Droplet growth in atmospheric turbulence

A direct numerical simulation study

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Abstract

This Ph.D. thesis examines the challenging problem of how turbulence affects the growth of cloud droplets in warm clouds. Droplets grow by either condensation or collision. Without turbulence, the condensation process driven by a uniform supersaturation field is only efficient when droplets are smaller than 15 µm (radius). Gravitational collision becomes effective when the radius of droplets is larger than 50 µm. The size gap of 15–50 µm, in which neither condensation nor collision processes dominate droplet growth, has puzzled the cloud microphysics community for around 70 years. It is the key to explaining the rapid warm rain formation with a timescale of about 20 minutes. Turbulence has been proposed to bridge this size gap by enhancing droplet growth processes, and thereby, to explain rapid warm rain formation. Since cloud–climate interaction is one of the greatest uncertainties for climate models, the fundamental understanding of rapid warm rain formation may help improve climate models.

The condensational and collisional growth of cloud droplets in atmospheric turbulence is essentially the problem of turbulence-droplet interaction. However, turbulence alone is one of the unresolved and most challenging problems in classical physics. The turbulence–droplet interaction is even more difficult due to its strong nonlinearity and multi-scale properties in both time and space. In this thesis, Eulerian and Lagrangian models are developed and compared to tackle turbulence–droplet interactions. It is found that the Lagrangian superparticle model is computationally less demanding than the Eulerian Smoluchowski model.

The condensation process is then investigated by solving the hydrodynamic and thermodynamic equations using direct numerical simulations with droplets modeled as Lagrangian particles. With turbulence included, the droplet size distribution is found to broaden, which is contrary to the classical theory without supersaturation fluctuations, where condensational growth leads to progressively narrower droplet size distributions. Furthermore, the time evolution of droplet size distributions is observed to strongly depend on the Reynolds number and only weakly on the mean energy dissipation rate. Subsequently, the effect of turbulence on the collision process driven by both turbulence and gravity is explored. It is found that the droplet size distribution depends moderately on the mean energy dissipation rate, but is insensitive to the Reynolds number. Finally, the effect of turbulence on the combined condensational and collisional growth is investigated. In this case, the droplet size distribution is found to depend on both the Reynolds number and the mean energy dissipation rate. Considering small values of the mean energy dissipation rate and high Reynolds numbers in warm clouds, it is concluded that turbulence enhances the condensational growth with increasing Reynolds number, while the collision process is indirectly affected by turbulence through the condensation process. Therefore, turbulence facilitates warm rain formation by enhancing the condensation process, which predominantly depends on the Reynolds number.

Keywords: *cloud micro-physics, turbulence, inertial particles, DNS, condensation, collision, coalescence.*

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DROPLET GROWTH IN ATMOSPHERIC TURBULENCE

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To my beloved family

献给我可爱的家人

Sammanfattning

I denna avhandling utreds hur turbulens påverkar dropptillväxt i moln. Droppar kan växa genom kondensation eller kollision. Utan turbulens sker kondensationstillväxten under konstant övermättnad och är bara effektiv när dropparnas radie är mindre än 15 μm. Kollisioner som sker genom tyngdkraftens påverkan blir effektiv när dropparnas radie är större än 50 μm. Hur dropptillväxten sker i intervallet mellan dessa storlekar har förbryllat molnfysikforskare i 70 år. Att förstå detta är nyckeln till hur det kan börja regna inom ca 20 minuter från ett moln som bara har vattendroppar. Turbulens har föreslagits vara en process som kan hjälpa molndroppstillväxten och därmed förklara hur det kan regna så snabbt från ett vattenmoln. Moln är en integrerad del av klimatsystemet och klimatmodeller är känsliga för hur dessa processer är beskrivna och leder till osäkerheter. Förståelsen av hur regn bildas i moln utan iskristaller kan bidra till förbättrad processbeskrivning i klimatmodeller.

Tillväxt av molndroppar i en turbulent atmosfär, genom kondensation och kollision, kan beskrivas som frågan hur turbulens interagerar med droppar. Turbulens är ett olöst och utmanande problem inom den klassiska fysiken. Att studera droppars interaktion med turbulens är ännu mer utmanande eftersom det är ickelinjärt och sträcker sig över många skalor i både tid och rum. För att studera detta så har Euleriska och Lagrangska modeller utvecklats i denna avhandling. Tester visar att den Lagrangska superpartikelmedtoden är mindre beräkningstung än den Eulerska Smoluchowskimodellen.

Kondensationsprocessen är också studerad genom att inkludera de hydrodynamiska och termodynamiska ekvationerna i direkta numeriska simuleringar med dropparna beskrivna som Lagrangska partiklar. Storleksfördelningen av dropparna breddas genom fluktuationerna i övermättnad, utan turbulens leder kondensationsprocessen till en smalare storleksfördelning med tiden. Utvecklingen av storleksfördelningen med tiden är starkt beroende på Reynoldstalet, men bara svagt beroende på energidissipationshastigheten. Därefter studerades turbulensens effekt på kollisionsprocessen då dropparna kolliderar genom påverkan av både turbulens och tyngdkraften. I detta fall är dropparnas storleksfördelning måttligt beroende av energidissipations hastigheten men oberoende av Reynolds tal. Slutligen studeras effekten för tillväxt genom både kondensation och kollision. I detta fall är breddningen av dropparnas storleksfördelning beroende av både Reynolds tal och energidissipationshastigheten. Om vi antar ett litet värde på energidissipationshastigheten och ett högt värde på Reynoldstalet, som vi finner i vattenmoln, så är slutsatsen att turbulensen förstärker kondensationstillväxten då Reynoldstal ökar. Kollisionsprocessen är indirekt påverkad av turbulensen genom påverkan på kondensationsprocessen. Turbulens kan genom sin förstärkning av kondensationsprocessen, som mest beror på Reynoldstalet dvs de stora turbulenta skalorna, leda till snabbare bildning av regndroppar i vattenmoln.

Papers included in this thesis

The following papers, referred to in the text by their Roman numerals, are included in this thesis.

PAPER I: **Xiang-Yu Li**, A. Brandenburg, N. E. L. Haugen, and G. Svensson (2017), **Eulerian and Lagrangian approaches to multidimensional condensation and collection**, *J. Adv. Modeling Earth Systems*, **9**, 1116–1137.

PAPER II: **Xiang-Yu Li**, G. Svensson, A. Brandenburg, N. E. L. Haugen (2018), **Cloud-droplet growth due to supersaturation fluctuations in stratiform clouds**, *Atmosph. Chem. Phys.*, submitted, *arXiv:1806.10529*.

PAPER III: **Xiang-Yu Li**, A. Brandenburg, G. Svensson, N. E. L. Haugen, B. Mehlig, and I. Rogachevskii (2018), **Effect of turbulence on collisional growth of cloud droplets**, *J. Atmosph. Sci.*, in press, *arXiv:1711.10062*.

PAPER IV: **Xiang-Yu Li**, B. Mehlig, G. Svensson, A. Brandenburg, and N. E. L. Haugen (2018), **Fluctuations and growth histories of cloud droplets: superparticle simulations of the collision-coalescence process**, *Phys. Rev. E.*, to be submitted, *DiVA:diva2:1237427*.

PAPER V: **Xiang-Yu Li**, A. Brandenburg, G. Svensson, N. E. L. Haugen, B. Mehlig, and I. Rogachevskii (2018), **Condensational and collisional growth of cloud droplets in a turbulent environment**, *J. Atmosph. Sci.*, under review, *arXiv:1807.11859*.

The following paper is not included in this thesis:
A. Brandenburg, N. E. L. Haugen, **Xiang-Yu Li**, and K. Subramanian (2018), Varying the forcing scale in low Prandtl number dynamos, *Month. Not. Roy. Astron. Soc.* **479**, 2827–2833.

Author’s contribution

The project started from scratch. I am one of the core members of the project and actively involved in all the developments, discussions, and simulations, having designed all of them either by myself or in consultation with coauthors.

Paper **I** is the start of the project. I implemented the Lagrangian algorithm and improved the Eulerian algorithm with inputs from coauthors. I wrote the main part of the text and revised the other parts written by coauthors. I performed all the simulations and produced most of the plots. I conducted all the revisions from the reviewers with input from coauthors.

The ideas of Paper **II**, **III**, and **V** were developed by myself. I implemented the modules for Paper **II** from scratch. I designed all the simulations with input from coauthors. I conducted all the simulations and produced all the plots. I wrote the manuscripts with comments from coauthors. I conducted all the revisions from the reviewers with inputs from coauthors.

Paper **IV** is initiated from the primary results of Paper **III** during a visit to Bernhard Mehlig. I implemented the history-tracking mechanism, designed, and conducted all the simulations with input from coauthors. I wrote the manuscript with comments from coauthors.

All my additions to the PENCIL CODE are in the public domain and can now be utilized by others under `https://github.com/pencil-code`.

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1. Introduction

1.1 Cloud micro-physics

When viewed from space, about 70% of Earth's surface is covered by clouds (Schneider et al., 2017). Clouds, the regulator of the radiative heating of the planet (Ramanathan et al., 1989), represent a major complication in the current modeling of the climate system (Stevens and Bony, 2013; Bony et al., 2017; Schneider et al., 2017). One of the most challenging problems of cloud–climate interactions is to understand how cloud microscopic processes affect macroscopic properties, such as precipitation efficiency and radiative properties (Shaw, 2003), which largely depend on the physical mechanisms of cloud-particles formation (Shaw, 2003; Schneider et al., 2017). Therefore, it is fundamentally important to understand the temporal and spatial variation of the cloud droplet (Shaw, 2003; Grabowski and Wang, 2013) in a highly turbulent environment.

1.2 Turbulence–droplet interactions in clouds

Observations reveal that warm clouds are highly turbulent, which are characterized by large Reynolds numbers ($\mathrm{Re}_\lambda \approx 10^4$) and relatively small mean energy dissipation rates ($\bar{\varepsilon} \approx 10^{-3}\,\mathrm{m^2 s^{-3}}$; see Siebert et al., 2006a). With this Reynolds number, turbulence exhibits multi-scale interactions with energy transfers from energy injection scales ($\sim 100\,\mathrm{m}$) to the smallest scales in three-dimensional (3-D) turbulence. Thus, it affects the cloud micro-physics from large to small scales. Since the typical size[1] ($\sim 10\,\mu\mathrm{m}$) of cloud droplets is about 100 times smaller than the Kolmogorov length scale ($\sim 1\,\mathrm{mm}$) in clouds, droplet dynamics and droplet-droplet interactions are influenced by the smallest scales of turbulence. Cloud droplets transported and dispersed by turbulence are inertial particles because of the large mass density ratio between liquid water and the dry air, due to which trajectories of droplets deviate from that of tracers. This makes the droplet dynamics and droplet-droplet interaction more complicated than the small-scale dynamics of turbulence alone. Cloud thermodynamics is influenced by the largest scales of turbulence, which affects the latent heat release due to the evaporation of droplets (Li et al., 2018c). The latent heat release in turn influences the motion of turbulence. Therefore, turbulence–droplet

[1] In this thesis, size of cloud droplets is in radius.

interactions are multi-scale, which leads to the coupling of cloud macro-physics and micro-physics.

1.3 Effect of turbulence on warm rain formation

In warm clouds, the typically observed timescale for rain formation is about 20 minutes (Stephens and Haynes, 2007), which is significantly shorter than the theoretically predicted timescale of about 8 hours according to the Saffman-Turner collision model (Saffman and Turner, 1956). Naturally, turbulence was proposed to explain the rapid warm rain formation (Saffman and Turner, 1956; Shaw, 2003; Bodenschatz et al., 2010; Devenish et al., 2012; Grabowski and Wang, 2013), which is fundamentally the question of how turbulence interacts with cloud droplets. The topic of this thesis is to explore how turbulence influences the development of droplet size distributions in warm clouds.

2. Physical mechanisms and models

The droplet size distribution $f(r,\boldsymbol{x},t)$ is the key and most challenging quantity in cloud microphysics (Shaw, 2003). It is often described by two numerical models originated from statistical physics: the Eulerian and the Lagrangian model. The Eulerian model treats $f(r,\boldsymbol{x},t)$ as a field, which is described in a mean-field manner, i.e., the particle density is assumed to be spatially uniform (Pumir and Wilkinson, 2016). The Lagrangian model handles $f(r,\boldsymbol{x},t)$ either in a deterministic or a stochastic fashion. In this chapter, the motion of turbulence and droplets, and the physical processes that determine $f(r,\boldsymbol{x},t)$ for raindrop formation are introduced. Descriptions of the two numerical models used in this thesis are then followed.

2.1 Turbulence

2.1.1 Momentum equation

The motion of fluid is governed by the Navier-Stokes equations (Pope, 2001),

$$\frac{D\boldsymbol{u}}{Dt} = -\rho^{-1}\boldsymbol{\nabla} p + \rho^{-1}\boldsymbol{\nabla}\cdot(2\nu\rho\mathsf{S}) + \boldsymbol{f}, \tag{2.1}$$

$$\frac{\partial\rho}{\partial t} + \boldsymbol{\nabla}\cdot(\rho\boldsymbol{u}) = 0, \tag{2.2}$$

where $D/Dt = \partial/\partial t + \boldsymbol{u}\cdot\boldsymbol{\nabla}$ is the material derivative, $\boldsymbol{f}$ is the forcing, ν is the kinematic viscosity of air, $\mathsf{S}_{ij} = \frac{1}{2}(\partial_j u_i + \partial_i u_j) - \frac{1}{3}\delta_{ij}\boldsymbol{\nabla}\cdot\boldsymbol{u}$ is the traceless rate-of-strain tensor (Li et al., 2017), p is the gas pressure, and ρ is the gas density obeying the equation of state:

$$p = \rho c_{\mathrm{s}}^2/\gamma, \tag{2.3}$$

where $\gamma = c_{\mathrm{p}}/c_{\mathrm{v}} = 7/5$ is the ratio between specific heats at constant pressure and constant volume, c_{p} and c_{v}, respectively. The sound speed c_s is set to a small value to render the flow nearly incompressible. A flow becomes turbulent when the Reynolds number $\mathrm{Re} \gg 1$, which is defined as $\mathrm{Re} \equiv UL_f/\nu$. Here U and L_f are the velocity and length scales where the fluid is forced, respectively. The Reynolds number is used to characterize the ratio between the nonlinear term $(\boldsymbol{u}\cdot\boldsymbol{\nabla})\boldsymbol{u}$ and the viscosity term $\boldsymbol{\nabla}\cdot(2\nu\rho\mathsf{S})$. In this thesis, the Taylor-scale

Reynolds number, defined as $\mathrm{Re}_\lambda \equiv u_{\mathrm{rms}}^2 \sqrt{5/(3\nu\bar{\varepsilon})}$, is adopted to measure the intensity of 3-D turbulence, where u_{rms} is the rms turbulent velocity, and $\bar{\varepsilon} = 2\nu \overline{\mathrm{Tr}\,\mathsf{S}_{\mathrm{ij}}\mathsf{S}_{\mathrm{ji}}}$ is the mean energy-dissipation rate per unit mass and Tr denotes the trace. At high Re_λ, analytical solutions of the Navier-Stokes equations have not yet been found (Pope, 2001). Therefore, direct numerical simulations have become an essential tool for studying turbulence.

2.1.2 Energy cascade in turbulence

The classic understanding of turbulence is based on the phenomenological Kolmogorov theory (Kolmogorov, 1941), which introduced the notion that small eddies are almost memoryless of the history of the flow. Under this assumption, Kolmogorov argued that a fully-developed, steady, and homogeneous flow can be described by a single quantity $\bar{\varepsilon}$ over a wide range of length scales (termed as the inertial range of turbulence). This results in the direct energy cascade of turbulence, i.e., energy is transferred from large eddies to small eddies in 3-D hydrodynamic[1] turbulence. The largest scale in 3-D turbulence is the turbulent integral length scale (energy injection scale) and the smallest scale is the Kolmogorov length scale. Energy cascades from the integral length scale to the Kolmogorov length scale, then dissipates due to viscosity. The Kolmogorov theory assumes that the energy dissipation rate is isotropic, which was, however, found to be anisotropic (She and Leveque, 1994). This is a phenomenon termed as intermittency in the turbulence-community and small-scale intermittency in meteorological context (Mahrt, 1989). Kolmogorov theory with the intermittency constitutes the state-of-art understanding of turbulence.

2.2 Motion of particles in fluids

Determining the motion of particles in flow is in general a difficult task (Pumir and Wilkinson, 2016) due to the nonlinear interactions between particles and fluid. It is also affected by the geometry of particles. Cloud droplets are small compared with the Kolmogorov length scale $\eta = (\nu^3/\bar{\varepsilon})^{1/4}$ and have a large mass density contrast with the air as discussed in Chapter 1.2. Thus, its motion is only subjected to the viscous drag and gravity settling. Each droplet is treated as a Lagrangian point-particle, where one solves for the particle position $\boldsymbol{x}_i$,

$$\frac{d\boldsymbol{x}_i}{dt} = \boldsymbol{V}_i, \tag{2.4}$$

[1] Energy can transfer from small eddies to large eddies in magnetic hydrodynamic turbulence in the presence of magnetic helicity due to its conservation property.

and its velocity $\boldsymbol{V}_i$ via

$$\frac{d\boldsymbol{V}_i}{dt} = \frac{1}{\tau_i}(\boldsymbol{u} - \boldsymbol{V}_i) + \boldsymbol{g}, \tag{2.5}$$

where i is the index of particles. Here, $\boldsymbol{u}$ is the fluid velocity at the position of the droplet, $\boldsymbol{g}$ is the gravitational acceleration, and τ_i is the inertial response (or stopping) time given by

$$\tau_i = 2\rho_l r_i^2/[9\rho_a \nu D(\mathrm{Re}_i)], \tag{2.6}$$

where $\rho_l = 1000\,\mathrm{kg}\cdot\mathrm{m}^{-3}$ is the mass density of liquid water and $\rho_a = 1\,\mathrm{kg}\cdot\mathrm{m}^{-3}$ is the reference mass density of dry air. The correction factor (Schiller and Naumann, 1933; Marchioli et al., 2008)

$$D(\mathrm{Re}_i) = 1 + 0.15\,\mathrm{Re}_i^{2/3} \tag{2.7}$$

is used to approximate the effect of non-zero particle Reynolds number $\mathrm{Re}_i = 2r_i|\boldsymbol{u} - \boldsymbol{V}_i|/\nu$.

The motion of inertial particles is determined by a single parameter, the Stokes number, which is defined as the ratio between the particle response time τ_i and the Kolmogorov time scale $\tau_\eta = (\nu/\bar{\varepsilon})^{1/2}$: $\mathrm{St} = \tau_i/\tau_\eta$. When $\mathrm{St} \ll 1$, particles behave like tracers; when $\mathrm{St} \geq 1$, the trajectories of particles deviate from that of tracers.

2.3 Condensation

When a cloud droplet is exposed to a supersaturated environment, it grows due to a net water vapor flux towards its surface by diffusion (Pruppacher and Klett, 2012). The flux is coupled by conservation of energy, conservation of mass, and the Clausius-Clapeyron equation (Shaw, 2003), resulting in the condensational growth law (Pruppacher and Klett, 2012),

$$\frac{\mathrm{d}r^2}{\mathrm{d}t} = 2Gs(\boldsymbol{x},t), \tag{2.8}$$

where $s(\boldsymbol{x},t)$ is the supersaturation, and G (with unit of $\mathrm{m}^2\mathrm{s}^{-1}$) is a thermodynamical parameter that weakly depends on temperature and pressure (Lamb and Verlinde, 2011a). From Equation (2.8), one obtains $dr/dt = Gs/r$, namely, the growth rate of cloud droplets is inversely proportional to the radius, suggesting that larger droplets grow slower than smaller ones. Condensational growth narrows the droplet size distribution under the assumption that s is positive and uniform, which results in a diffusive behavior $r \sim t^{1/2}$. However, supersaturation fields in clouds are spatially and temporally fluctuating, which are determined by the temperature $T(\boldsymbol{x},t)$ and water vapor mixing ratio $q_v(\boldsymbol{x},t)$ transported by turbulence. Turbulent motion is in turn affected by the buoyancy force due to condensation and evaporation of cloud droplets. For details of the model, see Li et al. (2018c). The effect of supersaturation fluctuations on condensational growth will be discussed in Chapter 3.2.

2.4 Collision

To form raindrops, smaller droplets need to grow from around $2\,\mu$m to a critical size such that it can fall out of the cloud. This critical size depends on the type of clouds, that gives differences in the vertical velocity of the uplifting parcel. For example, it is around $100\,\mu$m in stratocumulus clouds. Condensation dominates the growth in the size range of around 2–15 μm (Lamb and Verlinde, 2011a). However, as discussed in Section 2.3, the condensational growth is too slow. Therefore, collection, another microscopical mechanism, has been proposed to explain the rapid formation of raindrops (Yau and Rogers, 1996). The collection process consists of collision and coalescence.

2.4.1 Collision rate

The collision rate between a particle with radius r and other particles with radius r' is given by (Saffman and Turner, 1956),

$$R_c(r) = \int_0^\infty K(r,r')f(r')dr'E_c, \tag{2.9}$$

where $K(r,r')$ is the collision probability (often termed as the collision kernel) of two colliding particles. The collision rate R_c has a dimension of inverse time:$[R_c] = T^{-1}$. To obtain R_c, the preliminary task is to determine the collision kernel $K(r,r')$ with dimension of L^3T^{-1}. The collision rate between two spherical particles is the rate a particle crosses the spherical surface (Sundaram and Collins, 1997). Thus, $K(r,r')$ is proportional to the surface area $4\pi(r+r')$ and the radial relative velocity $\langle|\Delta w|\rangle$. If particles tend to cluster, $K(r,r')$ would be proportional to the two-point correlation function $g(r+r')$ characterizing the clustering effect (Reade and Collins, 2000; Gustavsson and Mehlig, 2016). When two droplet collide with each other, the droplet-droplet aero-hydrodynamics (Wang et al., 2005, 2007; Wang and Grabowski, 2009; Chen et al., 2018) reduce the collision rate, which can be taken into account by introducing the collision efficiency E_c. Thus, $K(r,r')$ can be expressed as (Sundaram and Collins, 1997),

$$K(r,r') = \frac{1}{2}4\pi(r+r')^2 g(r+r')\,\langle|\Delta w|\rangle\,E_c. \tag{2.10}$$

In this thesis, the collision efficiency E_c is assumed to be unit, which may overestimate the collision rate. The collision efficiency is difficult to tackle since it is a combination of particle dynamics and deformation of droplets. Comprehensive investigations of the collision efficiency in a turbulent environment by laboratory experiments are appealing.

2.4.2 Continuous collision from different terminal fall velocities

The most classical mechanism causing collisions among cloud droplets is gravity-generated collision (Lamb and Verlinde, 2011a; Li, 2016). Consider the following continuous collision process that two particles with different size (r_L and r_S) settle in still air due to gravity. There is no clustering, so $g(r+r')=1$. Also, the geometry is reduced from 3-D to 2-D. Therefore, Equation (2.10) can be written as,

$$K(r_L, r_S) = \pi(r_L + r_S)^2|V_L - V_S|E_c, \tag{2.11}$$

where V_L and V_S are the terminal velocity of the larger droplet and the smaller droplet, respectively. When $r_L \gg r_S$, the collision rate can be simplified as $K(r_L, r_S) = \pi r_L^2|V_L|$ assuming $E_c = 1$. The descent speed of the cloud droplet is roughly proportional to its size if the droplet remains spherical, i.e. $|V_L| \sim r_L$ (Lamb and Verlinde, 2011b; Li et al., 2018b). The mass growth rate of the collector $\mathrm{d}m_\mathrm{d}/\mathrm{d}t$ is proportional to K. Thus, $\mathrm{d}m_\mathrm{d}/\mathrm{d}t \approx \pi r_L^3$, giving an exponential growth behavior: $r(t) \sim \exp(\alpha t)$ (Pruppacher and Klett, 2012), where α is a constant.

2.4.3 Stochastic collision driven by gravity and turbulence

Continuous collisions become effective only if there are large enough collectors (Lamb and Verlinde, 2011b). To start the process, a triggering collector is needed, which is referred to as the "lucky" droplet (Telford, 1955; Kostinski and Shaw, 2005). Where do the lucky ones come from? The "lucky" droplet is assumed to emerge from the stochastic collision process (Yau and Rogers, 1996; Li, 2016). Recall that the Reynolds number of cloud-like turbulence is $\mathrm{Re}_\lambda \approx 10^4$. This is rather large, so one expects turbulence to play an important role in the stochastic collision process (Li, 2016). For example, the vigorous turbulence eddies may facilitate the formation of lucky droplets (Kostinski and Shaw, 2005). A more detailed discussion of the collisional growth in turbulence and gravity is presented in Chapter 3.3.

2.5 Eulerian model

The Eulerian model is widely used in both meteorological (Saffman and Turner, 1956; Lamb and Verlinde, 2011a; Pruppacher and Klett, 2012) and astrophysical (Drazkowska et al., 2014) contexts to simulate the condensation and collision processes (Li, 2016). The evolution of the droplet size distribution $f(r)$ is governed by the continuity equation with additional coupling terms due to condensation and collision (Li et al., 2017),

$$\frac{\partial f}{\partial t} + \boldsymbol{\nabla} \cdot (f\boldsymbol{v}) + \nabla_r(fC) = \mathcal{T}_{\text{coll}} + D_{\text{p}}\nabla^2 f, \tag{2.12}$$

where $\nabla_r = \partial/\partial r$ is the derivative with respect to r, $C \equiv \mathrm{d}r/\mathrm{d}t = Gs/r$, as given in Equation (2.8), and $\mathcal{T}_{\text{coll}}$ describes the change of the number density of particles for smaller and larger radii (Li et al., 2017), as will be defined below. Furthermore, $\boldsymbol{v}(\boldsymbol{x}, r, t)$ is the particle velocity within the resolved grid cell, which is governed by the momentum equation of particles. The diffusion term $D_{\text{p}}\nabla^2 f$ is for numerical stability, where D_{p} is the artificial viscosity.

The spatial distribution of cloud droplets varies dramatically because of collisions. For a given number density of cloud droplets with different size, the collision process yields an increasing number of larger cloud droplets and a decreasing number of smaller ones (Li, 2016). Thus the rate of change of the number density can be expressed by the gain of larger cloud droplets and the loss of smaller ones (mass conservation), which is referred to as the Smoluchowski equation (a form of Boltzmann transport equation) (Von Smoluchowski, 1916),

$$\begin{aligned}\mathcal{T}_{\text{coll}} = \tfrac{1}{2}\int_0^m & K(r-r', r')\, f(r-r')\, f(r')\, \mathrm{d}r' \\ & - \int_0^\infty K(r, r')\, f(r)\, f(r')\, \mathrm{d}r'.\end{aligned} \tag{2.13}$$

The Smoluchowski equation will be revisited in Chapter 3.3.3.

2.6 Lagrangian model

In the Lagrangian model, each individual droplet is tracked. Compared with the Eulerian model, Lagrangian modeling of collisions of inertial particles is closer to the nature of real physical collisions in the sense that it takes fluctuations into account (Li, 2016). As will be discussed in Chapter 3.3.3, fluctuations are significantly important for the collisional growth of cloud droplets, which, however, may not be captured by the Eulerian model. Nevertheless, the direct Lagrangian-detected collision approach is computationally demanding. Therefore, a Monte Carlo-type (Bird, 1978, 1981; Jorgensen et al., 1983) Lagrangian

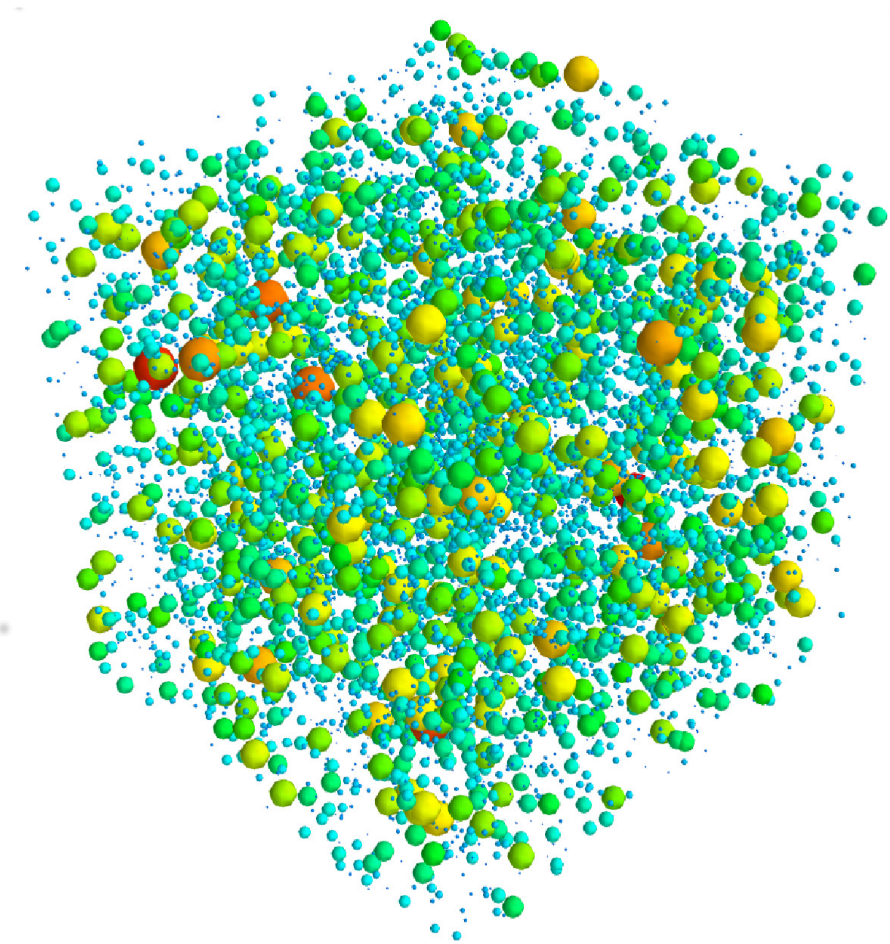

Figure 2.1: A snapshot of the spatial distribution of droplets from 3-D turbulence simulation of Paper **III** (Li et al., 2018b). Darker and redder color represent larger droplets.

tracking approach is employed to model collisions between numerical superparticles, which is a statistical approach to represent physical particles (Zsom and Dullemond, 2008). For details of the superparticle approach, see Li et al. (2017) and Li et al. (2018b). The superparticle approach is advantageous over the direct Lagrangian-detected collision approach in the following perspectives. First, it is technically easier to implement the collision processes and numerically less demanding compared with the direct Lagrangian-detected collision approach since collision only happens if two superparticles reside in the same grid cell (Li, 2016). This avoids handling with colliding pairs between the two neighboring numerical grid cells. Second, the superparticle approach is able to deal with large domain sizes, allowing us to follow collisions together with hydrodynamics in a larger domain at a moderate computational cost (Johansen et al., 2012), which also makes it easy to be adapted to large-eddy simulations. Figure 2.1 shows a snapshot of droplets simulated in 3-D turbulence.

In Paper **I**, the Eulerian approach is compared with two Lagrangian approaches. Good mutual agreement of the droplet size distribution is found for both condensational growth and collisional growth. The Lagrangian schemes are found to be superior to the Eulerian model. Therefore, the Lagrangian model is mainly used in this thesis.

3. Growth of cloud droplet in a turbulent environment

In this chapter, a state-of-art summary of the growth of cloud droplet in a turbulent environment is presented.

3.1 Observations of droplet size distributions

Observations provide us realistic information about cloud micro-physics, which also pose challenging problems about the mechanisms for the growth of droplets. Among all the challenging problems, effect of turbulence on condensational and collisional growth are the most notorious but significant ones (Shaw, 2003; Devenish et al., 2012; Grabowski and Wang, 2013).

Observing the droplet size distribution is challenging due to the fact that cloud droplets are around $10\,\mu$m, which requires high spatial and temporal resolutions to be observed. The Airborne Cloud Turbulence Observation System provides the first relatively accurate measurement of the small scales of cloud turbulence and the droplet size distribution (Grabowski and Wang, 2013), which showed that the small-scale properties of cloud-turbulence obey the Kolmogorov flow with intermittency (Siebert et al., 2006b) and the droplet size distribution is affected by the entrainment (Lehmann et al., 2009). A more accurate measurement by Beals et al. (2015) demonstrated that the local droplet size distribution is strongly inhomogeneous. Siebert and Shaw (2017) observed droplets with diameter up to $20\,\mu$m in shallow cumulus clouds, while the adiabatic diameter is about $10\,\mu$m. They attributed this to supersaturation fluctuations, whose standard deviation is on the order of 1%. Observing the dynamical collision process is not feasible.

Compared with observations, laboratory studies of condensational and collisional growth of cloud droplets are still in a primary stage. Chandrakar et al. (2016) measured the effect of aerosol concentration on droplet size distributions in a laboratory cloud-chamber, where moist convection was generated. They found that higher aerosol concentrations result in a narrow droplet size distribution and lower aerosol concentrations lead to a wider droplet size distribution. Nevertheless, how different turbulence intensities affect supersaturation fluctuations and therefore the broadening of cloud droplet is still unknown. Rapid development of the particle velocimetry tracking method has advanced our understanding of the Lagrangian properties of turbulence (Toschi and Bodenschatz, 2009). However, detecting collisions of cloud droplets in turbulence has not been achieved so far (Pumir and Wilkinson, 2016).

Overall, numerical simulations of condensation and collision in turbulence become significantly important due to the difficulties in investigating them by observations or laboratory experiment. As discussed in Chapter 1, turbulence–droplet interaction in turbulence is a strong nonlinear and multi-scale problem. To resolve into the smallest scales, direct numerical simulations (DNS) are required, which is challenging using the most powerful supercomputers hitherto due to the large range of scales and a numerous number ($\sim 10^8$) of droplets involved. Here DNS is the numerical method of solving the nonlinear physical equations directly without adopting any sub-grid models, and thus to resolve the dissipation scale of turbulence (Li, 2016). DNS help us understand the cloud microphysics from first principles. However, the Reynolds number reached in the DNS is still two orders of magnitude smaller than the one in clouds. In this thesis, the large scales refer to scales close to the integral length scale in DNS.

3.2 Condensational growth of cloud droplets

As discussed in Chapter 2.3, the condensational growth is influenced by turbulence. On the other hand, turbulence is affected by the buoyancy force due to the latent heat release of cloud droplets. The coupling of turbulence, droplet dynamics, and the thermodynamics renders the condensational growth untouchable by the analytical analysis. Well-controlled laboratory experiments are not feasible so far. Therefore, numerical simulations became an essential tool.

Nonuniform supersaturation for condensational growth of cloud droplets was first recognized by Srivastava (1989). Vaillancourt et al. (2002) conducted the fist DNS study of the condensational growth and found that the effect of turbulence is negligible. This is contrary to what others (Paoli and Shariff, 2009; Lanotte et al., 2009; Sardina et al., 2015; Siewert et al., 2017; Grabowski and Abade, 2017) observed that droplet size distributions broaden due to supersaturation fluctuations resulted from turbulence.

To tackle the contradiction among different DNS results, Paper **II** scrutinizes the effect of turbulence on condensational growth by solving the complete set of thermodynamic equations governing the supersaturation field. It is shown that droplet size distributions broaden dramatically with increasing Reynolds number and decrease slightly with increasing mean energy dissipation rate (Figure 3.1, in which r_{ini} is the initial radius of droplets). Therefore, Paper **II** suggests that the contradiction between the results of Vaillancourt et al. (2002) and others (Paoli and Shariff, 2009; Lanotte et al., 2009; Sardina et al., 2015; Siewert et al., 2017) could be due to the fact that the scale separation is too small in the DNS of Vaillancourt et al. (2002). More importantly, Paper **II** finds that the standard deviation of the surface area of cloud droplets increases as $t^{1/2}$,

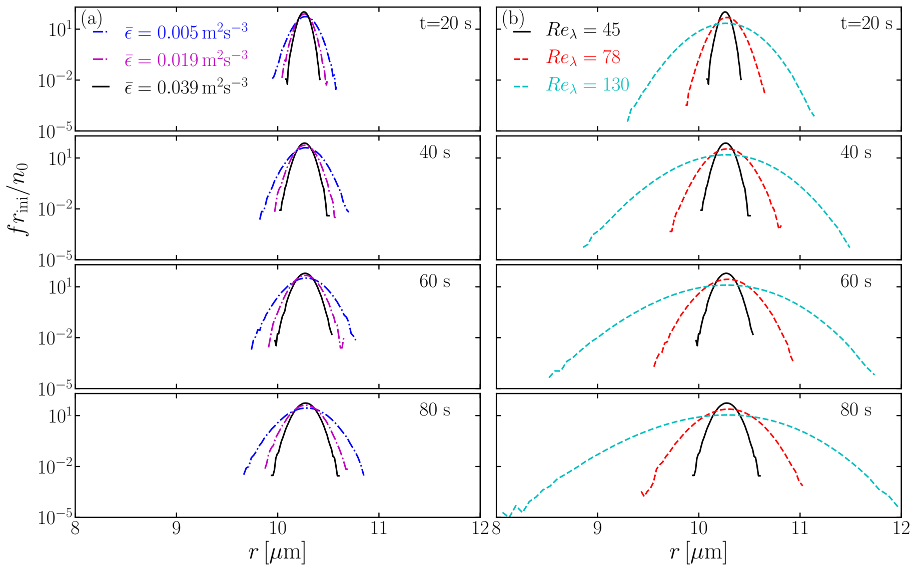

Figure 3.1: Time evolution of droplet size distributions resulted from condensation in different turbulence configurations; adapted from Paper **II** (Li et al., 2018c).

which is consistent with the Lagrangian stochastic model (Sardina et al., 2015).

3.3 Collisional growth of cloud droplets

The classical gravity-generated collisional growth of cloud droplets discussed in Chapter 2.2.1 was found to be inefficient to overcome the growth barrier. This is the main topic of Paper **III**. We return to computational aspects of the collisional growth of cloud droplets as part of a broader study later in Paper **IV**.

As discussed in Chapter 2.2, the dynamics of inertial particles is determined by the single parameter, the Stokes number, which determines the collision rate as well. When the inertia of particles is very small, i.e., St $\ll 1$, they are essentially advected by the flow. In this case, the collision rate is strongly influenced by turbulent shear. Saffman and Turner (1956) were the first to model the collision rate of small particles advected by turbulent shear. The key idea of the Saffman-Turner model is that the collision rate is dominated by small scales of turbulence (shearing motion) since the size of cloud droplets is three orders of magnitude smaller than the Kolmogorov length. Using dimensional analysis, Saffman-Turner suggested the following mean collision rate according to Equation (2.9),

$$\bar{R}_c = \frac{\sqrt{8\pi/15}\, n_0\, (2r)^3}{\tau_\eta}\,, \tag{3.1}$$

when there is no intermittency and the particle inertia is small. Giving $\tau_\eta = 0.04\,\mathrm{s}$, $n_0 = 1\times 10^8\,\mathrm{m}^{-3}$ (initial mean number density of cloud droplets), and $r = 10\,\mu\mathrm{m}$, we obtain $\bar{R}_c \approx 2.6\times 10^{-5}\,\mathrm{s}^{-1}$, which is negligible (Wilkinson, 2016). However, turbulence is highly intermittent at small scales and cloud droplets are inertial particles. When the inertia becomes significant, the collision rate is increased due to clustering (Maxey, 1987; Sundaram and Collins, 1997) and caustics (Falkovich et al., 2002; Wilkinson et al., 2006). Clustering (preferential concentration) is a phenomenon that inertial particles accumulate at the low vorticity region due to the centrifugal force (Maxey, 1987; Gustavsson and Mehlig, 2016), which becomes pronounced when $St \approx 0.6$ (Bec et al., 2007). Caustics (sling effect) are the singularities in the droplet dynamics giving rise to multi-valued droplet velocities, resulting in large velocity differences between nearby droplets (Wilkinson et al., 2006; Gustavsson and Mehlig, 2014; Li et al., 2018b). The caustics becomes efficient when $St \geq 0.3$ (Voßkuhle et al., 2014). However, $St \approx 0.02$ for $10\,\mu$m-sized droplet, suggesting that clustering and caustics are inefficient to affect the collisional growth of cloud droplets (Wilkinson, 2016). How does turbulence affect the collisional growth of cloud droplets?

Telford (1955) proposed the role of fluctuations on collisional growth, "*that for drops beginning growth at twice the volume of their neighbors, random fluctuations in the times taken for different drops to effect captures can lead to the formation of the complete raindrop in time shorter than that required for growth to raindrop size by the continuous growth processes*". Kostinski and Shaw (2005) developed a model (referred to as the lucky-droplet model) to explain how the fluctuations can overcome the growth barrier. Using large deviation theory, Wilkinson (2016) demonstrated theoretically that the cumulative collision time is a small fraction of the mean collision time. However, neither Kostinski and Shaw (2005) nor Wilkinson (2016) addressed explicitly the role of turbulence in random fluctuations of collisional growth (Li et al., 2018b).

As part of this thesis, collisional growth in a combined turbulence and gravity environment using DNS are investigated. The obtained droplet size distributions from DNS are a result of both stochastic and continuous collisions. It is indeed artificial to distinguish between stochastic and continuous collisions. Nevertheless, disentangling them helps us better understand the collisional growth in the state-of-art theoretical framework.

3.3.1 Collision

As a first step towards understanding the collisional growth of cloud droplets, collision without coalescence has been investigated intensively. Due to the lack of satisfactory theoretical prediction of collision and inaccessible laboratory experiment about collision, numerical simulation has again become favorable. The turbulence effect on the geometrical collision kernel was investigated by Franklin et al. (2005), Ayala et al. (2008a), Rosa et al. (2013) and Chen et al. (2016). Ayala et al. (2008b) developed a comprehensive parameterizations of the turbulent collision rate and concluded that turbulence enhances the geometrical collision rate with increasing energy dissipation rate. They also found that the dependence of the collision rate on the Reynolds number is minor. Rosa et al. (2013) and Chen et al. (2016) confirmed the secondary dependency of the collision statistics on the Reynolds number.

3.3.2 Collision-coalescence

Droplets grow by collision and coalescence, which is more complicated than the pure collision process due to the unresolved mechanism of coalescence and the dynamical Stokes number. First, the physical mechanism of coalescence is not well-established. When two droplets get close and contact with each other, they may deform depending on the size, shape, and relative velocity of the two colliding droplets. Also, the fluid trapped between them leads to a lubrication film (Pumir and Wilkinson, 2016). The deformation and lubrication film can be counted into the coalescence efficiency. Unit coalescence efficiency has been widely used due to the fact that the Weber number is only about 10^{-2}, defined as the ratio between the droplet inertia and its surface tension (Perrin and Jonker, 2015). However, more robust laboratory experiments are needed to investigate the dynamical process of coalescence so that a realistic coalescence efficiency can be obtained. Second, growth of cloud droplets leads to different Stokes numbers, which makes the collision process more complicated.

We first review the DNS studies of the collision-coalescence processes, after which, discussions of the lucky-droplet model and the Smoluchowski equation follow.

DNS

The Smoluchowski equation discussed in Chapter 2.5 has been regarded as the master equation to model the collision-coalescence process for both cloud droplets (Yau and Rogers, 1996) and planet formation (Johansen and Lambrechts, 2017). DNS has been widely used to solve the Smoluchowski equation. The outcome by solving the Smoluchowski equation largely depends on the collision kernel. However, there is no analytical formula for the collision kernel. Therefore, most of the DNS work first obtained the geometrical collision kernel of colliding pairs. Then, the parameterized collision kernel was used in the Smoluchowski equation (Equation (2.13)). For example, Franklin (2008) investigated collision-coalescence processes by solving the Smoluchowski equation together with the Navier-Stokes equation using DNS and found that the droplet size distribution is significantly enhanced by turbulence with increasing $\bar{\varepsilon}$. Using a similar approach, Xue et al. (2008) and Wang and Grabowski (2009) concluded that turbulence ($\bar{\varepsilon}$) enhances the collisional growth and the dependence on Reynolds number is uncertain due to its small value in their simulations. Onishi and Seifert (2016) updated the collision kernel model of Wang and Grabowski (2009) and found that the collisional growth of cloud droplets also depends on the Reynolds number.

In Paper **III**, to quantify the role of small-scale turbulence on the time evolution of droplet size distributions, the collision-coalescence process are investigated using DNS. Each droplet is tracked in a Lagrangian manner. The droplet size distribution is determined directly from numerical simulations, thus avoiding the use of a parameterized kernel. It is found that the time evolution of droplet size distributions is enhanced moderately with increasing mean energy dissipation rate $\bar{\varepsilon}$ (Figure 3.2, in which Sv is the non-dimensional terminal velocity defined as the ratio between the terminal velocity of the droplet and the Kolmogorov velocity). Its dependency on Reynolds number is weak in the range explored.

The lucky-droplet model

The DNS studies provide a comprehensive investigation of the collisional growth. However, it is unclear if the broadening of droplet size distributions results from the effect of turbulence on the mean collision rate or on fluctuations of collision, which is explored in Paper **IV**.

Assume that $10\,\mu$m-sized cloud droplets with $n_0 = 10^8\,\mathrm{m}^{-3}$ are randomly distributed. One larger droplet with radius $12.6\,\mu$m (twice the mass of $10\,\mu$m-sized droplets) falls through the $10\,\mu$m-sized droplets due to gravity. Tracing the collision sequences, the collision time intervals t_k of the larger droplet is assumed to follow an exponential distribution with a mean collision rate λ_k,

$$p_k(t_k) = \lambda_k \exp(-\lambda_k t_k), \tag{3.2}$$

where k denotes the k-th collision. The cumulative time $\mathscr{T}$ of collisions can then be expressed as

$$\mathscr{T} = \sum_{k=1}^{\mathscr{N}} t_k. \tag{3.3}$$

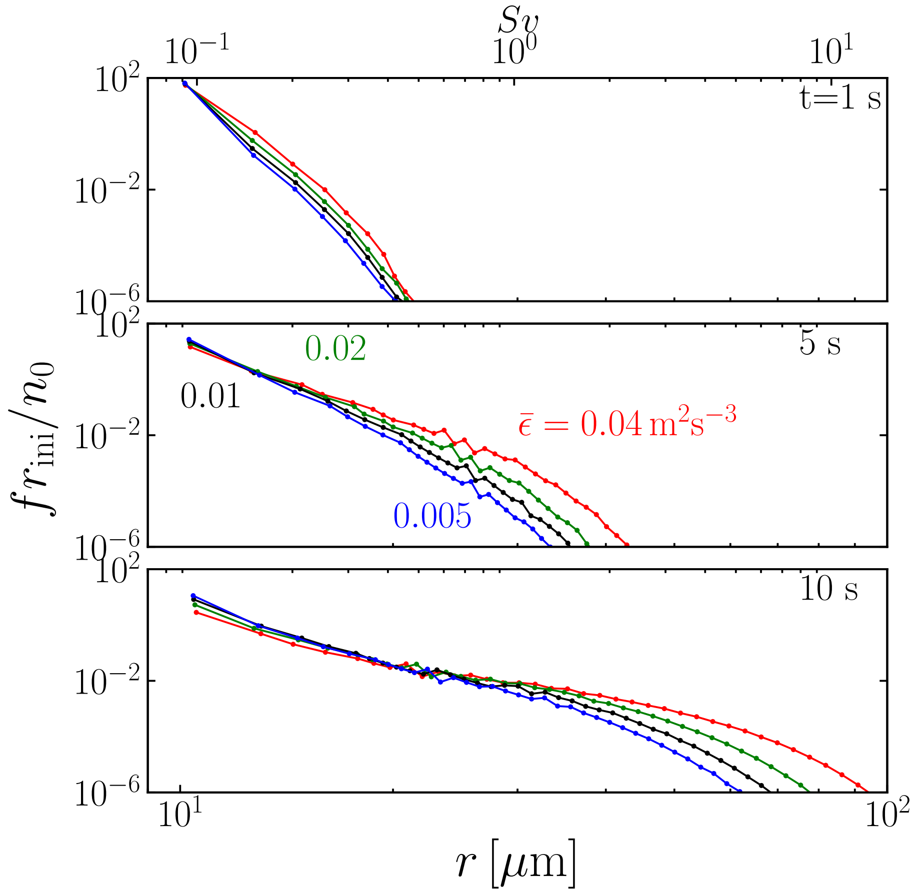

Figure 3.2: Time evolution of the droplet size distribution resulted from collisions for different $\bar{\varepsilon}$ in combined turbulence and gravity environment. $Re_\lambda = 100$. Droplets are all with radius $10\,\mu$m initially. Figure is adapted from Paper **III** (Li et al., 2018b).

The problem here is to determine the statistics of $\mathscr{T}$ in the limit as $\mathscr{N} \to \infty$ (Wilkinson, 2016). The mean collision rate λ_k between droplets of radii r_i and r_j and velocity difference $|\mathbf{V}_i - \mathbf{V}_j|$ can be expressed as

$$\lambda = E_c \cdot \pi\,(r_i + r_j)^2\,|\mathbf{V}_i - \mathbf{V}_j| n_{\mathrm{p/s}}, \tag{3.4}$$

where $n_{\mathrm{p/s}}$ is the number density of droplets per superparticle. Since droplets grow by collisions, after the k-th collision, the droplet volume increases by a factor of approximately k, thus the radius increases by a factor of $k^{1/3}$. Therefore the radius of the larger droplet scales as $r_k \sim r_0 k^{1/3}$. Thus, the collision rate obeys a power law (Wilkinson, 2016),

$$\lambda_k = \lambda_1 k^\gamma, \tag{3.5}$$

where λ_1 is the mean collision rate for the first collision determined by Equation (3.4). When $E_c \sim r^2$ and $|\mathbf{V}| \sim r^2$ (Pruppacher and Klett, 2012), the collision rate grows as the sixth power of the droplet size, i.e., as the second power of the droplet volume, which is the case of $\gamma = 2$. If one neglects the droplet size dependence on the collision efficiency, i.e., $E_c = 1$, then $\gamma = 4/3$. Since $\gamma > 1$, only the first few t_k determine $\mathscr{T}$. That is to say, the cumulative collision time $\mathscr{T}$ of the first few lucky droplets is a small fraction of the mean cumulative collision time $\langle \mathscr{T} \rangle$.

Mathematically, $P(\mathscr{T})$ follows an exponential distribution if λ_k is the same in Equation (3.2) for different k. However, λ_k is different for different time intervals, which results in a different shape of $P(\mathscr{T})$. Since $\tau = \mathscr{T}/\langle \mathscr{T} \rangle$ is independent of the first mean collision rate λ_1, $P(\tau)$ will be determined in the following study. Figure 3.3 shows the excellent agreement of $P(\tau)$ between the superparticle simulation and the numerical simulation of Equation (3.2), Equation (3.3) and Equation (3.5) with $\lambda_1 = 1\,\mathrm{s}^{-1}$ and $\gamma = 4/3$ ($\mathscr{N} = 128$), where 10^8 realizations are conducted. The excellent agreement demonstrates that the superparticle approach is able to capture the fluctuations of collision correctly even though a local mean collision rate (Equation (3.4)) is adopted.

3.3.3 Smoluchowski equation is not stochastic

The Smoluchowski equation is classically referred to as a stochastic equation in the meteorology community (Saffman and Turner, 1956; Berry and Reinhardt, 1974; Yau and Rogers, 1996; Pruppacher and Klett, 2012; Lamb and Verlinde, 2011a). At first glance, Smoluchowski equation is a mean-field equation without stochastic terms. Is Smoluchowski equation stochastic? First of all, when solving the Smoluchowski equation with mass binning method, one considers the mass and momentum transfer from small bins to large bins. Each mass bin is a representation of an ensemble of droplets, whose motion is treated in a Eulerian manner. In contrast, for the Lagrangian scheme, the phase and radius of each droplet (superparticle) are tracked. Second, to answer this question, we revisit the lucky-droplet model. The PDF of the cumulative collision time $P(\tau)$ in Figure 3.3 is calculated from the same numerical setup with different random seeds using the superparticle approach. However, by solving the Smoluchowski equation, one gets the same cumulative collision time for different random seeds. This suggests that the Smoluchowski equation is a mean-field equation instead of a stochastic equation, which should be used in caution.

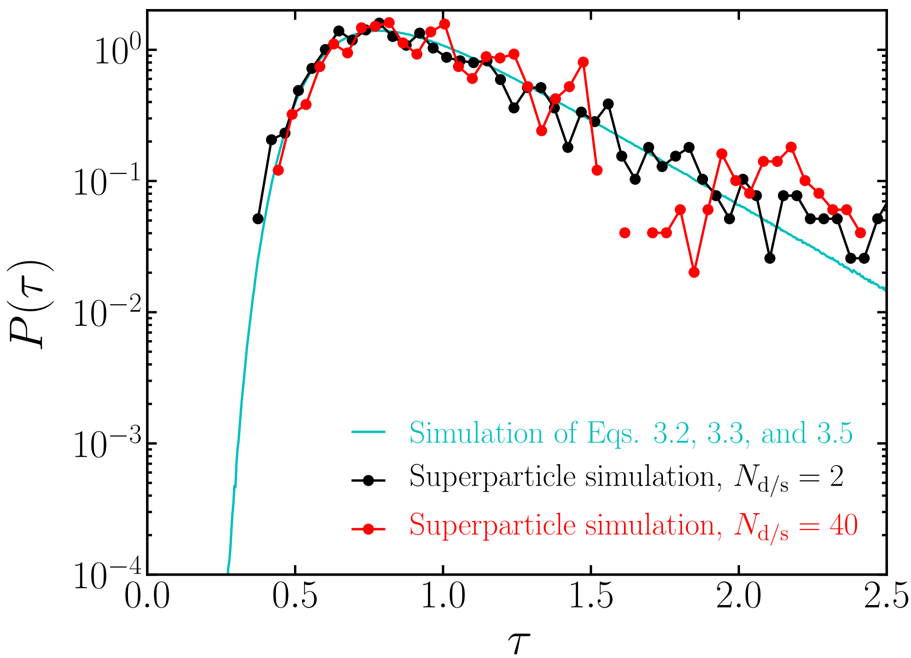

Figure 3.3: Distribution of τ calculated from 1059 simulations with different random seeds. The black ($N_{\mathrm{d/s}} = 2$) and red ($N_{\mathrm{d/s}} = 40$) curves represent the superparticle simulation results, where $N_{\mathrm{d/s}}$ is the initial number of droplets in each superparticle. The cyan curve represents the numerical simulation of Equation (3.2), Equation (3.3) and Equation (3.5) with $\lambda_1 = 1\,\mathrm{s}^{-1}$, $\gamma = 4/3$ ($\mathscr{N} = 128$), where 10^8 realizations are conducted. Figure is adapted from Paper **IV** with modifications of the legends.

Fluctuations are essential for collisional growth of cloud droplets (Kostinski and Shaw, 2005; Wilkinson, 2016). Can the Smoluchowski equation capture these fluctuations? To investigate this, the time evolution of droplet size distributions calculated from the Smoluchowski equation and the superparticle approach in 3-D turbulence are compared. As shown in Figure 3.4, the tail from the superparticle approach is about twice as wide as that from the Smoluchowski equation. Whether the widening tail is due to the fluctuations captured by the superparticle approach, which may not be represented by the Smoluchowski equation, requires further investigation. In Paper **I**, good agreement of droplet size distributions are observed between the Eulerian approach and the Lagrangian approach when the collision is driven by either gravity or 2-D turbulence, which is different from the case of 3-D turbulence. This could be due to the different initial droplet size distributions and the diluteness of the

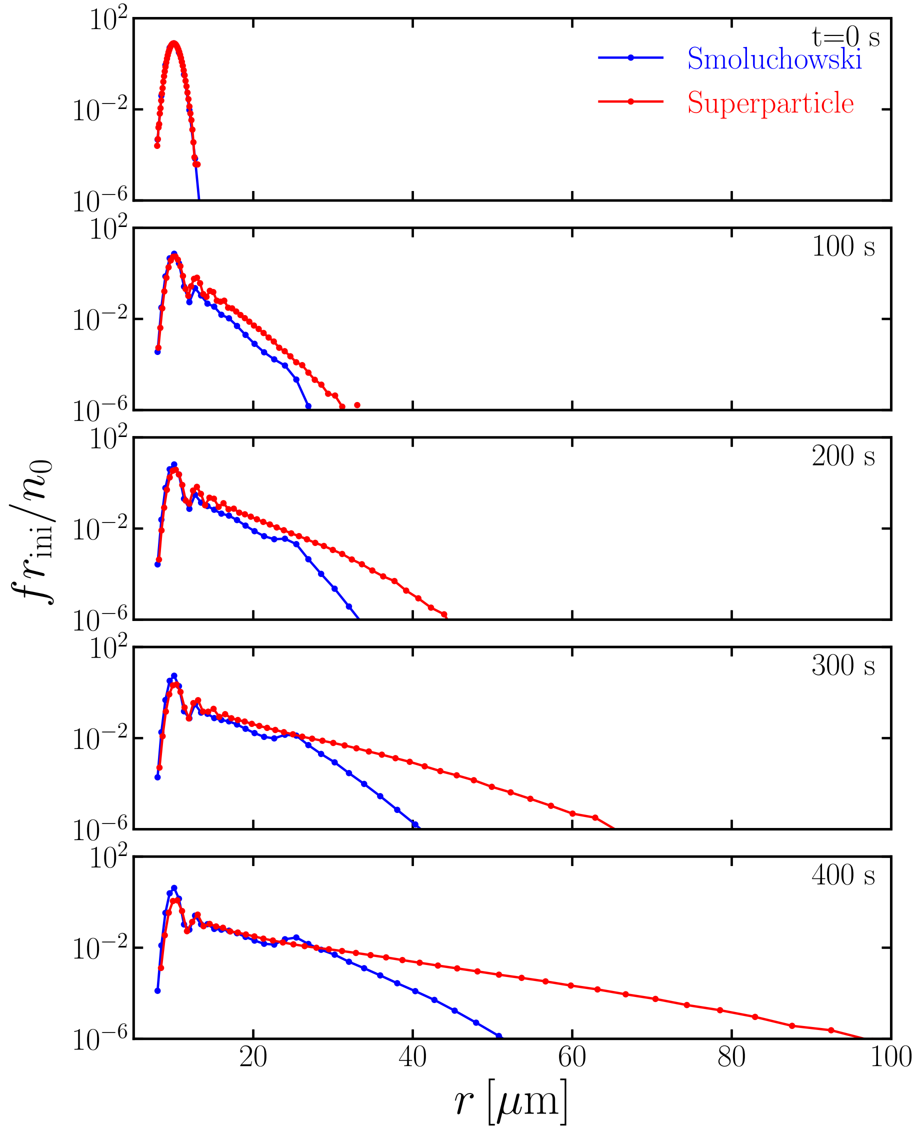

Figure 3.4: Comparison of the droplet size distribution simulated from the Eulerian approach (Smoluchowski equation) and the superparticle approach, in which collisions are driven by both turbulence and gravity. Figure is adapted from Paper **IV**.

system, which needs to be explored in the future.

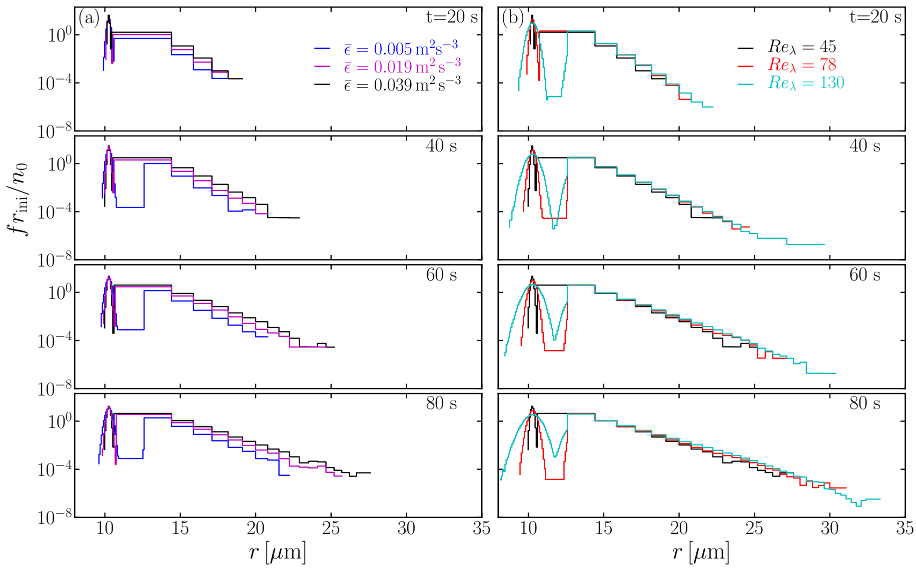

Figure 3.5: Time evolution of droplet size distributions resulted from condensation and collision for (a) different $\bar{\varepsilon} = 0.005\,\mathrm{m^2s^{-3}}$ (blue solid lines), 0.019 (magenta solid lines) and 0.039 (black solid line) at fixed $\mathrm{Re}_\lambda = 45$ and for (b) different $\mathrm{Re}_\lambda = 45$ (black solid lines), 78 (red solid lines), and 130 (cyan solid line) at fixed $\bar{\varepsilon} = 0.039\,\mathrm{m^2s^{-3}}$. Figure is adapted from Paper **V** (Li et al., 2018a).

3.4 Combined processes

Raindrop formation is a result of multiple cloud microphysical processes. As discussed in Section 3.2 that condensational growth due to supersaturation fluctuations broadens the droplet size distribution. This broadening is enhanced with increasing Reynolds number. Collisional growth is very sensitive to the tail of droplet size distributions and depends on the mean energy dissipation rate. Examining the effect of turbulence on the combined processes is computationally challenging because both large and small scales of turbulence need to be well resolved.

In Paper **V**, the effect of turbulence on the combined condensational and collisional growth of cloud droplets at different scales of turbulence are investigated using DNS. Thermodynamic equations governing the supersaturation field are solved. Droplets are tracked in a Lagrangian manner, whose trajectories differs from that of gas flow tracers due to inertia. Their motion is subjected to both turbulence and gravity. It is found that the combined condensational and collisional growth depends on both the mean energy dissipation rate and Reynolds number (Figure 3.5). Since the turbulence in warm clouds is characterized by high Reynolds number ($\mathrm{Re}_\lambda \approx 10^4$) and relatively small mean energy dissipation rate ($\bar{\varepsilon} \approx 10^{-3}\ \mathrm{m^2\,s^{-3}}$) as discussed in Chapter 1.2, turbulence mainly affects the condensational growth and enhances the collisional growth indirectly through condensation.

4. Summary of papers included in this thesis

This thesis investigates how turbulence affects the time evolution of droplet size distributions in warm clouds by means of DNS, aiming to explain the rapid warm rain formation. Since the problem of coupled droplet dynamics, collision-coalescence, thermodynamics, and turbulence is multi-scale and strongly nonlinear, the Eulerian and the Lagrangian schemes are developed and compared to find a physically and numerically optimal scheme for this problem. Then the effect of turbulence on the condensation, the collision-coalescence, and the combined processes are investigated, respectively.

In Paper **I**, two Lagrangian schemes are compared with an Eulerian scheme based on the mean-field Smoluchowski equation for the cases of pure condensation and pure collision, respectively, using DNS. Assuming a uniform supersaturation field, excellent agreement is observed between the Lagrangian scheme and the Eulerian scheme regarding droplet size distributions for the pure condensation case. Droplet size distributions resulted from collisional growth are also in good agreement either in the case of gravity or in 2-D turbulence in a dense system. The good agreement between the Lagrangian scheme and the Eulerian scheme paves the path towards investigating how turbulence affects the growth of cloud droplets. It is also found that the Lagrangian scheme is computationally about ten times more efficient than the Eulerian scheme. Therefore, the Lagrangian scheme has been adopted for the following studies (Li et al., 2017).

In Paper **II**, effect of turbulence upon condensational growth is investigated by solving the coupled thermodynamics, droplet dynamics, and turbulence using DNS (Li et al., 2018c). Contrary to the classical theory, that condensational growth leads to a narrow droplet size distribution considering a uniform supersaturation field (Pruppacher and Klett, 2012), it results in a wide droplet size distribution due to supersaturation fluctuations. Also, the width of the droplet size distribution increases as $t^{1/2}$, which is consistent with the Lagrangian stochastic model (Sardina et al., 2015). More importantly, the droplet size distribution broadens with increasing Reynolds number and is insensitive to the mean energy dissipation rate, which is due to the fact that supersaturation fluctuations are enhanced with increasing Reynolds number. Condensational growth due to supersaturation fluctuations may explain the broadening of droplet size distributions in stratiform clouds, where the updraft velocity is almost zero.

In Paper **III**, collisional growth of cloud droplets is explored in a turbulent environment (Li et al., 2018b). Momentum equations of the gas flow and droplets are solved using high-resolution DNS. The collision process is approximated by superparticle approach developed in Paper **I**. The collision and coalescence efficiency are assumed to be unit such that droplets coalescence upon collision. For a more general application, the collision process in a turbulent environment without gravity is studied, which is important for the dust growth of interstellar (Mathis et al., 1977). It is found that droplet size distributions depend strongly on the mean energy dissipation rate $\bar{\varepsilon}$ and weakly on the Reynolds number, which is explained by the Saffman-Turner collision model (Saffman and Turner, 1956). Remarkably, the mean collision rate from the DNS increases as $\bar{\varepsilon}^{1/2}$ as predicted by Saffman and Turner (1956), even though coalescence and droplet inertia are invoked in the DNS. More interestingly, the droplet size distribution exhibits power law behavior with a slope of -3.7, which is close to the observed size distribution of interstellar dust (Mathis et al., 1977). When collision is driven by both gravity and turbulence, it is found that the strong dependency of droplet size distributions on $\bar{\varepsilon}$ becomes weakened, which depends on the width σ of the initial distribution. Turbulence enhances the broadening of droplet size distributions efficiently when $\sigma = 0$ and weakly when $\sigma = 0.2$. As investigated in Paper **II**, condensational growth leads to broadening of droplet size distributions, after which collision can be triggered. Therefore, the effect of turbulence on the collisional growth of cloud droplets should be handled with caution. The mean collision rate resulted from collisions driven by both gravity and turbulence is shown to grow exponentially, which is consistent with the theoretical prediction of the continuous collision theory. Even though the continuous collision theory excludes turbulence. In Paper **IV**, how the superparticle approach represents fluctuations in the collision process is investigated. The superparticle approach is found to be optimal to capture the fluctuations in the collision process due to its generic stochastic property. Open questions regarding how to analyze the role of turbulence fluctuations on collisional growth are addressed.

In Paper **V**, the combined condensational and collisional growth of cloud droplets is scrutinized (Li et al., 2018a). It is observed that turbulence affects the combined processes from large to small scales. Considering the relative small values of $\bar{\varepsilon}$ and large Reynolds numbers of turbulence in clouds, it is concluded that turbulence predominantly enhances the condensational growth with increasing Reynolds number, while the collision process is indirectly affected by turbulence through condensational growth. Overall, it is suggested that turbulence facilitates the warm rain formation by enhancing the Reynolds number-dependent condensation process.

5. Outlook

Stay hungry, stay foolish
Steve Jobs

Ph.D. is an exciting journey. Many interesting scientific ideas have come out during this journey, which will be explored in the future!

5.1 High resolution simulations of cloud microphysics: a path towards the future climate model

DNS study is very limited by the domain size. Fortunately, the superparticle approach can be used in the large-eddy simulation (LES) with appropriate subgrid-scale scheme. LES models compute approximate solutions of Navier-Stokes equations and equations of thermodynamics, while using simplified representations for still unresolved cloud microphysical processes such as droplet and ice crystal formation (Schneider et al., 2017). The superparticle approach used in the present DNS can easily be adapted to large-eddy simulations (LES) with appropriate subgrid-scale models. The combined condensational and collisional growth of cloud droplets largely depends on the Reynolds number. DNS are limited by the state-of-art supercomputer power. LES can be used to simulate the condensational and collisional growth in high Reynolds number turbulence. This may shed some light on explaining the rapid warm rain formation, which will be explored in future studies.

5.2 Quantum computer for climate model

A physical and accurate future climate projection is urgent for humankind. It is almost impossible to resolve the entire scales of climate model from micrometer-sized aerosols to hundreds of kilometer-sized scales. Quantum mechanics is supposed to explain every physical law, including thermodynamics and turbulence, which are the cornerstones of atmospheric science. Quantum computer (Nielsen and Chuang, 2002), based on quantum mechanics, may shed some light on fully resolving the smallest scales in the climate model.

Modern computers are Turing machine, which is based on the masterpiece *Church-Turing thesis*. It states that: "*Any algorithm process can be simulated efficiently using a Turing machine*" (Nielsen and Chuang, 2002). After Turing's paper in 1936, John Von Neumann developed a theoretical model for how to put together all the components necessary for a computer in a practical fashion such that the computer can be as capable as the Universal Turing Machine (Nielsen and Chuang, 2002). When John Bardeen, Walter Brattain, and Will Shockley developed transistors, hardware for a computer has grown in a extraordinary pace, which has come to be known as the Moore's law (Nielsen and Chuang, 2002). The Turing machine is based on classical physics. Since the basic law of physics is ultimately quantum mechanics, a computer device based upon the principles of quantum mechanics naturally fits into the development of a truly new era of computer.

The bit is a fundamental concept of classical computer, where it can be in a state of either 0 or 1. For a quantum computer, analogy to a classical computer, qubit is the basic element (Nielsen and Chuang, 2002). Obeyed to principles of quantum mechanics, qubit can be in a state other than $|0\rangle$ or $|1\rangle$, where "$|\rangle$" is the Dirac notation. The qubit is the core to render the quantum computer faster than the classical computer (Biamonte et al., 2017).

The ultimate question for us is how much faster the quantum computer compared with the classical computer is when simulating turbulence or other time-accurate simulations of classical dynamical systems with chaotic behavior. This is referred to as the "quantum computational supremacy", which is still unknown (Boixo et al., 2018). When quantum computer are commercialized, perhaps in five years (Mohseni et al., 2017), it will be a great opportunity for us climate scientists or physicists to embrace for the future climate model.

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