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 parents

My mother, Sandhya Desai

My father, Uday Desai

For encouraging me to follow my dreams.

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Acknowledgments

*For once you have tasted flight
you will forever walk the earth
with your eyes turned skywards,
for there you have been
and there you will long to return.*

- Leonardo da Vinci

I would first like to thank my parents, without whom I would not be here, defending my thesis as a PhD student. They strove to provide me with everything they did not have as children and allowed me the freedom to pursue my dreams.

I would like to thank my advisor Dr. Raymond Shaw for agreeing to guide a novice in the field of atmospheric sciences, like me, so that I can fulfill my dream of knowing more about the skies above us. This dissertation would not have been possible without his continuous guidance, support and patience as I worked on my research and I will be forever grateful to him for that.

I am also grateful to my committee members Dr. Will Cantrell, Dr. Claudio Mazoleni and Dr. Jacob Fugal. Each of them guided me in different aspects of my research, the culmination of which is presented here. Without their support, this

thesis would not have been possible.

I have had the pleasure to collaborate with researchers outside Michigan Tech such as Dr. Dennis Niedermeier, Dr. Jens Voigtländer, and Dr. Frank Stratmann at the Leibniz Institute for Tropospheric Research (TROPOS) in Leipzig, Germany. I'm also grateful to Dr. Jan Henneberger and the entire HoloSuite group from ETH Zurich and Mainz, Germany for their constant discussions, guidance and improvements to the HoloSuite package. These collaborations have enriched me with experience and knowledge during my graduate studies.

Over the course of my time at Michigan Tech, I have had the pleasure to be in the company of an amazing group of researchers and colleagues from the cloud physics group. Dr. Kelken Chang, Dr. Jiang Lu, Dr. Colin Gurganus, Mr. Dave Ciochetto, Mr. Kamal Kant Chandrakar, Mr. Subin Thomas, Mr. Gregory Kinney and Mr. Eduardo Rodriguez. A special thanks to Ms. Susanne Glienke for repeated brainstorming through the mysteries of holography.

I also thank Dr. Ravindra Pandey, Mr. Wil Slough and everyone from the Physics department for their support during my research. Also thanks to the MTU Atmospheric Science Program, MTU Graduate School, The Elizabeth and Richard Henes Center for Quantum Phenomena (CQP) and The Earth, Planetary, and Space Sciences Institute (EPSSI) for allowing me to participate in conferences and present my research to the community.

Last but not least, I would like to sincerely thank the people closest to my life away from work. Shaizeen, for her constant care, affection and pushing me to be better every second of my life. My friends Ameya, Nupur, Kishan and Ninad for providing me with amazing company, support and laughter throughout my days in Houghton.

Abstract

In this study, we attempt to perform in-cloud measurements, both in the laboratory using the Michigan Tech II-chamber and in the atmosphere via the CSET field campaign. Atmospheric turbulence is believed to play a critical role in the growth, development and dissipation of clouds and it is important to study its effect in order to better understand and predict cloud properties such as albedo and lifetime. We use digital in-line holography to measure the effect of turbulence on cloud microphysical properties such as variations in droplet number concentration and droplet or ice particle size.

In the first half, we study warm clouds and investigate how cloud droplets grow between the regimes dominated by diffusional growth and growth by collision-coalescence. We propose that microphysical variability in droplet number concentration will lead to local increase in supersaturation fluctuations. Prior stochastic condensation theory assumed constant cloud properties such as phase relaxation time. We create a steady state warm turbulent mixing cloud in the laboratory and use digital in-line holography to obtain local instantaneous droplet number concentrations. We show using these measurements that phase relaxation time distributions are considerably broad leading to additional increase in droplet spectral width. We then

compare these results with in-situ measurements of marine stratocumulus clouds during the CSET campaign. We find certain signatures which we define as microphysical fingerprints of stochastic condensation occurring in these clouds.

In the second half, we focus our attention on mixed phase clouds which are ubiquitous in the atmosphere but very difficult to measure. By creating steady state mixed phase clouds in the laboratory, using digital in-line holography, we were able to measure cloud droplet and ice particle properties for extended period of time. We show that cloud glaciation is a function of the steady state supersaturation and can be altered by varying the ratio between ice nuclei and cloud condensation nuclei. By varying this ratio, we were able to generate steady state mixed phase clouds with varying ice fraction and study their properties.

Chapter 1

Introduction

1.1 Overview

Carl Sagan described the earth as seen from space as a “pale blue dot” [1]. While the water provides the blue, the paleness primarily is due to cloud cover over the Earth. Satellite images of the Earth show that our planet is visually dominated by atmospheric clouds. Clouds represent the highest mass concentration of any aerosol [2] and therefore are of great importance for heterogeneous chemistry and aerosol processing and removal. They play an important role in the Earths radiation balance by redistributing incoming solar radiation by way of transmission, absorption, reflection

and scattering. [3]. Clouds also play a major role in maintaining the Earth’s hydrological cycle, essential for nearly all life on Earth [4]. With climate change in effect, precipitation intensity and frequency likely will change globally as well as regionally. Accurately predicting these changes will be crucial to anticipating vulnerabilities, aiding adaptation, and understanding consequences of possible mitigation efforts.

One of the primary issues plaguing accurate prediction and modeling of cloud lifetimes and precipitation is the cloud droplet “growth gap”. Once cloud droplets are activated from cloud condensation nuclei (CCN), they grow rapidly to about $20\ \mu m$ radius through diffusion of water vapor [5]. But further growth by collision-coalescence, only becomes effective after the droplet radius increases to $40\ \mu m$ [6]. Thus, it is not clear how the droplets efficiently grow from $20\ \mu m$ to $40\ \mu m$ in radius since neither of the above mentioned mechanisms are effective during this growth period. Turbulence is considered to have a major role in cloud formation, structure, albedo and lifetime through entrainment and mixing [7]. Recent studies also indicate that turbulence affects cloud droplet growth in the previously mentioned growth gap.

In order to measure and quantify the possible effects of turbulence, we need measurements inside clouds of droplets with size range in this ‘growth gap’. However, this is a challenging problem due to the small size (micrometers) of the droplets and the relative inaccessibility of atmospheric clouds for long measurements. In-situ aircraft measurements have a drawback since typical aircraft speeds are $\approx 50 - 100\text{ m s}^{-1}$,

making it very difficult to obtain data of the turbulence acting on the droplets at the centimeter scale. Typical cloud measuring instruments such as the Cloud Droplet Probe and 2D precipitation probe measure single droplets at a time and require long time averaging over hundreds of meters to measure statistically significant measurements. Hence, they cannot provide any data regarding the local centimeter scale environment that the droplet exists in. Such measurements also suffer from poorly constrained boundary conditions due to lack of detailed measurements of the surrounding environment. This also leads to multiple processes such as adiabatic ascent and entrainment, affecting the droplets at the same time and an inability to separate the effects of each process. Another aspect plaguing in-situ atmospheric measurements of cloud droplets, is the transient nature of clouds. The timescales at which some of the previously mentioned processes occur and affect cloud properties are quite short and airborne instruments, due to their flight speed, are unable to measure for a long enough time period in order to obtain statistically significant measurements.

1.2 Methods

1.2.1 Π -chamber

Regarding the issue of clouds being not easily accessible for measurements and their environment being complex, the Michigan Tech Π -chamber allows us to create a turbulent cloud under controlled laboratory conditions [8]. A schematic of the chamber is shown in Fig. 1.1; this unique laboratory facility has internal dimensions $1 \times 2 \times 2 \text{ m}^3$, which can be further reduced to 3.14 m^3 by using a 2-m diameter cylindrical insert. The top, bottom and side walls are temperature controlled with a range of 55 to -55 °C to simulate different environmental temperature conditions. The pressure inside the chamber can also be varied between 60 and 1000 hPa simulating cloud properties at different altitudes. The Π -chamber allows us to decouple the multiple processes that affect airborne microphysical measurements, thereby making their measurement and interpretation a little easier. The clouds generated in the Π -chamber have the added advantage of repeatability and steady-state conditions allowing data collection of long periods of time to obtain statistically significant datasets. To characterize the properties of the cloud generated, the chamber is equipped with a number of instruments such as: a Phase Doppler Interferometer (PDI), which provides droplet properties such as size and velocity, a sonic anemometer which provides turbulent

flow velocities, and a LI-COR[®] H_2O analyzer providing humidity measurements.

Figure 1.1: A schematic of the Michigan Tech II-chamber with the cylinder inside.

1.2.2 Holography

To measure local scale droplet microphysics, a new in-line holographic system named ‘HoloPi’ is assembled in order to measure droplet properties of clouds generated inside the II-chamber. This work follows on the work of [9, 10], who used digital in-line holography to visualize cloud droplets through diffraction of light. An in-line hologram is an interference pattern resulting from the superposition of an incident plane wave and the diffraction pattern obtained due to the interaction of the coherent beam with suspended particles. When reconstructed, this gives us a volume measurement, allowing the measurement of multiple droplets at the same time. Thereby we can

obtain cloud properties such as size distributions and number concentrations without the need for large-scale spatial averaging. However, this advantage comes along with intensive computation requirements. For this purpose, a data processing and analyzing software package named HOLOSUITE was developed by Dr. Jacob Fugal in collaboration with colleagues at Michigan Technological University, Johannes Gutenberg University of Mainz, National Center for Atmospheric Research (NCAR) and ETH Zürich. This package is used for the purposes of processing and analysis of the results in this dissertation.

1.3 Organization of dissertation

The objectives of the research in this dissertation are

1. To measure and understand the effect of turbulence in droplet growth by condensation in the cloud droplet growth gap in the laboratory.
2. To compare the laboratory measurements with in-situ measurements.
3. To extend the measurements to temperatures below freezing and explore the growth of particles in such conditions through the formation and growth of ice in mixed-phase clouds.

In Chp. 2 we use HoloPi to make laboratory measurements in the Π -chamber for a steady state turbulent mixing cloud. The measurements are compared with a stochastic condensation model in order to evaluate stochastic condensation as a possible mechanism for cloud droplet growth. The above mentioned facility and instrumentation allows us to measure the effects of turbulence on cloud droplet growth by condensation in the laboratory. However, for a better understanding of these effects on atmospheric clouds, a comparison with in-situ measurements is essential.

In Chp. 3 we compare our results from the laboratory experiment with airborne measurements made using an in-line holographic instrument named HOLODEC (Holographic Detector for Clouds). The in-situ aircraft measurements are made over the Pacific Ocean between California and Hawaii. Due to the similarity between the instruments, a comparison between the two results gives us new insight into similar processes which may broaden cloud droplet sizes in atmospheric clouds. However, this comparison is only valid for warm clouds and only 30% of all clouds over Earth are in warm or liquid phase. Which means that 70% of all clouds are either ice or mixed phase clouds and they must contribute significantly to Earth's total radiation budget.

In Chp. 4, we extend the measurement procedure used in Chp. 1 to measure mixed phase cloud properties. Mixed phase clouds consist of a mixture of water droplets and ice particles and are notoriously difficult to measure in the atmosphere due to

their unstable nature and dangerous icing conditions for aircraft attempting to study them. By creating a steady state mixed phase cloud in the Π -chamber, we were able to collect droplet data for long periods of time and understand the properties of these clouds.

Chapter 2

Influence of Microphysical Variability on Stochastic Condensation in a Turbulent Laboratory Cloud

This chapter details the holographic measurements for a steady state warm mixing cloud generated in the Π -chamber. This work was published in full form in the Journal of Atmospheric Sciences. ¹ ²

¹Desai N, Chandrakar KK, Chang K, Cantrell W, Shaw RA. Influence of Microphysical Variability on Stochastic Condensation in a Turbulent Laboratory Cloud. Journal of the Atmospheric Sciences. 2018 Jan;75(1):189-201.

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2.1 Abstract

Diffusional growth of droplets by stochastic condensation and a resulting broadening of the size distribution has been considered as a mechanism for bridging the cloud droplet growth gap between condensation and collision-coalescence. Recent studies have shown that supersaturation fluctuations can lead to a broadening of the droplet size distribution at the condensational stage of droplet growth. However, most studies using stochastic models assume the phase relaxation time of a cloud parcel to be constant. In this paper we ask how do variability in droplet number concentration and radius influence the phase relaxation time? And what effect does it have on the droplet size distributions? To answer these questions, we created steady state cloud conditions in the laboratory and used digital inline holography to directly observe the variations in local number concentration and droplet size distribution and, thereby, the integral radius. We also extend the stochastic equations to account for fluctuations in integral radius and obtain new terms that are compared with the laboratory observations. We find that the variability in integral radius is primarily driven by variations in the droplet number concentration and not the droplet radius. This variability does not contribute significantly to the mean droplet growth rate, but contributes significantly to the rate of increase of the size distribution width.

2.2 Introduction

The cloud physics community has grappled with the ‘growth gap’ problem in warm rain formation for a few decades now. Turbulence has been considered as one of the mechanisms to overcome this bottleneck between cloud droplet growth by diffusion and growth by collision-coalescence [7, 11]. Early studies in the 1960s had proposed already that stochastic condensation itself may be able to bridge this growth gap by means of turbulence induced fluctuations in supersaturation and diffusive mixing (e.g., see the recent summary by Mazin [12]). This can produce a broad size distribution and depart from conventionally imagined narrow droplet size distributions due to uniform condensational growth. However, [13] argued that such a process cannot explain broadening of the size distribution since the required supersaturation fluctuations within clouds are closely correlated with updrafts. Hence, a droplet experiencing higher supersaturations will be in a stronger updraft, thereby reducing the amount of time it will get to grow before reaching a level with steady-state supersaturation again. [14] proposed that the broadening of size distributions may then be primarily due to turbulence induced by entrainment and mixing instead of vertical velocity. [15] argued that cloud turbulence due to vertical velocity variations may still induce broadening of droplet size distributions by considering variations in the integral radius (and thus the phase relaxation time of droplets, both of which we shall consider in this paper). Further refinements to the theory of stochastic condensation have been made,

and it has been suggested that these approaches are attractive for implementation within a computational framework, for example, for large eddy simulation [16, 17].

Other approaches linked to stochastic condensation, have dealt in particular with the source of supersaturation variability. [18] put forward the argument that the local supersaturation around a drop and not the average supersaturation of a cloud parcel should be taken into account while calculating droplet growth rates. This local supersaturation may be significantly different for every drop from the average value due to local variability in the droplet number concentration and vertical air velocity. Since then, some attention has been given to the possible influence of spatial and temporal variability in droplet concentration due to finite droplet inertia [e.g., 19, 20, 21]. Furthermore, [22] and [23] have shown that broadening due to supersaturation variability from isobaric mixing, or even mixing between vertically cycling parcels [24] can be important. Whatever the source, careful in-situ measurements by [15], [25] and [26] have found broadening of the droplet size distribution even in cumulus cloud cores.

Recent studies by [27] showed that turbulence causes an increase in the variance of the droplet size distribution with time, producing a broad droplet size distribution. This broadening may provide enough large droplets to cross the growth gap and start the collision-coalescence process. They used the stochastic approach suggested by [28] followed by DNS and LES simulations to show that the variance grows as $t^{1/2}$,

but did not include any effects of changing aerosol or droplet number concentration. [29] performed a study using three dimensional DNS and compared it with a similar stochastic Lagrangian model. They found different regimes of broadening depending on how the droplet growth timescale and supersaturation field response timescale compared with the turbulent mixing timescale. However, most of these modeling approaches assume a constant phase relaxation time. Using laboratory measurements, [30] produced a steady state, warm turbulent cloud and showed that the width of the droplet size distribution increases with a decrease in the aerosol concentration due to larger variability in supersaturation fluctuations. However, to characterize cloud microphysics, they used a Phase Doppler Interferometer, which requires averaging times which are much greater than the large eddy time scale. Thus, they too obtained a droplet size distribution that is assumed to be spatially constant within the entire chamber leading to a spatially constant phase relaxation time (τ_c).

Variation in τ_c , with the resulting fluctuations in the local supersaturation, can occur for a large number of reasons. [31] found that inhomogeneous mixing events due to entrainment can greatly reduce the droplet number concentration in some regions. [19] suggested that turbulent mixing can cause clustering of particles away from regions of high vorticity causing broadening of the droplet size distributions due to supersaturation fluctuations. [32] combined three dimensional cloud model with a Lagrangian microphysical parcel model which mapped droplet trajectories that ended up at the same point. They found large size widths along with small droplets high in the clouds,

due to different entrainment and mixing zones the trajectories encountered. [33] further investigated this mechanism for size distribution broadening by imposing droplet mixing and different growth histories, resulting in significant increase in broadening compared to adiabatic parcel models.

In this paper, we ask the questions, how reasonable is the approximation of constant τ_c ? How does the size distribution vary spatially? Can spatial variability in droplet number and size lead to a significant change in τ_c , and how does this affect the mean and width of the droplet size distribution? We will not directly address the causes of variations in number concentrations, but rather study their influence on the droplet size distribution through condensation growth. We will therefore not consider possible effects of variations in number concentration on droplet growth by collision-coalescence. We attempt to answer these questions by creating a turbulent cloud in a laboratory cloud chamber called the Π chamber, similar to [30]. (See [8] for details about the chamber.) Such a controlled experiment is crucial for evaluating the theory because long-time averages are required, thereby necessitating statistically stationary conditions which are difficult to attain in the atmosphere. The theory validated under such idealized conditions can then be applied to more complex atmospheric conditions. Furthermore, in the laboratory we can adjust the parameter space to simulate desired cloud conditions optimal for the analysis. For example, since our cloud exists in an enclosed space, we can be assured that observed broadening of the size distribution will not be due to entrainment effects. The estimated cloud droplet collision times

for droplet sizes obtained during the experiment are of the order of 1 μm , while the droplet residence times are of the order 1-10 minutes. Hence we are also confident that the effects on droplet size are not due to collision-coalescence but primarily due to stochastic condensation. LES calculations have also confirmed this result [30]. Finally, a crucial aspect of the experimental approach is the ability to measure the droplet size distribution from a spatially-localized volume without temporal averaging, using digital in-line holography. This approach allows the questions about microphysical variability, as expressed through the integral radius and the phase relaxation time, to be investigated.

The paper proceeds as follows: first, in section 2, we present the theoretical approach based on stochastic differential equations (SDEs) for supersaturation and droplet growth, with non-constant phase relaxation time. In section 3, we describe the laboratory experiments, emphasizing the holographic system and how it enables measurement of the quantities that come out of the SDEs. In section 4, we present the results of the experiments, and finally in section 5 we discuss the findings and implications of those results.

2.3 Stochastic Condensation for variable τ_c

Motivated from the stochastic differential equation approach of [27] and [30], we extend the theoretical framework for cloud droplet growth in the II chamber to allow for variable phase relaxation time τ_c . Growth of a single cloud droplet due to condensation in a supersaturated environment can be expressed as [34]:

$$\frac{dr^2}{dt} = 2\xi s, \quad (2.1)$$

where r is the droplet radius, ξ is a thermodynamic growth parameter, and s is the water vapor supersaturation. In our experiment, the system is in a steady state, but individual droplets grow and shrink depending on the supersaturation field they encounter, according to Eq. 2.1. For a collection of cloud droplets exposed to a fluctuating supersaturation field $s = \bar{s} + s'$, we can express the mean droplet size as:

$$\frac{d\bar{r}^2}{dt} = 2\xi\bar{s}. \quad (2.2)$$

The variance of r^2 can then be found as

$$2r^{2'} \frac{dr^{2'}}{dt} = 4\xi s' r^{2'} \quad (2.3)$$

Or

$$\frac{d\sigma_{r^2}^2}{dt} = 4\xi \overline{s'r'^2}. \quad (2.4)$$

Here, over-bars and primes indicate mean and fluctuating quantities. Eq. 2.4 suggests that the growth of the width of the droplet size distribution depends upon correlations between fluctuations in the supersaturation field and droplet size. Finally, note that Eq. 2.2 refers to the average growth rate experienced by individual droplets in the dynamic steady state: the cloud itself has a constant size distribution consisting of droplets that are continuously activated, grow by condensation, and are removed by sedimentation.

To obtain values for $\overline{dr^2}/dt$ and $d\sigma_{r^2}^2/dt$, we will need to convert the right side of Eqs. 2.2 and 2.4 into variables which can be experimentally measured. Let us first obtain an equation for $\bar{s}$ for our experiment, where we have imposed a temperature gradient between the top and bottom boundaries resulting in isobaric mixing. Details of the experiment will follow in Section 3. The governing equation for the turbulent supersaturation field in the cloud chamber, without any cloud droplets, can be expressed as [35, p.484-487]:

$$\begin{aligned} ds(t) &= s(t+dt) - s(t) \\ &= \left[\frac{s_0 - s}{\tau_t} \right] dt + \left(\frac{2\sigma_{s_0}^2 dt}{\tau_t} \right)^{1/2} \eta(t). \end{aligned} \quad (2.5)$$

This is the Langevin equation for supersaturation as the stochastic variable where first term on the right side of Eq. 2.5 expresses the tendency of turbulent mixing to relax the system back to the equilibrium supersaturation s_0 that exists without cloud droplet formation (i.e., when no aerosol particles are present in the chamber). The last term is the differential of the Wiener process, with $\eta(t)$ being the Gaussian white noise term (with mean of 0 and standard deviation of 1) representing turbulent fluctuations, and $\sigma_{s_0}^2$ being the variance of the supersaturation (in the absence of cloud formation). The time scale τ_t is the Lagrangian correlation time, effectively equivalent to the turbulence large eddy turnover time [28, 35]. This equation assumes homogeneous turbulence and supersaturation fields. Cloud droplet growth is included in Eq. 2.5 as a term causing exponential relaxation of the supersaturation field with time scale τ_c :

$$\begin{aligned} ds(t) &= s(t + dt) - s(t) \\ &= \left[\frac{s_0 - s}{\tau_t} - \frac{s}{\tau_c} \right] dt + \left(\frac{2\sigma_{s_0}^2 dt}{\tau_t} \right)^{1/2} \eta(t). \end{aligned} \tag{2.6}$$

The phase relaxation time is defined as $\tau_c = (4\pi D_v I)^{-1}$, where D_v is a constant that accounts for competing roles of diffusion and latent heat release [15, 34] and $I = \int r n_d(r) dr$ is the integral radius [15]. The integral radius is the product of the droplet number concentration n_d and the mean droplet radius.

At this juncture, [30] assumed τ_c to be constant and obtained simplified relationships

for $\overline{r^2}$, and $\sigma_{r^2}^2$. In this paper, we aim to check whether the assumption of constant τ_c is reasonable and whether the contribution from the terms obtained if τ_c is not assumed to be constant are significant. Equations for the mean and fluctuations of supersaturation can be obtained from Eq. 2.6 as:

$$d\bar{s}(t) = \left[\frac{s_0 - \bar{s}}{\tau_t} - 4\pi D_v (\bar{I}\bar{s} + \overline{I's'}) \right] dt \quad (2.7)$$

and

$$ds'(t) = \left[-\frac{s'}{\tau_t} - 4\pi D_v (\bar{I}s' + \bar{s}I' + I's' - \overline{I's'}) \right] dt + \left(\frac{2\sigma_{s_0}^2 dt}{\tau_m} \right)^{1/2} \eta(t) \quad (2.8)$$

Here, $\bar{I}$ is the integral radius averaged over different cloud parcels and I' is the fluctuation about the mean value.

$$ds'(t) = \left[-\frac{s'}{\tau_t} - 4\pi D_v (\bar{I}s' + \bar{s}I') \right] dt + \left(\frac{2\sigma_{s_0}^2 dt}{\tau_t} \right)^{1/2} \eta(t). \quad (2.9)$$

In Eq. 2.9 we have neglected the term $(I's' - \overline{I's'})$, since it is a second order fluctuation and is therefore expected to be small. We have chosen to follow the approach of [15] and allow the phase relaxation time to vary solely through fluctuations in I . Separate fluctuations of n_d and r could also be considered, but atmospheric observations

often show that n_d dominates the variations in I , for example through the process of inhomogeneous mixing [10].

For steady state conditions, which are achievable in the laboratory, we can say that the time derivative of $\bar{s}$ will be zero. Thus we get,

$$\bar{s} = s_0 \left[1 + \frac{\tau_t}{\tau_{c,avg}} \left(1 - \frac{\overline{I'^2}}{\bar{I}^2} \right) \right]^{-1} \quad (2.10)$$

In this equation we have defined $\tau_{c,avg}$ as the mean value obtained with $\bar{I}$. Similarly, with Eq. 2.4 in mind, we can also obtain an expression for $\overline{s'r^{2'}}$ as:

$$d(s'r^{2'}) = s'(t+dt)r^{2'}(t+dt) - s'(t)r^{2'}(t) \quad (2.11)$$

Substituting the expression for $s'(t+dt)$ and $r^{2'}(t+dt)$ from Eq. 2.9 and Eq. 2.3 in the above equation results in:

$$\begin{aligned} d(s'r^{2'}) = & \left(s'(t) + \left[-\frac{s'}{\tau_t} - 4\pi D_v(\bar{I}s' + \bar{s}I') \right] dt + \left(\frac{2\sigma_{s_0}^2 dt}{\tau_m} \right)^{1/2} \eta(t) \right) \\ & \times \left(r^{2'}(t) + 2\xi s'(t)dt \right) - s'(t)r^{2'}(t) \quad (2.12) \end{aligned}$$

Taking an average eliminates all the cross-correlation terms with $\eta(t)$ since $s'(t)$ and $r^{2'}(t)$ are independent from the Gaussian noise increment $\eta(t)$. Now, if we take a time derivative, the final average differential equation will be (or using Ito calculus [36]):

$$\frac{d\overline{s'r^{2'}}}{dt} = 2\xi\overline{s'^2} - \frac{\overline{s'r^{2'}}}{\tau_t} - 4\pi D_v(\overline{\bar{I}s'r^{2'}} + \overline{\bar{s}I'r^{2'}}) \quad (2.13)$$

$$\overline{s'r^{2'}} = 2\xi\overline{s'^2}\tau_s - 4\pi D_v\overline{\bar{s}I'r^{2'}}\tau_s, \quad (2.14)$$

where $\tau_s = \tau_c\tau_t/(\tau_c + \tau_t)$ is a system time scale [30]. The stochastic variable $r^2(t)$ is a function of $s(t - dt)$ which is independent of the current increment $ds(t)$ and corresponding white noise term. Hence, all correlations of r' with the noise term can be neglected [37, p.69]. However, we also have two new terms $\overline{s'^2}$ and $\overline{I'r^{2'}}$, which can be evaluated from Ito calculus as:

$$ds'^2(t) = 2s'(t)ds'(t) + (ds'(t))^2 \quad (2.15)$$

In the above expression, substituting the value of $ds'(t)$ from Eq.2.9:

$$\frac{d\overline{s'^2}}{dt} = -2 \left[-\frac{\overline{s'^2}}{\tau_t} + 4\pi D'(\overline{I s'^2} + \overline{s I' s'}) \right] + \left(\frac{2\sigma_{s_0}^2}{\tau_t} \right) \quad (2.16)$$

to give,

$$\overline{s'^2} = \frac{\sigma_{s_0}^2 \tau_s}{\tau_t} - \overline{s I' s'} (4\pi D_v \tau_s) \quad (2.17)$$

Similarly, assuming the fluctuations in integral radius are also because of the turbulence and can be modeled using Gaussian random process, the governing equation for the integral radius fluctuation can also be modeled as:

$$dI'(t) = -\frac{I'}{\tau_t} dt + (1 - C_{Is}^2)^{1/2} \left(\frac{2\sigma_{I_0}^2 dt}{\tau_t} \right)^{1/2} \zeta(t) + C_{Is} \left(\frac{2\sigma_{I_0}^2 dt}{\tau_t} \right)^{1/2} \eta(t). \quad (2.18)$$

Here σ_{I_0} represents the magnitude of the RMS integral radius fluctuation, $C_{Is} = \frac{\overline{I' s'}}{\sigma_{s_0} \sigma_{I_0}}$ is the supersaturation-integral radius correlation coefficient, and $\zeta(t)$ is a independent Gaussian random noise similar to $\eta(t)$. With the use of the equation above and 2.3, an averaged equation for $\overline{I' r^{2'}}$ is given by Ito calculus as:

$$\frac{d\overline{I' r^{2'}}}{dt} = -\frac{\overline{I' r^{2'}}}{\tau_t} + 2\xi \overline{I' s'} \quad (2.19)$$

to obtain,

$$\overline{I'r^{2'}} = 2\xi \overline{I's'} \tau_t. \quad (2.20)$$

Both Eq. 2.17 and 2.20 are functions of $\overline{I's'}$, which itself can be evaluated similar to Eq. 2.13, by using Eq. 2.9 and Eq. 2.18:

$$\frac{d\overline{I's'}}{dt} = -2\frac{\overline{I's'}}{\tau_t} - 4\pi D_v(\overline{II's'} + \overline{I'^2\bar{s}}) + C_{Is} \left(\frac{2\sigma_{I_0}^2 dt}{\tau_t} \right)^{1/2} \left(\frac{2\sigma_{s_0}^2 dt}{\tau_t} \right)^{1/2} \quad (2.21)$$

Ensemble averaging results in all terms associated with a Gaussian random noise terms and second order fluctuations to be zero. Assuming steady state conditions for $\overline{I's'}$, we get:

$$\overline{I's'} = -\bar{s} \left(\frac{\overline{I'^2}}{\bar{I}} \right). \quad (2.22)$$

Thus eq. 2.17 becomes

$$\overline{s'^2} = \frac{\sigma_{s_0}^2 \tau_s}{\tau_t} + \frac{\bar{s}^2 \tau_s}{\tau_{c,avg}} \left(\frac{\overline{I'^2}}{\bar{I}} \right) \quad (2.23)$$

and

$$\overline{I'r^{2'}} = -2\xi \bar{s} \left(\frac{\overline{I'^2}}{\bar{I}} \right) \tau_t. \quad (2.24)$$

Combining these last equations with Eqs. 2.4 and 2.14 gives us the expression for the

growth of a droplet size distribution width in a turbulent environment:

$$\frac{d\sigma_{r^2}^2}{dt} = 8\xi^2 \bar{s}^2 \tau_s \left[\frac{\sigma_{s_0}^2}{\bar{s}^2} \frac{\tau_s}{\tau_t} + \frac{\overline{I'^2}}{\bar{I}^2} \frac{\tau_s + \tau_t}{\tau_{c,avg}} \right]. \quad (2.25)$$

Similarly, the expression for the mean size is obtained using Eqs. 2.2, 3.1):

$$\frac{d\bar{r}^2}{dt} = 2\xi s_0 \left[1 + \frac{\tau_t}{\tau_{c,avg}} \left(1 - \frac{\overline{I'^2}}{\bar{I}^2} \right) \right]^{-1} \quad (2.26)$$

We have now obtained expressions for the mean and fluctuation in droplet size in terms of measurable microphysical quantities $\bar{I}$, and $\overline{I'^2}$. We will now discuss the approach for obtaining these measurements, as well as needed turbulence and thermodynamics quantities, in a laboratory setting.

2.4 Experimental approach

2.4.1 Chamber setup and instruments

The microphysical variability of a steady-state cloud in a turbulent environment is studied by creating a turbulent mixing cloud in the Michigan Tech II chamber. A

buoyancy driven convective flow environment (Rayleigh-Bénard convection) is created by applying an unstable temperature difference between the top and bottom surfaces of the chamber. To achieve supersaturated conditions for cloud formation and growth, the top and bottom boundaries are kept saturated with liquid water. The turbulent Rayleigh-Bénard convection mixes air parcels from the top and bottom boundaries; this isobaric mixing of air parcels containing different water vapor concentrations at distinct temperatures creates a supersaturated environment for aerosol activation and subsequently cloud droplet growth. Stated in a slightly different way, in the context of Eq. 2.5: the top and bottom boundaries are held at fixed temperatures and equilibrium vapor pressures, and the resulting heat and vapor fluxes tend to force a background supersaturation s_0 . Turbulent fluctuations or droplet growth can drive the system away from this value, but it always tends to relax to s_0 (cf., first term on right side of Eq. 2.5). The turbulent mixing cloud formation process in the Michigan Tech II chamber is described by [8]. A balance between the source (droplet activation by continuous aerosol injection) and sink (settling of droplets) gives steady-state cloud microphysical conditions after a starting transition period [30]. The cloud microphysical and thermodynamic conditions during this steady-state period are used for the current study.

In this set of experiments, a 19-K temperature difference between the top ($T_t = 7\text{ }^\circ\text{C}$) and bottom ($T_b = 26\text{ }^\circ\text{C}$) boundaries is used to drive the turbulent moist convection. Both these boundaries are saturated with water and the side walls are maintained

at the mean temperature between these boundaries. The side walls are also covered by 3.2-mm-thick polycarbonate sheets to minimize heat flux (relative to the top and bottom boundaries). Once the turbulent and thermodynamic properties, such as temperature gradient between the boundaries, water vapor concentration and velocity fields reach a steady state, salt aerosol injection is started at a constant rate (5000 cm^{-3} at 2 lpm of inlet air flow). A fraction of injected aerosol is activated once they experience supersaturation more than the critical value and then they grow in this fluctuating supersaturated environment. As the droplets grow, gravitational sedimentation becomes a significant loss mechanism, and this limits the lifetime of individual cloud droplets inside the chamber. The Chamber is allowed to run in this state for a couple of hours until the rate of activation of new droplets matches the rate of droplet sedimentation, giving us a steady state droplet number concentration and droplet size distribution as measured by the Phase Doppler Interferometer. As noted in the previous section, the fact that the cloud is in a dynamic steady state implies that even though collective properties are constant in time, individual droplets experience growth by condensation at an average rate given by Eq. 2.2.

Thermodynamic properties required for this study are air temperature and water vapor concentration. Resistance thermometers (RTDs, Minco) and a LI-COR hygrometer (7500A) are used to measure these properties. To characterize the turbulent flow properties, a sonic anemometer is employed, which measures the flow velocity in all

three directions at a high frequency (20 Hz). Aerosol particles are generated by atomizing an NaCl-water solution using an atomizer (3076 TSI) and subsequently passing it through a diffusion dryer. During this experiment, the measured Turbulent Kinetic Energy (TKE) was approximately $0.004 \text{ m}^2\text{s}^{-2}$, dissipation rate $(\epsilon) = 10^{-3}\text{m}^2\text{s}^{-3}$, Reynolds Number (Re) = 80 and $\text{Ra}(\text{dry}) = 2 \times 10^9$. Most of the values are comparable to atmospheric clouds, whereas TKE and Re are smaller due to the chamber dimensions compared to typical large eddy lengths in the atmosphere [8]. Details of the microphysical measurement approach are discussed in the subsequent section.

Figure 2.1: (Top) Optical assembly above the II chamber. (Bottom Left) A side-view schematic of the digital in-line holographic setup, not to scale. (Bottom Right) The imaging assembly below the II chamber. A steady state warm mixing cloud exists throughout the volume of the chamber. Cloud parcels passing the measurement volume are imaged and analyzed for the study.

2.4.2 Microphysical measurements and holographic setup

The key measurements in this study are of the cloud droplet size distribution and number concentration, in order to assess microphysical variability and its contribution to stochastic condensation. Specifically, from Eqs. 2.25 and 2.26 it can be seen that $\bar{I}$ and $\overline{I'^2}$ are required in order to assess the rate of growth of the mean and variance of the droplet size distribution within a turbulent environment. Typical measurement approaches, which involve single droplet detection and integration over time, will not provide the instantaneous variability of the integral radius, I' . We have therefore developed a holographic system specifically designed for making instantaneous, spatially-localized measurements of the droplet concentration and size distribution [10].

The holographic setup consists of a collimated CryLaS 532 *nm* laser which is passed through the top access port of the Π chamber (Fig. 2.1). This beam passes through the chamber and is received by a K2 Distamax Lens and a Photron Fastcam SA2 camera looking up through the bottom access port of the chamber. The lens provides a magnification of $2.85\times$ at the focal plane, which is flush with the bottom of the inner chamber. The camera has a 2048×2048 pixel detector with a single pixel pitch of 10 μm . The equivalent pixel size thus obtained is 3.5 μm , however, at least two pixels are needed to confidently distinguish a particle from the background image. Hence, we

have rejected any possible particle smaller than $7\text{ }\mu\text{m}$. A burst of seven consecutive images is taken every 30 seconds for 2.5 hours at 500 fps. The time interval of 30 s was chosen since it was sufficiently longer than the turbulence decorrelation timescale ($\approx 10\tau_t$) measured by the sonic anemometer. The seven images at every instant are used for averaging and background division used for improvement of signal-to-noise ratio. Bench tests showed that the resolution of the holograms does not depreciate much through 20 cm beyond the focal plane. This gives us a total measurement volume of approximately 10 cm^3 and an average number concentration of 80 droplets per cm^3 . This measurement volume is of the order of the large eddy length scale, and therefore is consistent with the ability to resolve the turbulent fluctuations in microphysical properties. The digital holograms are then reconstructed numerically at every $100\text{ }\mu\text{m}$ depth into the focal plane using the convolution method in Fourier space [9]. This procedure allows us to find droplets and their diameters using a light intensity threshold [38]. [38] and [39] also showed that the error in diameter calculation using digital holography is approximately the square root of the equivalent pixel size. In our case this is approximately $1.87\text{ }\mu\text{m}$.

The uncertainty in the mean radius (r) for each hologram can be obtained using a normal distribution as $\delta r/\sqrt{N} = 0.07\mu\text{m}$. where N is the average number of droplets in one hologram. Thus $r = 8.1 \pm 0.07\text{ }\mu\text{m}$ with the uncertainty being very small compared to the mean and can be neglected. The uncertainty in the droplet number concentration for each hologram can be obtained also using Poisson statistics as $\sqrt{N} =$

Figure 2.2: Variation in droplet size distribution within consecutive holograms taken 30 seconds apart. Steady state cloud conditions had already been attained at $t = 0$ s when the measurement was started. We see considerable variation in the droplet size distribution even for this small sample set of eight measurements with respect to the mean over the same eight holograms.

28.28 per hologram or 3 cm^{-3} . In this paper, we primarily concentrate on the variation in the Integral radius (I). The uncertainty in I comes out as 20.1 m^{-2} while the measured RMS fluctuations $I'_{rms} = 317.6 \text{ m}^{-2}$. Thus the uncertainty is very small (6%) compared to the measured rms fluctuations.

2.5 Results

2.5.1 Local microphysical variability

Fundamentally, this study is about a quantity very difficult to access in both naturally occurring and laboratory clouds: what does the cloud droplet size distribution look like on the ‘local’ scale at which droplets are interacting through vapor and temperature fields? As discussed already, this information is inaccessible to typical droplet-by-droplet measurements, which require averaging over long times or, in the case of field measurements, over long distances. For example, [30] needed a minimum of 10 minute average to obtain reasonable estimate of the size distribution but used a 100 minute average for better statistics. However, measurements provided by the digital holographic system described in Sec. 2.4 allow us to observe the instantaneous and spatially local size distribution and how it varies in time, as illustrated in Fig. 2.2. Later we will consider the consequences of that microphysical variability. Fig. 2.2 shows eight examples of instantaneous droplet size distributions compared to an average over the eight frames. The time separating each realization is several large eddy turnover times, and indeed, we observe significant changes in the number concentration and mean size of the droplets.

Figure 2.3: Probability Density Function of τ_c shows a long tail for higher values of τ_c . These higher values correspond to holograms with smaller droplet number concentrations and corresponding higher supersaturation fluctuations.

How does this variability affect the value of the ‘local’ phase relaxation time τ_c ? It is worth noting again that τ_c has typically been assumed to be constant in theoretical treatments, with relatively few exceptions [15]. We can construct a probability density function (pdf) of τ_c by calculating τ_c from the droplets in each single hologram, and repeating for a large number of statistically independent realizations, under statistically stationary conditions. We emphasize that the simultaneous achievement of statistically independent samples and statistically stationary conditions is difficult to achieve in the atmosphere [40, Chpt. 2] and can therefore be considered an advantage of the laboratory approach. The pdf thus obtained is displayed in Fig. 2.3, and has a mean of approximately 17 s and a standard deviation of 10 s. Significantly, the pdf has a positive skewness with the right tail exceeding 60 s. The parcels with such

large τ_c lie in the extreme ‘slow microphysics’ limit and may therefore be important for broadening of the droplet size distribution [30].

The cause of variability in τ_c comes from some combination of fluctuations in the droplet number density and the mean droplet radius. Fig. 2.4 shows the local-mean droplet radius $\langle r \rangle$ as a function of n_d with each data point corresponding to one hologram sample volume. The observations tell us that the main contribution to fluctuations in τ_c is the variability in n_d , and in fact the mean droplet radius is quite steady. This result is reminiscent of the inhomogeneous mixing scenario, in which entrainment and dilution leads to a reduction in the liquid water content solely by reducing n_d . With that in mind, we plot $\langle r^3 \rangle$ versus n_d in Fig. 2.5. Contours of constant liquid water content (LWC) reveal that the data points do not exactly follow a single curve, but stay relatively close to a mean value of $\text{LWC} = 0.2 \text{ g m}^{-3}$, varying from approximately 0.1 to 0.5 g m^{-3} . We find it intriguing to consider whether an upward trend in $\langle r^3 \rangle$ with decreasing n_d could be considered a signature of stochastic condensation in naturally occurring clouds, but that will remain the topic of future work.

Finally, the relative dispersion of cloud droplet radius, $\sigma_r / \langle r \rangle$, is also plotted in Fig. 2.4, and is observed to have a slight trend showing a decrease with increasing number concentration. Since collision-coalescence and entrainment have been ruled out, and the classical theory of growth due to diffusion predicts that the standard

Figure 2.4: (Top) Observed values for mean radius $\langle r \rangle$ as a function of number concentration n_d , showing that it does not vary significantly over large n . (Bottom) Relative dispersion of r versus n_d . Here, $\langle r \rangle$ refers to the mean radius for each hologram and is plotted against the number concentration of droplets in the respective holograms.

Figure 2.5: Cubed radius against number concentration with constant Liquid Water Content (gm^{-3}) lines. Here, $\langle r^3 \rangle$ refers to the averaged cubed radius for each hologram. The red point and associated uncertainties indicate mean $\langle r^3 \rangle$ at the mean n_d with standard deviations along each axis.

deviation of droplet size will decrease with increasing radius, this trend agrees well with stochastic condensation theory which predicts an increase in standard deviation with increasing radius. It is also interesting that under fixed experimental conditions as shown here, the strong trend with n_d reported by [30] is not observed. The local relative dispersion (*i.e.* measured for a single hologram) for a given set of boundary conditions exhibits a relatively weak correlation with the local n_d . Presumably it is because n_d shown in Fig. 2.4 are only local, transient realizations whereas the droplet concentrations and associated relative dispersions discussed in [30] are global.

2.5.2 Quantities relevant to stochastic condensation

We now move from a general view of microphysical variability to the specific quantities relevant to the theory of stochastic condensation described in Sec. ???. In order to evaluate the growth rates for mean and variance of r^2 (Eqs. 2.2 and 2.4), it is apparent that in addition to measuring $\bar{I}$ and $\overline{I'^2}$, we also need to determine $\bar{s}$. We cannot use Eq. 3.1 directly, however, because we are not able to measure s_0 with required accuracy. Instead, we see that $\bar{s}$ can be expressed in terms of microphysical variables by rearranging Eq. 2.24:

$$\bar{s} = \frac{-\overline{I' r'^2}}{2\xi \left(\overline{I'^2} / \bar{I} \right) \tau_t}. \quad (2.27)$$

With the resulting estimate of $\bar{s}$ and the measurements of the other microphysical contributions to the correlation $\overline{s' r'^2}$, we can calculate $d\overline{r^2}/dt$ and $d\sigma_{r^2}^2/dt$. Furthermore, we can compare the values of $d\overline{r^2}/dt$ and $d\sigma_{r^2}^2/dt$ with the corresponding values obtained under the assumption that τ_c is a constant. Here we must be precise in defining the two types of averages being considered. Thus far in this section, results have been presented based on average values obtained from individual holograms, such as mean radius or mean τ_c for a given local cloud volume. In the treatment of

stochastic condensation, in contrast, ensemble averages are required — that is, averages over the data obtained from many holograms. In this paper the ensemble average is denoted by an overline. Ensemble averaging the microphysical observations from the holographic system gives the results summarized in Table 3.1. We have written most of the terms in dimensionless form so that the numbers have a meaning that can be interpreted in context of the physics. For example, the quantity $\overline{I'^2}/\overline{I}^2$ appears in both Eqs. 3.1 and 2.26, and can be considered the dimensionless quantity that captures the relative importance of local microphysical variability for condensation growth. Under the measured conditions, it has a value of 0.32. So $I_{\text{RMS}}/\overline{I} = 0.56$ and the RMS fluctuations in local phase relaxation time are therefore quite significant relative to the mean τ_c .

We now continue working towards estimating Eqs. 3.1 and 2.26, and how much they change for assumed constant or variable τ_c . Measurements from the LICOR give us an initial variance of the supersaturation distribution $\sigma_{s_0} = 0.0095$ and turbulence measurements give $\tau_t = 2.8$ s. Assuming constant phase relaxation time, we obtain $\tau_c = 17.7$ s and $\tau_s = 2.4$ s. The $\overline{s}$ resulting from Eq. 2.27 is 0.020. Using that value in Eq. 3.1 then allows us to estimate $s_0 = 0.022$. In turn, assuming constant τ_c and therefore using Eq. 3.1 with $\overline{I'^2} = 0$, we obtain an estimated $\overline{s} = 0.019$. The ratio of those two mean supersaturations is listed in Table 3.1 and provides a sense for the degree to which microphysical variability influences mean properties. It is consistent with the ratio of the growth rate for mean droplet radius (Eq. 2.26) for variable

Measured quantity	Measured value
$\overline{r^2}$	72.5
σ_{r^2}	49.0
$\sigma_{r^2}/\sqrt{\overline{r^2}}$	0.68
$\overline{I'^2}/\overline{I}^2$	0.32
$\overline{I'r^{2'}}/\overline{I}r^2$	-0.06
$\overline{s} _{\tau_c, \text{var}}/\overline{s} _{\tau_c, \text{const}}$	1.05
$\left.\frac{d\overline{r^2}}{dt}\right _{\tau_c, \text{var}}/\left.\frac{d\overline{r^2}}{dt}\right _{\tau_c, \text{const}}$	1.04
$\left.\frac{d\sigma_{r^2}^2}{dt}\right _{\tau_c, \text{var}}/\left.\frac{d\sigma_{r^2}^2}{dt}\right _{\tau_c, \text{const}}$	1.56

Table 2.1

The units for the first two quantities are in μm^2 while the rest of the quantities have been non-dimensionalized. The last two ratios are evaluated using the stochastic model and are not measured directly. From the table, we see very little change due to variability for the mean size but a significant change for the width of the size distribution.

and constant τ_c , also shown in the table. Both ratios suggest that changes to mean properties are modest, at the few percent level. Finally, although the combined water vapor and temperature measurements do not have the absolute accuracy sufficient to estimate supersaturations directly due to inherent uncertainties and measurement error from the LICOR and RTDs, the measured difference $s_0 - \overline{s} = 0.007$ is within a factor of 2 or 3 of the indirectly estimated values above. However, similar to [30], supersaturation fluctuations can be compared and the s' value obtained from holographic measurements (Eq. 2.23) is 0.0089 and agrees well with 0.0095 obtained from the LICOR and RTDs.

The resulting values for Eq. 2.25, however, show that the growth rate of the width of

the droplet size distribution is significantly enhanced when turbulent fluctuations in τ_c are taken into account (by approximately 60%, as shown in the last line of Table 3.1). Thus, we may expect a significantly larger width in the droplet size distribution than predicted using constant τ_c . Qualitatively this seems straightforward, since fluctuations in τ_c will affect the local supersaturation response; but quantitatively, this can be considered a rare instance in which stochastic condensation theory has been directly linked to measurements.

2.5.3 Comparison between theory and measurements

The logic thus far has been to use the holographic system to measure $\overline{I}^2$, $\overline{I'^2}$, and other needed quantities, and then to use the stochastic theory to calculate $d\overline{r^2}/dt$ and $d\sigma_{r^2}^2/dt$. This evaluation of the contributions of microphysical variability to mean and variance in growth rate is the focus of this paper. A further step can be taken, however, by comparing the measured droplet size distribution to that arising from the predicted growth rates, thereby evaluating the success or failure of the stochastic condensation theory. This is achieved by using the holographic system to measure $\overline{r^2}$ and σ_{r^2} , and then using an estimate of the mean droplet residence time τ_{res} to evaluate $d\overline{r^2}/dt \approx \overline{r^2}/\tau_{res}$ and $d\sigma_{r^2}^2/dt \approx \sigma_{r^2}^2/\tau_{res}$. Methods for determining τ_{res} are rather indirect and uncertain because of its Lagrangian nature, but this is an aspect we hope to improve with future developments in measurement capabilities.

We start with a rough approximation of a droplet with mean radius $\bar{r}$ starting at the top of the chamber and falling straight to the bottom with a Stokes settling velocity of $w = kr^2$, where $k = 1.2 \times 10^8 \text{ m}^{-1} \text{ s}^{-1}$ is the Stokes coefficient. To fall over 1 m height of the chamber, we get $\tau_{\text{res}} = 66 \text{ s}$. However, here we have neglected any change in the droplet size and the presence of turbulence. If we integrate the fall speed over the depth H of the chamber taking into account steady droplet growth from the mean supersaturation, we obtain [41]

$$\tau_{\text{res}} = \left(\frac{H}{k\xi\bar{s}} \right)^{1/2}. \quad (2.28)$$

With this, we get $\tau_{\text{res}} = 62 \text{ s}$, which is not very far from our previous approximation. However, for the measured value of $\overline{r^2}$ to match the calculated value of $d\overline{r^2}/dt$, a residence time of $\tau_{\text{res}} = 15 \text{ s}$ is required. To match the measured mean value of $\sigma_{r^2}^2$ with the calculated $d\sigma_{r^2}^2/dt$, we need $\tau_{\text{res}} = 85 \text{ s}$. Thus the rough estimates of residence time fall within those estimated from the growth rates themselves. Overall, the results are highly uncertain due to the presence of turbulence and what is likely a broad distribution of residence times (e.g., [42]). Nevertheless, at least to a factor of 4 the numbers show consistency, and it provides motivation for Lagrangian residence time measurements in the future.

2.6 Discussion

Most previous studies on stochastic condensation [15, 27, 33, 43] introduced supersaturation fluctuations by implementing the Langevin noise term through vertical velocity similar to [44]. This approach had the drawback as mentioned by [13] that larger vertical velocity fluctuations will lead to larger supersaturation values but the time spent by droplets in these regions will be smaller. This negates the effect of higher supersaturations and the droplets will not grow as large. Here we have implemented the noise term directly through supersaturation similar to [28] and [30]. The fluctuations in droplet number concentration are likely due to a combination of localized droplet activation in regions of high supersaturation, and differential droplet sedimentation. The number concentration fluctuations, in turn, influence the supersaturation field independently from vertical velocity.

In this paper, we have described an experiment to investigate the importance of the assumption of constant phase relaxation time τ_c on the process of stochastic condensation. We produced a steady state warm turbulent mixing cloud and observed the effect of droplet number and size variations within the cloud on the phase relaxation time, using a digital holographic measurement method. The data are analyzed in the context of stochastic theory with and without constant τ_c . The results suggest that for our conditions, the assumption of constant τ_c is a good enough approximation

for the growth rate of the mean squared radius, but not such a good approximation for calculating the rate of growth of the width of the droplet size distribution. The result is interesting because it suggests that in addition to the importance of supersaturation variability, which was observed by [30], the microphysical variability can also play an important role.

Physically, we can interpret the finding as follows. Consider a small parcel of cloudy air that suddenly finds itself with a lower droplet number concentrations compared to the average due to turbulent fluctuations. Let us assume that most of the droplets in this parcel begin with a size close to the mean. This parcel has different properties from its surroundings and hence, soon gets mixed with neighboring parcels bringing the droplet number concentration back to the mean. The few droplets that existed within this parcel were able to grow for a small amount of time during which the parcel was isolated, due to the decreased competition for available water vapor. But the enhanced growth halted as soon as the parcel underwent turbulent mixing. When averaged together with other droplets, including those which may have experienced suppressed growth on account of being contained in a different parcel with higher than mean droplet concentration, a significant change in $\overline{r^2}$ is not observed. However, the different response times and correspondingly different changes in the droplet size, will affect the width of the size distribution. With sufficiently numerous fluctuations in I , we can expect to observe a significant increase in the droplet size distribution width σ_{r^2} over time.

With respect to the applicability of these results to clouds, this study motivates a comparison with in-situ cloud measurements where the fluctuations in integral radius can be recorded. The conditions created in the laboratory are very close to those expected within nearly steady state stratocumulus clouds. We expect that large variations in supersaturation will exist in clouds with low droplet number concentrations [30]. But what level of microphysical variability is observed? This motivates the need for in situ observations of fluctuation terms like $\overline{I'^2}$, which require spatially localized measurement of the droplet size distribution [e.g., 10]. Separating the two effects, plus the effect of entrainment, will be challenging, but perhaps insight will come from dependence on droplet number concentration and comparison to suitable stochastic theories [27, 29, 30]. Once we are able to compare these results with in-situ measurements, we can estimate conditions suitable for large variability in integral radius. Current cloud models can then be modified for these conditions to incorporate the effect of broadening due to stochastic condensation and may be able to better predict cloud properties such as lifetime, albedo etc.

Acknowledgments We thank D. Ciochetto for assistance in setting up the holographic system, J. Fugal for mentoring with the hologram reconstruction, and J. Lu and F. Yang for helpful discussions on the theory. This work was supported by the National Science Foundation, grant AGS-1623429.

Chapter 3

Search for microphysical signatures of stochastic condensation in marine boundary layer clouds using airborne digital holography

This chapter details the airborne holographic measurements from the Cloud Systems Evolution in the Trades (CSET) campaign. This work has been submitted in full form to the Journal of Geophysical Research and is currently in review. ¹

¹Desai N, Glienke S, Fugal J, Shaw RA. Search for microphysical signatures of stochastic condensation in marine boundary layer clouds using airborne digital holography. Journal of Geophysical Research.

3.1 Abstract

Droplet growth due to stochastic condensation has been considered as one of the mechanisms to cause broadening of cloud droplet size distributions and jump the bottleneck between droplet growth due to diffusion and collision-coalescence. Digital in-line holography is used to measure variations in droplet number concentration and droplet size in marine boundary layer clouds. Distributions of phase relaxation times are quite broad for some clouds. Turbulence correlation times are estimated, and the comparison of these with phase relaxation times suggests that clouds exist in both fast and slow microphysical regimes. Signatures of stochastic condensation, such as increasing relative size dispersion and increasing droplet size with decreasing number density, are observed.

3.2 Introduction

Warm clouds such as tropical and sub tropical stratocumulus clouds account for roughly 30 percent of cloud coverage over the earth [45] and hence contribute significantly to the earth's radiation and energy balance. They also account for roughly 70 percent of total rainfall in the tropics, which makes modeling the lifetimes and radiative properties of such clouds correctly very important [4]. However, these properties

depend significantly on droplet size distributions and droplet growth rates within the clouds. And one of the major debated topics in the cloud physics community is the droplet growth gap between droplet growth due to condensation and collision coalescence. Standard condensation theory predicts that the droplet size spectra will become narrow with time. However, in-situ measurements [6, 46, 47, 48] often show that droplet size distributions in clouds are broader than expected in an adiabatic updraft.

Various mechanisms such as entrainment with environmental air [49, 50, 51, 52], stochastic condensation [16, 44], particle-turbulence interactions [7, 11], etc. have been considered possible mechanisms to explain this broadening. Recently, numerical studies [27, 28, 29, 43, 53] and observations of supersaturation variability [54, 55] have reignited the debate that stochastic condensation may play an important role in broadening of the size distribution. These build on the work by [15] showing that stochastic condensation may still produce broadening despite the limitations pointed out by [13]. These theoretical models show promise that stochastic condensation may help in solving the broad droplet size distribution problem and await further experimental validation.

Based on experimental studies, [30] argued that under low aerosol conditions, stochastic condensation due to supersaturation fluctuations can broaden the droplet size distribution. Although the role of mean supersaturation also likely plays a role in

the interpretation of those experiments, in this work we will focus on the role of fluctuations. It was argued that the cloud response to fluctuations depends on the relationship between the phase relaxation time (τ_c) and the turbulence correlation time (τ_t). It was shown that the droplets in a cloud parcel and their size distribution behave differently depending on the regime they find themselves in. When $\tau_c < \tau_t$, known as fast microphysics regime, the droplets are able to adjust to the surrounding supersaturation fluctuations quickly and we obtain a narrow size distribution of droplets. Whereas when $\tau_c > \tau_t$, known as the slow microphysics regime, the droplets take longer to adjust and we obtain a broad size distribution. [56] added to this study and showed that the phase relaxation time can vary spatially within a cloud due to variations in the integral radius [15]. This spatial variability in phase relaxation time can further increase the width of the droplet size distribution, allowing some droplets to jump the droplet growth gap and possibly begin collision-coalescence. Both of these laboratory measurements were made in the Michigan Tech II-chamber with idealized steady state conditions with regards to droplet number concentration and other thermodynamic variables. These laboratory studies provided a unique perspective regarding droplet growth behavior and stochastic condensation by correlating supersaturation fluctuations with the variance in droplet size. The mean supersaturation, coupled with size-dependent droplet removal through sedimentation, also likely contributes to the broadening of size distributions in the experiments. Supersaturation fluctuations were measured, however, and it is their contribution that is

explored here in the atmospheric context. We therefore aim to make connections to actual in-situ measurements, building on other recent work in this direction [55, 57].

In this paper, we attempt to extend the approach of [30, 56] by analyzing in-situ measurements of warm boundary-layer clouds to search for indications of stochastic condensation in the microphysical properties. We use the same experimental method as [56] used in the laboratory to measure phase relaxation time in real clouds: Digital in-line holography allows us to measure local, instantaneous droplet number concentrations and droplet size in clouds on the centimeter scale [10]. This provides a phase relaxation time for every hologram (image) without the need for spatial or temporal averaging. We analyze data from the Cloud Systems Evolution in the Trades (CSET) study in which research flights with the holographic instrument HOLODEC on board, flew through marine stratocumulus and cumulus clouds between California and Hawaii. We consider the distribution of phase relaxation times τ_c relative to turbulence correlation times τ_t and ask whether microphysical properties are connected to the relative values of those two time scales in real clouds. The in situ results are compared to and interpreted in the context of the cloud chamber stochastic condensation observations. This study is centered on the word ‘search’, in the sense that we ask whether microphysical signatures expected for stochastic condensation are found in the observations. Correspondence does not imply proof, but it adds to the plausibility that the conceptual picture is reasonable.

To help guide the data analysis, we summarize a few key results from the stochastic differential equation approach of [30] and [56] for cloud droplet growth in a supersaturated environment. The rate of increase in the size of a single cloud droplet is given by [34] $dr^2/dt = 2\xi s$, where r is the droplet radius, ξ is a thermodynamic growth parameter, and s is the water vapor supersaturation. For a collection of cloud droplets exposed to a fluctuating supersaturation field $s = \bar{s} + s'$, where over-bars and primes indicate mean and fluctuating quantities. The growth of the droplet size distribution width depends upon correlations between fluctuations in the supersaturation field and droplet size, and following [56] the mean and variance of r^2 can be expressed as functions of $\tau_{c,avg}$, τ_t , and $\overline{I'^2}/\bar{I}^2$.

Here, $I = \int_0^\infty rn(r)dr$ is the integral radius, defined as the product of the droplet number concentration n and the mean droplet radius [15]. The phase relaxation time is defined as $\tau_c = (4\pi D_v I)^{-1}$ and $\tau_{c,avg}$ refers to its mean value obtained using the ensemble mean value of I for a series of hologram sample volumes. D_v is a modified water vapor diffusion coefficient which considers diffusion as well as latent heat release [15, 34]. The time scale τ_t is the Lagrangian correlation time, effectively equivalent to the turbulence large eddy time. When cloud droplet growth is occurring the supersaturation mean and variance adjust according to [56]

$$\bar{s} = s_0 \left[1 + \frac{\tau_t}{\tau_{c,avg}} \left(1 - \frac{\overline{I'^2}}{\bar{I}^2} \right) \right]^{-1} \quad (3.1)$$

and

$$\overline{s'^2} = \left[\sigma_{s_0}^2 + \frac{\bar{s}^2 \tau_t}{\tau_{c,avg}} \left(\frac{\overline{I'^2}}{\overline{I^2}} \right) \right] \left[1 + \frac{\tau_t}{\tau_{c,avg}} \right]^{-1}, \quad (3.2)$$

where s_0 and σ_{s_0} correspond to the supersaturation mean and variance that the system would relax toward if there were no cloud droplets.

Equations 3.1 and 3.2 largely determine the mean radius and width of the droplet size distribution in a cloud. The ratio $\tau_t/\tau_{c,avg}$ shows up in both, and in part determines the magnitudes of $\bar{s}$ and $\overline{s'^2}$. This ratio is the Damkohler number Da and it defines whether the cloud parcel exists in a fast (polluted) or slow (clean) microphysics regime, corresponding to $Da \gg 1$ and $Da \ll 1$, respectively [30, 55]. The dependence of these quantities on Da indicates that the supersaturation variability and hence the mean radius and width of the droplet size distribution depends on which regime the cloud exists in. In the results section, we will compare some of the above quantities with varying Da to better understand how these quantities vary. But before that, the next section describes how the dataset for this analysis was obtained.

3.3 In-situ measurements

Stratocumulus clouds provide the ideal cloud system for comparison to the cloud chamber results and the stochastic condensation theory because they often exist in a quasi-steady state. Marine stratocumulus lifetimes tend to be very long compared to

Figure 3.1: Two flight segments taken in closed-cellular stratocumulus clouds. (Left) Flight position overlaid with satellite imagery showing where the research aircraft was and a small inset image from one of the wings [<http://catalog.eol.ucar.edu/maps/cset>]. The yellow and red lines show total flight track and distance covered in last hour respectively. The blue circles indicate dropped radiosondes and times are in UTC. (Right) Altitude, droplet number density and droplet size along the flight path, a subsection of which is analyzed for current study. Top panels are for RF02 and bottom panels are for RF10-5. Analyzed data segment times for each of the flights were: RF02 - 17:21:30 to 17:23:30, RF10-5 - 17:22:30 to 17:24:30

the turbulence large-eddy correlation time, and the same is true in the steady-state cloud chamber experiments. In this paper, we analyze data from the Cloud System Evolution of the Trades (CSET) measurements taken during July and August 2015. The field campaign included 16 research flights off the coast of California between Sacramento and Hawaii using the NSF/National Center for Atmospheric Research

(NCAR) Gulfstream-V High-performance Instrumented Airborne Platform for Environmental Research (GV HIAPER) aircraft. Data was analyzed from sections of flights RF02, 10, 15 and 16. These flight sections were selected to satisfy certain criteria we needed to compare with laboratory data obtained using the Π -chamber at Michigan Technological University. In the Π -chamber, we achieve a temporal equilibrium with respect to droplet size distribution by achieving a balance between droplet activation and settling [8]. This produces statistically homogeneous conditions using a stationary measurement instrument. By measuring over a long period of time, we obtain a statistically significant dataset needed for ensemble averaging. Hence, while selecting in-situ measurements for comparison, we needed similar statistically homogeneous conditions so that the notion of ensemble averaging can be used again. We select flight segments with duration of several minutes and corresponding distances of several 10s of km (this approximately replicates the number of data points used during the laboratory measurements). Furthermore, we select flight segments for which consecutive holograms show that the size distribution and number concentration do not change spatially during the time that the instrument is flown through the cloud. This allows us to obtain spatially homogeneous conditions to compare to statistically stationary conditions in the Π -chamber. The vertical velocity in these flight segments was checked so that we do not attempt to compare with regions affected by extreme up-drafts or down-drafts. We also tried to select cloud conditions that spanned from ultra-clean to moderately clean in order to explore a variety of phase relaxation times.

Figure 3.2: Three flight segments taken in open-cellular clouds. (Left panels) Flight position overlaid with satellite imagery showing where the research aircraft was and a small inset image from one of the wings [<http://catalog.eol.ucar.edu/maps/cset>]. The yellow and red lines show total flight track and distance covered in last hour respectively. The blue circles indicate dropped radiosondes and times are in UTC. (Right panels) Altitude, droplet number density and droplet size along the flight path, a subsection of which is analyzed for current study. Top panels are for RF10-10, middle panels are for RF15, and bottom panels are for RF16. Analyzed data segment times for each of the flights were: RF10-10 - 19:20:00 to 19:24:00, RF15 - 18:23:00 to 18:24:30, RF16 - 18:29:17 to 18:30:47.

Subsequently in this paper, we shall refer to these flight segments by their flight names RF02, RF10, etc. Average flight speed was approximately 150 m s^{-1} , hence a 2 minute sampling duration corresponds to around 18 km of cloud path. We obtained approximately four hundred holograms during each section and each hologram was then reconstructed and analyzed to obtain droplet sizes and number concentrations. Two flight segments in stratocumulus layers are shown in Fig. 3.1. The figure shows (left panels) a satellite view with the aircraft track and an inset view from the aircraft camera, and (right panels) a time series of flight altitude and droplet size distribution. Both of these flight segments were through unbroken cloud, and it is expected that entrainment effects are confined primarily to the cloud top. Three flight segments through open-cellular clouds are shown in Fig. 3.2. These flight segments are distinctive compared to the two in Fig. 3.1 on account of the broken cloud characteristics seen in the right panels. The topmost panels belong to RF10-10 and was studied by [58] as an ultraclean and optically thin cloud. The middle and bottom panels show low altitude broken cumulus clouds with no, or minimal, drizzle present.

To measure the key microphysical quantities like droplet size and number concentration, among a variety of measurement instruments installed on the GV HIAPER we used HOLODEC, Holographic Detector for Clouds, which measures cloud droplets of diameters exceeding $6 \mu\text{m}$ up to mm range [9, 59]. A comparison with other instruments on board the research flight for this campaign by [57] showed good agreement

for droplet size distributions and number concentrations. This allows a robust analysis of the cloud properties without loss of information due to the size detection limit. Using HOLODEC also allows for measurements within local sample volumes of about $1 \text{ cm}^2 \times 13 \text{ cm}$ for each hologram. This sample volume is sufficiently large that an estimate of the local droplet size distribution can be obtained from a single hologram. The image sampling rate is 3.3 Hz at a mean flight speed of 150 m s^{-1} , so that holograms are spatially separated by approximately 45 m . All data from HOLODEC used in this study was reconstructed using high performance computing to search each hologram individually for potential particles. After reconstruction, the particles were classified using both supervised and unsupervised machine learning. The unsupervised classification was then checked manually and has proven to yield scientifically reliable results for CSET [57] and other in-situ measurement campaigns [10].

3.4 Results

Using HOLODEC, which is a volume measurement tool, we can estimate τ_c on a local scale by calculating the mean droplet size and the number of droplets in each hologram. This is in contrast to typical in-situ measurements that rely on droplet-by-droplet counting, and therefore require large-scale spatial averaging to estimate τ_c . We repeat this procedure for each subsequent hologram within a particular cloud

Figure 3.3: (Left) Comparing the PDFs of τ_c , we see broad distributions in clouds similar to the one we saw in the laboratory denoted by HoloPi. (Right) Dimensionless variables like supersaturation, relative variance of integral radius and relative variance of droplet size are averaged over all the holograms for a particular flight and plotted against Damkohler number for each of the 5 in-situ cloud measurements. These quantities show an increase with decreasing Da .

knowing that the holograms are statistically independent of each other due to the sample rate and mean flight speed. Fig. 3.3 (left panel) shows distributions of τ_c calculated from five such cloud passes. For reference, we also display the holographic measurements from the Π -chamber (HoloPi instrument). We see that τ_c has a broad distribution, both within individual flights, as well as between flights. Within individual flights τ_c can vary by more than a factor of 10, and for the five flight segments studied here τ_c spans approximately three orders of magnitude. RF10-5, RF15 and RF16 have similar droplet number concentrations and mean diameter to the HoloPi measurements (Table 3.1) and hence similar τ_c distributions. RF02 had a much higher droplet number concentration while RF10-10 had a much lower value and hence the τ_c distributions have much smaller and larger mean values, respectively. RF15 and RF16 consisted of measurements taken at low altitude and through shallow cumulus

Table 3.1

Mean number concentration, diameter, supersaturation, turbulence correlation time, average phase relaxation time, Damkoehler number and mean vertical wind speed for the respective cloud segments. The mean value is calculated by averaging over all the holograms from the cloud pass segment. The HoloPi vertical velocity is the rms value.

Variable	Units	RF02	RF10-5	RF15	RF16	RF10-10	HoloPi
$\bar{n}$	cm^{-3}	368	100	70	67	8	72
$\bar{d}$	μm	15	18	19	20	29	16
$\bar{s}$	-	7.6e-4	7.7e-4	1.2e-3	1.0e-3	1.2e-3	0.02
τ_t	s	88	113	133	147	133	2.8
$\tau_{c,avg}$	s	1.4	4.4	5.7	5.2	40.2	7.0
Da	—	62.8	25.7	23.6	28.5	3.3	0.4
w	ms^{-1}	-0.004	-0.12	-0.06	0.17	-0.10	0.10

clouds. A number of data gaps seen in Fig. 3.2 indicate clear air and lack of cloud droplets. We therefore expect that RF15 and RF16 clouds may be microphysically distinct due to strong entrainment of non-cloudy air.

3.4.1 Damkoehler number

With the wide range of τ_c in mind, we proceed to consider the Damkoehler number. By definition, this number gives us the ratio of the mixing time scale to the phase relaxation time scale. Prior studies have used it primarily as a measure of entrainment mixing [51, 60]. However, in this study we use it as a measure of mixing without active entrainment with unsaturated air [30, 55]. Variation in the integral radius (I) due

to a variation in the droplet number concentration affects the phase relaxation time (τ_c) [15] and can result in a cloud existing in a new Da regime. But in order to estimate Da , an estimate of the turbulence correlation time (τ_t) is required. For that purpose, we use the high resolution wind velocity data measured at 25 Hz with the GPS-corrected wind vector measurements on board the aircraft. However, due to the flight speed (150 m s^{-1}), the velocity measurements were still separated by 5 m. This meant that the velocity autocorrelation time scale could not be used due to the lack of spatial resolution. The vertical and horizontal wind velocity fluctuations (u') are instead used to create respective longitudinal structure functions. These are normalized by the expected $x^{2/3}$ scaling, where x is lag-distance along the flight path. The plateau value for the resulting compensated structure functions provides an estimate of the mean turbulent kinetic energy dissipation rate ε . Then using Kolmogorov scaling, we approximate the turbulence correlation time as

$$\tau_t = \frac{u'^2}{\varepsilon}. \quad (3.3)$$

Due to the flight speed and required averaging to obtain ε , we are limited to flight-segment-averaged values of τ_t (Table 3.1). Comparing these values with the obtained phase relaxation times for these clouds gives us the Damkohler numbers. We can see that, in general, $\tau_{c,avg}$ values are less than or approaching τ_t , suggesting that most of the clouds reside in the fast-microphysics regime. But the larger values of τ_c observed

in clouds like RF10-5, RF15, RF16, and RF10-10, suggest that Da can approach or become smaller than unity, and the cloud therefore can reside in the slow microphysics regime. The prior laboratory work suggests that as $Da \sim 1$ is approached, effects of stochastic condensation will become more pronounced and size distributions will become broader [30, 56, 61].

Figure 3.3 (right panel) shows three quantities that the stochastic theory suggests should have a Da dependence, namely relative variance of droplet radius, relative variance of integral radius and the inferred quasi-steady supersaturation. We obtain droplet radius and integral radius directly from the holographic measurements of droplet size distribution, as described in Sec. 3.3. The flight-averaged Da is obtained by using $\tau_{c,avg}$ and τ_t . The ratio s/s_0 is obtained from Eq. 3.1 using the holographic measurements and the value for τ_t .

All three quantities are observed to increase monotonically with decreasing Da . A possible exception in the integral radius trend are the two highest points near $Da \approx 30$, corresponding to the strongly-entraining clouds in RF15 and RF16. This is not unexpected since entrainment is known to cause large variations, predominantly a decrease in number and droplet size. The three points corresponding to RF02 and RF10 (Fig. 3.3) show a steady increase in variance due to decrease in number and increase in droplet size, consistent with the expectation for stochastic condensation. The relative dispersion in size and supersaturation measurements agree well with [30]

who showed a similar result using laboratory measurements of cloud droplet growth by condensation in a turbulent flow. This correspondence is suggestive that the processes occurring in the chamber leading to broad droplet distributions may not be confined to the laboratory but can be found in real clouds as well. However, the effects of entrainment in RF15 and RF16 and the effects of collision-coalescence in RF10-10 make it difficult to directly compare the stochastic condensation behavior with these cloud segments. The likely absence of strong entrainment and collision-coalescence affecting droplet properties in RF02 and RF10-5 suggest that their microphysical properties can reasonably be considered indications of stochastic condensation. In order to further explore this possibility we look at data properties we consider as microphysical fingerprints of stochastic condensation.

3.4.2 Relative dispersion

A key suggestion of the stochastic condensation theory is that droplet size distributions should become broader as Da decreases. The observations suggest that this is indeed the case for mean cloud properties (cf., Fig. 3.3). We now consider how size distribution width varies at the local microphysical level measured in individual holograms. The left panel of Fig. 3.4 shows the relative dispersion in droplet radius $\sigma_r/\bar{r}$ versus n . Each point corresponds to $\sigma_r/\bar{r}$ calculated from one hologram, with color denoting the flight segment. The data show a remarkably uniform progression

Figure 3.4: (Left) The relative dispersion in radius for each hologram is obtained by dividing the standard deviation in radius by the mean radius in each hologram. It is then plotted against the droplet number concentration in that hologram. We can see that the relative dispersion increases with decreasing droplet number concentration for laboratory as well in-situ measurements. (Right) Mixing diagrams comparing in-situ measurements with those from the Π -Chamber. The dotted lines represent constant liquid water content (LWC) lines with the ratio of w and w_0 corresponding to the ratio of LWC in the holograms and the LWC corresponding to the least diluted sample respectively. The HoloPi points are plotted behind the in situ results, but can be seen to extend over a similar range as RF10-5, RF15, and RF16. The points for RF10-10 actually extend up to nearly 10 on the Y axis, but we have cropped the figure to show clearly the similarity in trend between the HoloPi and RF02, RF10-5 measurements.

to increasing relative dispersion with decreasing n . Furthermore, the cloud chamber stochastic condensation results show a similar trend. We take this as one of the hologram-scale microphysical fingerprints of stochastic condensation. While the overlapping of some of the data points does show the similarity in behavior, it also obscures the trends visually. Hence, we calculated correlation coefficients of linear regression (R) and conducted a test of statistical significance (p -value) of the data. The R values are -0.49, -0.68, -0.40, 0.05, -0.13, -0.22 and the p values are 0.00, 0.00, 0.00, 0.45, 0.02, 0.00 for HoloPi, RF02, RF10-5, RF15, RF16, RF10-10 respectively. The negative R values indicate an inverse relationship between $\sigma_r/\bar{r}$ and n . The p

values below 0.05 indicate statistical significance at the 95% significance level [62]. The values show a robust and statistically significant negative correlation between $\sigma_r/\bar{r}$ and n for HoloPi, RF02, RF10-5, and RF10-10. Flight segments from RF15 and RF16 are affected by entrainment mixing and do not show the same robustness.

3.4.3 Mixing diagrams

A second hologram-scale microphysical fingerprint of stochastic condensation, pointed out by [56] for the Π -chamber studies, is the tendency of mean-volume diameter ($\overline{d^3}$) to increase with decreasing n . This is in contrast to the more typically-observed steady or decreasing $\overline{d^3}$ for entrainment mixing [60]. Mixing diagrams for the in-situ observations are shown in the right panel of Fig. 3.4. The HoloPi laboratory measurements are again shown for comparison (blue circles). Mixing diagrams are constructed as follows: The hologram with the maximum number concentration (n_o) was considered as the least diluted sample for each flight segment and the corresponding mean volume diameter as d_0 [10, 63]. While these values may not represent the sample with maximum liquid water content, and other normalization approaches exist, the method used here is typical and shows the variation of the mean volume diameter with variation in the droplet number concentration, which is the focus of this study. We can see that RF15 and RF16 are broken, entraining clouds (cf., Fig. 3.2) and indeed the mixing diagrams exhibit the typical signature of inhomogeneous mixing

[10]: a steady, but slightly-decreasing diameter with decrease in n . The other flight segments, however, show an increase in diameter with decreasing droplet number concentration, consistent with the HoloPi measurements [56].

For the three cloud cases that are less influenced by entrainment, and that show increasing $\overline{d^3}$ with decreasing n in the right panel of Fig. 3.4, we now consider the possible role of collision-coalescence growth. The mean diameters for RF02 and RF10-5 clouds are approximately 15 and 18 μm respectively, which are smaller than the critical radius to begin the autoconversion process for a marine cloud ($r_{cr} = 10.3 \mu\text{m}$) [64]. If we consider the Long collision kernel [65] and follow [66], we can obtain an autoconversion timescale for cloud liquid water content as $\tau \approx L/P_l$ where P_l is the Long collision kernel coefficient and L is the liquid water content. For RF02 and RF10-5, τ is 214 and 27 hrs respectively which is much larger than typical collision-coalescence timescale. Hence, we can be fairly confident that this increase in diameter is unlikely due to collision-coalescence. This can be contrasted with the data from RF10-10, which was an extremely clean cloud [58] with a mean droplet number concentration of 8 cm^{-3} and a mean diameter of 29 μm . This size is larger than $r_{cr} = 10.3 \mu\text{m}$ for marine clouds and the autoconversion timescale is nearly 3 hrs. If we estimate a turnover timescale (τ_0) for a stratocumulus cloud by considering a $H = 1\text{-km}$ -deep boundary layer and a mean vertical velocity of $w = 0.1 \text{ m s}^{-1}$, we get $\tau_0 = H/w \approx 3$ hours. This value is within an expected diurnal cycle timescale for stratocumulus

Figure 3.5: (Top) The relative dispersion points are now colored according to the mean vertical velocity for each hologram. Top left shows relative dispersion values for RF02, RF10-5 and RF10-10. Top right shows relative dispersion values for RF15 and 16. (Bottom) Mixing diagram points also colored according to the mean vertical velocity for each hologram. The bottom left and right sequence follows that for relative dispersion so that the points do not overlap each other. We can see from the top and bottom figures that the mean volume diameter as well as the relative dispersion is not influenced by vertical velocity.

clouds [67]. The extremely large values along the y-axis can then be confidently attributed to onset of collision-coalescence. In prior literature, an observed increase of $\overline{d^3}$ with decreasing n has been interpreted as dilution by inhomogeneous mixing and subsequent growth [e.g., 63]. The HoloPi measurements are consistent with that view, in the sense that a strongly fluctuating τ_c , primarily from variability in n from whatever source, leads to favored growth of the diluted regions of cloud.

3.4.4 Vertical velocity

As an alternate interpretation of these microphysical signatures from the effects of stochastic condensation and its dependence on droplet number concentration, we consider the effect of vertical velocity with its implications for local-mean supersaturation. This is not a consideration for the Π -chamber data since supersaturation variability there is not directly linked to vertical velocity through adiabatic cooling, but rather to turbulent mixing alone. As seen in Table 3.1, the vertical velocity during these cloud segments is very small and has no clear effect on droplet size properties. Fig. 3.5 shows that the largest mean diameters and the largest relative dispersion do not correlate with the largest updraft velocities. The mean vertical wind speed for most of the cloud segments was of similar magnitude as the RMS vertical wind speed measured in the Π -chamber (Table 3.1). Indeed, one of the reasons this data set was chosen for a comparison with the Π -chamber was the weakly-turbulent nature typical of stratocumulus clouds. An interesting observation is that early studies considering stochastic condensation suggested that supersaturation fluctuations leading to broadening may be caused by vertical velocity fluctuations [44]. This was shown to be ineffective by [13] since a correlation between higher supersaturation values and updrafts would also mean that the droplets would spend lesser time experiencing these higher supersaturation values. The CSET results show no correlation between the vertical velocity and droplet diameters at least for marine stratocumulus clouds,

and this suggests that a local-mean-supersaturation view does not account for the observed microphysical signatures.

3.5 Discussion

Broadening of the cloud droplet size distribution has been a central problem in the cloud physics community ever since early observations by [49]. While the exact causes for this broadening have been intensely debated, many studies suggest that the broadening occurs at the condensation-growth stage and is caused by spatial and temporal variability in supersaturation [e.g., 15, 22, 68]. The cause of this variability has been attributed to a variety of mechanisms, including: variation in local droplet number concentration due to entrainment mixing [51, 52, 69, 70, 71], ripening combined with de-activation of cloud droplets [48, 72, 73, 74], microscale variability of supersaturation [75, 76, 77], vertical cycling of air in clouds [78, 79], etc. We note that in these studies, the variability in supersaturation is sometimes considered as fluctuations, and other times it is considered as variation in the local mean supersaturation; the perspective usually depends on the way the problem is posed. Many of the proposed mechanisms for supersaturation variability have a conceptual overlap, especially those considering vertical cycling, entrainment, and ripening.

Even in the cloud chamber experiments, the mean supersaturation coupled with

droplet removal by sedimentation, likely contribute to the overall width of the droplet size distributions. In this paper, however, we focus our attention on the role of variability in supersaturation, but viewed from the perspective of stochastic condensation [e.g., 15]. This perspective can be understood through the equation for rate of change of the width of the cloud droplet size distribution through Lagrangian supersaturation fluctuations [e.g., 27, 28, 30]: $d\sigma_a^2/dt \propto \overline{s'a'}$, where $a \equiv r^2$ and primed quantities are fluctuations from the mean. Any process that generates variability s' that can result in correlations with a' is capable of broadening the mean size distribution. That could include entrainment and vertical cycling combined with mixing. In the cloud chamber measurements, variability in supersaturation and microphysical properties results from randomness of scalar fields (temperature and water vapor mixing ratio) inherent in turbulent moist Rayleigh-Bénard convection, as well as randomness in droplet activation rate due to local droplet condensation growth and its coupling to the supersaturation field. Heat fluxes to or from the sidewalls and water vapor fluxes to the sidewalls also effectively serve as weak entrainment. Nevertheless, the system is closed in the sense that no injection of clear air occurs, and because of its limited vertical extent, there is no significant role of collision-coalescence. The cloud chamber observations therefore serve as a relatively simple system to which observations from the much more complex natural clouds can be compared. Similarities may suggest common, underlying physics, which may play a dominant role in some environments.

In this work, we have looked for microphysical signatures of stochastic condensation

in marine boundary layer clouds with both closed and open cellular patterns, observed during the CSET campaign. The expected signatures are motivated by the laboratory observations and physical interpretations of [30] and [56]. We analyze the full range of sampled clouds, having high to low number concentrations and different mixing conditions. Spatially localized droplet size distributions are obtained by the holographic instrument HOLODEC on board the GV aircraft during this campaign. The laboratory microphysical observations used here were obtained with a similar in-line digital holographic system. We have identified several microphysical signatures in the laboratory observations that can be interpreted in the context of stochastic condensation theory. We seek for common microphysical signatures in the in situ observations. The observed similarity does not constitute proof of a common mechanism, but at least suggests consistency with both the conceptual picture and some quantitative expectations for stochastic condensation.

We find that phase relaxation time distributions for the range of sampled clouds have considerable widths, varying by more than a factor of 10 for individual cloud systems. This has implications for the width of droplet size distributions [56]. Microphysical quantities such as supersaturation, relative dispersion in integral radius and droplet size are expected to increase with decreasing Damkoehler number, which in turn depends on phase relaxation time. In light of the laboratory observations of [30] this is relevant because the slow microphysical regime, with onset near $Da \sim 1$, leads to an increase in the width of droplet size distribution. The in situ observations confirm

that the $Da \sim 1$ regime is commonly approached in marine boundary layer clouds.

At the single-hologram scale, we observe a clear increase in the relative dispersion of droplet radius with decrease in droplet number concentration. Relative dispersion in radius has been a focus in cloud physics over the last 20 years, with condensation theory predicting a increase in relative dispersion with increase in number concentration [34, 80, 81, 82]. Some in-situ measurements [38, 83] show unclear signals for the relative dispersion, while many [78, 84, 85, 86, 87] show an increase in relative dispersion with decrease in number concentration, consistent with the findings presented here. Recent studies by [88, 89] suggest that our flight segments could be in an updraft limited regime and the relative dispersion should depend on the aerosol concentration. Turbulent fluctuations may lead to considerable variability in droplet number concentration thereby increasing the spectral width of the droplet size distribution, consistent with the systems theory approach by [90, 91]. Other explanations for the increased size-distribution width with decreasing n in marine stratocumulus clouds include variability in updraft velocity combined with internal mixing [92, 93] and repeated vertical cycling, entrainment, and mixing [78]. In particular, the observations of [78] indicating negative correlations for mean diameter and relative dispersion with n are consistent with the our results, and conceptual connections between their vertical-cycling mechanism and the stochastic-condensation approach should be explored.

Mixing diagrams constructed with single-hologram-scale data suggest three microphysical regimes. Open-cellular cloud segments such as RF15 and RF16 show typical behavior expected for entrainment and inhomogeneous mixing, with a slightly decreasing mean diameter with decrease in droplet number concentration. Although this is distinct from the mixing diagram behavior observed in the cloud chamber, it is interesting to note that these clouds still exhibit a weak increase in relative dispersion in Fig. 3.4 (left). This may be indicative of some effects of stochastic condensation together with the dominant effects of entrainment mixing. Other sampled clouds such as RF02 and RF10-5 do show an increase in mean droplet size with a decrease in number concentration. The in situ observations exhibit a lot of scatter, but the points seem to align with constant liquid water content contours. Variability in up-draft speed near cloud base where activation occurs could account for this behavior. It is also the behavior expected for inhomogeneous mixing followed by subsequent growth [63]. In any case, Figs. 3.1 and 3.2 suggesting these clouds were sampled relatively far from regions of active entrainment. Cloud-top mixing is sure to have been occurring, and we can speculate that the weak entrainment effects are similar to sidewall effects in the cloud chamber. Once regions of with slight variations in microphysical properties such as temperature, number concentration and supersaturation exist, isobaric mixing can lead to preferred droplet growth [23, 69]. This may also act as a source of microphysical variability in droplet number concentration resulting in droplet growth by stochastic condensation as proposed by [56].

The mean droplet diameters in these clouds being 15 and 18 μm and the large autoconversion time scales makes it unlikely that this behavior may be a result of collision-coalescence. The similarity in this trend with the data obtained from laboratory measurements in the Π -chamber suggests that stochastic condensation may be occurring in these clouds. The behavior of an ultra-clean cloud RF10-10, which had the lowest droplet number concentration and largest mean droplet diameter (29 μm) has a distinct and scattered mixing-diagram signature compared to the other clouds, indicating that collision-coalescence is occurring. The behavior of RF10-10 in both relative dispersion and mixing diagrams provides a comparison between the observed microphysical signatures of collision-coalescence and stochastic condensation.

The observations taken together show consistency between microphysical signatures from the cloud chamber and from solid, extensive (but non-precipitating) stratocumulus clouds far from regions of strong entrainment. Implications of the stochastic condensation perspective are therefore worth considering. The dependence of Da , as defined in this paper, on liquid water content $L \propto n\bar{r}^3$ is of particular relevance. Because $Da = \tau_t/\tau_c$ and $\tau_c = (4\pi D_v n \bar{r})^{-1}$, we can say that approximately $\tau_c \propto r^2/L$. Thus, for small variations in radius, Da is directly proportional to L . This suggests that the optimal process leading to strong stochastic condensation is dilution through turbulent mixing with pre-humidified, cloud-free air. This results in no change in r , but reduction in n and therefore in L , and a proportional decrease in Da . Entrainment at cloud top, along with inhomogeneous mixing leading to complete evaporation

of droplets, is a process capable of generating the air that, through subsequent mixing events, can lead to cloud dilution without evaporation. This sequence of events, followed by adiabatic ascent, would lead to super-adiabatic droplet sizes [94]. It therefore has the potential to be a powerful mechanism for size-distribution broadening. This sequential view of mixing is consistent with the concept of stochastic condensation, which simply postulates that fluctuations in supersaturation will lead to broader size distributions. Those fluctuations may arise from inhomogeneous mixing and subsequent dilution, variability in vertical velocity, or direct isobaric mixing. One suggestion from the observations presented here is that when entrainment becomes dominant, then r decreases as part of the reduction in L , with the result that the change in Da is ambiguous and the associated microphysical signatures for stochastic condensation are less clear.

The observed dependence of mean microphysical properties on Damkohler number, and the two identified local-scale microphysical signatures (i.e., for relative dispersion and for mean-volume radius versus n), are all consistent with recent laboratory and theoretical studies of cloud droplet growth in a turbulent environment with supersaturation fluctuations. This correspondence does not amount to proof, but builds on the growing evidence that turbulence, as well as aerosol properties, modulates the microphysical properties of clouds. The observations suggest that clouds can possess slow versus fast microphysical response times relative to turbulent mixing times, which has implications for droplet-size-distribution broadening, drizzle formation, and aerosol

indirect effects.

3.6 Acknowledgments

This research was supported by U.S. National Science Foundation (NSF) grant AGS-1623429. The National Center for Atmospheric Research is sponsored by NSF. We are grateful to the CSET science team and the NCAR Earth Observing Laboratory and its Research Aviation Facility for the project organization and implementation. Field data used in the study are available from the CSET data archive hosted by NCAR: http://data.eol.ucar.edu/master_list/?project=CSET and cloud chamber data are available at: <https://digitalcommons.mtu.edu/physics-fp/136/>.

Chapter 4

Aerosol mediated glaciation and persistence of mixed phase clouds: steady-state laboratory experiments

This chapter details the laboratory measurements of steady state mixed phase clouds generated in the Michigan Tech Π -chamber. This work is in preparation for publication.¹

¹Desai N, Chandrakar KK, Kinney G, Cantrell W, Shaw RA. Aerosol mediated glaciation of mixed-phase clouds using laboratory measurements. To be submitted.

4.1 Abstract

Mixed phase clouds are not in thermodynamic equilibrium, and yet are observed to persist for long periods of time. Their microphysical properties and the processes that sustain them are essential to understanding their influence on Earth’s radiation budget, as well as practical problems such as aircraft icing. In this paper, it is demonstrated that mixed phase clouds can be sustained in steady-state within a laboratory cloud chamber. For constant externally-controlled thermodynamic and turbulence properties, the ice to liquid ratio is modulated by varying the ice-nucleating-particle concentration. Microphysical properties are measured for the various ice fractions and are consistent, to within experimental uncertainties, with theoretical predictions for the Wegener-Bergeron-Findeisen process.

4.2 Introduction

Cloud cover significantly influences the Earth’s radiation budget and allows higher, more comfortable temperatures to exist at the surface. Nearly 70% of all cloud cover consists of either ice or mixed phase clouds which contain some combination of liquid and ice particles [45, 95]. As a specific example, in the western Arctic nearly 40% of the time cloud cover consists of mixed phase clouds [96]. Understanding these clouds

is thus vital in our attempt to model Earth’s changing climate using Global Climate models or to predict aircraft icing conditions [97].

A number of field observations such as the Beaufort and Arctic Seas Experiment (BASE) [98], First International Satellite Cloud Climatology Project Regional Experiment - Arctic Clouds Experiment (FIRE-ACE) [99], Mixed Phase Arctic Cloud Experiment (MPACE) [100] and others have attempted to measure microphysical properties of mixed phase clouds using in-situ and remote sensing observation platforms. Aircraft measurements of mixed phase clouds give the most detailed microphysical information, but are sparse in time and space, and can be challenging due to hazardous aircraft icing conditions. Numerical studies have explored the duration and microphysical properties of mixed phase clouds (e.g. [101, 102, 103, 104, 105]. All these studies have found that these clouds can persist for a number of days (e.g. [106]) or rapidly glaciate within a few minutes (e.g. [105, 107]). This variability in cloud lifetime significantly affects their radiative properties and relevance [108, 109]. It also makes mixed phase clouds difficult to represent using climate models [110]. At least part of the variability may result from the response of mixed phase clouds to aerosol properties, through what has been termed the glaciation indirect effect [111].

Laboratory measurements have the distinct advantage of carefully controlled conditions and ease of measurements access to generated conditions. The possibility of detailed measurements and well-characterized boundary conditions facilitates direct

comparison to theory and numerical models. The Π cloud chamber has the ability to generate conditions conducive to the formation of mixed phase clouds [8], and this study is the first effort at quantitatively exploring the microphysical properties that can be generated. Specifically, we ask the questions, can mixed phase clouds be sustained in steady state in spite of being out of thermodynamic equilibrium, and how do microphysical properties of the mixed phase clouds respond to varying concentrations of ice nucleating particles (INP) and cloud condensation nuclei (CCN)? To answer these questions, we generate a steady-state supercooled cloud in the Π -chamber through turbulent isobaric mixing [8] and study its behavior for varying the aerosol input. By varying the ratio of CCN to INP concentration, different distributions of supercooled cloud droplets and ice particles are produced. The resulting ice-to-total mass ratios or ‘ice fractions’ [112] are quantified and compared to theoretical expectations. Finally, the measured microphysical properties are analyzed to provide insight into the centimeter-scale phase partitioning within these turbulent clouds.

The paper proceeds as follows: In section 4.3, we explain the experimental setup to generate steady state mixed phase clouds. In section 4.4 we extend theoretical predictions to the context of the laboratory experiments. In section 4.5 we present the microphysical results of the experiments and, where possible, compare to theoretical predictions. And finally, in section 4.6, we summarize the main results and discuss implications for the atmospheric context.

4.3 Experimental approach

4.3.1 Chamber setup

To generate a steady state mixed phase cloud, a turbulent mixing cloud is created in the Michigan Tech II-chamber [8]. The bottom surface of the chamber is maintained at $T_b = 4\text{ }^{\circ}\text{C}$ and saturated with liquid water. The top surface is maintained at $T_t = -16\text{ }^{\circ}\text{C}$ and also water-saturated conditions. The unstable temperature gradient between these boundaries creates turbulent convection, and the associated isobaric mixing between the bottom and top saturated surfaces leads to a supersaturated environment. For these experiments it is important that there is a constant water vapor flux into the chamber. The side boundaries are maintained at $T_w = -8\text{ }^{\circ}\text{C}$, close to the mean temperature between the top and bottom surfaces. To create the cloud, we first begin a steady injection rate of NaCl aerosols (20000 cm^{-3} at 3.2 lpm of inlet air flow). These aerosol particles are generated by atomizing an NaCl-water solution using an atomizer (3076 TSI) and subsequently passing it through a diffusion dryer. A fraction of these salt aerosols act as cloud condensation nuclei and a cloud of supercooled liquid droplets forms (fig. 4.1 left). This cloud is allowed to run in this state for a couple of hours until the rate of droplet activation equals that of droplet settling and steady state conditions with respect to droplet size and number

concentration are achieved.

After achieving this stage, Snowmax aerosols generated using a shaking-flask are injected into the chamber at a steady rate at 4 lpm of inlet air flow. Clouds were allowed to relax to steady state for a range of Snowmax injection rates, from 150 cm^{-3} to 5000 cm^{-3} , all at the same air flow rate. These concentrations were chosen since they were the smallest and largest accurately measurable concentrations we could inject into the chamber, for the available equipment. Each of these Snomax injection rates is maintained individually for a time period of 2.5 h and data are recorded for the particular Snomax injection rate under steady state conditions. This entire process starting from the supercooled liquid cloud is then repeated for the next Snomax injection rate keeping all other variables constant. Since Snomax is the only ice nucleating particle (INP) used in this experiment, we will refer to it as INP from here on. Example of the resulting clouds, illuminated by a vertical light sheet, are shown in Fig. 4.1: the left panel is the supercooled liquid cloud obtained with no injection of INP, and the right panel is a mixed phase cloud resulting when INP are injected.

Figure 4.1: (Left) Supercooled liquid cloud produced by injecting NaCl aerosols into the chamber with mean temperature $T_m = -6^\circ\text{C}$. (Right) Mixed phase cloud generated after introduction of Snomax aerosols to the previously generated liquid cloud. We can see that the mixed phase cloud has lower droplet number concentration and visually apparent sparkles from ice crystals.

4.3.2 Holographic setup

We use digital in-line Holography for particle sizing and related microphysical measurements. The holographic setup used for the measurements is the same as used by [56]. A burst of nine consecutive images recorded at 500 frames per second (fps) were taken every 30 seconds for 2.5 h. The image groups were used to eliminate background noise by averaging to produce a single data point every 30 seconds. The obtained digital holograms were then reconstructed numerically at every $100\text{-}\mu\text{m}$ position along the focal plane over 20 cm using the convolution method in Fourier space

[9]. This gives each hologram a measurement volume of 10 cm^3 . The reconstructed images are then analyzed to find particles and their equivalent diameters using a light intensity threshold [38]. Cloud droplets and ice crystals can be distinguished through particle shape [113]. Examples of ice crystals detected in the experiments are shown in Fig. 4.2.

Figure 4.2: Some ice particles as seen in the holograms. The size and the shape of these particles is not the same as ice crystals found in atmospheric measurements due to the small particle lifetimes inside the laboratory chamber.

4.4 Theory

A mixed phase cloud containing all three phases of water is an unstable system due to the difference in saturation vapor pressure over ice particles compared to

water droplets [95, 103]. However, to consider the effect of INP and CCN on the supersaturation inside a cloud parcel, we first start by considering the differential equation for supersaturation over ice as [114]

$$\frac{ds_i}{dt} = \frac{s_{i,0} - s_i}{\tau_t} - \frac{s_i}{\tau_{c,i}} - \frac{s_l}{\tau_{c,l}}. \quad (4.1)$$

Here, s_i, s_l are the supersaturations over ice and liquid, respectively. The ice supersaturation that would be achieved in the chamber in absence of any hydrometeors is $s_{i,0}$. Similarly, $\tau_{c,i}, \tau_{c,l}$ are the phase relaxation times for ice and liquid water particles, respectively, and τ_t is the turbulence mixing time. The first term on the right side of Eq. 4.1 indicates the relaxation of the supersaturation toward the equilibrium value, while the second and third terms are sinks for supersaturation due to ice crystal growth and cloud droplet growth, respectively. The last term can be written in terms of ice supersaturation using $s_l = (e_i/e_l)(s_i + 1) - 1 = A(s_i + 1) - 1$ [114]. Assuming that the system is in steady state, we can solve for s_i :

$$s_i = \left(\frac{s_{i,0}}{\tau_t} + \frac{1 - A}{\tau_{c,l}} \right) \left(\frac{1}{\tau_t} + \frac{1}{\tau_{c,i}} + \frac{A}{\tau_{c,l}} \right)^{-1}. \quad (4.2)$$

Now since this can be equated to $s_i = (s_l + 1)/A - 1$, we can solve for the condition $s_l \geq 0$, which is the condition for coexistence of liquid water and ice. After some

rearranging, this results in

$$\tau_{c,i} \geq \tau_t \left(\frac{e_l - e_i}{e_0 - e_l} \right) = \tau_t \frac{s_l^*}{s_{l,0}}, \quad (4.3)$$

where we have defined $s_l^* \equiv -(e_i - e_l)/e_l$, the magnitude of the liquid-water supersaturation deficit that exists at the ice-saturation level, and denoted the liquid-water supersaturation corresponding to $s_{i,0}$ as $s_{l,0}$. Alternatively, we can write this as a condition for the ice integral radius:

$$n_i \bar{r}_i \leq \frac{1}{4\pi D_v \tau_t} \frac{s_{l,0}}{s_l^*}. \quad (4.4)$$

This expression is analogous to Eq. 4 from [95], and expresses the bound on ice integral radius for a given source of water vapor. [103] and [95] considered vertical velocity as the source of supersaturation, whereas for the cloud chamber, the source is the turbulent flux of water vapor and heat from the bottom to the top boundaries. Equation 4.4 shows that for a given liquid-water supersaturation $s_{l,0}$, relative to the supersaturation deficit that drives the WBF-process (i.e., s_l^*), a critical integral radius exists at which the amount of ice in the cloud chamber should lead to complete glaciation and disappearance of the liquid phase. This analogy allows us to make a one-to-one comparison between the mixed phase cloud produced in the laboratory

and those found in the atmosphere. In the next section we will study the measured microphysical properties of the laboratory mixed phase cloud.

4.5 Results

4.5.1 Size distribution

Probability distribution functions of the measured equivalent diameter of hydrometeors for several INP injection rates are shown in Fig. 4.3 (henceforth this combination of liquid droplets and ice crystals together will be referred to simply as ‘particles’). When INP are not being injected in to the chamber (black curve), a steady state supercooled liquid droplet distribution is observed inside the chamber. The mean supersaturation within the chamber should be close to the saturated vapor pressure over the liquid phase. However, with increasing INP injection rates, the size distribution begins to develop another mode close to $30\text{ }\mu\text{m}$. This corresponds to the appearance of ice particles within the chamber and is consistent with the visual impression of ‘scintillating’ particles illuminated in the cloud chamber (cf., Fig. 4.1). With production of ice particles, the mean water vapor mixing ratio should decrease steadily toward ice saturation. The size PDFs suggest that supercooled liquid is never completely eliminated. At the highest INP injection rate, however, the majority of the

Figure 4.3: Probability density functions of particle equivalent diameter are shown for various INP injection rates. As the INP injection rate increases, larger particles begin to appear and the size distribution becomes bi-modal in nature. The plot would be too busy with all uncertainties shown; for reference, the approximate counting uncertainties for the ranges 10-20, 20-30 and 30-40 μm are 21, 16, 10 particles, which corresponds to 0.008, 0.005, 0.003 on the PDF scale, respectively.

water mass is in the ice phase and therefore the cloud is mostly glaciated. In general, the mean supersaturation will maintain a steady state value somewhere between the two extremes, depending on the number of ice particles compared to liquid droplets (or the ice mass fraction). Consistent behavior can also be seen in the PDFs as a continuous decrease of the mean diameter of the supercooled liquid droplets with increasing INP injection rates.

For the purposes of this analysis, supercooled liquid droplets and ice particles are

classified according to size. All particles with diameters smaller than $20\ \mu\text{m}$ are considered to be water droplets and those with larger effective diameters are considered to be ice particles. This approach is taken because for such small particle sizes, it is challenging to distinguish liquid and ice particles based on shape alone (cf., Fig. 4.2). The $20\ \mu\text{m}$ cutoff is chosen based on the large-diameter tail of the zero INP case, and on the minimum between the two modes of the maximum INP case. Similar approaches have been used in other studies such as [115] where a size step classification function was used to distinguish between ice and water droplet. Using this approach, and assuming ice particles are spheres, the ice mass fraction can be obtained for the control case with no INP being injected. This value is 0.09 corresponds to the supercooled liquid droplets falsely classified as ice particles due to the step size cutoff described above. We subtract this value from the rest of the steady state INP injection rate to obtain the following ice mass fractions 0.41, 0.56, 0.60, 0.81 and 0.82 respectively for the injection rates shown in fig. 4.3. The values are within the mixed phase cloud range for measured ice fraction, with the last two close to the full-glaciation threshold of 0.9 as defined by [116].

4.5.2 Phase relaxation time

The phase relaxation time is defined as

$$\tau_c = (4\pi D_v I)^{-1} \quad (4.5)$$

Where D_v is a constant of diffusivity that also accounts for competing roles of diffusion and latent heat release [15, 34] and $I = \int_0^\infty r n(r) dr$ is the integral radius [15]. The integral radius is the product of the particle number concentration n and the mean particle radius r . Since in a mixed phase cloud, we have two different types of particles, we can find their individual phase relaxation times as

$$\tau_{c,i} = (4\pi D_{v,i} I_i)^{-1} \quad (4.6)$$

and

$$\tau_{c,l} = (4\pi D_{v,l} I_l)^{-1}. \quad (4.7)$$

With $I_i = \int r_i n_i(r_i) dr_i$ and $I_l = \int r_l n_l(r_l) dr_l$ being the integral radius of ice and liquid, respectively. Here, for ice particles, we consider their mean equivalent radius and for water droplets, their mean radius for each hologram. Thus, each hologram gives us a mean radius and number concentration for each phase, which is then

inserted into Eqs. 4.6 and 4.7 to give us instantaneous phase relaxation times per hologram for the entire time period of the steady state experiment. Thus, with digital in-line holography we can measure τ_c at the cm scale instantaneously as compared to most other instruments which rely on droplet-by-droplet counting and thus require a long time average. This allows us to measure not only the mean value of τ_c but also the variation in their instantaneous values [56]. Fig. 4.4 shows that due to turbulent fluctuation in number concentration and size, the probability distributions of the phase relaxation time are quite broad for each phase as well as for both of the phases together.

Figure 4.4: Distributions of phase relaxation time for liquid ($\tau_{c,l}$) in blue, ice ($\tau_{c,i}$) in red and the liquid-ice mixture in gray. Left shows the distributions for an INP injection rate of $0.15 \text{ cm}^{-3}\text{min}^{-1}$. Right shows the distributions for an INP injection rate of $5.0 \text{ cm}^{-3}\text{min}^{-1}$. The comparison shows that the mean τ_c values for liquid may exceed those for ice for higher INP injection rates.

In our experiments, we vary INP injection rates in order to vary the ratio between the number of ice particles and water droplets. We also observe that for low INP injection rates, the mean value for $\tau_{c,i}$ is much larger compared to the mean value

for $\tau_{c,l}$. This is mainly a result of τ_c being a function of the number concentration of the particles. For low INP injection rates, the number of ice particles is very low compared to the number of water droplets and hence ice particles have a larger mean $\tau_{c,i}$. The variation in particle size between ice particles and water droplets, say a factor of 2 or 3, is not as large as the variation in number concentrations between the two. However, for higher INP injection rates, the number of ice particles increases and eventually becomes of the same order, if not more than the concentration of water droplets. This results in mean $\tau_{c,l}$ becoming larger than the mean $\tau_{c,i}$. Hence the phase relaxation times for each phase in a mixed phase cloud depend on the ice fraction of the cloud. Alternatively, phase relaxation time could be used to distinguish different glaciation stages within mixed phase clouds. For example, if mean $\tau_{c,i} > \tau_{c,l}$, the cloud response to humidity fluctuations is liquid dominated, while if mean $\tau_{c,i} < \tau_{c,l}$ it is ice dominated and closer to glaciation. A liquid dominated cloud would be expected to react differently to external processes like entrainment mixing, adiabatic ascent, stochastic condensation, etc. compared to an ice dominated cloud.

4.5.3 INP activated fraction and cloud glaciation

In order to calculate the INP injection rate that will lead to complete glaciation of the cloud, we need to calculate the relation between INP input and ice concentration. For the purposes of this study, we are assuming that 100% of the injected INP are

Figure 4.5: The calculated ice number concentration in blue is compared with the INP injection rate and the measured ice number concentration in red along with the uncertainty calculated as the variability in measured value. The black line with measurement uncertainty shows background ice concentration measured when no INP was being injected. The agreement between the blue and red symbols show that our assumption of nearly 100% of INP being converted to ice was close and the activated fraction was slightly smaller than 100%.

converted to ice particles, which for given conditions and chamber supersaturations, we believe is not a bad assumption to begin with. For steady state conditions, it can then be assumed that the input of INP per unit time is balanced by the removal of ice crystals through sedimentation, where that removal rate is equal to the ice concentration divided by the particle residence time inside the cloud:

$$\left. \frac{dn_i}{dt} \right|_{injected} = \frac{n_i}{\tau_{res}} \quad (4.8)$$

Since most of the ice particle are close to spherical, we can approximate τ_{res} of a particle having mean radius $\bar{r}$ and falling over the height of the chamber $H = 1m$ as, [41]

$$\tau_{res} = \left(\frac{H}{k\xi\bar{s}_i} \right)^{1/2}. \quad (4.9)$$

Here, $\bar{s}$ is the mean supersaturation experienced by the particle as it falls. Assuming that the conditions are saturated with respect to water, $\bar{s}_i$ experienced by the particle is approximately 5% [34] for a mean temperature of -5°C . This gives us a residence time of $\tau_{res} = 62$ s. The corresponding expected ice particle concentration is shown in Fig. 4.5. It is then compared to the measured ice crystal concentration with decent agreement to within the experimental uncertainty with a slight over estimation. This tells us that the fraction of INP that end up forming ice particles should be less than our assumption of 100%. The black line shows background ice concentration measured when no INP was being injected. We believe this is a measurement artifact due to our size step classification between ice and water droplets.

In Sec. 4.4 we obtained a bound on the ice phase relaxation time or integral radius required to sustain coexistence of ice and supercooled liquid water, i.e., mixed phase conditions. Those bounds are given by Eqs. 4.3 and 4.4. The ice integral radius is a directly-measured quantity in our experiments, so we can test the predicted bound if

we know the required supersaturation $s_{l,0}$. Measuring liquid-water supersaturation is challenging, and we instead use the stochastic condensation approach outlined in [56]. We use the zero-INP case since the method is defined for liquid cloud conditions. The method also requires an estimate of the turbulence mixing time τ_t for the Π -chamber, which we obtain from measurements of the time required by the chamber to relax back to an equilibrium condition after a perturbation. That results in $\tau_t = 61.2$ s, which then leads to an inferred supersaturation of $s_{l,0} = 0.031$. That is the liquid-water supersaturation that would exist in the chamber when no CCN or INP are present.

Figure 4.6: Ice integral radius required for glaciation of mixed phase cloud is shown in red while measured values of ice integral radius are shown in blue. The variability in measured values are shown as corresponding shaded regions. This figure shows that for a sufficiently high INP input rate, one can achieve glaciation of a mixed phase cloud.

With estimates of $s_{l,0}$ and s_l^* we can obtain a predicted value for the ice integral

radius required for complete glaciation of the cloud. Fig. 4.6 shows this value in red, along with its uncertainty. The measured ice integral radii are shown in blue. We can see that for the INP injection rates in our experiment, we are just able to approach high enough INP input rates in order to glaciate the cloud. This is consistent with the ice mass fractions estimated earlier, which indicated that at the highest INP injection rate, the cloud is nearly glaciated. Although the expression is in a somewhat different form, we take this as direct experimental validation of the analogous expression derived in [103].

4.5.4 Phase partitioning

Observations by [117, 118] showed phase partitioning in mixed phase clouds as separation of liquid tops above precipitating ice. However, these measurements were typically made using instruments with approximately 100-m spatial resolution. In this study, we are able to observe cloud phase behavior at the centimeter scale. While it has been argued by [114] that below 100 m, ice and liquid particles can be assumed to be well mixed, Fig. 4.7 shows that ice and liquid droplets are not well mixed at the centimeter scale in the Π -chamber. The top frames show holograms containing single phase particles while the bottom frames show a mixture of observed liquid and ice particles. All four frames were obtained during a steady state mixed phase cloud for the highest INP injection rate. This shows that turbulent mixing in mixed phase

Figure 4.7: Particle spatial positions are shown in a 3-dimensional holographic volume representation for four different holographic frames recorded during steady state mixed phase cloud conditions with INP injection rate $= 5.0 \text{ cm}^{-3} \text{ min}^{-1}$. The ice crystals and liquid droplets are shown as red diamonds and blue circles respectively. Phase partitioning at the centimeter scale shows that ice particles and liquid droplets may not be well mixed.

clouds may disrupt the well mixed assumption between ice and liquid phases leading to microphysical variability in number concentrations of each phase. This may lead to variability in local supersaturation value allowing different mixing ratios or ice number fractions to coexist at the centimeter scale.

4.5.5 Mixing diagrams

Mixing diagrams are generally used to understand the effect of entrainment on cloud microphysical properties [63, 119]. The abscissa consists of the particle number concentration normalized by the adiabatic or maximum concentration while the ordinate

Figure 4.8: Mixing diagram with particle number concentration (n) on the abscissa normalized by the highest number concentration (n_0) measured in a hologram. On ordinate is the mean volume diameter (d^3) from each hologram normalized by the mean volume diameter (d_0^3) corresponding to the hologram with largest number concentration. The colors represent ice number fraction which corresponds to the ratio of number of ice particles to total particles (ice + liquid) in a hologram. The number fraction provides a larger range of values for comparison and hence is used here instead of mass fraction. The dotted lines represent constant LWC lines with the ratio of w and w_0 corresponding to the ratio of LWC in the holograms and the LWC corresponding to the least diluted sample respectively. (Left) INP injection rate = $0.15 \text{ cm}^{-3} \text{ min}^{-1}$. (Right) INP injection rate = $5.0 \text{ cm}^{-3} \text{ min}^{-1}$

consists of the measured mean cubed diameter normalized by the mean cubed diameter of the adiabatic parcel. Hence, following [120], if the points lie on the line for $d^3/d_0^3 = 1$ for varying n/n_0 , inhomogeneous mixing is said to occur and if the points lie below $d^3/d_0^3 = 1$, homogeneous mixing is said to occur. For our experiment, we do not have external, unsaturated air mixing with the cloud generated within the chamber. However, we use mixing diagrams to understand the deviation of the droplet mean volume diameter (mvd) with respect to the adiabatic value. In our case, the number concentration and diameter correspond to the values for the least diluted hologram, or in other words, the hologram with the largest particle number concentration. In Fig. 4.8 we see that the mvd in a hologram increases with decreasing particle number

concentration. This behavior does not change with a change in the INP injection rate. This indicates that in holograms with lower number concentrations, the particles are able to grow larger than those at higher number concentrations. The colors indicate the ice number fraction defined within a single hologram, which is defined as

$$\phi = \frac{\text{number of ice particles}}{\text{number of ice particles} + \text{number of liquid droplets}}. \quad (4.10)$$

Here a value of 1 corresponds to all particles being ice and a value of 0 corresponds to all particles being water. Each hologram then has a value of ϕ associated with it depending on the ice number concentration for that hologram. The ice number fractions show that the holograms with the largest ice fractions also tend to have smaller local particle number concentrations.

4.5.6 Relative dispersion

While mixing diagrams show us how the mean diameter changes with respect to fluctuations in number concentration, relative dispersion, defined as $\sigma_r/\bar{r}$, tells us how the particle size distribution shape responds to these fluctuations. The relative dispersion is calculated for each hologram by dividing the standard deviation in radius for all the particles within the hologram, by the mean radius for the hologram. Fig. 4.9 (left) shows that we see an increase in relative dispersion with decrease in number

Figure 4.9: (Left) Shows Relative dispersion values when the INP injection rate = $0.15 \text{ cm}^{-3} \text{ min}^{-1}$. (Right) Shows when the INP injection rate = $5.0 \text{ cm}^{-3} \text{ min}^{-1}$. The colors represent ice number fraction (ϕ) for each hologram. We can see that for low INP injection rates, the relative dispersion increases with decreasing particle number concentrations. But for higher INP injection rates, the trend reverses due to the bimodal size distribution.

concentration for low INP injection rates, where most of the particles are liquid. However when the INP injection rate is increased Fig. 4.9 (right) we see an increase in relative dispersion with an increase in number concentration. The increase in INP injection rate increases the mean ice number fraction (ϕ) within the cloud which indicates more ice particles. And as Fig. 4.3 shows, higher injection rates correspond to a bi-modal size distribution.

4.6 Discussion

Understanding the microphysical processes within mixed phase clouds is essential in our attempt to understand the effect of mixed phase clouds on the Earth's radiation budget as well as practical considerations like aircraft icing. In this paper, we have

described experiments in which we created a steady state mixed phase cloud in the Michigan Tech II-chamber and studied the microphysical properties within this controlled environment. We were able to control the supersaturation using INP to CCN concentrations within the cloud. Equation 4.4 shows that for a given forcing capable of producing a liquid water supersaturation, there exists a critical ice integral radius that will lead to complete cloud glaciation. By increasing the rate of INP injection, we were able to quantitatively verify this glaciation condition (cf., Fig. 4.6).

This provides a different perspective on the Wegener-Bergeron-Findeisen (WBF) process occurring in clouds. The WBF process is most commonly associated with growth of ice crystals at the expense of liquid droplets [103], i.e., cloud glaciation occurs as supercooled cloud droplets are evaporated. However, that is true for a transition state between a glaciated cloud from a supercooled liquid cloud with a few ice particles. The perspective for a steady state mixed phase cloud requires a slightly different interpretation. The larger the concentration of ice particles in a steady state cloud, the lower the mean supersaturation will be within the range defined by the difference in the saturation vapor pressure between ice particles and water droplets. Hence, for a certain ice number concentration, the mean supersaturation of the cloud parcel can be reduced to a value that will prevent any CCN from activating in the first place, and thereby achieve cloud glaciation without evaporation of cloud droplets (Eq. 4.4).

In the atmosphere, achieving such glaciation through large INP concentration is likely

rare, given typical INP concentrations of 0.01 to 100 L^{-1} [34, pp.244]. Only in certain extreme scenarios may give rise to such high INP concentrations [121, 122]. The effect of such high INP concentrations is similar to the cloud ‘overseeding’ concept explored by [123, p.369] and [124]. This may lead to a reduction in particle size and shutting down of the WBF process [125, 126]. We use this exact concept in our experiments to maintain a steady state mixed phase cloud with variable ice mass fraction, by varying the INP concentration. The steady state size distributions achieved within the chamber using the approach, indicate a steady decrease in mean supersaturation due to increase in ice number concentration. This can be seen as a steady reduction in liquid droplet size peak.

Phase partitioning in mixed phase clouds is important since ice and liquid drops have different radiative properties due to their refractive indices, shape, size etc. [108]. Observations have shown the phase composition to have a significant contribution to precipitation and cloud lifetimes [127]. Measurements indicated that turbulent fluctuations in particle number concentrations may cause variability in local supersaturation. This may allow variations in ice number fraction thereby affecting local cloud properties. Similarly, in the atmosphere, cloud ice fraction may change due to a number of factors such as turbulence, variations in ice nuclei [128] or vertical velocity [43, 104]. This variability in the ice concentration at the centimeter scale showed that liquid water and ice particles may not well mixed at the these scales.

It was then important to study how the properties of these clouds changed with change in the ice number fraction. This variability in phase composition leads to broad phase relaxation time distributions. For ice concentrations larger than the their liquid counterparts, the mean value for $\tau_{c,l}$ may exceed the mean value for $\tau_{c,i}$. This time scale could be possibly used to distinguish different glaciation stages within mixed phase clouds. If mean $\tau_{c,i} < \tau_{c,l}$, the cloud may be considered as liquid dominated while if mean $\tau_{c,i} > \tau_{c,l}$, as ice dominated and closer to glaciation. Depending on the dominant phase or glaciation stage, the reaction to external processes like entrainment mixing, adiabatic ascent, stochastic condensation etc. may vary. Mean volume diameter shows an increase with decreasing particle concentration similar to warm clouds [56], indicating that the droplet growth due to higher supersaturation variability caused by reduction in particle concentration is not confined to warm clouds but affects mixed phase clouds as well. Parcels with larger ice fractions benefited the most from these supersaturation fluctuations due to the low saturation pressure over ice particles compared to water. Relative dispersion showed a similar trend to that of warm clouds for lower INP injection rates [56]. However, for higher INP injection rates, the relative dispersion showed an opposite trend indicating the formation of a second (ice) peak in the size distributions.

This study provides a unique perspective on glaciation in mixed phase clouds, through the creation of steady state mixed phase clouds in the laboratory. The similarity with atmospheric mixed phase clouds allows us to study these clouds under controlled

conditions and study their properties for different ice fractions. The results motivate a comparison with in-situ measurements so that we can obtain a better understanding of how ice fraction affects cloud properties. Current cloud models can then incorporate the studied microphysics to evaluate cloud macrophysical properties such as stability, lifetime, albedo, etc.

4.7 Acknowledgments

This work was supported by the National Science Foundation, grant AGS-1754244.

Chapter 5

Additional research topics and conclusions

This chapter begins by briefly describing several other, ongoing projects, all related to the question of how turbulence may affect processes governing the growth of cloud droplets. They all utilize digital in-line holography to obtain laboratory measurements useful in studying the underlying processes. The chapter then concludes with a brief summary of the key findings within this thesis.

5.1 Additional research topics

5.1.1 Effect of turbulence on collision-coalescence

The primary objective of this dissertation is to explore how cloud droplets cross the droplet growth gap between condensation and collision-coalescence. We hypothesized that turbulence plays a role in this process and presented observations to validate it. However, turbulence may continue to play an important role in other aspects of cloud droplet growth beyond condensation. [7, 11] review a wide range of work demonstrating that turbulence makes a significant contribution to the collision-coalescence of droplets and can increase the collision kernel for droplets up to a factor of 5. For example, a predominant understanding of collision and coalescence of cloud droplets is based on the assumption that droplets are randomly distributed in space at small (sub-grid) scales, and that relative droplet speeds are driven solely by differential gravitational sedimentation. However, studies have shown that droplets tend to be clustered [129] on various scales, likely giving rise to a large range of relative velocities depending upon the effects of gravitational settling and turbulence induced motion. This clustering may increase the collisional probability thereby changing the droplet size distribution as well. Larger droplets in turbulent flow can fall at a greater speed compared to droplets in a more quiescent flow. These droplets can then collide with

Figure 5.1: (Left) The Michigan tech turbulence chamber. (Right) a schematic of the holographic assembly to study effect of turbulence and gravity on collision coalescence.

slow moving droplets, assimilating the water and further growing in size. Thus we see that turbulence can significantly affect the droplet growth rates as well as other microphysical processes, but obtaining clear physical understanding and representation across the full range of droplet sizes relevant to the problem is still a challenge [130].

During the course of this PhD work, attention has also been devoted to studies of droplet dynamics in homogeneous, isotropic turbulence, with the objective of understanding the relative velocity between droplets. That relative velocity directly influences the droplet collision rate. In the laboratory, we can explore the transition between two extreme limits: the cloud physics limit, which assumes relative velocity completely dominated by differential gravitational settling; and the mechanical engineering limit, which assumes relative velocity completely dominated by turbulence. In fact, many cloud systems lie within the two limits, and both gravity and turbulence

play a role [131]. To observe these variations in droplet distributions and their relationship to turbulence, we can use a digital holographic imaging system and a chamber capable of producing homogeneous, isotropic turbulence of known energy dissipation rate [132]. A photograph and schematic view of the setup are shown in Fig. 5.1. That enables us to measure droplet spatial correlations, relative velocity distributions and Lagrangian acceleration distributions in a laboratory turbulence chamber. We can obtain Lagrangian droplet tracks over a range of droplet sizes, concentrations and turbulence dissipation rates. By varying these quantities, we can get a better understanding of the underlying microphysics and also make comparisons with field results such as those by [133, 134]. However, it is to be noted that Lagrangian tracking is currently extremely difficult through field measurements due to flight speed and hence we can only compare droplet diameter distributions, concentrations and dissipation rates similar to those observed in atmospheric clouds.

5.1.2 Effect of turbulence on droplet activation

Turbulence can affect cloud droplet growth even before cloud droplets are formed. In order for the water vapor in the atmosphere to form droplets, the ambient supersaturation needs to be sufficiently higher than the saturation vapor pressure over the surface of spherical droplets. The presence of aerosol particles in the atmosphere such as sodium chloride (NaCl) help in the formation of cloud droplets by dissolving with

water and forming a solution. Considering solute effects reduces the required ambient supersaturation. An approximation of the combination of curvature and solute effects on the saturation vapor pressure of a droplet is given by [34, pp.88]

$$\frac{e_s(r)}{e_s(\infty)} = 1 + \frac{a}{r} - \frac{b}{r^3}. \quad (5.1)$$

This is known as the Köhler equation, where $e_s(r)$, $e_s(\infty)$ correspond to saturation vapor pressure over a droplet of radius r and that over bulk water, respectively. The curvature term a/r indicates the increase in saturation ratio over a droplet compared to a plane surface. The solute term b/r^3 shows a decrease in saturation ratio due to the presence of a solution instead of pure water. Using Eq. 5.1 for an NaCl solution droplet radius of $0.2 \mu m$, the supersaturation required for the droplet to grow is nearly 0.4 %. However, most studies have shown that a supersaturation of 0.4 % in atmospheric clouds is rare with the typical value being in the range of 0.1-0.2 % [6, 21]. Although some studies have measured high supersaturation in clouds [135], the low probabilities of obtaining such values may cause cloud models to incorrectly predict cloud formation and lifetimes. However, turbulence may influence droplet activation by producing fluctuations in supersaturation leading to droplet activation and growth.

In the laboratory, we can explore this effect of turbulence on droplet activation by

Figure 5.2: Holographic assembly installed onto the LACIS chamber. (Left) Image of the assembly. (Right) Schematic of the assembly.

using aerosols of a known size and chemical composition. Such aerosols will require a particular supersaturation in order to grow into droplets. The LACIS-LEAK chamber provides an ideal infrastructure for carrying out such investigations, as it allows for the generation of cloud droplets under well-defined and reproducible conditions with a well-characterized range of droplet sizes and concentrations [136]. By mixing two saturated air flows of a known temperature, an environment with a mean supersaturation just below the critical value can be produced. Digital in-line holography can be used to obtain volume measurements of the particles in this mixture. The lens system can be altered to extend the resolution to resolve droplets in the size range $2\text{-}10\ \mu\text{m}$. The overall setup is depicted in Fig. 5.2. If the aerosols do grow to form droplets that are detected by the holographic system, we will know that turbulent supersaturation fluctuations exceeded the mean supersaturation and caused the aerosols to form droplets. By varying the air flow rates and aerosol injection rates, we can determine how the supersaturation fluctuations respond to these variations.

5.2 Conclusions

The objective of the research in this dissertation is

1. to measure and understand the effect of turbulence in droplet growth by condensation in the cloud droplet growth gap in the laboratory.
2. to compare the laboratory measurements with in-situ measurements.
3. To extend the measurements to temperatures below freezing and explore the growth of particles in such conditions through the formation and growth of ice in mixed-phase clouds.

In Chp. 2 we created a steady state warm turbulent mixing cloud in a controlled environment of a laboratory chamber to study the effect of turbulence on the cloud droplet size distribution. We found that turbulent fluctuations in droplet number concentrations even for a steady state cloud may lead to fluctuations in supersaturation. Measurements showed that these fluctuations may lead to local broadening of the droplet spectrum and associated broadening of the phase relaxation time. Holographic measurements allowed us to measure instantaneous droplet size distributions and phase relaxation times. These measurements when combined with the stochastic

condensation model showed that turbulent fluctuations in phase relaxation time will lead to a broader size distribution of droplets compared to when the phase relaxation time is assumed to be a constant. Some of these larger droplets can jump the growth gap and begin collision-coalescence.

Motivated by the laboratory measurements, we explored whether similar processes occur in the atmosphere, leading to a broadening of the droplet size distribution. In Chp. 3 we look for signatures of stochastic condensation occurring in marine stratocumulus clouds over the Pacific Ocean. While relative dispersion is seen to increase with decreasing droplet number concentration for all the flight segments analyzed, mixing diagrams show these processes do not always follow the same trend as seen in the Π -chamber. This allows us to separate droplet size-distribution broadening due to other processes like entrainment and collision-coalescence, from stochastic condensation. Hence, we define an increase in relative dispersion as well as a corresponding increase in mean volume diameter with decreasing droplet number concentration as microphysical fingerprints of stochastic condensation. These common signatures are suggestive that stochastic condensation is occurring in certain clouds, leading to some droplets growing large enough to start collision-coalescence.

While Chp. 2 and 3 dealt with warm clouds and the cloud droplet growth gap for the warm rain process, it is also known that precipitation can be initiated through the formation of ice in mixed phase clouds. Indeed, a significant fraction of total

cloud cover over Earth consists of mixed phase clouds. While these clouds and the ice crystals within them are difficult to measure, digital in-line holography and the Michigan Tech *Pi*-chamber allowed us the unique opportunity to measure mixed phase cloud microphysical properties and growth of ice crystals under steady state and controlled laboratory conditions. In Chp. 4 we describe the creation of steady state mixed phase clouds with varying concentrations of ice nucleating particles (INP) compared to cloud condensation nuclei. We showed that a mixed phase cloud can reach glaciation if the ice integral radius exceeds a certain threshold value. This threshold value is primarily a function of the source of supersaturation within the cloud. By varying the number of INP inside the chamber, the mean supersaturation value can be varied to produce different ice fractions at steady state conditions, and the measurements are consistent with theoretical prediction of the threshold. Lastly, phase partitioning and distributions of cloud droplet and ice crystal phase relaxation times were explored.

References

- [1] C. Sagan, *Pale blue dot: A vision of the human future in space*. Random House Digital, Inc., 1997.
- [2] S. Twomey, “Atmospheric aerosols,” 1977.
- [3] G. L. Stephens, S.-C. Tsay, P. W. Stackhouse Jr, and P. J. Flatau, “The relevance of the microphysical and radiative properties of cirrus clouds to climate and climatic feedback,” *Journal of the atmospheric sciences*, vol. 47, no. 14, pp. 1742–1754, 1990.
- [4] P. J. Webster, “The role of hydrological processes in ocean-atmosphere interactions,” *Reviews of Geophysics*, vol. 32, no. 4, pp. 427–476, 1994.
- [5] K. V. Beard and H. T. Ochs III, “Warm-rain initiation: An overview of microphysical mechanisms,” *Journal of Applied Meteorology*, vol. 32, no. 4, pp. 608–625, 1993.

- [6] H. Pruppacher and J. Klett, “Microphysics of clouds and precipitation, atmos,” *Oceanogr. Sci. Libr., Kulwer Acad., Norwell, Mass*, 1997.
- [7] R. A. Shaw, “Particle-turbulence interactions in atmospheric clouds,” *Ann. Rev. Fluid Mech.*, vol. 35, pp. 183–227, 2003.
- [8] K. Chang, J. Bench, M. Brege, W. Cantrell, K. Chandrakar, D. Ciochetto, C. Mazzoleni, L. Mazzoleni, D. Niedermeier, and R. Shaw, “A laboratory facility to study gas-aerosol-cloud interactions in a turbulent environment: The π chamber,” *Bull. Amer. Meteor. Soc.*, vol. 97, 2016.
- [9] J. P. Fugal, R. A. Shaw, E. W. Saw, and A. V. Sergeyev, “Airborne digital holographic system for cloud particle measurements,” *Appl. Opt.*, vol. 43, pp. 5987–5995, 2004.
- [10] M. J. Beals, J. P. Fugal, R. A. Shaw, J. Lu, S. M. Spuler, and J. L. Stith, “Holographic measurements of inhomogeneous cloud mixing at the centimeter scale,” *Science*, vol. 350, pp. 87–90, 2015.
- [11] W. W. Grabowski and L.-P. Wang, “Growth of cloud droplets in a turbulent environment,” *Ann. Rev. Fluid Mech.*, vol. 45, pp. 293–324, 2013.
- [12] I. Mazin and V. Merkulovich, “Stochastic condensation and its possible role in liquid cloud microstructure formation,” in *Some Problems of Cloud Physics*, pp. 217–267, Russ. Acad. Sci., 2008.

- [13] J. Bartlett and P. Jonas, “On the dispersion of the sizes of droplets growing by condensation in turbulent clouds,” *Quart. J. Roy. Meteor. Soc.*, vol. 98, pp. 150–164, 1972.
- [14] M. Manton, “On the broadening of a droplet distribution by turbulence near cloud base,” *Quart. J. Roy. Meteor. Soc.*, vol. 105, pp. 899–914, 1979.
- [15] W. A. Cooper, “Effects of variable droplet growth histories on droplet size distributions. Part I: Theory,” *J. Atmos. Sci.*, vol. 46, pp. 1301–1311, 1989.
- [16] V. I. Khvorostyanov and J. A. Curry, “Toward the theory of stochastic condensation in clouds. Part I: A general kinetic equation,” *J. Atmos. Sci.*, vol. 56, pp. 3985–3996, 1999.
- [17] C. A. Jeffery, J. M. Reisner, and M. Andrejczuk, “Another look at stochastic condensation for subgrid cloud modeling: Adiabatic evolution and effects,” *J. Atmos. Sci.*, vol. 64, pp. 3949–3969, 2007.
- [18] R. Srivastava, “Growth of cloud drops by condensation: A criticism of currently accepted theory and a new approach,” *J. Atmos. Sci.*, vol. 46, pp. 869–887, 1989.
- [19] R. A. Shaw, W. C. Reade, L. R. Collins, and J. Verlinde, “Preferential concentration of cloud droplets by turbulence: Effects on the early evolution of cumulus cloud droplet spectra,” *J. Atmos. Sci.*, vol. 55, pp. 1965–1976, 1998.

- [20] P. Vaillancourt, M. Yau, P. Bartello, and W. Grabowski, “Microscopic approach to cloud droplet growth by condensation. Part II: Turbulence, clustering, and condensational growth,” *J. Atmos. Sci.*, vol. 59, pp. 3421–3435, 2002.
- [21] A. S. Lanotte, A. Seminara, and F. Toschi, “Cloud droplet growth by condensation in homogeneous isotropic turbulence,” *J. Atmos. Sci.*, vol. 66, pp. 1685–1697, 2009.
- [22] H. Gerber, “Supersaturation and droplet spectral evolution in fog,” *J. Atmos. Sci.*, vol. 48, pp. 2569–2588, 1991.
- [23] A. V. Korolev and G. A. Isaac, “Drop growth due to high supersaturation caused by isobaric mixing,” *J. Atmos. Sci.*, vol. 57, pp. 1675–1685, 2000.
- [24] A. Korolev, M. Pinsky, and A. Khain, “A new mechanism of droplet size distribution broadening during diffusional growth,” *J. Atmos. Sci.*, vol. 70, pp. 2051–2071, 2013.
- [25] M. K. Politovich, “A study of the broadening of droplet size distributions in cumuli,” *J. Atmos. Sci.*, vol. 50, pp. 2230–2244, 1993.
- [26] J.-L. Brenguier and L. Chaumat, “Droplet spectra broadening in cumulus clouds. Part I: Broadening in adiabatic cores,” *J. Atmos. Sci.*, vol. 58, pp. 628–641, 2001.

- [27] G. Sardina, F. Picano, L. Brandt, and R. Caballero, “Continuous growth of droplet size variance due to condensation in turbulent clouds,” *Phys. Rev. Lett.*, vol. 115, p. 184501, 2015.
- [28] R. Paoli and K. Shariff, “Turbulent condensation of droplets: direct simulation and a stochastic model,” *J. Atmos. Sci.*, vol. 66, pp. 723–740, 2009.
- [29] C. Siewert, J. Bec, and G. Krstulovic, “Statistical steady state in turbulent droplet condensation,” *J. Fluid Mech.*, vol. 810, pp. 254–280, 2017.
- [30] K. K. Chandrakar, W. Cantrell, K. Chang, D. Ciochetto, D. Niedermeier, M. Ovchinnikov, R. A. Shaw, and F. Yang, “Aerosol indirect effect from turbulence-induced broadening of cloud-droplet size distributions,” *Proc. Natl. Acad. Sci. (USA)*, vol. 113, pp. 14243–14248, 2016.
- [31] M. Baker and J. Latham, “The evolution of droplet spectra and the rate of production of embryonic raindrops in small cumulus clouds,” *J. Atmos. Sci.*, vol. 36, pp. 1612–1615, 1979.
- [32] S. G. Lasher-Trapp, W. A. Cooper, and A. M. Blyth, “Broadening of droplet size distributions from entrainment and mixing in a cumulus cloud,” *Q. J. Royal Meteorol. Soc.*, vol. 131, p. 195220, 2005.
- [33] W. W. Grabowski and G. C. Abade, “Broadening of cloud droplet spectra through eddy hopping: Turbulent adiabatic parcel simulations,” *J. Atmos. Sci.*, vol. 74, no. 5, pp. 1485–1493, 2017.

- [34] R. Rogers and M. Yau, *A Short Course in Cloud Physics*. Butterworth-Heinemann, 1989.
- [35] S. B. Pope, *Turbulent Flows*. Cambridge University Press, 2001.
- [36] K. Jacobs, *Stochastic processes for physicists: understanding noisy systems*. Cambridge University Press, 2010.
- [37] C. W. Gardiner *et al.*, *Handbook of stochastic methods*, vol. 3. springer Berlin, 1985.
- [38] J. Lu, J. P. Fugal, H. Nordsiek, E. W. Saw, R. A. Shaw, and W. Yang, “Lagrangian particle tracking in three dimensions via single-camera in-line digital holography,” *New J. Phys.*, vol. 10, p. 125013, 2008.
- [39] J. Henneberger, J. Fugal, O. Stetzer, and U. Lohmann, “Holimo ii: a digital holographic instrument for ground-based in situ observations of microphysical properties of mixed-phase clouds,” *Atmos. Meas. Tech.*, vol. 6, pp. 2975–2987, 2013.
- [40] J. C. Wyngaard, *Turbulence in the Atmosphere*. Cambridge University Press, 2010.
- [41] F. Yang, M. Ovchinnikov, and R. A. Shaw, “Microphysical consequences of the spatial distribution of ice nucleation in mixed-phase stratiform clouds,” *Geophys. Res. Lett.*, vol. 41, pp. 5280–5287, 2014.

- [42] F. Yang, M. Ovchinnikov, and R. A. Shaw, “Long-lifetime ice particles in mixed-phase stratiform clouds: Quasi-steady and recycled growth,” *J. Geophys. Res.*, vol. 120, pp. 11,617–11,635, 2015.
- [43] P. Field, A. Hill, K. Furtado, and A. Korolev, “Mixed-phase clouds in a turbulent environment. Part 2: Analytic treatment,” *Quart. J. Roy. Meteor. Soc.*, vol. 140, no. 680, pp. 870–880, 2014.
- [44] I. Mazin and V. Smirnoff, “On the theory of cloud drop size spectrum formation by stochastic condensation,” *Proc. CAO*, vol. 89, pp. 92–94, 1969.
- [45] K. Lau and H. Wu, “Warm rain processes over tropical oceans and climate implications,” *Geophysical Research Letters*, vol. 30, no. 24, 2003.
- [46] I. R. Paluch and C. A. Knight, “Mixing and the evolution of cloud droplet size spectra in a vigorous continental cumulus,” *Journal of the atmospheric sciences*, vol. 41, no. 11, pp. 1801–1815, 1984.
- [47] J.-L. Brenguier and L. Chaumat, “Droplet spectra broadening in cumulus clouds. part i: Broadening in adiabatic cores,” *Journal of the atmospheric sciences*, vol. 58, no. 6, pp. 628–641, 2001.
- [48] S. Romakkaniemi, G. McFiggans, K. Bower, P. Brown, H. Coe, and T. Choulaton, “A comparison between trajectory ensemble and adiabatic parcel modeled cloud properties and evaluation against airborne measurements,” *Journal of*

- Geophysical Research*, vol. 114, no. D6, pp. D06214, doi:10.1029/2008JD011286, 2009.
- [49] J. Warner, “The microstructure of cumulus cloud. part ii. the effect on droplet size distribution of the cloud nucleus spectrum and updraft velocity,” *Journal of the Atmospheric Sciences*, vol. 26, no. 6, pp. 1272–1282, 1969.
- [50] J. W. Telford and S. K. Chai, “A new aspect of condensation theory,” *pure and applied geophysics*, vol. 118, no. 2, pp. 720–742, 1980.
- [51] M. Baker, R. Corbin, and J. Latham, “The influence of entrainment on the evolution of cloud droplet spectra: I. a model of inhomogeneous mixing,” *Quarterly Journal of the Royal Meteorological Society*, vol. 106, no. 449, pp. 581–598, 1980.
- [52] I. R. Paluch and C. A. Knight, “Does mixing promote cloud droplet growth?,” *Journal of the atmospheric sciences*, vol. 43, no. 18, pp. 1994–1998, 1986.
- [53] W. W. Grabowski and G. C. Abade, “Broadening of cloud droplet spectra through eddy hopping: Turbulent adiabatic parcel simulations,” *J. Atmos. Sci.*, vol. 74, no. 5, pp. 1485–1493, 2017.
- [54] F. Ditas, R. A. Shaw, H. Siebert, M. Simmel, B. Wehner, and A. Wiedensohler, “Aerosols-cloud microphysics-thermodynamics-turbulence: evaluating supersaturation in a marine stratocumulus cloud,” *Atmospheric Chemistry and Physics*, vol. 12, no. 5, pp. 2459–2468, 2012.

- [55] H. Siebert and R. A. Shaw, “Supersaturation fluctuations during the early stage of cumulus formation,” *Journal of the Atmospheric Sciences*, vol. 74, no. 4, pp. 975–988, 2017.
- [56] N. Desai, K. Chandrakar, K. Chang, W. Cantrell, and R. Shaw, “Influence of microphysical variability on stochastic condensation in a turbulent laboratory cloud,” *Journal of the Atmospheric Sciences*, vol. 75, no. 1, pp. 189–201, 2018.
- [57] S. Glienke, A. Kostinski, J. Fugal, R. Shaw, S. Borrmann, and J. Stith, “Cloud droplets to drizzle: Contribution of transition drops to microphysical and optical properties of marine stratocumulus clouds,” *Geophysical Research Letters*, vol. 44, no. 15, pp. 8002–8010, 2017.
- [58] R. Wood, K.-T. O, S. Bretherton, Christopher, J. Mohrmann, B. A. Albrecht, P. Zuidema, V. Ghatge, C. Schwartz, E. Eloranta, S. Glienke, *et al.*, “Ultraclean layers and optically thin clouds in the stratocumulus to cumulus transition: part i. observations,” *Journal of the Atmospheric Sciences*, no. 2018, 2018.
- [59] S. M. Spuler and J. Fugal, “Design of an in-line, digital holographic imaging system for airborne measurement of clouds,” *Applied optics*, vol. 50, no. 10, pp. 1405–1412, 2011.
- [60] K. Lehmann, H. Siebert, and R. A. Shaw, “Homogeneous and inhomogeneous mixing in cumulus clouds: Dependence on local turbulence structure,” *Journal of the Atmospheric Sciences*, vol. 66, no. 12, pp. 3641–3659, 2009.

- [61] K. Chandrakar, W. Cantrell, D. Ciochetto, S. Karki, G. Kinney, and R. Shaw, “Aerosol removal and cloud collapse accelerated by supersaturation fluctuations in turbulence,” *Geophysical Research Letters*, vol. 44, no. 9, pp. 4359–4367, 2017.
- [62] R. A. Fisher, *Statistical methods for research workers*. Genesis Publishing Pvt Ltd, 2006.
- [63] S. S. Yum, J. Wang, Y. Liu, G. Senum, S. Springston, R. McGraw, and J. M. Yeom, “Cloud microphysical relationships and their implication on entrainment and mixing mechanism for the stratocumulus clouds measured during the vocals project,” *Journal of Geophysical Research: Atmospheres*, vol. 120, no. 10, pp. 5047–5069, 2015.
- [64] Y. Liu, P. H. Daum, and R. McGraw, “An analytical expression for predicting the critical radius in the autoconversion parameterization,” *Geophysical research letters*, vol. 31, no. 6, 2004.
- [65] Y. Liu, P. H. Daum, R. L. McGraw, M. A. Miller, and S. Niu, “Theoretical expression for the autoconversion rate of the cloud droplet number concentration,” *Geophysical Research Letters*, vol. 34, no. 16, 2007.
- [66] K. Chandrakar, W. Cantrell, and R. Shaw, “Influence of turbulent fluctuations on cloud droplet size dispersion and aerosol indirect effects,” *Journal of the Atmospheric Sciences*, vol. 75, no. 9, pp. 3191–3209, 2018.

- [67] R. Wood, “Stratocumulus clouds,” *Monthly Weather Review*, vol. 140, no. 8, pp. 2373–2423, 2012.
- [68] M. K. Politovich and W. A. Cooper, “Variability of the supersaturation in cumulus clouds,” *Journal of the atmospheric sciences*, vol. 45, no. 11, pp. 1651–1664, 1988.
- [69] S. G. Lasher-trapp, W. A. Cooper, and A. M. Blyth, “Broadening of droplet size distributions from entrainment and mixing in a cumulus cloud,” *Quarterly Journal of the Royal Meteorological Society: A journal of the atmospheric sciences, applied meteorology and physical oceanography*, vol. 131, no. 605, pp. 195–220, 2005.
- [70] H. Morrison and W. W. Grabowski, “Modeling supersaturation and subgrid-scale mixing with two-moment bulk warm microphysics,” *Journal of the Atmospheric Sciences*, vol. 65, no. 3, pp. 792–812, 2008.
- [71] W. A. Cooper, S. G. Lasher-Trapp, and A. M. Blyth, “The influence of entrainment and mixing on the initial formation of rain in a warm cumulus cloud,” *Journal of the Atmospheric Sciences*, vol. 70, no. 6, pp. 1727–1743, 2013.
- [72] A. V. Korolev, “The influence of supersaturation fluctuations on droplet size spectra formation,” *Journal of the atmospheric sciences*, vol. 52, no. 20, pp. 3620–3634, 1995.

- [73] F. Çelik and J. D. Marwitz, “Droplet spectra broadening by ripening process. part i: Roles of curvature and salinity of cloud droplets,” *Journal of the atmospheric sciences*, vol. 56, no. 17, pp. 3091–3105, 1999.
- [74] F. Yang, P. Kollias, R. A. Shaw, and A. M. Vogelmann, “Cloud droplet size distribution broadening during diffusional growth: ripening amplified by deactivation and reactivation,” *Atmospheric Chemistry and Physics*, vol. 18, no. 10, pp. 7313–7328, 2018.
- [75] R. Srivastava, “Growth of cloud drops by condensation: A criticism of currently accepted theory and a new approach,” *Journal of the atmospheric sciences*, vol. 46, no. 7, pp. 869–887, 1989.
- [76] R. A. Shaw, “Supersaturation intermittency in turbulent clouds,” *Journal of the atmospheric sciences*, vol. 57, no. 20, pp. 3452–3456, 2000.
- [77] A. Kostinski, “Simple approximations for condensational growth,” *Environmental Research Letters*, vol. 4, no. 1, p. 015005, 2009.
- [78] J. Wang, P. H. Daum, S. S. Yum, Y. Liu, G. I. Senum, M.-L. Lu, J. H. Seinfeld, and H. Jonsson, “Observations of marine stratocumulus microphysics and implications for processes controlling droplet spectra: Results from the marine stratus/stratocumulus experiment,” *Journal of Geophysical Research: Atmospheres*, vol. 114, no. D18, 2009.

- [79] A. Korolev, M. Pinsky, and A. Khain, “A new mechanism of droplet size distribution broadening during diffusional growth,” *Journal of the Atmospheric Sciences*, vol. 70, no. 7, pp. 2051–2071, 2013.
- [80] Y. Liu and P. H. Daum, “Anthropogenic aerosols: Indirect warming effect from dispersion forcing,” *Nature*, vol. 419, no. 6907, p. 580, 2002.
- [81] S. S. Yum and J. G. Hudson, “Adiabatic predictions and observations of cloud droplet spectral broadness,” *Atmospheric research*, vol. 73, no. 3-4, pp. 203–223, 2005.
- [82] Y. Liu, P. H. Daum, and S. S. Yum, “Analytical expression for the relative dispersion of the cloud droplet size distribution,” *Geophysical research letters*, vol. 33, no. 2, 2006.
- [83] C. Zhao, X. Tie, G. Brasseur, K. J. Noone, T. Nakajima, Q. Zhang, R. Zhang, M. Huang, Y. Duan, G. Li, *et al.*, “Aircraft measurements of cloud droplet spectral dispersion and implications for indirect aerosol radiative forcing,” *Geophysical research letters*, vol. 33, no. 16, 2006.
- [84] N. L. Miles, J. Verlinde, and E. E. Clothiaux, “Cloud droplet size distributions in low-level stratiform clouds,” *Journal of the atmospheric sciences*, vol. 57, no. 2, pp. 295–311, 2000.
- [85] H. Pawlowska, W. W. Grabowski, and J.-L. Brenguier, “Observations of the

- width of cloud droplet spectra in stratocumulus,” *Geophysical research letters*, vol. 33, no. 19, 2006.
- [86] M.-L. Lu, W. C. Conant, H. H. Jonsson, V. Varutbangkul, R. C. Flagan, and J. H. Seinfeld, “The marine stratus/stratocumulus experiment (mase): Aerosol-cloud relationships in marine stratocumulus,” *Journal of Geophysical Research: Atmospheres*, vol. 112, no. D10, 2007.
- [87] C. Lu, Y. Liu, S. Niu, and A. M. Vogelmann, “Observed impacts of vertical velocity on cloud microphysics and implications for aerosol indirect effects,” *Geophysical Research Letters*, vol. 39, no. 21, 2012.
- [88] J. Chen, Y. Liu, M. Zhang, and Y. Peng, “New understanding and quantification of the regime dependence of aerosol-cloud interaction for studying aerosol indirect effects,” *Geophysical Research Letters*, vol. 43, no. 4, pp. 1780–1787, 2016.
- [89] J. Chen, Y. Liu, M. Zhang, and Y. Peng, “Height dependency of aerosol-cloud interaction regimes,” *Journal of Geophysical Research: Atmospheres*, vol. 123, no. 1, pp. 491–506, 2018.
- [90] Y. Liu and J. Hallett, “On size distributions of cloud droplets growing by condensation: A new conceptual model,” *Journal of the atmospheric sciences*, vol. 55, no. 4, pp. 527–536, 1998.

- [91] Y. Liu, P. H. Daum, and J. Hallett, “A generalized systems theory for the effect of varying fluctuations on cloud droplet size distributions,” *Journal of the atmospheric sciences*, vol. 59, no. 14, pp. 2279–2290, 2002.
- [92] J. G. Hudson and G. Svensson, “Cloud microphysical relationships in california marine stratus,” *Journal of Applied Meteorology*, vol. 34, no. 12, pp. 2655–2666, 1995.
- [93] J. G. Hudson and S. S. Yum, “Droplet spectral broadening in marine stratus,” *Journal of the atmospheric sciences*, vol. 54, no. 22, pp. 2642–2654, 1997.
- [94] F. Yang, R. Shaw, and H. Xue, “Conditions for super-adiabatic droplet growth after entrainment mixing,” *Atmospheric Chemistry and Physics*, vol. 16, no. 14, pp. 9421–9433, 2016.
- [95] A. Korolev, “Limitations of the wegener–bergeron–findeisen mechanism in the evolution of mixed-phase clouds,” *Journal of the Atmospheric Sciences*, vol. 64, no. 9, pp. 3372–3375, 2007.
- [96] M. D. Shupe, S. Y. Matrosov, and T. Uttal, “Arctic mixed-phase cloud properties derived from surface-based sensors at sheba,” *Journal of the atmospheric sciences*, vol. 63, no. 2, pp. 697–711, 2006.
- [97] S. G. Cober, G. A. Isaac, and J. W. Strapp, “Characterizations of aircraft icing environments that include supercooled large drops,” *Journal of Applied Meteorology*, vol. 40, no. 11, pp. 1984–2002, 2001.

- [98] J. Curry, J. Pinto, T. Benner, and M. Tschudi, “Evolution of the cloudy boundary layer during the autumnal freezing of the beaufort sea,” *Journal of Geophysical Research: Atmospheres*, vol. 102, no. D12, pp. 13851–13860, 1997.
- [99] G. M. McFarquhar and S. G. Cober, “Single-scattering properties of mixed-phase arctic clouds at solar wavelengths: Impacts on radiative transfer,” *Journal of climate*, vol. 17, no. 19, pp. 3799–3813, 2004.
- [100] J. Verlinde, J. Y. Harrington, G. McFarquhar, V. Yannuzzi, A. Avramov, S. Greenberg, N. Johnson, G. Zhang, M. Poellot, J. H. Mather, *et al.*, “The mixed-phase arctic cloud experiment,” *Bulletin of the American Meteorological Society*, vol. 88, no. 2, pp. 205–222, 2007.
- [101] J. O. Pinto, “Autumnal mixed-phase cloudy boundary layers in the arctic,” *Journal of the atmospheric sciences*, vol. 55, no. 11, pp. 2016–2038, 1998.
- [102] I. Zawadzki, W. Szyrmer, and S. Laroche, “Diagnostic of supercooled clouds from single-doppler observations in regions of radar-detectable snow,” *Journal of Applied Meteorology*, vol. 39, no. 7, pp. 1041–1058, 2000.
- [103] A. Korolev and G. Isaac, “Phase transformation of mixed-phase clouds,” *Quarterly Journal of the Royal Meteorological Society: A journal of the atmospheric sciences, applied meteorology and physical oceanography*, vol. 129, no. 587, pp. 19–38, 2003.

- [104] A. Hill, P. Field, K. Furtado, A. Korolev, and B. Shipway, “Mixed-phase clouds in a turbulent environment. part 1: Large-eddy simulation experiments,” *Quarterly Journal of the Royal Meteorological Society*, vol. 140, no. 680, pp. 855–869, 2014.
- [105] M. Pinsky, A. Khain, and A. Korolev, “Theoretical analysis of liquid–ice interaction in the unsaturated environment with application to the problem of homogeneous mixing,” *Journal of the Atmospheric Sciences*, vol. 75, no. 4, pp. 1045–1062, 2018.
- [106] M. D. Shupe, V. P. Walden, E. Eloranta, T. Uttal, J. R. Campbell, S. M. Starkweather, and M. Shiobara, “Clouds at arctic atmospheric observatories. part i: Occurrence and macrophysical properties,” *Journal of Applied Meteorology and Climatology*, vol. 50, no. 3, pp. 626–644, 2011.
- [107] R. P. Lawson, S. Woods, and H. Morrison, “The microphysics of ice and precipitation development in tropical cumulus clouds,” *Journal of the Atmospheric Sciences*, vol. 72, no. 6, pp. 2429–2445, 2015.
- [108] Z. Sun and K. P. Shine, “Parameterization of ice cloud radiative properties and its application to the potential climatic importance of mixed-phase clouds,” *Journal of climate*, vol. 8, no. 7, pp. 1874–1888, 1995.

- [109] T. J. Garrett and C. Zhao, “Increased arctic cloud longwave emissivity associated with pollution from mid-latitudes,” *Nature*, vol. 440, no. 7085, p. 787, 2006.
- [110] A. Illingworth, R. Hogan, E. O’connor, D. Bouniol, M. Brooks, J. Delanoë, D. Donovan, J. Eastment, N. Gaussiat, J. Goddard, *et al.*, “Cloudnet: Continuous evaluation of cloud profiles in seven operational models using ground-based observations,” *Bulletin of the American Meteorological Society*, vol. 88, no. 6, pp. 883–898, 2007.
- [111] U. Lohmann, “A glaciation indirect aerosol effect caused by soot aerosols,” *Geophysical Research Letters*, vol. 29, no. 4, pp. 11–1, 2002.
- [112] A. Korolev, G. McFarquhar, P. R. Field, C. Franklin, P. Lawson, Z. Wang, E. Williams, S. J. Abel, D. Axisa, S. Borrmann, *et al.*, “Mixed-phase clouds: Progress and challenges,” *Meteorological Monographs*, vol. 58, pp. 5–1, 2017.
- [113] J. P. Fugal, T. J. Schulz, and R. A. Shaw, “Practical methods for automated reconstruction and characterization of particles in digital in-line holograms,” *Measurement Science and Technology*, vol. 20, no. 7, p. 075501, 2009.
- [114] A. V. Korolev and I. P. Mazin, “Supersaturation of water vapor in clouds,” *Journal of the atmospheric sciences*, vol. 60, no. 24, pp. 2957–2974, 2003.
- [115] D. Niedermeier, S. Hartmann, R. Shaw, D. Covert, T. Mentel, J. Schneider, L. Poulain, P. Reitz, C. Spindler, T. Clauss, *et al.*, “Heterogeneous freezing of

- droplets with immersed mineral dust particles—measurements and parameterization,” *Atmospheric Chemistry and Physics*, vol. 10, no. 8, pp. 3601–3614, 2010.
- [116] A. V. Korolev, G. A. Isaac, S. G. Cober, J. W. Strapp, and J. Hallett, “Microphysical characterization of mixed-phase clouds,” *Quarterly Journal of the Royal Meteorological Society*, vol. 129, no. 587, pp. 39–65, 2003.
- [117] P. V. Hobbs and A. L. Rangno, “Microstructures of low and middle-level clouds over the beaufort sea,” *Quarterly Journal of the Royal Meteorological Society*, vol. 124, no. 550, pp. 2035–2071, 1998.
- [118] P. Field, R. Hogan, P. Brown, A. Illingworth, T. Choularton, P. H. Kaye, E. Hirst, and R. Greenaway, “Simultaneous radar and aircraft observations of mixed-phase cloud at the 100 m scale,” *Quarterly Journal of the Royal Meteorological Society: A journal of the atmospheric sciences, applied meteorology and physical oceanography*, vol. 130, no. 600, pp. 1877–1904, 2004.
- [119] H. Pawlowska, J. Brenguier, and F. Burnet, “Microphysical properties of stratocumulus clouds,” *Atmospheric research*, vol. 55, no. 1, pp. 15–33, 2000.
- [120] F. Burnet and J.-L. Brenguier, “Observational study of the entrainment-mixing process in warm convective clouds,” *Journal of the atmospheric sciences*, vol. 64, no. 6, pp. 1995–2011, 2007.

- [121] J. L. Stith, P. V. Hobbs, and L. F. Radke, “Airborne particle and gas measurements in the emissions from six volcanoes,” *Journal of Geophysical Research: Oceans*, vol. 83, no. C8, pp. 4009–4017, 1978.
- [122] P. V. Hobbs, L. F. Radke, M. W. Eltgroth, and D. A. Hegg, “Airborne studies of the emissions from the volcanic eruptions of mount st. helens,” *Science*, vol. 211, no. 4484, pp. 816–818, 1981.
- [123] B. Mason, “The physics of clouds, 369 pp,” 1971.
- [124] A. J. Durant, R. Shaw, W. I. Rose, Y. Mi, and G. Ernst, “Ice nucleation and overseeding of ice in volcanic clouds,” *Journal of Geophysical Research: Atmospheres*, vol. 113, no. D9, 2008.
- [125] M. L. Rokicki and K. C. Young, “The initiation of precipitation in updrafts,” *Journal of Applied Meteorology*, vol. 17, no. 6, pp. 745–754, 1978.
- [126] K. C. Young *et al.*, *Microphysical processes in clouds*. Oxford University Press, 1993.
- [127] H. Jiang, W. R. Cotton, J. O. Pinto, J. A. Curry, and M. J. Weissbluth, “Cloud resolving simulations of mixed-phase arctic stratus observed during base: Sensitivity to concentration of ice crystals and large-scale heat and moisture advection,” *Journal of the atmospheric sciences*, vol. 57, no. 13, pp. 2105–2117, 2000.

- [128] U. Lohmann, “Possible aerosol effects on ice clouds via contact nucleation,” *Journal of the atmospheric sciences*, vol. 59, no. 3, pp. 647–656, 2002.
- [129] A. B. Kostinski and R. A. Shaw, “Scale-dependent droplet clustering in turbulent clouds,” *Journal of fluid mechanics*, vol. 434, pp. 389–398, 2001.
- [130] E. Bodenschatz, S. P. Malinowski, R. A. Shaw, and F. Stratmann, “Can we understand clouds without turbulence?,” *Science*, vol. 327, no. 5968, pp. 970–971, 2010.
- [131] H. Siebert, S. Gerashchenko, A. Gylfason, K. Lehmann, L. Collins, R. Shaw, and Z. Warhaft, “Towards understanding the role of turbulence on droplets in clouds: in situ and laboratory measurements,” *Atmospheric Research*, vol. 97, no. 4, pp. 426–437, 2010.
- [132] J. Lu, H. Nordsiek, E. W. Saw, and R. A. Shaw, “Clustering of charged inertial particles in turbulence,” *Physical review letters*, vol. 104, no. 18, p. 184505, 2010.
- [133] S. Risius, H. Xu, F. Di Lorenzo, H. Xi, H. Siebert, R. Shaw, and E. Bodenschatz, “Schneefernerhaus as a mountain research station for clouds and turbulence,” *Atmospheric Measurement Techniques*, vol. 8, no. 8, pp. 3209–3218, 2015.
- [134] H. Siebert, R. Shaw, J. Ditas, T. Schmeissner, S. Malinowski, E. Bodenschatz, and H. Xu, “High-resolution measurement of cloud microphysics and turbulence

- at a mountaintop station,” *Atmospheric Measurement Techniques*, vol. 8, no. 8, pp. 3219–3228, 2015.
- [135] W. A. Cooper, D. Baumgardner, and J. E. Dye, *Preprints, Conf. on Cloud Physics, Snowmass, CO, Amer. Meteor. Soc.* 1986.
- [136] D. Niedermeier, J. Voigtländer, H. Siebert, N. Desai, R. Shaw, K. Chang, S. Krueger, J. Schumacher, and F. Stratmann, *APS Meeting Abstracts*. 2017.
- [137] W. W. Grabowski and P. Vaillancourt, “Comments on preferential concentration of cloud droplets by turbulence: effects on the early evolution of cumulus cloud droplet spectra,” *J. Atmos. Sci.*, vol. 56, pp. 1433–1436, 1999.
- [138] P. Squires, “The spatial variation of liquid water and droplet concentration in cumuli,” *Tellus*, vol. 10, pp. 372–380, 1958.
- [139] B. Kumar, J. Schumacher, and R. A. Shaw, “Cloud microphysical effects of turbulent mixing and entrainment,” *Theor. Comput. Fluid Dyn.*, pp. 1–16, 2013.
- [140] S. G. Warren, C. J. Hahn, J. London, R. M. Chervin, and R. L. Jenne, “Global distribution of total cloud cover and cloud type amounts over the ocean,” tech. rep., USDOE Office of Energy Research, Washington, DC (USA). Carbon Dioxide Research Div.; National Center for Atmospheric Research, Boulder, CO (USA), 1988.

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