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Deshmukh, Akash

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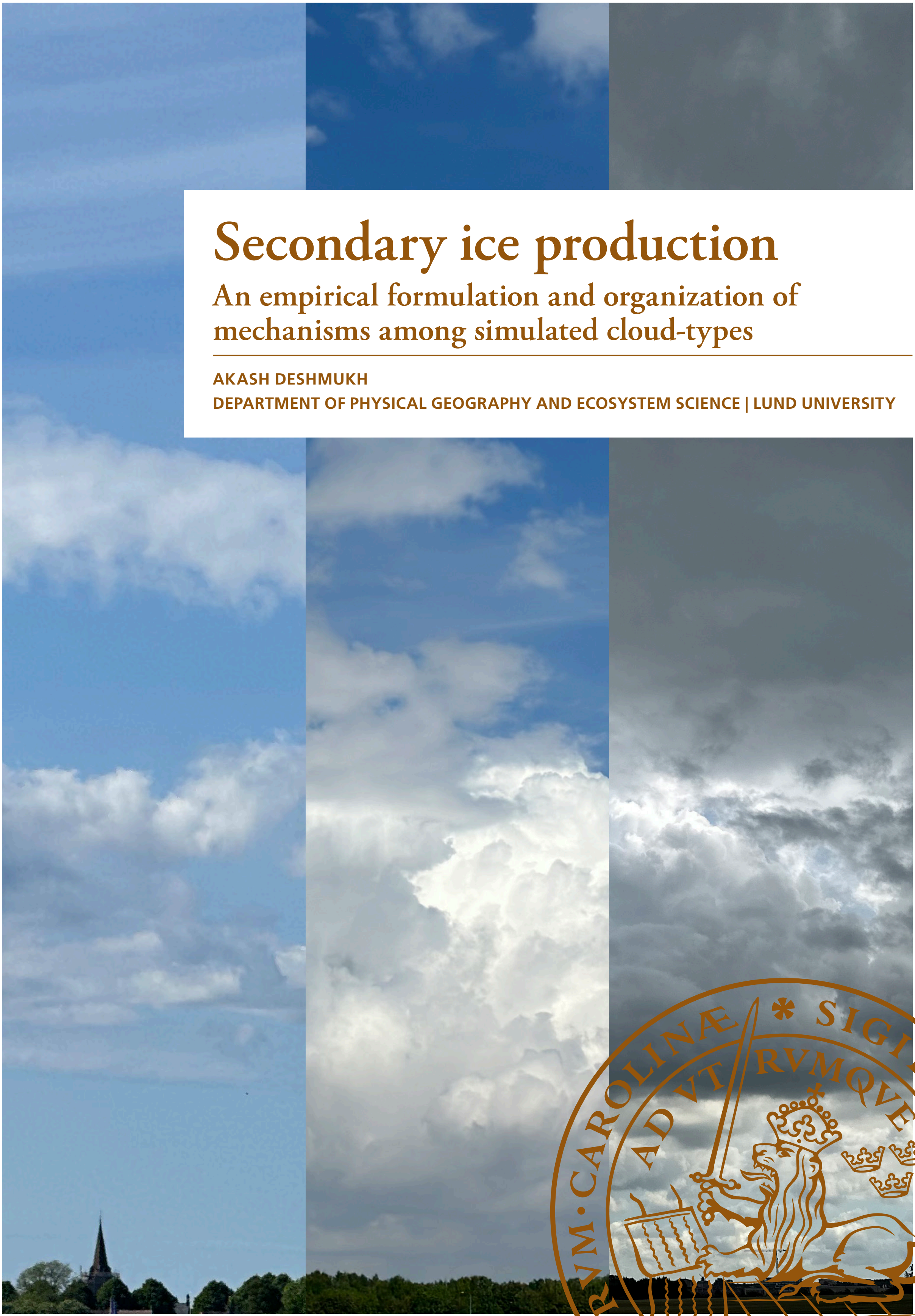

Secondary ice production

An empirical formulation and organization of mechanisms among simulated cloud-types

AKASH DESHMUKH
DEPARTMENT OF PHYSICAL GEOGRAPHY AND ECOSYSTEM SCIENCE | LUND UNIVERSITY

LUND UNIVERSITY

Department of Physical Geography
and Ecosystem Science
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Secondary ice production: An empirical formulation and organization of mechanisms among simulated cloud-types

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Akash Deshmukh

LUND
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DOCTORAL DISSERTATION

Doctoral dissertation for the degree of Doctor of Philosophy (PhD) at the Faculty of Science at Lund University to be publicly defended on Thursday, the 7th December 2023, at 10.00 AM in Världen Auditorium, Geocentrum I, Department of Physical Geography and Ecosystem Science.

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Abstract:

Clouds are essential elements within Earth's atmosphere, posing a challenge for cloud-resolving models in understanding the creation of new cloud ice particles from existing ice and liquid phases. Such ice initiation determines cloud microphysical and radiative properties, influencing cloud phase, precipitation and cloud extent/properties. To address this challenge effectively, it proves beneficial to differentiate the fundamental microphysical properties of various cloud types, considering their basic classifications.

A few historical experimental studies a few decades ago delved into how sublimation of ice crystals causes the emission of fragments. These fragments subsequently grow into crystals, and in some cases, may evolve into snow or graupel. This sublimational breakup represents a form of secondary ice production, capable of causing ice multiplication in natural clouds. The origins of the high ice concentrations observed in clouds are becoming better understood, but still have some uncertainty.

In this study, an empirical numerical formulation for sublimation breakup in cloud models is introduced. This formulation is based on comprehensive laboratory data gathered from previous studies. By analyzing experiments that measured the number of ice fragments generated through sublimation, considering factors such as relative humidity and initial ice particle size, we derived essential parameters for a sublimation breakup scheme. The research findings highlight the prevalence of size dependency in smaller particles, while larger particles exhibit comparable dependencies.

Ice initiation in clouds has primarily focused on specific cloud systems, revealing that the majority of ice particles in the mixed-phase region result from secondary ice production mechanisms. However, these studies have been limited to individual cloud types. The objective of this thesis is to broaden the understanding of each secondary ice production mechanism's contribution across various fundamental cloud types with a more all-inclusive approach. To achieve this, numerical simulations are conducted utilizing our ‘Aerosol-Cloud model’ for different cloud categories. These simulations are then validated against in-situ cloud observations obtained from four distinct cloud observational campaigns, each representing a different cloud type for comprehensive analysis.

In this study, the roles of various secondary ice production processes, including the HM process, ice-ice collisional breakup, raindrop-freezing fragmentation, and sublimational breakup were meticulously examined. These analyses are conducted through controlled simulations for different fundamental cloud types. Within warm cloud convective clouds, the HM process is particularly notable near the freezing level, making contributions within specific temperature ranges. Ice-ice collisional breakup emerges as the predominant secondary ice production mechanism across all cloud types, being the only one with appreciable activity in cold-based convection. Additionally, in slightly warm-based convective clouds, the breakup resulting from ice-ice collision takes precedence within the convective updrafts.

Key words: fragmentation, sublimation, ice, secondary ice, ice enhancement, ice production, collision

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The two most powerful warriors are patience and time.

- Leo Tolstoy

To my Parents…

Table of Contents

Abstract

Clouds are essential elements within Earth's atmosphere, posing a challenge for cloud-resolving models in understanding the creation of new cloud ice particles from existing ice and liquid phases. Such ice initiation determines cloud microphysical and radiative properties, influencing cloud phase, precipitation and cloud extent/properties. To address this challenge effectively, it proves beneficial to differentiate the fundamental microphysical properties of various cloud types, considering their basic classifications.

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all cloud types, being the only one with appreciable activity in cold-based convection. Additionally, in slightly warm-based convective clouds, the breakup resulting from ice-ice collision takes precedence within the convective updrafts.

Popular Summary

Within Earth's atmosphere, clouds serve as vital components, yet they present a challenge to understand the intricate process of forming new ice particles from existing ice and liquid phases. This ice initiation process significantly impacts cloud properties, including their microphysical and radiative characteristics, as well as cloud phase, precipitation patterns, and overall cloud extent.

To tackle this complexity, it is essential to discern the fundamental microphysical properties of various cloud types, considering their basic classifications. Several decades ago, pioneering experimental studies explored the sublimation of ice crystals, revealing that this process emits fragments which can transform into crystals, snowflakes, or graupel. This sublimational breakup represents a secondary ice production mechanism, enhancing ice concentration within natural clouds. While the origins of high ice concentrations in clouds are gradually becoming clearer, uncertainties remain.

This study introduces an empirical numerical formulation for sublimation breakup in cloud models, grounded in comprehensive laboratory data. By analyzing experiments measuring ice fragments generated through sublimation, considering factors like relative humidity and initial ice particle size, essential parameters for a sublimation breakup scheme were derived. Notably, smaller particles exhibited size dependency, while larger particles displayed comparable trends.

Unlike prior studies limited to specific cloud systems, this research adopts a comprehensive approach, aiming to broaden understanding across various fundamental cloud types. Utilizing the 'Aerosol-Cloud model,' numerical simulations were conducted for different cloud categories. The study meticulously examined several secondary ice production processes, such as the HM process, ice-ice collisional breakup, raindrop-freezing fragmentation, and sublimational breakup. This comprehensive exploration sheds light on the intricate dynamics of cloud ice particle formation, contributing valuable insights to our understanding of atmospheric processes.

Populärvetenskaplig sammanfattning

Moln fungerar som viktiga komponenter i atmosfären. Samtidigt skapar de utmaningar när man försöker studera de processer där nya ispartiklar bildas. Denna process för påbörjad skapelse av is påverkar molnens egenskaper avsevärt, inklusive deras mikrofysikaliska och strålningsegenskaper, samt molnfas, nederbördsformation och molnets övergripande omfattning.

För att hantera denna komplexitet är det nödvändigt att skilja de grundläggande mikrofysikaliska egenskaperna hos olika molntyper, med hänsyn till deras grundläggande klassificeringar. För några årtionden sedan utforskade banbrytande experimentella studier sublimering av iskristaller och avslöjade att denna process avger fragment som kan omvandlas till kristaller, snöflingor eller mjukt hagel. Denna sublimerande uppdelning representerar en sekundär mekanism för isproduktion som ökar koncentrationen av is inom naturliga moln. Medan ursprunget till höga iskoncentrationer i moln gradvis blir tydligare finns det fortfarande osäkerheter.

Denna studie introducerar en empirisk numerisk formulering för sublimeringens uppdelning i molnmodeller, baserad på omfattande laboratoriedata. Genom att analysera experiment som mäter de isfragment som genereras genom sublimering, med hänsyn till faktorer som relativ luftfuktighet och ursprunglig ispartikelstorlek, härleddes väsentliga parametrar för ett schema över sublimeringens uppdelning. Intressant nog visade sig mindre partiklar vara storleksberoende, medan större partiklar uppvisade jämförbara trender.

I motsats till tidigare studier som var begränsade till specifika molnsystem, antar denna forskning ett omfattande tillvägagångssätt och syftar till att bredda förståelsen av olika grundläggande molntyper. Genom att använda 'Aerosol-Cloud'-modellen utfördes numeriska simuleringar för olika molnkategorier. Studien undersökte noggrant flera sekundära isproduktionsprocesser, såsom HM-processen, uppdelning orsakad av is-is-kollision, regnkärnbildning genom frysning och sublimerande uppdelning. Denna omfattande undersökning synliggör komplex dynamik hos molnens ispartikelbildning och bidrar med värdefulla insikter till vår förståelse av atmosfäriska processer.

सारांश

पृथ्वीच्या वातावरणात मेघ महत्त्वपूर्ण घटक म्हणून काम करतात. हे मेघ, बर्फ आणि द्रव यांच्यापासून नवीन बर्फाचे कण तयार करण्याची जटिल प्रक्रिया समजून घेण्याच्या आव्हानाने भरलेले आहेत. या प्रक्रिया त्याच्या सूक्ष्म जैविक आणि उत्सर्जन वैशिष्ट्यांसह काम करतात. तसेच हे घटक ढगांचा जीवनकालीन टप्पा, पर्जन्याचे आकृतीबंध आणि एकूण ढगाच्या व्याप्तीसह ढगांच्या गुणधर्मांवर लक्षणीय परिणाम करतात. या जटिल प्रक्रिया समजून घेण्यासाठी, विविध मेघ प्रकारांचे मूलभूत सूक्ष्म भौतिक गुणधर्म ओळखणे आणि त्यांच्या मूलभूत वर्गीकरणात ध्यान देणे अत्यंत आवश्यक आहे. अनेक दशकांपूर्वी, अग्रगण्य प्रायोगिक अभ्यासकांनी केलेल्या अभ्यासांतून, बर्फ-स्फटिकांच्या उदात्तीकरणाची प्रक्रिया पुढे आली, ज्यातून हे स्पष्ट झाले की ही प्रक्रिया तुकड्यांचे उत्सर्जन करते, ज्यामुळे हे स्फटिकांचे अंश किंवा तुकडे पुढे बर्फामध्ये रूपांतरित होऊ शकतात. हे उदात्तीकरणात्मक विघटन, दुय्यम बर्फ उत्पादन प्रकियांमध्ये गणले जाते. या प्रकियांद्वारे, नैसर्गिक नभांमध्ये वाढणारी बर्फ संख्या विशद करता येते. बर्फाच्या उच्च संख्या आणि बर्फाची उत्पत्ती संशोधनातून हळूहळू स्पष्ट होत आहे तरीही या प्रक्रियेतील अनिश्चितता कायम असल्याचे दिसून येते.

या अभ्यासामध्ये, मेघ प्रणालीमध्ये उदात्तीकरणाद्वारे होणाऱ्या विखंडनासाठी एक संख्यात्मक सूत्रीकरण करण्यात आले आहे, जे की पूर्वी प्रकाशीत झालेल्या सर्वसमावेशक प्रयोगांच्या अभ्यासावर आधारित आहे. उदात्तीकरणाद्वारे उत्पन्न झालेल्या बर्फाच्या तुकड्यांचे विश्लेषण करून, सापेक्ष आर्द्रता आणि प्रारंभिक बर्फाच्या कणांचा आकार यासारख्या घटकांचा विचार करून, उदात्तीकरण विभाजन योजनेसाठी आवश्यक मापदंड प्राप्त करण्यात आले आहेत. विशेषतः, लहान कणांचे उदात्तीकरणाद्वारे होणारे विखंडन हे कणांच्या आकारावर अवलंबून आहे.

विशिष्ट मेघ प्रणालीच्या संदर्भात, पूर्वीच्या अभ्यासांच्या विपरीत, सर्वसमावेशक दृष्टिकोण अवलंबून, समज वाढवण्याचा प्रयत्न केला गेला आहे. संख्यात्मक मेघ प्रतिरूपाचा वापर करून, विविध मेघ श्रेणींसाठी संख्यात्मक प्रतिकृती तयार करण्यात आली. अनेक दुय्यम बर्फ उत्पादन प्रक्रियांचे बारकाईने परीक्षण केले गेले आहे, जसे की एच-एम प्रक्रिया, बर्फ-बर्फाचा आदळाव, पावसाच्या थेंबाचे अतिशीत विखंडन आणि उदात्तीकरणपासून विखंडन. या अभ्यासाद्वारे मेघांतील बर्फ कण निर्मितीच्या गुंतागुंतीच्या प्रक्रियांवर प्रकाश टाकला गेला आहे, ज्यामुळे आपल्या वातावरणातील प्रक्रियांबद्दल जाणून घेण्यासाठी मौल्यवान दृष्टिकोण मिळेल. वरील सर्व प्रक्रियांचा तपशील ह्या प्रबंधामध्ये सादर केलेला आहे.

List of Scientific Publications

This thesis incorporates findings from the following publications:

I. **New Empirical Formulation for the Sublimational Breakup of Graupel and Dendritic Snow**

Akash Deshmukh, Vaughan TJ Phillips, Aaron Bansemer, Sachin Patade, Deepak Waman
Journal of the Atmospheric Sciences,2022,79, 317-336

II. **Organization of ice multiplication mechanisms among basic cloud types. Part I: Validation and analysis of the control simulations**

Akash Deshmukh, Deepak Waman, Sachin Patade, Arti Jadav, Martanda Gautam, Vaughan Phillips, Aaron Bansemer, Ashok Gupta, Paul Connolly, Greg McFarquhar, Paul J. DeMott, Jiwen Fan, Yun Lin, Jann Schrod, Heinz Bingemer
Manuscript submitted to the Journal of the Atmospheric Sciences.

III. **Organization of ice multiplication mechanisms among basic cloud types. Part II: Sensitivity Tests**

Akash Deshmukh, Vaughan Phillips
Manuscript submitted to the Journal of the Atmospheric Sciences.

IV. **The microphysics of the warm-rain and ice crystal processes of precipitation in simulated continental convective storms**

Ashok Kumar Gupta, **Akash Deshmukh**, Deepak Waman, Sachin Patade, Arti Jadav, Vaughan TJ Phillips, Aaron Bansemer, Jorge A Martins, Fabio LT Gonçalves
Communications Earth & Environment, 2023,4.

Author’s contribution to the papers

Akash Deshmukh (**AD**), Vaughan TJ Phillips (VTJP), Aaron Bansemer (AB), Ashok Kumar Gupta (AKG)

Paper I

AD and VTJP designed the research and lead and implemented by **AD**. The parcel model simulations performed by **AD** and AB provided the campaign data. **AD** prepared the paper with contributions from all co-authors.

Paper II

AD lead the research and implemented the formulation in model. This research is designed by VTJP. The simulations are set-up by VTJP and **AD** (e.g., ACAPEX and GOAmazon). AB provided the field campaign data. **AD** prepared the manuscript with contributions from all co-authors.

Paper III

AD and VTJP designed the research. This research lead by **AD**. **AD** prepared the manuscripts with contribution from VTJP.

Paper IV

AKG and VTJP designed the research. This research lead by AKG. **AD** set-up the simulation of GOAmazon and contributed in the preparation of the manuscript.

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I humbly recognize and deeply cherish the collaborative endeavors of those whose hands and hearts guided this expedition, expertly placing each piece in its destined space. Your support has been invaluable, and I am profoundly grateful for the opportunity to have worked with such exceptional individuals.

Acronyms

AC	Aerosol-cloud model
AAF	ARM Aerial Facility
ACAPEX	ARM Cloud Aerosol Precipitation Experiment
AMF2	ARM Mobile Facility 2
ARM	Atmospheric Radiation Measurement
AR	Atmospheric Rivers
CF	Central Facility
CCN	Cloud condensation nuclei
CRM	Cloud-resolving model
CAPE	Convective Available Potential Energy
DoE	Department of Energy
GPM	Global Precipitation Measurement
GOAmazon	Green Ocean Amazon
HM process	Hallett-Mossop process
IE ratio	Ice enhancement ratio
INP	Ice nucleating particle
IN	Ice nuclei
IMPROVE	Interagency Monitoring of Protected Visual Environments
IPCC	Intergovernmental Panel on Climate Change
MSL	Mean sea level
MCS	Mesoscale Convective System
MC3E	Mid-latitude Continental Convective Clouds Experiment
NASA	National Aeronautics and Space Administration
SIP	Secondary ice production
STEPS	Severe Thunderstorm Electrification and Precipitation Study

SGP	Southern Great Plains
UTC	Universal Time Coordinated
WRF	Weather research and Forecast

1. Introduction

Clouds are an integral part of the Earth's atmosphere, covering a significant portion of the globe, typically ranging from 60% to 70% (Lohmann et al. 2016). They have a profound impact on the climate by regulating the temperature and moisture distribution in the atmosphere. Therefore, it is essential to consider clouds in studies that address climate-related issues, such as in the prediction of future climate changes resulting from human activities like the release of greenhouse gases and aerosols. However, there are still many aspects of the cloud-climate relationship that remain not fully understood or cannot be accurately quantified or modelled. The Intergovernmental Panel on Climate Change (IPCC) identifies clouds and their associated feedbacks as one of the primary uncertainties when it comes to predicting future climate patterns (Solomon et al. 2007).

The quest to understand the processes of initiation of atmospheric ice particles is a noteworthy problem in cloud physics. The formation of clouds in the atmosphere occurs when aerosol particles undergo activation and transform into liquid cloud droplets or ice crystals within ascending air parcels. Once cloud particles reach a specific size threshold, they grow to become precipitation that descends in the form of raindrops, snow crystals, graupel or hailstones. The formation of ice crystals holds the potential to impact a cloud's microphysical characteristics, its interaction with radiation, and its ability to generate precipitation. Therefore, the process of ice initiation plays a pivotal role in influencing the Earth's climate.

1.1 Cloud nomenclature and classification

Clouds have long captivated human imagination and held a prominent place in literature, art, and science. Clouds high above the Earth's surface, constantly change shape, size, and appearance. Meteorologists have categorized clouds based on their appearance, altitude, and weather patterns, providing valuable insights into atmospheric dynamics and weather forecasting. Cloud classifications are vital tools for meteorologists, weather forecasters, and scientists studying atmospheric phenomena. They facilitate a comprehensive understanding of the Earth's climate, weather patterns, and the various factors influencing them. By observing and categorizing clouds, we gain valuable insights into the complex interactions between the atmosphere and the Earth's surface. Understanding cloud formations helps us predict weather patterns, assess potential hazards, and appreciate the mesmerizing beauty of the skies above. As we continue to delve into the mysteries of the atmosphere, cloud classifications will remain an essential aspect of meteorology and climate science, shaping the way we interpret and engage with the dynamic world of weather.

The system of cloud classification was introduced in 1802 by meteorologist Luke Howard (Hamblyn 2002). He is renowned for his talk addressing the Askesian Society in London about cloud classification based on his sketches of cloud forms from trips outdoors. This was the first successful classification of clouds using observable criteria for their visual appearance. Howard’s original system of classification consists of three basic types of clouds, namely: stratiform (stratus or strato-), cumuliform or convective (cumulus or cumulo-) and cirriform (cirrus or cirro-) (Jardine 2014). Stratiform clouds are thin-layer clouds that can extend for more than many tens of kilometers horizontally with vertical velocities of less than 1m/s. The stratiform clouds may persist from several hours to several days. The stratiform precipitation rate is around 2-3 mm/hr.

Convective clouds extend vertically up to the tropopause and can even penetrate into the stratosphere. Convection arises due to an unstable vertical thermodynamic structure of the troposphere around the clouds (Rogers and Yau 1996; Lohmann et al. 2016). The vertical velocities in convective clouds are stronger than 1 m/s. The lifetime of convective clouds is generally shorter than that of stratiform clouds, typically ranging from 30 minutes to a few hours. Convective clouds can cover up to about one or two tens of kilometers in width. Convective clouds are always associated with rainfall rates exceeding approximately 25 mm/h.

These fundamental cloud types have been further categorized based on various characteristics. These characteristics include rainfall (indicated by terms like "nimbus" or "nimbo-"), the presence of ice, cloud shape (e.g., "congestus"), and the level of cloud base (e.g., "alto-" for clouds with a high base). In the advanced lexicon of cloud classification, the nomenclature of a particular cloud type involves amalgamating Latin terminology that encapsulates these qualities with the fundamental cloud type's appellation. Hybrid cloud types occasionally appear, combining traits from two or more basic cloud types. For instance, "stratocumulus" merges characteristics from both stratiform and convective clouds, and the same goes for "cirrocumulus." In modern cloud classification, basic types are also classified according to cloud base temperature and altitude, such as "low," "middle," or "high" clouds (Houze 1993).

Clouds take shape as a consequence of the ascent and subsequent cooling of warm, moisture-laden air, leading to the condensation of water vapor into minuscule water droplets or ice crystals encompassing particulate matter. The ever-changing atmospheric dynamics and geographical settings yield a remarkable array of cloud structures, prompting the establishment of intricate cloud classification frameworks.

Clouds typically form when a parcel of air ascends vertically and cools to reach its dew point. This process is driven by four distinct lifting mechanisms: Orographic lifting, convection, convergence, and large-scale ascent. Orographic lifting occurs when air encounters a mountain barrier and is compelled to flow over it due to the inability to penetrate through it. Frontal lifting transpires when less dense warm air is obliged to ascend over cooler, denser air as weather fronts move, with this phenomenon being most prevalent during the winter season. Convection takes place as solar energy penetrates the atmosphere, heating the Earth's surface, causing the air near the surface to become less dense than the surrounding air, leading to its upward ascent. Convergence lifting transpires when surface-level air converges, resulting in compression and an upward push, propelling it upwards. Large-scale ascent occurs in weather systems, such as in the warm sector of a synoptic-scale mid-latitude cyclonic depression with warm moist air rising near a front. In all these lifting mechanisms, the rising air generates updrafts inside the clouds that creates a deep layer of condensation. If the cloud is precipitating, then the updrafts can support precipitation particles allowing time for them to grow.

1.2 Microphysical processes

Warm-based clouds are typically characterized by the presence of relatively large cloud droplets, where precipitation does not solely occur through condensation. In the diffusional growth of droplets, water vapor in the atmosphere condenses on airborne aerosol particles referred to as cloud condensation nuclei (CCN). During this phase transition, water vapor transforms from a gaseous state to a liquid state, initiating the formation of minute cloud droplets. However, the primary mechanism responsible for precipitation in these clouds is the collision-coalescence process, often referred to as the "warm rain process" (Rogers and Yau 1989). After diffusional growth of cloud droplets, cloud droplets start to collide with each other and merge through collisions, this process described as the collision-coalescence process. Larger cloud droplets have slightly higher terminal velocities compared to smaller droplets. The larger droplets fall faster and collide with smaller cloud droplets. Sometimes, the cloud droplets adhere and merge, forming larger droplets. This phenomenon initiates a positive feedback loop wherein these larger droplets descend at an accelerated rate. During their descent, they collide with an increasing number of smaller droplets in their path, fostering further aggregation. This process leads to the continual amalgamation of cloud droplets, creating a cascading effect of coalescence and growth within the cloud. However, collision between cloud drops does not always mean coalescence will occur. Sometimes drops will bounce apart during collision if their surface energy is too weak compared with their collision kinetic energy (e.g., Low and List 1982). For collision-coalescence to begin, a cloud needs to have a wide spectrum of cloud droplet sizes. In principle, cloud droplet diameters range from 2 μm to about 100 μm and raindrop diameters range from about 0.1 mm to 5 mm (drizzle drops are at about 0.1 mm). However, there is a statistical distribution of drop diameters, with most cloud-droplets having sizes comparable with a mean diameter (e.g., 10-30 microns typically).

Clouds are classified as cold clouds when in-cloud temperatures are usually below freezing (0ºC). Such clouds possibly will be composed of liquid water, supercooled water, and/or ice. In cold clouds, precipitation can form by growth of ice crystals, first by diffusion of vapor and later by riming or aggregation to yield snow and this snow may rime to form graupel that may melt during fall-out. This precipitation mechanism is termed the 'ice crystal process' (Rogers and Yau 1989).

Similar to how cloud droplets need a surface on which to condense, the initial formation of ice crystals also needs a nucleus or ice embryo upon which to freeze. Without an ice nucleus, liquid water drops can remain supercooled at temperatures as low as about −36°C. It is much more difficult for the first ice crystals to form in the atmosphere (in the absence of homogeneous freezing) as compared to cloud droplets: they require the solid surface of aerosol material to initiate growth - called an ice nucleus (IN). However, such solid aerosol material typically needs to have optimal properties (e.g., shape, chemical composition of molecular lattice structure, size) for ice crystals to grow on them. Any liquid droplets will remain in a supercooled state down to temperatures as low as about −36°C, beyond which all droplets turn to ice crystals. Between 0℃ and −36℃, water can exist in a supercooled or ice state. The majority of cold clouds, therefore consist of a mixture of ice crystals and supercooled liquid droplets. The supercooled droplets massively outnumber the number of ice crystals. Ice crystals have a lower saturation vapor pressure at their surface than liquid water droplets. This means that ice crystals require less water vapor in the atmosphere than liquid droplets to grow by deposition (diffusional growth).

In many cold clouds with sufficiently weak ascent, the environment is subsaturated with respect to liquid water, but supersaturated with respect to ice, due to the vapor growth of ice particles (Korolev 2007). This results in the liquid droplets evaporating, while the ice crystals grow at their expense. These ice particles can grow large enough that they begin to fall out of the cloud. They may continue to grow by aggregation with other crystals, or by riming - where small supercooled cloud-droplets freeze directly onto the ice crystal surface. These ice crystals can aggregate together to form snowflakes, and can reach the surface as snow if the atmosphere remains below freezing. However, they can also melt to form rain at the surface. This process is known as Wegener–Bergeron–Findeisen process and is a special type of the ice crystal process of precipitation production noted above.

1.2.1 Nucleation

Nucleation represents a phase transformation and takes place within the surrounding phase to form a stable form. In contrast with the formation of liquid droplets, which does not occur homogeneously in the atmosphere, both homogeneous nucleation of ice crystals from the liquid phase (i.e., homogenous freezing) and heterogeneous nucleation of ice crystals from the vapor and liquid phase occur in the atmosphere.

1.2.1.1 Homogenous nucleation

Homogenous nucleation means the spontaneous formation of the new phase without the action of any foreign material (not water). Homogeneous nucleation of liquid from vapor, requires more than a factor of 20 for the super-saturation to induce homogeneous nucleation at 0ºC; it does not occur in the real atmosphere due to the high super-saturation required. Homogeneous freezing yields ice crystals in appreciable numbers when the temperature drops below about −35ºC.

1.2.1.2 Heterogeneous nucleation

Cloud-droplets form by activation of aerosols with soluble material, namely CCN if the supersaturation with respect to liquid slightly exceeds zero. Heterogeneous nucleation of ice means formation of ice crystals from the activity of foreign particles. Foreign particles are significant in the formation of ice crystals and known as 'ice nucleating particles' (INPs), (an active IN). INPs are analogous to CCN in the formation of water droplets (warm clouds). After contact with most solid materials, supercooled droplets freeze before temperatures can become colder than −36ºC. There are four heterogeneous ice nucleation modes distinguished in the literature as follows:

- Immersion freezing: Immersion freezing refers to freezing that originates within a cloud or solution droplet. It requires that the INP is already immersed in the droplet. Afterwards, freezing is initiated by cooling of the droplet. Immersion freezing can occur at all supersaturations but it requires pre-existence of cloud droplets and water saturation.

- Condensation freezing: Condensation freezing refers to a different path where an air parcel holding INPs from the beginning of subsaturated (dry) conditions. Generally, an INP contains a small amount of soluble salts/compounds in accumulation to the insoluble compound. This mixture of compounds increases the possibility that an INP can act as a CCN for small change in supersaturation. Consequently, condensation freezing can only occur in the slim band of supersaturations before the activation of CCN.

- Deposition nucleation: Deposition nucleation involves the direct deposition of vapor on INPs and requires supersaturation with respect to ice. Deposition nucleation is initiated in cirrus clouds, when vapor is deposited on INP (Cziczo et al. 2013). Previous laboratory studies proposed that deposition nucleation only occurs for temperature below −18ºC (Schaller and Fukuta 1979).

- Contact freezing: Contact freezing describes the process of collision of an INP with a supercooled cloud droplet which is followed by freezing. It depends on collisions between the supercooled droplet and INPs, and it is counted as a limitation of this process. The collision rate and efficiency are the most vital parameters here.

1.2.2 Growth processes

After nucleation, these ice crystals can grow by diffusion, aggregation or accretion. Depending on the temperatures and saturation levels in the cloud, the ice crystals can grow in various forms or crystal habits. In mixed-phase clouds, diffusional growth happens due to differences in the saturation vapor pressure between ice and water. At a specific temperature, the saturated vapor pressure over a water surface exceeds that over an ice surface. In mixed-phase conditions (water droplets and ice crystals co-exist), a vapor pressure gradient develops between the droplets and crystals. Due to the vapor pressure gradient, water vapor moves from the higher pressure surrounding the droplets to the lower pressure surrounding the crystals. This process generates sub-saturation with respect to water in the ambient air, and the droplets evaporate to maintain the ambient humidity near water saturation, making additional water vapor available for ice crystal growth. Finally, the cloud becomes glaciated.

‘Cloud-ice’ crystals are smaller than 0.3 mm and mostly can only grow by diffusion of vapor. Large ice crystals (‘snow’; 0.3mm to few centimetres) can grow through collision with supercooled droplets, this process being named accretion. The supercooled cloud-droplet freezes on contact and sticks to the ice crystal, which is known as ‘riming’. Extreme riming ultimately causes the formation of graupel (0.3 to 5 mm) and then hail (> 5mm). Graupel/hail particles are dense particles of ice precipitation because they are formed predominantly of rime. The fully-grown ice crystals can collide and stick together, a process is known as ‘aggregation’. Crystals with branches can mechanically interlock as well. This forms even bigger aggregates (Pruppacher and Klett 1978). Such unrimed (or weakly rimed) ice precipitation is termed ‘snow’ (large crystals or aggregates), which has maximum diameters greater than 0.3 mm.

1.3 Secondary ice production

In present-day times, there has been a renewed emphasis on understanding and quantifying primary ice nucleation (e.g., DeMott et al. 2011; Kanji et al. 2017; Knopf et al. 2018). Unexpectedly, the process of Secondary Ice Production (SIP) has not garnered equivalent attention, despite its overwhelming impact on in ice crystal concentrations in deep precipitating clouds, as inferred from aircraft observations (e.g., Field et al. 2017). Hence, the pioneering investigations into SIP carried out during the 1970s and 1980s retain their lasting pertinence (e.g., Hallett et al. 1978; Hobbs et al. 1980), exerting a continued influence on modern cloud modelling approaches.

Airborne observations reveal high concentrations of ice particles in precipitating ice clouds that are typically greater than those of active ice nuclei (IN) by orders of magnitude (Hallett et al. 1978; Mossop 1985; Hobbs and Rango 1985, 1990; Beard 1992; Blyth and Latham 1993). The reported high concentrations of ice are generally attributed to pathways of fragmentation of pre-existing ice, known as SIP processes (e.g., Hobbs and Alkezweeny 1968; Hallett and Mossop 1974; Griggs and Choularton 1986; Oraltay and Hallett 1989; Bacon et al. 1998; Sullivan et al. 2017,2018a). These occur in positive feedbacks involving the growth of ice fragments to become precipitation ('ice multiplication'), (Langmuir 1948). Therefore, it has long been proposed that a SIP process must able to quickly increase the number concentration of the ice following early primary ice nucleation events. SIP is vital for the prediction of ice concentrations.

The initial exploration of SIP through laboratory experiments, chiefly undertaken in the 1970s by pioneers such as Hallett and Mossop (1974) and Takahashi and Yamashita (1977), represents a foundational phase in this research. Laboratory experiments dedicated to SIP have unveiled numerous mechanisms pertaining to ice precipitation, frequently elucidating instances of ice fragmentation, often linked to the process of riming (Brewer and Palmer 1949; Hallett and Mossop 1974).

The prevailing consensus is that the Hallett-Mossop (HM) process operates within a temperature range spanning approximately −3 to −8 ºC. This phenomenon necessitates the coexistence of liquid cloud droplets with diameters smaller than 13 mm and liquid drops larger than 25 mm. Upon encountering substantial ice particles, these larger drops possess the capacity to undergo freezing.

In the laboratory, Vardiman (1978) presents compelling evidence suggesting that collisions between ice crystals have the potential to induce fragmentation. These findings receive substantial corroboration from a multitude of in-situ images taken within cloud environments, as exemplified by the research of Cannon et al. (1974). These images notably emphasize scenarios where dendrites have been observed to fracture due to collisions.

Observations have been made regarding the fragmentation of ice during the process of sublimation (Schaefer and Cheng, 1971; Oraltay and Hallett 1989; Bacon et al. 1998). These observations entailed an examination of individual ice crystals at different temperatures and in conditions of subsaturated humidity relative to ice.

Though several mechanisms have been postulated to explain the breakup of supercooled water droplets, the HM process of rime splintering stands as the prevailing choice in numerical models. The rate at which splinters are generated is contingent upon the quantity of accumulated supercooled liquid, giving rise to a mass-dependent relationship that is easy to include in models (Scott and Hobbs 1977; Beheng 1987; Mason 1996; Blyth and Latham 1997; Phillips et al. 2001; Clark et al. 2005; Connolly et al. 2006; Fridlind et al. 2007; Phillips et al. 2007; Dearden et al. 2016). Vardiman (1978) introduced a parameterization for supercooled droplet impact by crystal-to-crystal collisions, which has been utilized exclusively by Fridlind et al. (2007). Yano and Phillips (2011) incorporated secondary ice production into a theory about a cold cloud where collisions among large graupel particles (2 mm) cause glaciation, based on laboratory experiments reported by Takahashi et al. (1995). Ferrier (1994) proposed an empirical parameterization method to augment ice formation, providing the flexibility to tailor its impact within defined temperature ranges.

Ice particles are commonly found in significant amounts within convective clouds where temperatures are below 0°C. However, it is notable that cloud-top temperatures persist above approximately −12°C altitude (e.g., Hobbs and Rangno 1985; Cooper 1986; Lawson et al. 2015). Taylor et al. (2016) investigated aircraft measurements within maritime cumulus clouds characterized by colder cloud-base temperatures of approximately +11°C, which had formed over the southwestern region of the United Kingdom. Their research revealed that during the early stages of cloud development, nearly all initial ice particles consisted of frozen raindrops, measuring around 0.5 to 1 millimeter in size. As the clouds evolved, vapor-grown ice crystals became the dominant form of ice particles. These observations suggest that the freezing of drizzle and raindrops plays a crucial role in the formation of larger ice particles during the intermediate stages of cloud development.

Furthermore, in the more mature phase of cloud development, the study identified high concentrations of small ice particles within the HM process temperature range. Heymsfield and Willis (2014) observed a robust correlation between elevated concentrations of secondary ice particles within the HM temperature range in tropical cumulus clouds over the Caribbean and West Africa, particularly in regions with relatively weak vertical motions around ±2 m/s, and their findings indicated that the initial ice particles in these clouds were predominantly large frozen raindrops. Observations (in-situ) of ice fragments and non-pristine ice have led to the proposition that the augmentation of ice concentrations might involve the significant mechanisms of drop shattering and ice-ice collisional breakup, in addition to the previously recognized rime splintering within Arctic stratus clouds, as explored by Rangno and Hobbs (2001). Lawson et al. (2015) established a correlation between freezing supercooled drops with diameters in the millimeter range and the subsequent rise in ice number concentrations (secondary ice mostly). Furthermore, their findings indicate that the instantaneous freezing observed within vigorous updraft cores, with speeds reaching approximately 10 m/s, takes place at temperatures colder, and at a significantly faster rate, than can be explained by the HM process.

Hitherto, numerical modelling of clouds has usually included representation of only one process of ice multiplication, namely the HM process. This might be a reason for results about the effect of SIP on precipitation seeming small in some simulations of cases and seeming large for other cases (Field et al. 2017). If the other SIP processes, which are typically not adequately represented (e.g., sublimational breakup), indeed play a substantial role in certain types of clouds, their omission must be introducing substantial biases into the existing numerical models used for numerical weather forecasting.

2 Objectives

The significant increase often exceeding a thousand-fold in observed ice crystal number concentrations compared to natural INP concentrations highlights the critical role of SIP in cloud development. SIP has been observed in natural cloud formations, replicated in controlled laboratory settings, and incorporated into cloud models. In future, there will be a necessity to achieve a more comprehensive understanding and precise characterization of SIP from laboratory experiments and atmospheric observations, to improve its incorporation into models. Nevertheless, the current state of the science allows considerable insights from a combined approach of high-resolution modelling, lab observations to inform SIP treatment and field observations (e.g., by aircraft) in a given study such as ours.

This thesis encompasses the overarching goals of utilizing numerical cloud modelling to investigate secondary ice production. The aims of this research endeavour are as follows:

1. Create an empirical formulation of fragmentation during sublimational breakup on the basis of previous experimental studies and also record the performance and overall phase-space relationships of dependencies within this empirical formulation.
2. Simulate four basic cloud-types and validate the predicted cloud properties. Also, investigate whether the dominant warm rain or ice crystal precipitation mechanisms in a specific cloud types dictates the functions of distinct SIP mechanisms and rank their importance for each cloud-type.
3. Perform sensitivity tests with these simulations of four cloud-types (as outlined in Objective 2) to scrutinize the impact of environmental and microphysical properties on SIP for each cloud-type (ACAPEX, STEPS, GOAmazon and MC3E field Campaigns).
4. Are the coexistence of warm rain and ice crystal processes in clouds, and the equilibrium between these processes, influenced by cloud base temperature and solute aerosol conditions?

2.1 Hypotheses to test

In pursuit of Objective 3, the hypotheses to test are as follows:

1. Does any synergy exist among the various SIP mechanisms as they exchange secondary ice fragments? Is there a form of synergy wherein the positive feedback from one SIP mechanism could generate precipitation that interacts with other SIP processes?
2. How do these synergistic effects impact ice enhancement? Is the presence of such synergies influenced by the specific type of cloud?
3. How does altering the conditions of aerosol loading, such as those related to the land-ocean contrast, influence a given cloud type? Is the impact of this land-ocean contrast distinct for stratiform and convective clouds? Furthermore, does an increase in the abundance of INPs in the environment lead to greater ice enhancement within clouds?
4. Does the occurrence of homogeneous ice at higher altitudes influence SIP in the mixed-phase region (ranging from 0 to −36ºC) below? Additionally, is there an observable impact on the charge separation during storm electrification due to ice multiplication across a wide range of various cloud types? This was observed by Phillips et al. (2020) for a specific instance of continental cold-based convection.
5. The HM process has been observed in only a limited number of laboratory experiments. Do the aircraft data from a historical study (Harris-Hobbs and Cooper 1987), which claimed to observe anticipated correlations, still provide actual observational evidence for the existence of this phenomenon, in light of more recent findings (e.g., characterization of measurement biases and other SIP processes)?

3 Methods

3.1 Observational basis

In laboratory experiments, it has been noted that sublimational fragmentation is influenced by both the structure and size of the ice crystal (Oraltay and Hallett 1989; Dong et al. 1994; Bacon et al. 1998). Additionally, it is also affected by the ambient temperature and humidity levels during the sublimation process (Schaefer and Cheng 1971). Oraltay and Hallett (1989) exclusively considered dendritic crystals that were both larger than 3 mm in size and unrimed. Furthermore, they observed that plates and columns do not undergo fragmentation during sublimation when relative humidities are above approximately 80% and frost point temperatures range from 0 to -2°C. However, they did not investigate the sublimation of plates and columns at humidities below 70%. Currently, substantial knowledge gaps persist regarding the sublimational fragmentation of plates, columns, and needles.

Oraltay and Hallett (1989) demonstrated that the number of observed fragments for a specific initial size is influenced by the relative humidity in relation to ice (RH_i). Furthermore, there was no observation of fragmentation for dendritic crystals larger than approximately 3 mm when the relative humidity with respect to ice (RH_i) exceeded about 78% (Oraltay and Hallett 1989). Bacon et al. (1998) refrained from trying to discern the relationship between the breakup rate and RH_i because their frost particles were exceptionally small, each releasing only a single fragment and also, considerable variability existed in both the crystal shapes and RH_i levels within each experiment.

Dong et al. (1994) reported observations of rimed graupel sublimation, it was noted that the breakup rate tends to rise with the wind speed in the vicinity of the rimed particle. This phenomenon is in line with the definition, which states that the rate of mass change of an ice particle through vapor diffusion is directly linked to the ventilation coefficient.

3.2 Pooled datasets for dendritic snow and graupel

Based on insights garnered from previous laboratory investigations conducted by Oraltay and Hallett (1989) and Bacon et al. (1998), a combined dataset is constructed to examine the relationship between the number of fragments per parent particle, crystal size, and relative humidity (RH_i). Four distinct instances of observed fragmentation, stemming from sublimation processes without melting as observed by Oraltay and Hallett (1989) (Experiments 1-4 from Table 1). The initial crystal diameter for each case falls within the range of 3 to 8 mm, commonly classified as "snow" in cloud models. The measurement of the initial crystal diameter is based on the maximum dimension found in reported images by Oraltay and Hallett (1989). Besides that, Oraltay and Hallett (1989) reported that approximately half of the emitted fragments subsequently reattach to the same parent ice crystal.

In laboratory experiments, Oraltay and Hallett (1989) conducted controlled growth of dendritic crystals for experimental cases 1 to 4 in Table 1. They carefully regulated supersaturation, temperature, and wind speed to create ideal conditions. The dendritic crystals were grown within a temperature range of -14 and -17 ºC, with ice supersaturation levels ranging from 15% to 30%. However, due to the inability to vary wind speed to match the increasing fall speed during vapor growth and the lack of any realistic hydrodynamic aspects of fall behaviour, the crystal shape appeared unnatural. This was a result of diffusional growth and an unrealistic distribution of vapor density surrounding the crystal, causing branches to grow in the direction of the airflow. Each parent crystal was rotated at the start of the sublimation process to create an angle between 30° and 45° relative to the reverse wind direction to facilitate the counting of fragments released into the air during sublimation. The observations were reported at various relative humidity levels, ranging from 50% to 95%, and air temperatures between -6 and 5ºC (Oraltay and Hallett 1989).

Furthermore, an additional set of three cases (cases 5-7 from Table 1) were derived from the laboratory investigation conducted by Bacon et al. (1998). In this study, crystal sizes ranged from 50 to 250 μm, and we categorized them into three distinct size ranges: case 5 (200-300 μm), case 6 (100-200 μm), and case 7 (20-100 μm). Initially, ice (frost) formation occurred on the walls of the diffusion chamber due to supersaturated conditions. Subsequently, individual ice (frost) particles were cautiously extracted and transferred to another chamber with subsaturated conditions for the sublimation process. The crystals observed in this study were described as irregular and artificial dendrites (Bacon et al. 1998).

Tabel 1. The inferred number of ice fragments per crystal (N) represented here in pooled dataset from published laboratory studies (adapted from Deshmukh et al. 2022).

Experimental Studies	Experimental Cases	Relative humidity with respect to ice RH_i (%)	Parent crystal diameter or initial diameter d (mm)	Temperature T (K)	Duration of experiment t (s)	Equivalent to a natural crystal	
						Inferred sublimated mass M (kg)	Inferred number of fragments (N)
Oraltay and Hallett (1989)	1	57	6.8	274.35	320	6.99×10^{-7}	45
	2	70	5	274.15	780	9.14×10^{-7}	45
	3	75	5.5	274.15	780	1.17×10^{-6}	24
	4	78	3.6	270.65	480	4.01×10^{-11}	0
Bacon et al. (1998)	5	94	0.125	264.45	360	7.13×10^{-10}	0.4
	6	94	0.05	264.45	360	1.05×10^{-10}	0.24
	7	94	0.025	264.45	360	2.89×10^{-11}	0.1

In a previous laboratory study conducted by Dong et al. (1994), it was observed that 200 fragments were emitted from a graupel particle with an initial diameter of 5 mm. This observation took place under specific conditions, including a relative humidity of 70% with respect to ice and an air temperature of approximately −9°C. The duration of the sublimation process was recorded to be 320 seconds.

3.3 Observational Field Campaigns

With the aim of illustrating a diverse range of cloud types, four field campaigns were carefully chosen to encompass distinct categories including warm- and cold-based clouds exhibiting both convective and stratiform characteristics. The objective for incorporating these observed cloud instances is to enhance comprehension through numerical simulation. To accomplish this, the initial step involves comparing the simulations with observations to evaluate the model's authenticity. The subsequent sections provide a detailed explanation of each observational campaign along with its associated cloud features.

3.3.1 ACAPEX

The US Department of Energy (DoE) conducted an Atmospheric Radiation Measurement (ARM) Cloud Aerosol Precipitation Experiment (ACAPEX) field campaign was conducted. During the postfrontal scenario on February 7, 2015 and DOE G-1 aircraft collected cloud samples across the Sierra Nevada mountain range. Within this campaign, the ARM Mobile Facility 2 (AMF2) and the ARM Aerial Facility (AAF) Gulfstream-1 aircraft were strategically positioned, aligning with ACAPEX field study. The ACAPEX field campaign had a goal of enhancing comprehension and refining modelling of large-scale dynamics as well as the intricate coordination of cloud and precipitation processes entwined with atmospheric rivers (ARs). Additionally, the campaign explored into the interaction of aerosol-cloud interactions that shape the spectrum of precipitation patterns, from variability to extremes, across the enormous spread of the western USA.

Observations showed the occurrence of dispersed low-level clouds, mainly orographic and primarily stratiform in nature. These clouds revealed a dense cloud base situated approximately 1.2 km above the mean sea level (around 8°C), positioned ahead of the mountains as documented by Leung (2016). The mountains stretched up to a base elevation of approximately 2.1 km above mean sea level (MSL) around −2°C. The south-westerly flow from the Pacific Ocean crossed the central valley. It then ascended over the mountain range, generating a broad stratiform cloud layer. This cloud layer had a cloud base situated upstream over the valley and extending to meet the elevated terrain over the mountains.

3.3.2 GOAmazon

Green Ocean Amazon (GOAmazon) campaign was primarily focused on toward the study of the interactions between clouds and aerosols within clouds that develop in the Amazon Basin. Also, the ARM Climate Research Facility stationed within the Amazon Basin aimed to unravel the life cycles of aerosols and clouds. It particularly focused on understanding the vulnerability of clouds to interactions with aerosols that impact precipitation. The ARM Mobile Facility was positioned leeward of Manaus, Brazil (3° 6' 47" S, 60° 1' 31" W), near Manacapuru, from January 2014 to November 2015.

On the 19th of March 2014, a mesoscale convective system (MCS) manifested from 0500 to 1900 UTC. Within the Amazon region, the occurrence of MCSs is a frequent phenomenon during March (Romatscke and Houze 2013; Rehbein et al. 2018, 2019). The clouds that were studied during the GOAmazon initiative exhibited notably warm bases (~25°C), situated around 990hPa. Radiosonde data showed an exceptionally high Convective Available Potential Energy (CAPE) of approximately 5000 J kg^{-1} (Gupta et al. 2023). The vertical shear of the mean flow followed a south-westerly direction, increasing by 22 m/s from 3 to 13 km altitude above MSL. This increase was attributed to an easterly jet in the lower troposphere and a westerly return flow aloft.

3.3.3 STEPS

The Severe Thunderstorm Electrification and Precipitation Study (STEPS) was conducted near the border of Colorado and Kansas from May to July in the year 2000. The goal of this campaign was to enhance comprehension regarding the intricate interplay among kinematics, the generation of precipitation, and the electrification processes within severe thunderstorms.

On the 19th of June 2000, a cold-based multicellular convective storm was observed near the border of Kansas and Colorado, during STEPS campaign (Lang et al. 2004). This multicellular system exhibited a cloud base near 0°C, featuring various cloud-types including cirriform, stratiform, and cumuliform. In-flight observations unveiled a notable scarcity of supercooled raindrops at higher altitudes, hinting at precipitation stemming from the ice crystal process. The South Dakota School of Mines and Technology's armoured T-28 aircraft conducted measurements for the campaign, sampling the vigorous convective updrafts.

Phillips et al. (2017b) delineated the microphysical properties of this specific convective system as simulated by AC, comparing them with coinciding in-situ aircraft observations, satellite data, and ground-based observations. There was good agreement with observations including the ice concentration. The lower troposphere conditions were dry, caused a lower CAPE.

3.3.4 MC3E

The collaborative research campaign named, Midlatitude Continental Convective Cloud Experiment (MC3E) was co-led by the U.S. Department of Energy's Atmospheric Radiation Measurements (ARM) program and the National Aeronautics and Space Administration's (NASA) Global Precipitation Measurement (GPM) mission. The study focused on the vicinity of the DOE ARM's Southern Great Plains (SGP) Central Facility (CF) and the region of north-central Oklahoma. This extensive campaign collected data from 22 April 2011 to 6 June 2011, incorporating both aerial in-situ and ground-based measurements (Jensen 2016).

On May 11th, 2011, warm-based MCSs were observed over Oklahoma during MC3E campaign. These MCSs had a cloud base situated around 17ºC, located at an elevation of approximately 1.2 km above the ground. Radiosonde observations conducted around 0300 UTC on May 10th showed a CAPE of approximately about 3500 J kg^{-1}. The vertical shear has a modest increase of about 6 m/s from 3 to 13 km above the ground. However, the increase within the lowest 3 km was approximately 13 m/s. This multicellular convective line comprised a mixture of cloud types, with cumuliform (such as cumulonimbus) and stratiform clouds being predominant (Waman et al. 2022).

3.4 AC model

The AC model was developed by Phillips et al. (2009) that incorporates various aerosol species into their cloud-resolving model (CRM). Constructed within the framework of the Weather Research and Forecasting (WRF) software set-up, this model signifies the convergence of cloud microphysics and aerosol chemistry through a two-moment bulk microphysics scheme. The involved microphysical species are cloud liquid, cloud ice/crystals, snow, graupel/hail and rain.

From this “two-moment” scheme, the AC model proficiently predicts the advection and diffusion of total mass and number mixing ratios for hydrometeors within each microphysical category. Furthermore, the microphysical processes are facilitated through an emulated bin microphysics approach (Phillips et al. 2007, 2009, 2013, 2015, 2017a, b, 2020).

Within the framework of AC, the nuanced phenomenon of preferential evaporation of smaller cloud droplets in cloud liquid, particularly near −36°C during homogeneous freezing, is adeptly addressed (Phillips et al. 2007).

SIP processes are included in AC, described as follows:

- HM process of rime splintering
- Ice-ice collision fragmentation
- Raindrop-freezing fragmentation
- Sublimational breakup

Ice fragmentation is linked to the growth of ice particles through the process of riming. Graupel and hailstones are examples of ice particles that undergo riming, leading to increased density. The HM process encompasses the phenomenon of ice particle splintering and is operative within the temperature range of −3°C to −8°C. For this process to occur, there is a requirement for a diverse array of in-cloud droplets with sizes exceeding 24 μm and impacting at a minimum speed of 0.2 m/s within this specified temperature range, as described by Hallett and Mossop (1974).

Ice particles tend to undergo fragmentation as a consequence of their collisions, giving rise to the formation of secondary ice particles. Vardiman (1978) indicated that interactions between ice particles, specifically ice-ice collisions, have the potential to generate splintering within natural cloud environments. The initial stages of model development paved the way for simulating collisions between ice crystals in the context of Arctic cloud simulations (Fridlind et al. 2007). Phillips et al. (2017a) formulated a comprehensive framework for describing the fragmentation occurring during ice-ice collisions, elucidating the relationships among various microphysical elements, including crystals, snowflakes, graupel, and hailstones.

Regarding raindrop-freezing fragmentation, an empirical model was constructed to encompass all sizes of naturally occurring rain and drizzle drops across the entire spectrum of sub-zero temperatures within the mixed-phase region. This model was established based on data collected from laboratory experiments involving drops in free-fall, as reported in published studies (Phillips et al. 2018). Phillips et al. (2018) proposed two distinct modes of fragmentation that occur during collisions between substantial water droplets and ice particles (mode 1) and a collision of raindrops and massive ice particles (mode 2). As for Mode 1, the incidence of raindrop-freezing fragmentation is comparatively constrained when temperatures stray from the optimal range, typically situated around −15°C, or when dealing with tiny droplets. (e.g., drop size smaller than 0.1 mm). Their bin model and 0D theory show that it is possible to reach ice enhancement ratios ranging from 100 to 1000. Nonetheless, within Mode 2, raindrop-freezing fragmentation holds greater significance across a broader spectrum of freezing temperatures, extending well beyond −15°C when compared to Mode 1. In addition, it is important to note that Mode 2 shows an acute sensitivity to the size of the droplets involved.

Regarding sublimational breakup, Deshmukh et al. (2022) formulated an empirical model based on laboratory experiments, which was then integrated into the AC. The formulation effectively characterizes the sublimational fragmentation tendencies of graupel and dendritic snow particles, factoring in various influential factors, including particle size, temperature, and sublimation rate. A theoretical scaling analysis of a weak deep convective downdraft with a speed of 2 m/s, coupled with an initial population density of 3 L^{-1} and uniform-sized dendritic snow particles measuring 2 mm, and graupel particles, provides an estimated ice enhancement ratio of between 5 and 10, correspondingly.

3.5 Experimental setup

The particular orographic stratiform cloud scenario was reproduced using data from the ACAPEX campaign, which took place in the vicinity of the Sierra Nevada mountains near Sacramento airport. The simulation was conducted over a 360 × 80 km area for a duration of 3 hours, considering in the Sierra Nevada mountainous terrain. Commencing at 1915 UTC and concluding at 2215 UTC, the simulation domain was adjusted by a 31º counter clockwise rotation to align it with the prevailing wind direction from the western inflow boundary. Most of the simulation domain encompasses the mountain range where the stratiform cloud formation is occurred.

In the case of STEPS, the same multicellular convective system was recreated, employing the model configuration as detailed by Phillips et al. (2017b). In brief, the horizontal grid spacing in the model is set at 1 km, while the vertical grid spacing is approximately 0.5 km, and the time step used is 10 seconds. The convective line progresses downwind toward the east-northeast direction, which is 71º clockwise from the north. Consequently, the domain and coordinate system (x and y) within the model were adjusted to align the x-axis with this direction, thereby making the convective line parallel to the y-axis.

The observed case of MCS in MC3E campaign has been simulated within a domain comprising a grid of 80 × 80 km and using a time step of 10 seconds. The simulation extends for a duration of 72 hours, spanning from 0000 UTC on May 10th to 0000 UTC on May 13th, 2011. Across all altitude levels, a uniform vertical model resolution of approximately 0.5 km is maintained. For a more comprehensive understanding of the experimental setup for simulating MCS by AC, detailed information can be found in Waman et al. (2022).

For convective clouds with a very warm base in the GOAmazon region, the AC model employs a horizontal grid spacing of 2 km and a vertical spacing of approximately 0.5 km. Simulations are conducted with a time step of 10 seconds within a domain spanning 320 x 80 km. The simulation of the convective storm begins at 0500 UTC and concludes around 1900 UTC on March 19th, 2014. The initialization conditions are explained in papers II and IV. Vertical profiles of aerosol loadings for each species were initialized by rescaling global model output for month and location of each case, ensuring agreement with ground-based observations (e.g., IMPROVE). More details about every simulated case are given in papers II and IV.

3.6 Sensitivity tests

There were two methods for analyzing the simulations regarding the various types of SIP mechanisms. Firstly, the realism of the control simulation of each case was established by comparison with coincident observations by aircraft and ground-based instruments. This was followed by analysis of tagging tracers (Paper II). Secondly, sensitivity tests were performed to answer the question of how cloud properties are truly impacted by inclusion of the change in environmental factors. More details of the sensitivity simulations are given in Paper III.

Sensitivity experiments were conducted to explain the causal impact of ice multiplication, both in the context of its collective influence encompassing all SIP processes, and the influence of each individual SIP process in isolation. These tests aimed to answer questions regarding how cloud microphysical properties would manifest if, hypothetically, the target processes were absent or if the environmental aerosol conditions were very different. Through these sensitivity tests, we uncover the genuine causal effects within a broader context, considering the presence of the target multiplication mechanisms and the potential interaction with other processes, including any compensatory mechanisms.

One facet of these tests is the quantification of synergy among ice multiplication mechanisms, where they may share microphysical positive feedbacks, potentially leading to splinters. Additionally, some tests explore how the impact on cloud properties stemming from the contrast in aerosol conditions between land and ocean is either dampened or intensified by ice multiplication.

In control simulations, all SIP processes are active. Conversely, in a sensitivity simulation, all SIP processes are intentionally disabled artificially. Furthermore, we conduct runs where we individually assess the impact of each SIP mechanism by isolating them. Each perturbation simulation carried out using the AC model involves only modifying the target processes, while keeping all other aspects identical to their corresponding control runs.

A wide spectrum of atmospheric conditions, spanning from extremely pristine environments to scenarios featuring polluted air. To assess the contrast in CCN aerosol concentration between land and ocean regions, we apply a factor that varies with altitude, which changes the aerosol content in the environment used as the control run. The most extensive alterations are observed near the Earth's surface, with changes of up to an order of magnitude, both above and below, in each of the control simulations.

This study primarily delves into the sensitivity linked to the process of homogeneous freezing of cloud liquid during ascent. A novel sensitivity test simulation is designed to intentionally omit this process. By comparing the outcomes of this sensitivity test with those of the corresponding control run, the influence of homogeneous freezing on the ice multiplication processes can be determined.

4 Results

4.1 Empirical formulation of sublimation breakup

Fig. 1 depicts a comprehensive representation of the sublimated mass, denoted as "M," as a function of the number of fragments, referred to as "N". The data (from Table 1) encompasses various laboratory experiments conducted on an equivalent natural crystal, all of which have the same size. The curve that best aligns with the observations in Fig. 1 can be expressed by the following equation,

$$N = K\, M^{\alpha} \tag{1}$$

wherein the constants K and α have been empirically derived. This relationship is here termed as the 'sublimated mass activity spectrum'.

In this context, we establish a parameter $M = m_0 - m(t)$, where m_0 signifies the initial mass of the particle or crystal at the commencement of sublimation and $m(t)$ is its mass after that time 't'. Differentiating with respect to time, we have

$$\frac{dN}{dt} = \alpha K M^{(\alpha-1)} \frac{dM}{dt} \tag{2}$$

Nonetheless, the rate of mass transformation throughout sublimation exhibits a direct proportionality to the crystal's instantaneous size 'd', supersaturation and the ventilation factor (f_v) (Rogers and Yau 1989):

$$\frac{dM}{dt} = A\, d(100 - RH_i\,) f_v \tag{3}$$

Now, merging Eqs (2) and (3) give the following formulation of sublimation breakup:

$$\frac{dN}{dt} = \alpha \widehat{K}\, M^{(\alpha-1)}\;\; A\, d(100 - RH_i) f_v\;\; \Xi\, \nu \tag{4}$$

The emission factor, $\Xi = \Xi(d)$, has been introduced to symbolize the fraction denoting the number of fragments formed that undergo permanent emission into the surrounding air, where $K = \Xi \nu K\hat{}$. Also $\nu = \nu(RH_i, d)$ denotes the threshold of critical RH_i, signifying the point at which no fragmentation has been observed by Oraltay and Hallett (1989).

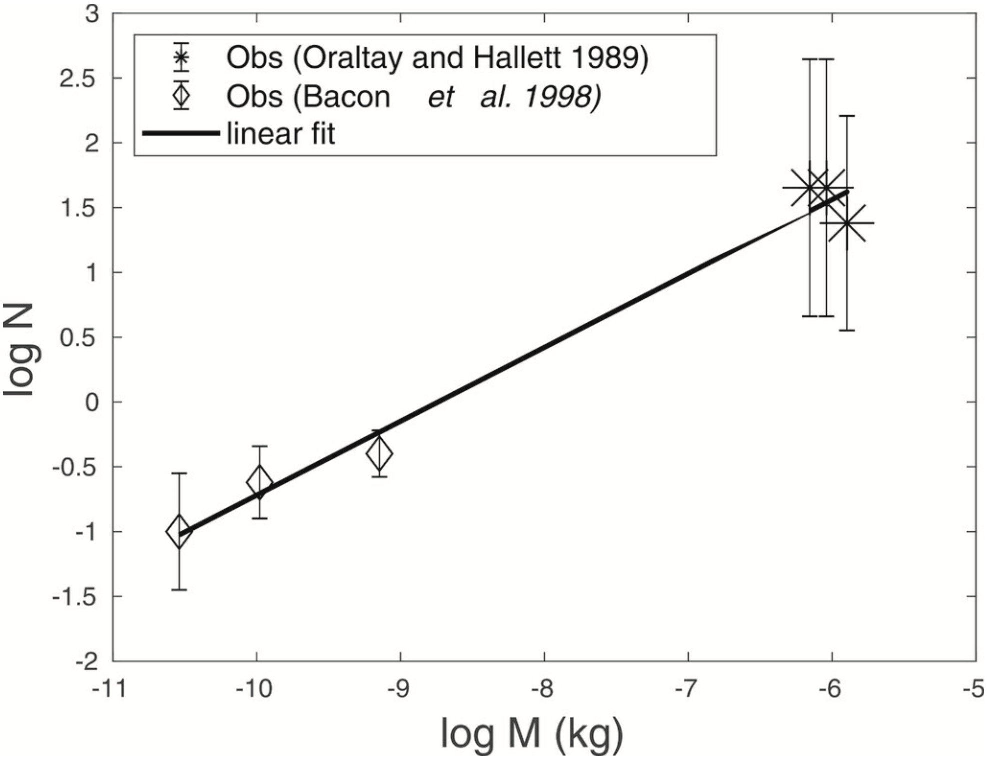

Figure 1 Relationship of sublimated mass '*M'* and the observed number of fragments '*N*' is displayed (adapted from Deshmukh et al. 2022).

In the scenario where the majority of fragments are actively being emitted, it can be inferred that M nearly approximates a constant multiplied by a certain exponent of d among an ensemble of sublimating particles spanning a broad spectrum of sizes. So, a simplified version of the empirical formulation can be expressed as follows:

$$\boxed{\frac{dN}{dt} \approx \beta d^{\gamma}\, d(100 - RH_i)f_v\ \Xi\, \nu} \tag{5}$$

Figure 2 presents a three-dimensional visualization illustrating the phase-space representation of the sublimational breakup formulation across various initial crystal diameter and RH_i conditions. $\frac{dN}{dt}$ demonstrated here with and without the inclusion of the emission factor Ξ in Figs. 2a and b respectively. Generally, $\frac{dN}{dt}$ shows a gradual intensification as initial crystal size increases with RH_i. Fig. 2c displays a

3D visualisation of the formulation for the set of conditions of graupel (rimed particles). Also, $\frac{dN}{dt}$ represented with an inclusion of emission factor Ξ. $\frac{dN}{dt}$ intensifies with increasing size of the rimed particles and with falling RH_i .

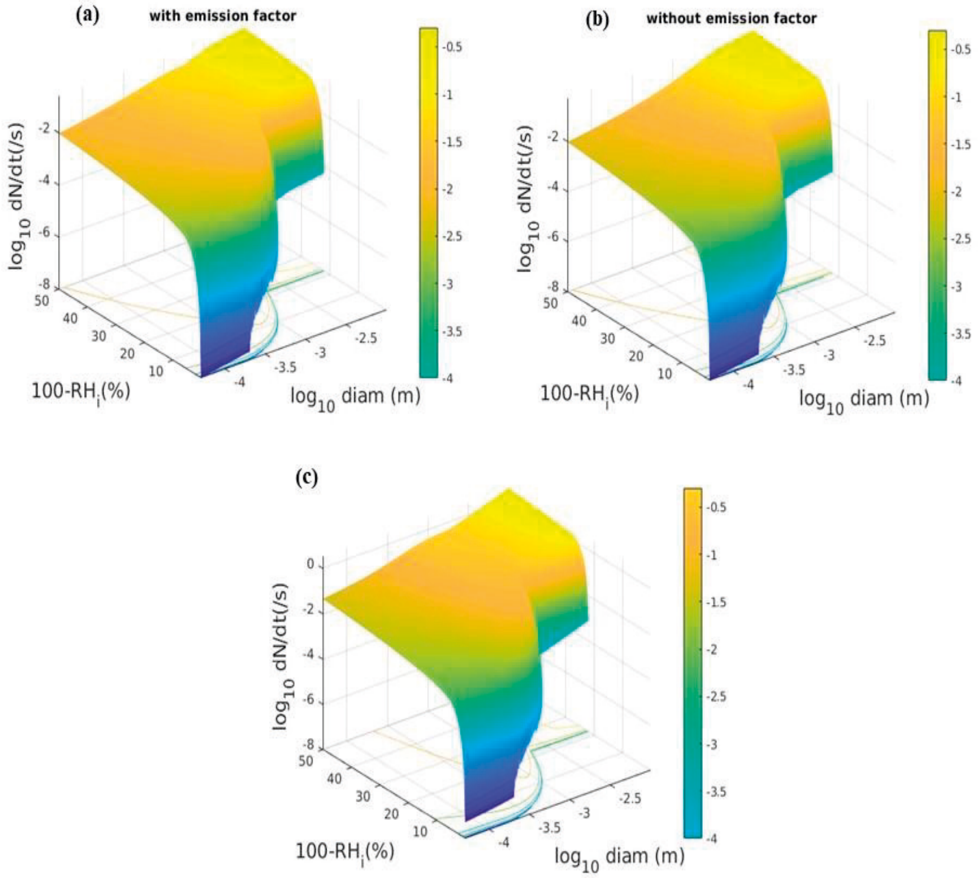

Figure 2 The 3D conception of the formulation for the over a wide range of conditions of d and RH_i (a) with the emission factor and (b) without the emission factor for ice crystals, (c) with emission factor for rimed particles (adapted from Deshmukh et al. 2022).

The new formulation is specifically applicable to dendritic crystals and heavily rimed particles. Moreover, an idealized scenario involving subsaturation during in-cloud descent is carried out using a thought experiment. An estimate is derived through the scale analysis, indicating an ice enhancement ratio of roughly 5 (for dendritic snow) or 10 (for graupel) within a moderately weak deep convective downdraft, approximately 2 m/s. This estimate pertains to an initial monodisperse population of dendritic snow or graupel particles, sized at 3 L^{-1} and 2 mm.

Throughout this descent, there exists a dynamic equilibrium between the continuous emission of fragments and their depletion via sublimation. A simplified bin microphysics parcel model effectively portrays this dynamic quasi-equilibrium, aligning with the thought experiment. The fragments exhibit average lifetimes of approximately 90 seconds for dendrites and 70 seconds for graupel. The projection is that sublimational breakup will lead to noteworthy secondary ice production. More detailed results are given in Paper I.

4.2 SIP and basic cloud types

In this part, the analysis shows the distribution of cloud glaciation in a two-dimensional space defined by sub-zero temperature and vertical velocity. This aims to facilitate the comparison of ice multiplication mechanisms across different basic cloud types. The $'w - T\ map'$ is used as an analytical tool to illustrate that how SIP mechanisms align with the fundamental thermodynamic variables that characterize cloud types. Specific microphysical regimes are observed to favor each of these mechanisms.

In Figure 3, the $w - T$ maps of the total IE ratio are presented. The IE ratio exhibits a minimum value in the stratiform cloud scenario with weak ascent, especially when ascent is nearly zero in the ACAPEX case. In the case of the slightly cold-based stratiform clouds of ACAPEX, the IE ratio shows a significant ice enhancement, exceeding a factor of 10^3 in the lower half of the mixed-phase region (from 0 to -15 ºC) and reaching a factor of 10^1 in the upper half (Fig. 3a). In the case of the exceptionally warm-based convective clouds observed in GOAmazon, a distinctive maximum region in the IE ratio spans the entire spectrum of vertical velocities, encompassing both ascent and descent (Fig. 3d). This maximum region results from a combination of processes, including the HM process, breakup in ice-ice collisions, and sublimational breakup.

The HM process, with rime splintering, greatly increases ice formation, especially between 0 and -10°C, reaching a peak enhancement ratio of up to 10^3 in the stratiform clouds of ACAPEX. However, in the upper half of the mixed-phase region, the HM process has a limited impact on ice enhancement. Comparatively, the HM process displays higher activity than both raindrop-freezing fragmentation and sublimational breakup across the entire $w - T$ map for slightly cold-based stratiform clouds. In the stratiform ascent, an IE ratio ranging from 3 to 10 is observed, primarily attributed to sublimational breakup splinters originating from the descent.

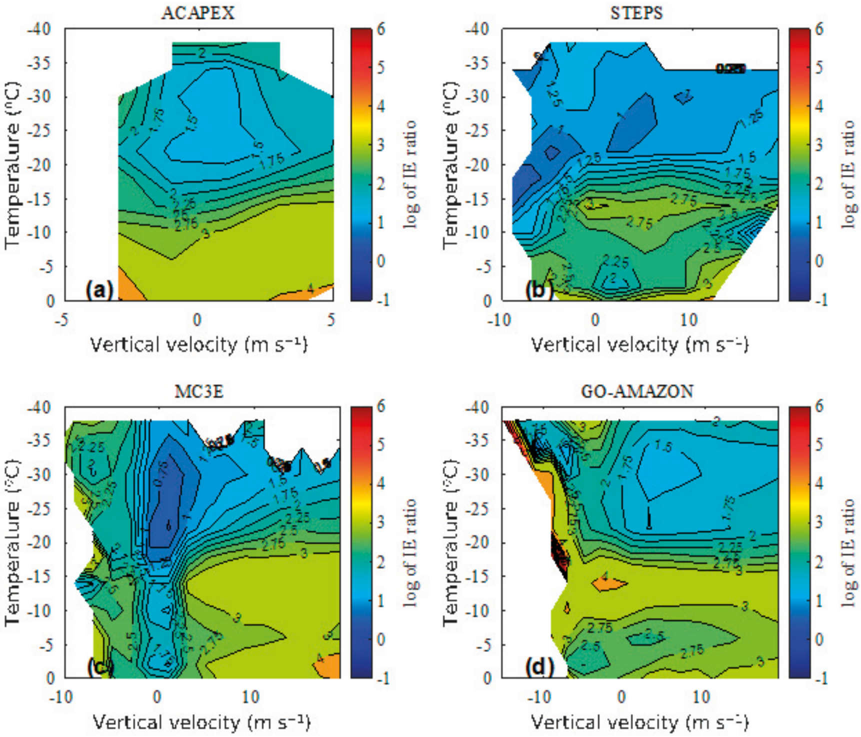

Figure 3 Total IE ratio for (a) ACAPEX, (b) STEPS, (c) MC3E and (d) GOAmazon are shown on the $w - T$ map (adapted from Paper II).

Breakup in ice-ice collisions is the dominant process for non-homogeneous ice initiation in these cold-based clouds, with the majority of fragments clustering around the −15°C peak, resulting in an IE ratio peak of approximately 10^2. In the case of cold-based convective clouds of STEPS, ice-ice collisions are the predominant SIP mechanism that makes a substantial and effective contribution.

In warm-based clouds of the MC3E case, IE ratios from raindrop-freezing fragmentation are less pronounced compared to that from the HM process. Breakup in ice-ice collisions reaches an IE ratio peak of approximately 10^3, similar to the other scenarios, occurring around −15ºC. Meanwhile, sublimational breakup leads to an IE ratio of up to around 10^2 in the convective downdrafts, particularly in the upper half of the mixed-phase region, but it contributes minimally to ice enhancement during ascent.

For very warm-based clouds of GOAmazon case, breakup due to ice-ice collisions exhibits a unique pattern in the IE ratio. It shows a minimum near 0 m/s vertical velocity and displays minimal dependence on updraft speed. Additionally, there is an IE ratio peak of 10^3 observed at approximately −15°C. Sublimational breakup yielded an IE ratio of 10^3 within convective downdrafts and the HM process portrays a maximum IE ratio of 10^3.

In Figure 4, a pie chart illustrates the distribution of relative numbers of ice particles that originate from primary ice and SIP mechanisms in all four simulated cloud cases. In cold-based convective clouds of STEPS, ice-ice collisions leading to breakup account for approximately 96% of the total ice particles initiated in the overall storm budget. In STEPS, sublimational breakup ranks as the second most significant contributor with respect to the initiation of ice particles, with other mechanisms being nearly negligible. The percentage of ice particles initiated by breakup in ice-ice collisions is decreased from cold to warm base clouds. The particles initiated by sublimational breakup are comparable to breakup in ice-ice collisions. The HM and raindrop-freezing fragmentation processes initiated less ice particles.

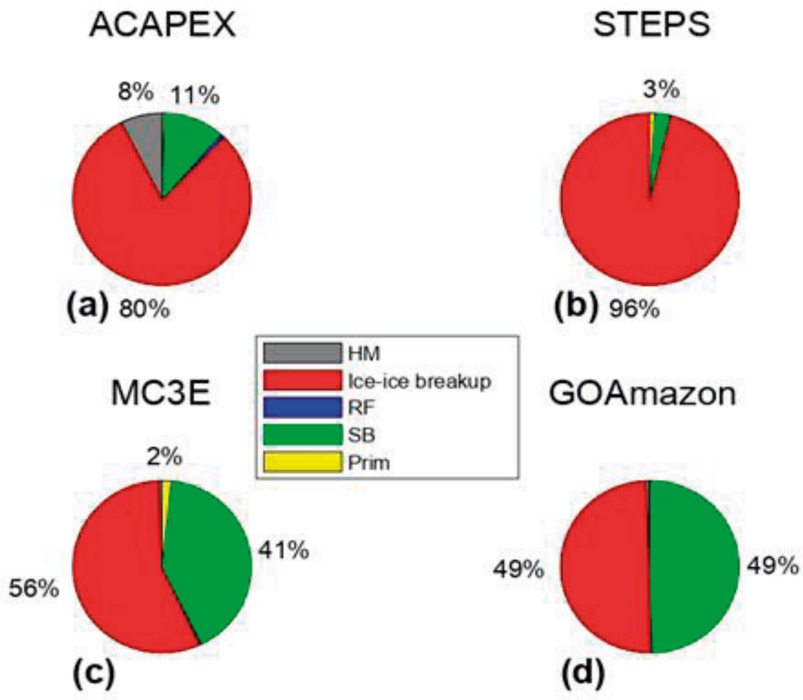

Figure 4 Pie chart represented the percentage contribution from different SIP processes and primary ice (heterogenous) for the following cloud-cases (a) ACAPEX, (b) STEPS, (c) MC3E and (d) GOAmazon (adapted from Paper II). The HM process ('HM'), breakup due to ice-ice collisions ('ice-ice breakup'), raindrop-freezing fragmentation ('RF'), sublimational breakup ('SB') and primary ice ('prim') included here.

Figure 5 depicts pie charts illustrating the distribution of rain components categorized into warm rain processes (represented by 'yellow') and ice crystal processes (depicted as 'grey'). In warm-based stratiform and convective clouds, warm rain processes contribute more significantly than ice crystal processes (as shown in Figure 5a and Figure 5d). However, in both cold-based and warm-based clouds, ice crystal processes take precedence in precipitation generation. This is because warm rain processes in the lower half of the mixed-phase region promote the HM process and raindrop-freezing fragmentation. Warm-based convective clouds support all SIP mechanisms, as processes like raindrop-freezing lead to the formation of additional graupel. However, in cold-based convective clouds, most SIP mechanisms are suppressed. The cloud base plays a crucial role in determining the intensity of warm rain and ice crystal processes within the clouds. Nevertheless, note that a warm cloud base alone is not sufficient for warm rain processes to prevail; aerosol conditions also exert significant influence, although aerosol influence is not addressed in this context. In our simulations, a cloud base exceeding about 20ºC is a necessary condition for a cloud system to be dominated by warm rain processes when continental aerosols are involved. In convective clouds with very warm bases and under continental aerosol conditions, ice enhancement resulting from all SIP mechanisms surpasses that observed in other types of clouds.

Also, we investigated the contribution of warm rain and cold in these clouds, distinguishing warm and cold components of surface precipitation and understanding the impact of different physical processes are crucial for enhancing the accuracy of global climate models. In STEPS and MC3E, cold precipitation components dominate significantly, outweighing warm components by orders of magnitude in mass and number mixing ratios for graupel and rain. Conversely, in the tropical GOAmazon case, warm precipitation components prevail, boosted by high cloud base temperature and maritime CCN conditions. The control simulations in STEPS and MC3E reveal a majority of surface precipitation from the ice crystal process, while warm rain contributes only 20%. However, in GOAmazon, warm rain dominates (70%) over the entire mesoscale domain. Sensitivity tests show a dynamic interplay between warm and cold precipitation components. Lowering cloud base enhances warm components significantly, and reducing both cloud base and CCN concentrations together leads to the most substantial increase in warm components. When secondary ice production mechanisms are turned off, warm components' contribution rises substantially, altering precipitation dynamics notably. In GOAmazon, heightened CCN and solid aerosol concentrations decrease warm rain contributions due to smaller cloud droplets and weaker LWC aloft (Gupta et al. 2023; Paper IV).

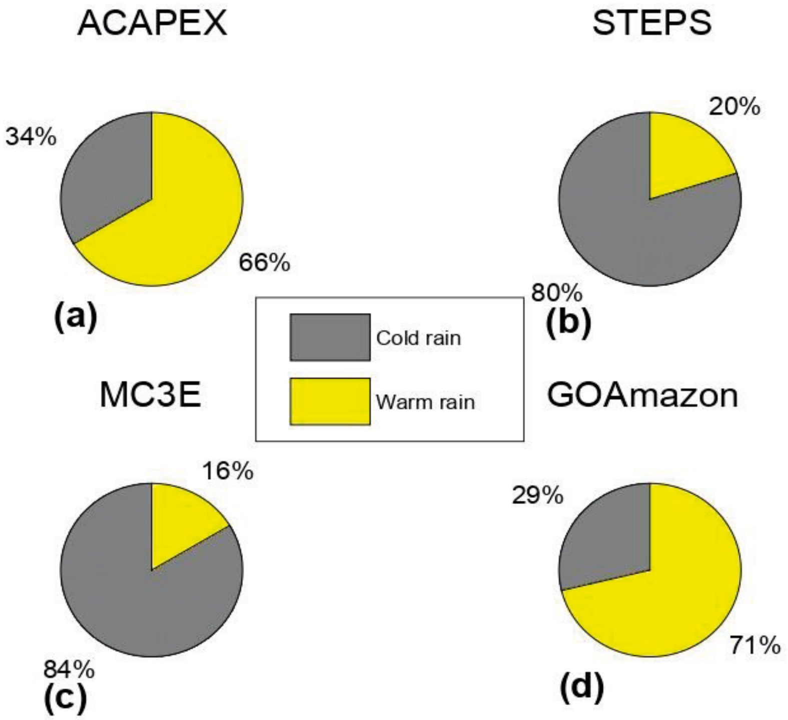

Figure 5 Pie chart represented the percentage of rain from warm rain (yellow) and ice crystal processes or cold rain processes (grey)for the following different cloud-cases (a) ACAPEX, (b) STEPS, (c) MC3E and (d) GOAmazon (adapted from Paper II).

The IE ratio for ice-ice collision breakup has a minimum near zero ascent (0 m/s vertical velocity) and a peak of 10^3 around -15°C in warm and very warm-based convective clouds regardless of ascent. Similarly, the total ice concentration dominated by ice-ice collision breakup in slightly cold-based stratiform clouds has a minimum at zero ascent, similar to weak ascent in convective cases. In all three convective cases, a deep peak in IE ratio centred around -15°C to -25°C intensifies with convective ascent due to the breakup in ice-ice collisions on $w - T$ maps. This peak is less pronounced in slightly cold-based stratiform clouds, showing a weak rise in ice concentrations with increasing vertical velocity and height (Figs. 18-20 from Paper II).

4.3 Sensitivity tests for ice multiplication

Sensitivity tests were conducted to assess the impact of environmental factors, including aerosol conditions and microphysical processes, on secondary ice formation, alongside the evaluation of the interplay of SIP processes and the connection between ice multiplication and storm electrification. When examining individual SIP mechanisms (the HM process, raindrop-freezing fragmentation, and breakup in ice-ice collisions) in isolation, ice number concentrations increase by approximately 0.5 to 2 orders of magnitude below the −25ºC level for ACAPEX, MC3E, and GO-Amazon cloud cases when each is included compared to the prohibited SIP process run (no-SIP run). However, their impact becomes minimal above the −25ºC level in the convective region of all four cases due to upper-level homogeneously nucleated ice. Conversely, excluding each SIP mechanism from the control run leads to a reduction in ice concentration by up to 1.0, 0.1, 1.5, and 0.3 orders of magnitude for the HM process, raindrop-freezing fragmentation, ice-ice collision breakup, and sublimational breakup, respectively (Figs. 2 and 3 in paper III).

Figure 6 presents a contrast between ice concentrations in the control-run and CCN-altered runs. The lower half of the mixed-phase region in all four cases exhibits minimal sensitivity. This is due to the fact that while CCN activation does influence cloud-droplet concentrations and mean sizes, the impact is overshadowed by the ice concentration being determined predominantly by ice-ice collision-induced breakup. In the lower portion of the mixed-phase region within GOAmazon clouds, a reduction in CCN leads to an elevation in ice concentrations, with an increase of up to a factor of 2. This phenomenon is primarily attributed to the HM process, which relies on the presence of large cloud droplets.

Figure 7 illustrates the impact of CCN sensitivity when all secondary ice formation processes are prohibited across all altered CCN simulations. Comparing Figures 6 and 7, within the context of SIP, changes in CCN concentrations have only a modest impact on the HM process and minimal influence on other SIP mechanisms. The main effect is observed in the concentrations of homogeneous ice within the upper portion of the mixed-phase region. In the absence of SIP mechanisms, ice concentrations in the lower half exhibit a fractional sensitivity for CCN variations as compare to no-SIP run but are significantly smaller in magnitude.

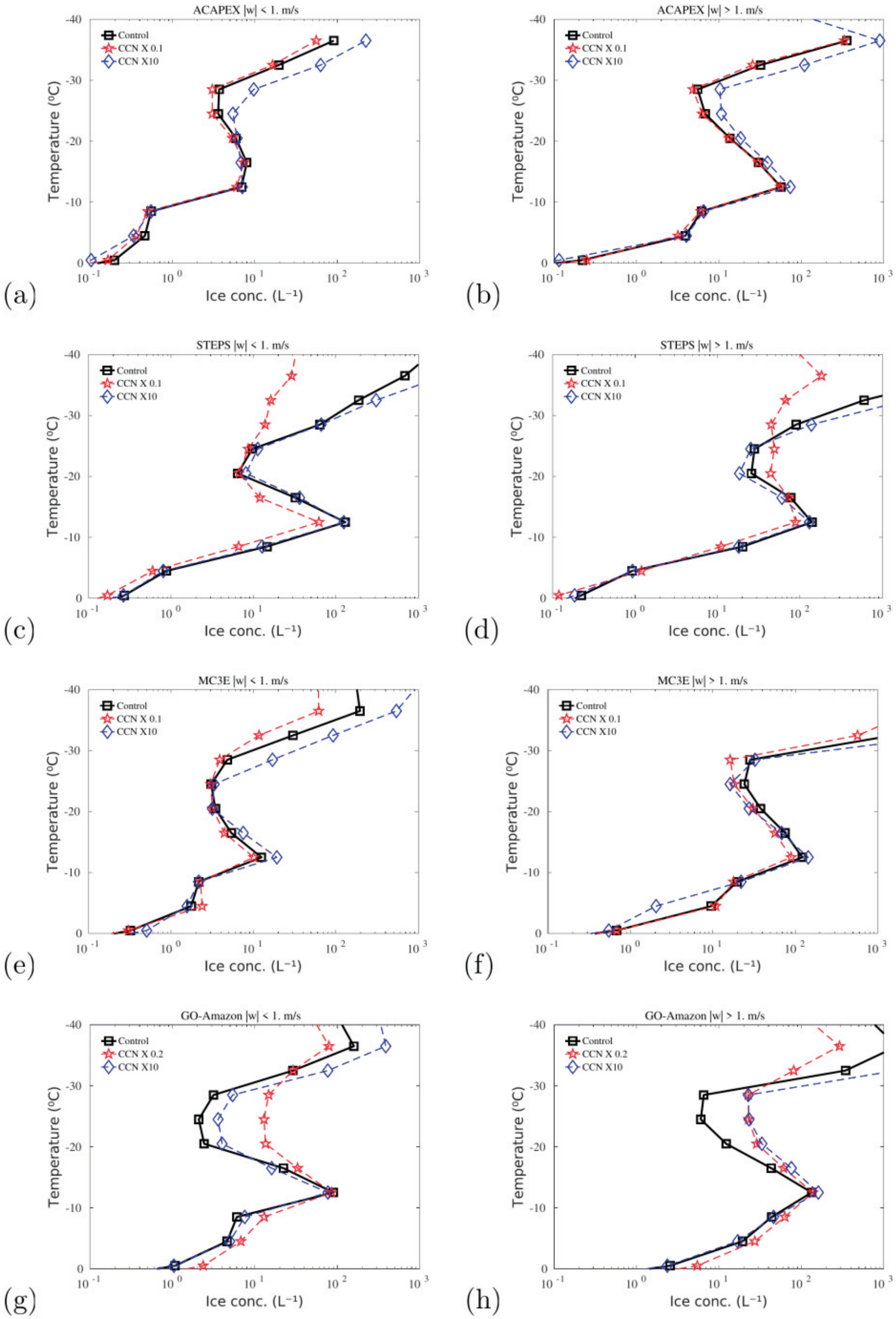

Figure 6 Vertical profiles of ice number concentrations for the stratiform and convective regions of the (a, b) ACAPEX, (c, d)STEPS, (e, f) MC3E, and (g, h) GOAmazon cases from No-SIP run (hollow squares), CCN× 0.1/0.2 No- SIP run (pentagram), CCN× 10 No- SIP run (diamond) represented here (adapted from paper III).

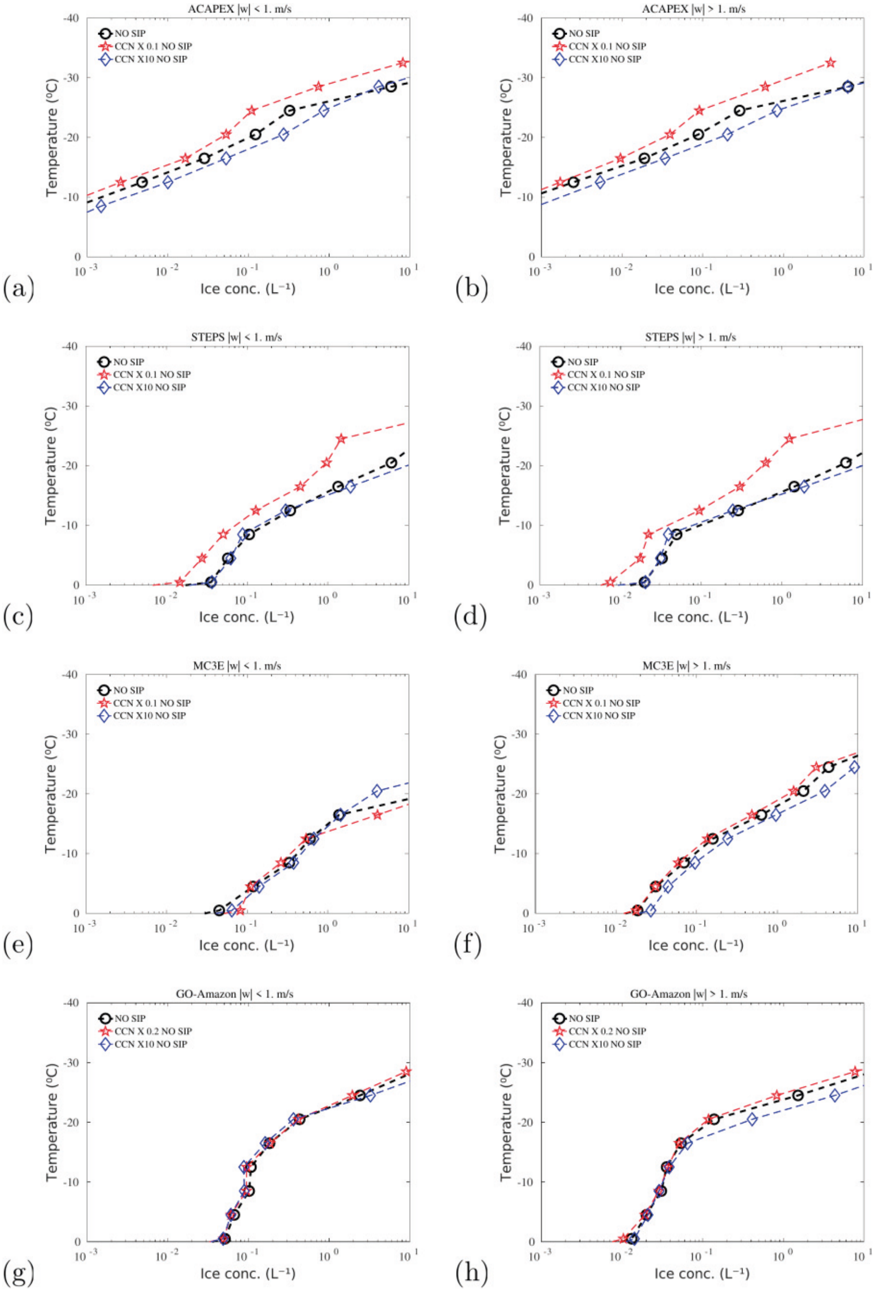

Figure 7 Vertical profiles of ice concentrations from no-SIP (hollow circles), CCN× 0.1/0.2 no-SIP run (pentagram), CCN× 10 no-SIP run (diamond) are presented here for the stratiform and convective regions of the ACAPEX (a, b), STEPS (c, d), MC3E (e, f), and GOAmazon (g, h). no-SIP indicates that the SIP processes are artificially prohibited (adapted from paper III).

Similarly, the sensitivity tests for altered IN concentrations are performed. Ice multiplication mechanisms damped the sensitivity to altered IN concentrations. Without ice multiplication, ice concentrations in the lower half of the mixed-phase region exhibit a strong response to changes in IN concentrations, leading to variations in ice concentration spanning from 0.5 to 3 orders of magnitude compared to the scenario with artificially prohibited SIP processes.

In summary, this assessment delves into the sensitivity of ice multiplication mechanisms concerning a range of factors, including microphysical and environmental conditions, as well as the influence of the presence or absence of one or more secondary ice production processes. More results are given in Paper III.

4.4 Revisiting historical measurements to detect HM process

An attempt was made to reproduce the correlation plots from aircraft observations by Harris-Hobbs and Cooper (1987) (‘HC87’). This aims to determine whether HC87 provided valid evidence of the HM process's activity or if the observed correlations could be attributed to alternative SIP mechanisms, in light of recent findings. The most important correlation in their study was between measured ice crystal production rate and predicted ice crystal production rate.

In our control simulation of MC3E, this correlation is predicted with qualitative realism. However, when the HM process is prohibited (No HM run), this correlation is predicted to remain almost unchanged. The correlation seems to indicate activity by a different SIP mechanism, possibly involving graupel. Total SIP prohibition causes measured rates to drop significantly, indicating that while the HC87 correlation technique detects SIP in general, it cannot distinguish the HM process specifically. This technique applied to the simulation without breakup in ice-ice collisions shows a minor reduction relative to the control run. Lastly, when both the HM process and ice-ice collisions are prohibited, the best fit line is much lower than the control run, consistent with the correlation being due to both combined SIP effects, predominantly graupel-related.

5 Discussion

Most cloud models do not incorporate the phenomenon of secondary ice production. Although certain processes have been glimpsed in laboratory experiments, their full characterization remains elusive, as highlighted by Field et al. (2017). As noted above (Sec. 4.1), in an effort to comprehensively account for secondary ice production mechanisms and bridge the existing gaps in CRMs, this study introduced an empirical formulation addressing fragmentation due to sublimation, a SIP process hitherto completely missed by all cloud models (Paper I). Notably, the challenge lies in the limited scope of previous laboratory studies' pooled datasets (as shown in Table 1), with only two published studies conducted by Bacon et al. (1998) and Oraltay and Hallett (1989). Additionally, there was an attempt to quantify ice fragments from dendritic-like ice crystals and one for rimed particles by Dong et al. (1994).

These past lab studies lack comprehensive data on ice particle conditions (size, morphology, temperature, and humidity). Sampling uncertainties arise due to small sample sizes and artificial partial crystals in prior studies (Bacon et al. 1998; Oraltay and Hallett 1989). Existing observations focus on dendritic and heavily rimed particles, leaving gaps in understanding for the sublimational breakup of plates, columns, and needles. The scarcity of laboratory data on graupel hinders precise characterisation of its SIP, particularly regarding sublimational breakup (only one measurement for graupel could be found, from Dong et al. (1994)).

Regarding the accuracy of the sublimational breakup formulation, the error in predicting number of fragments is approximately a factor of 2. For dendritic crystals, the primary error source is the uncertainty (±60%) in the inferred number of fragments per parent particle based on measurements from published studies. This includes the reported error of ±50% (Oraltay and Hallett 1989) and an instrumental error of 10% for temperature and relative humidity measurements. In the cloud model, tracking dendrites during advection and their fragments might introduce further errors. Gaps in knowledge due to lacking observations under specific conditions and sizes also contribute to uncertainties; for instance, the absence of time evolution data for number of fragments in observations introduces uncertainty in the formulation. The limited availability of laboratory data on graupel, especially regarding sublimational breakup, makes it difficult to quantify the associated error, since it is possible that the only data available might be unrepresentative for natural graupel in clouds. Graupel analysis, similar to dendrites, employed the parcel bin model under diverse sensitivity testing conditions. However, only a single experimental observation was accessible for graupel. This data scarcity, akin to dendrites, introduces significant uncertainty. Consequently, due to this limited dataset, a factor of 2 uncertainty is assumed for graupel. But it is ineluctable that more experimental data is needed.

Regarding Paper IV to assess the contribution from warm and cold rain processes, in specific simulations with a very warm cloud base and almost maritime CCN concentrations, warm rain processes dominate surface precipitation and graupel/hail formation aloft, despite the presence of ice (GOAmazon; Gupta et al. 2023). Raindrop-freezing quickly generates graupel, bypassing the usual slow process of crystal vapor growth into snow, followed by riming into graupel. This aligns with previous modelling findings by Phillips et al. (2002). For such situations (e.g., in the tropics), such results contradict some earlier studies (Mülmenstädt et al. 2015; Field and Heymsfield 2015) that assumed cold surface precipitation whenever there is ice aloft in cloudy columns. Validation of our simulation (GOAmazon) showed this assumption is not always true. This challenges the notion that warm-rain can dominate surface precipitation in deep convection worldwide due to the persistent presence of aloft ice, like in cirriform anvil outflows. However, the acuity of the validation of the model was limited: only total precipitation could be measured for comparison with the model, and some other quantities were not measured in GOAmazon.

Ladino et al. (2017) observed that secondary ice predominantly influences the formation of high concentration ice crystals, with INPs making a negligible contribution for measured ice concentrations during High Altitude Ice Crystals and High Ice Water Content (HAIC-HIWC) field campaign. Sotiropoulou et al. (2020) noticed that simulations incorporating SIP consistently yield ice number concentrations within observed ranges, whereas simulations with artificially prohibited SIP mechanisms significantly underestimate observations. This suggests that SIP can account for observed concentrations despite uncertainties in the breakup in ice-ice collisions. In Arctic cloud simulations, Fu et al. (2019) and Sotiropoulou et al. (2020) demonstrated the negligible impact of freezing-drop shattering (raindrop-freezing fragmentation) and this is true for the simulations represented in paper II also. Sotiropoulou et al. (2020) further revealed that only the combination of HM and breakup in ice–ice collisions could explain observed ice crystal number concentrations. Moreover, in simulations of summer Antarctic mixed-phase clouds, Sotiropoulou et al. (2021) proposed that ice–ice collision fragmentation accounted for high ice crystal number concentrations when the HM process was weak. In paper II, we found that the breakup in ice-ice collisions is most dominating mechanism, irrespective of cloud-types. If SIP mechanisms are important to predict the accurate number concentrations. In the HAIC-HIWC field campaigns, Huang et al. (2021) identified the necessity of incorporating multiple SIP mechanisms to achieve realistic ice number concentrations. In paper II, we found that SIP processes are important to represent in CRM to get better agreements with observations. There are several SIP mechanisms that are not represented in CRM.

Sullivan et al. (2018b) revealed that SIP processes contribute to the ice number concentrations as high as primary ice nucleation, and the HM process was the most prevalent process in the simulation of a cold frontal rainband observed during the Aerosol Properties, PRocesses And InfluenceS on the Earth's climate (APPRAISE) campaign in the UK. In the APPRAISE simulation using AC (Waman et al. 2023), we found that the persistent prevalence of HM is consistently projected to dominate in overall ice concentrations compared to other SIP mechanisms. We compare this with other results, we found the HM process is the second most prevailing process in the orographic stratiform clouds and breakup in ice-ice collision is the most dominating process for all four simulated cloud-types. Also, Hartmann et al. (2023) attempted to reproduce the results of the HM process. They found no efficient and reproducible SIP by rime splintering. In their experiment, the rimed ice particle was fixed to a small carbon fiber and not rotated during riming. By contrast, the original experiment by Hallett and Mossop (1974) involved a vertical rod being moved along a horizontal circular path and with its leading-edge riming with supercooled liquid in a mist. This difference in the two experimental setups between Hallett and Mossop (1974) and Hartmann et al. (2023) may have caused the difference in the outcomes.

If, in reality, there is no ice enhancement because of the HM process, one can speculate about possible reasons for the ice observed by Hallett and Mossop (1974). Could the centrifugal force in the experiments of Hallett and Mossop (1974) have broken any fragile fingers of rime that might formed on the rod, perhaps as seen by Dong et al. (1994)? Conversely, if the HM process is real, one can speculate about why it was not observed by Hartmann et al. (2023). Could there have been subsaturation with respect to ice in the ambient air, where ice splinters were emitted, which might have sublimated away any splinters before they could be detected by Hartmann et al. (2023)? This experimental issue should be more explored in the future to try to reproduce the lab data from almost a half-century ago.

Harris-Hobbs and Cooper (1987) observed by aircraft all the correlations expected from the HM process, when measuring ice concentrations in hundreds of warm-based clouds over USA. However, a problem is that we showed (Paper III) that these same correlations are also expected from other SIP mechanisms too, such as breakup in ice-ice collisions. When we switched off the HM process in our simulations, the same correlations observed by Harris-Hobbs and Cooper (1987) are still predicted. So, their 1987 study does not prove that the HM process exists. Nevertheless, our simulations indicate that it may be a key process to represent precisely in cloud models because it improves the agreement in observed and predicted ice concentrations from freezing level to −10ºC. More laboratory studies are needed to clarify the activity of this sensitive process.

Regarding the technology for simulations performed here, there are some limitations of accuracy. AC uses a hybrid bin/bulk microphysics scheme. It is crucial to note that the bin formulation offers more consistent accuracy in representing these processes compared to bulk schemes. Utilizing high-resolution mass grids, spectral bin schemes more precisely replicate observed cloud structures, particle size distributions, their moments, and the formation of raindrops and drizzles. Bin microphysical scheme studies shows a comprehensive perspective on how aerosols augment cloud cover, cloud top altitude, and radiative forcing. None of the tested bulk schemes managed to replicate these outcomes (Khain et al. 2015). In bin microphysical schemes, particle mass is the primary characteristic. However, in reality, particles of equal mass possess diverse properties like density, shape, fall velocity, etc. These parameters must be considered for various applications. In schemes by Phillips et al. (2014, 2015), we additionally characterize such size-dependent properties of ice particles, for example based on their rimed or liquid water fraction.

6 Conclusion and Future outlook

This thesis presents the formulation for an overlooked SIP process, namely sublimational breakup (paper I). By examining relationships between ice diameter and relative humidity, an empirical formula for estimating the quantity of secondary ice particles resulting from this fragmentation process was derived. This formula can be applied in atmospheric models to accurately replicate ice fragmentation due to sublimation in various cloud scenarios. The extent of ice fragmentation through sublimation is governed by three critical factors: the relative humidity with respect to ice, the initial size of ice particles, and the ventilation factor. These dependencies are incorporated into the formulation to precisely forecast the number of fragments produced per parent ice particle within a given timeframe.

When examining the phase space of number of fragments per parent particle as a function of relative humidity and its initial size, a threshold behavior becomes apparent, which is contingent on a size-specific critical humidity level. At humidities near ice saturation, sublimation does not occur. Nevertheless, under this threshold, the number of fragments increases as subsaturation intensifies and particle size grows.

In conclusion, these novel representations of SIP play a crucial role in reducing the uncertainty associated with ice concentration in cloud models. In particular, breakup in ice-ice collisions is essential for validation of predicted cloud properties (e.g., ice concentration) in simulation of observed cases. It is imperative that laboratory experiments are conducted to comprehensively characterize sublimational breakup, with particular emphasis on its behavior for graupel particles.

Warm-based or very warm-based convective clouds exhibit more ice enhancement from ice-ice collision breakup (IE ratio nearly 10^3) compared to cold-based convective clouds (IE ratio around 10^2). In all cases simulated, this SIP mechanism dominates over the lifetime of cloud systems. The ice enhancement due to the HM process increases with warmer cloud-base temperatures (Paper III), particularly in slightly cold-based stratiform clouds dominated by warm rain processes (66% cold rainfall, 34% warm rain, paper IV). In downdrafts of warm and very warm-based convective clouds, the contribution from sublimational breakup to IE ratio strengthens with higher cloud-base temperatures, while raindrop-freezing fragmentation lacks enhancement in cold-based clouds and shows slight enhancement (IE ratio $< 10^2$) in warm-based clouds. Sublimational breakup has the highest IE ratios in downdrafts, consistent with idealized parcel simulations and thought experiment in Paper I. The raindrop-freezing fragmentation and HM processes have a uniform distribution of ice enhancement over the lower mixed-phase region, in GOAmazon warm-based clouds, with the former being weaker by one or two orders of magnitude.

Lowering cloud base by approximately 18 K in STEPS and 11 K in MC3E, bringing it closer to warmer ground levels, amplifies the contributions from warm precipitation components. A moister lower troposphere helps condensation and coalescence, resulting in warm components constituting nearly half of the total surface precipitation in both STEPS and MC3E. In the GOAmazon case, higher CCN concentrations, mimicking polluted aerosol conditions, decreased both the fractional and absolute contributions of warm rain to overall surface precipitation. In STEPS and MC3E scenarios, simultaneous reductions in cloud base height and CCN concentrations led to the most significant rise in the fractional contributions from warm components to surface precipitation.

In ACAPEX, a synergistic relationship is observed between the HM process and raindrop-freezing fragmentation. However, no such synergy is detected for other pairs of SIP processes, including breakup in ice-ice collisions and sublimational breakup, as well as the HM process and breakup in ice-ice collisions. In MC3E, no synergy or dyssynergy is found in the lower half of mixed-phase clouds. The minor dyssynergies observed in all cases can be attributed to the competition among various ice multiplication mechanisms.

In the context of tropical deep convection, specifically observed in GOAmazon with very warm cloud bases, a reduction in CCN concentrations in the lower half of the mixed-phase region resulted in increased simulated ice concentrations. However, this IN reduction had limited impact on secondary ice and homogeneously nucleated ice. Ice multiplication processes mitigated the effects of altered IN and CCN aerosol loadings in the environment (Paper III).

The upper half of mixed-phase clouds exhibits lower ice concentrations when homogeneous ice is artificially prohibited. However, across all cloud scenarios, the prevention of homogeneous freezing of cloud liquid does not impact ice multiplication and charge separation.

In emulating the correlations noted by HC87 with our simulations, it is found that their study provides no unambiguous detection of the HM process. This is evident from similar correlations being predicted even when the HM process was disabled in MC3E simulations. It is plausible that both breakup in ice-ice collisions and the HM process together contributed to the observed correlations reported by HC87.

While conducting this thesis, we pinpointed several areas that warrant further exploration and development, such as:

1. SIP processes are not fully represented in the models. The question is whether all SIP processes are known or not? Still, the known SIP mechanisms are presented in the models starting to be used by the community. The challenge is to represent all discovered SIP mechanisms in all cloud models with adequate accuracy.

2. For sublimational breakup, the formulation can be improved and needs a wide range of experimental data for that. Will we be able to incorporate future observations of this behavior in the framework of the existing formulation?

3. The effect of SIP processes on the different domains (i.e., tropics, midlatitude and polar regions) should be studied for stratiform and convective clouds of the same cloud base temperatures? Arctic clouds are relatively unexplored, partly due to a lack of modern comprehensively observed cases to simulate.

4. How do the relative activities of SIP mechanisms change over time in the models and the atmospheres? What are the optimal conditions for a cloud to have SIP mechanisms? What is the threshold of ice precipitation for a cloud to begin ice multiplication?

5. Also, it will be interesting to do an intercomparison of various cloud models for a same case simulation, representing multiple SIP mechanisms.

We highlight the importance of enhanced collaboration between modelling and experimental scientists. This collaboration aims to enhance the representation of processes within atmospheric models, integrating cutting-edge insights from laboratory/observational studies into regional-scale processes and their intricate interaction.

Our study underlines the significance of delving into historically underexplored atmospheric processes. Addressing the existing knowledge gaps surrounding these aspects allows for the generation of more robust assessments regarding the resolving processes in natural clouds to evolve the atmospheric models.

References

Bacon, N. J., B. D. Swanson, M. B. Baker, and E. J. Davis, 1998: Breakup of levitated frost particles. J. Geophys. Res. Atmos., 103, 13763–13775, https://doi.org/10.1029/98JD01162.

Beard, K. V., 1992: Ice initiation in warm-base convective clouds. An assessment of microphysical mechanisms. Atmos. Res., 28, 125–152, doi:10.1016/0169-8095(92)90024-5.

Beheng, K. D., 1987: Microphysical properties of glaciating cumulus clouds: Comparison of measurements with a numerical simulation. Quart. J. Roy. Meteor. Soc., 113, 1377–1382, doi:10.1002/qj.49711347815.

Blyth, A. M., and J. Latham, 1993: Development of ice and precipitation in New Mexican summertime cumulus clouds. Q. J. R. Meteorol. Soc., 119, 91–120, https://doi.org/10.1002/qj.49711950905.

Blyth, A. M., and J. Latham, 1997: A multi-thermal model of cumulus glaciation via the Hallett-Mossop process. Quart. J. Roy. Meteor. Soc., 123, 1185–1198, doi:10.1002/qj.49712354104.

Brewer, A. W., and H. P. Palmer, 1949: Condensation processes at low temperatures, and the production of new sublimation nuclei by the splintering of ice [6]. Nature, 164, 312–313, https://doi.org/10.1038/164312a0.

Cannon, T. W., J. E. Dye, and V. Toutenhoofd, 1974: The mechanism of precipitation formation in northeastern Colorado cumulus II. Sailplane measurements. J. Atmos. Sci., 31, 2148–2151, doi:10.1175/1520-0469(1974)031<2148:TMOPFI>2.0.CO;2.

Clark, P., T. W. Choularton, P. R. A. Brown, P. R. Field, A. J. Illingworth, and R. J. Hogan, 2005: Numerical modelling of mixed-phase frontal clouds observed during the CWVC project. Quart. J. Roy. Meteor. Soc., 131, 1677–1693, doi:10.1256/qj.03.210.

Connolly, P. J., A. J. Heymsfield, and T. W. Choularton, 2006: Modelling the influence of rimer surface temperature on the glaciation of intense thunderstorms: The rime–splinter mechanism of ice multiplication. Q. J. R. Meteorol. Soc., 132, 3059–3077, https://doi.org/10.1256/qj.05.45.

Cooper, W. A., 1986: Ice initiation in nature clouds. Precipitation Enhancement—A Scientific Challenge, Meteor. Monogr., No. 43, Amer. Meteor. Soc., 29–32.

Cziczo, D.J., Froyd, K.D., Hoose, C., Jensen, E.J., Diao, M., Zondlo, M.A., Smith, J.B., Twohy, C.H. and Murphy, D.M., 2013: Clarifying the dominant sources and mechanisms of cirrus cloud formation. Science, 340 (6138), pp.1320-1324.

Dearden, C., G. Vaughan, T. Tsai, and J. Chen, 2016: Exploring the diabatic role of ice microphysical processes in two North Atlantic summer cyclones. Mon. Wea. Rev., 144, 1249–1272, doi:10.1175/MWR-D-15-0253.1.

DeMott, P.J., Möhler, O., Stetzer, O., Vali, G., Levin, Z., Petters, M.D., Murakami, M., Leisner, T., Bundke, U., Klein, H. and Kanji, Z.A., 2011: Resurgence in ice nuclei measurement research. Bulletin of the American Meteorological Society, 92(12), pp.1623-1635.

Deshmukh, A., V. T. J. Phillips, A. Bansemer, S. Patade, and D. Waman, 2022: New Empirical Formulation for the Sublimational Breakup of Graupel and Dendritic Snow. J. Atmos. Sci., 79, 317–336, https://doi.org/10.1175/JAS-D-20-0275.1.

Dong, Y. Y., and J. Hallett, 1989: Droplet accretion during rime growth and the formation of secondary ice crystals. Q. J. R. Meteorol. Soc., https://doi.org/10.1002/qj.49711548507.

Ferrier, B. S., 1994: A double-moment multiple-phase four-class bulk ice scheme. Part I: Description. J. Atmos. Sci., 51, 249–280, doi:10.1175/1520-0469(1994)051<0249:ADMMPF>2.0.CO;2.

Field, P.R. and Heymsfield, A.J., 2015: Importance of snow to global precipitation. Geophysical Research Letters, 42(21), pp.9512-9520.

Field, P.R., Lawson, R.P., Brown, P.R., Lloyd, G., Westbrook, C., Moisseev, D., Miltenberger, A., Nenes, A., Blyth, A., Choularton, T. and Connolly, P., 2017: Secondary ice production: Current state of the science and recommendations for the future. Meteorological Monographs, 58, pp.7-1.

Fridlind, A. M., A. S. Ackerman, G. McFarquhar, G. Zhang, M. R. Poellot, P. J. DeMott, A. J. Prenni, and A. J. Heymsfield, 2007: Ice properties of single-layer stratocumulus during the Mixed-Phase Arctic Cloud Experiment: 2. Model results. J. Geophys. Res. Atmos., 112, https://doi.org/10.1029/2007JD008646.

Fu, S., Deng, X., Shupe, M.D. and Xue, H., 2019: A modelling study of the continuous ice formation in an autumnal Arctic mixed-phase cloud case. Atmospheric Research, 228, pp.77-85.

Griggs, D., And T. Choularton, 1986: A laboratory study of secondary ice particle production by the fragmentation of rime and vapour-grown ice crystals. Q. J. R. Meteorol. Soc., 112, 149–163, https://doi.org/10.1256/smsqj.47108.

Gupta, A.K., Deshmukh, A., Waman, D., Patade, S., Jadav, A., Phillips, V.T., Bansemer, A., Martins, J.A. and Gonçalves, F.L., 2023: The microphysics of the warm-rain and ice crystal processes of precipitation in simulated continental convective storms. Communications Earth & Environment, 4(1), p.226.

Hallett, J., and S. C. Mossop, 1974: Production of secondary ice particles during the riming process. Nature, 249, 26–28, https://doi.org/10.1038/249026a0.

Hallett, J., R. I. Sax, D. Lamb, and A. S. R. Murty, 1978: Aircraft measurements of ice in Florida cumuli. Q. J. R. Meteorol. Soc., 104, 631–651, https://doi.org/10.1002/qj.49710444108.

Hamblyn, R., 2002: The invention of clouds: How an amateur meteorologist forged the language of the skies. Pan Macmillan.

Harris-Hobbs, R. L., and W. A. Cooper, 1987: Field evidence supporting quantitative predictions of secondary ice production rates. J. Atmos. Sci., 44, 1071–1082, doi:10.1175/1520-0469(1987)044<1071:FESQPO>2.0.CO;2.

Hartmann, S., Seidel, J., Keinert, A., Kiselev, A., Leisner, T. and Stratmann, F., 2023: Secondary ice production-No evidence of a productive rime-splintering mechanisms during dry and wet growth (No. EGU23-11199). Copernicus Meetings, https://doi.org/10.5194/egusphere-egu23-11199,2023.

Heymsfield, A. J., and P. Willis, 2014: Cloud conditions favoring secondary ice particle production in tropical maritime convection. J. Atmos. Sci., 71, 4500–4526, doi:10.1175/JAS-D-14-0093.1.

Hobbs, P. V., and A. J. Alkezweeny, 1968: The Fragmentation of Freezing Water Droplets in Free Fall. J. Atmos. Sci., 25, 881–888, https://doi.org/10.1175/1520-0469(1968)025<0881:tfofwd>2.0.co;2.

Hobbs, P. V., J. L. Stith, and L. F. Radke, 1980: Cloud-active nuclei from coal-fired electric power plants and their interactions with clouds. J. Appl. Meteorol., 19, 439–451, https://doi.org/10.1175/1520-0450(1980)019<0439:CANFCF>2.0.CO;2.

Hobbs, P. V., and A. L. Rangno, 1985: Ice particle concentrations in clouds. J. Atmos. Sci., 42, 2523–2549, https://doi.org/10.1175/1520-0469(1985)042<2523:IPCIC>2.0.CO;2.

Hobbs, P. V., and A. L. Rangno, 1990: Rapid development of high ice particle concentrations in small polar maritime cumuliform clouds. J. Atmos. Sci., 47, 2710–2722, https://doi.org/10.1175/1520-0469(1990)047<2710:RDOHIP>2.0.CO;2.

Houze, R. A., 1993: Cloud dynamics. Cloud Dyn., 432, https://doi.org/10.1016/0377-0265(87)90017-0.

Huang, Y., Wu, W., McFarquhar, G.M., Wang, X., Morrison, H., Ryzhkov, A., Hu, Y., Wolde, M., Nguyen, C., Schwarzenboeck, A. and Milbrandt, J., 2021: Microphysical processes producing high ice water contents (HIWCs) in tropical convective clouds during the HAIC-HIWC field campaign: evaluation of simulations using bulk microphysical schemes. Atmospheric Chemistry and Physics, 21(9), pp.6919-6944.

Jardine, B., 2014: Made real: artifice and accuracy in nineteenth-century scientific illustration. Sci. Museum Gr. J., 2, https://doi.org/10.15180/140208.

Jensen, M.P., Petersen, W.A., Bansemer, A., Bharadwaj, N., Carey, L.D., Cecil, D.J., Collis, S.M., Del Genio, A.D., Dolan, B., Gerlach, J. and Giangrande, S.E., 2016: The midlatitude continental convective clouds experiment (MC3E). Bulletin of the American Meteorological Society, 97(9), pp.1667-1686.

Kanji, Z.A., Ladino, L.A., Wex, H., Boose, Y., Burkert-Kohn, M., Cziczo, D.J. and Krämer, M., 2017: Overview of ice nucleating particles. Meteorological Monographs, 58, pp.1-1.

Khain, A.P., Beheng, K.D., Heymsfield, A., Korolev, A., Krichak, S.O., Levin, Z., Pinsky, M., Phillips, V., Prabhakaran, T., Teller, A. and Van Den Heever, S.C., 2015. Representation of microphysical processes in cloud-resolving models: Spectral (bin) microphysics versus bulk parameterization. Reviews of Geophysics, 53(2), pp.247-322.

Knopf, D.A., Alpert, P.A. and Wang, B., 2018. The role of organic aerosol in atmospheric ice nucleation: a review. ACS Earth and Space Chemistry, 2(3), pp.168-202.

Korolev, A., 2007: Limitations of the Wegener-Bergeron-Findeisen mechanism in the evolution of mixed-phase clouds. J. Atmos. Sci., 64, 3372–3375, https://doi.org/10.1175/JAS4035.1.

Ladino, L.A., Korolev, A., Heckman, I., Wolde, M., Fridlind, A.M. and Ackerman, A.S., 2017. On the role of ice-nucleating aerosol in the formation of ice particles in tropical mesoscale convective systems. Geophysical research letters, 44(3), pp.1574-1582.

Lang, T.J., Miller, L.J., Weisman, M., Rutledge, S.A., Barker III, L.J., Bringi, V.N., Chandrasekar, V., Detwiler, A., Doesken, N., Helsdon, J. and Knight, C., 2004: The severe thunderstorm electrification and precipitation study. Bulletin of the American Meteorological Society, 85(8), pp.1107-1126.

Langmuir, I., 1948: The production of rain by a chain reaction in cumulus clouds at temperatures above freezing. Journal of the Atmospheric Sciences, 5(5), pp.175-192.

Leung, L., 2016: ARM Cloud-Aerosol-Precipitation Experiment (ACAPEX) Field Campaign Report.

Lawson, R. P., S. Woods, and H. Morrison, 2015: The microphysics of ice and precipitation development in tropical cumulus clouds. J. Atmos. Sci., 72, 2429–2445, doi:10.1175/JAS-D-14-0274.1.

Lohmann, U., Lüönd, F. and Mahrt, F., 2016: An introduction to clouds: From the microscale to climate. Cambridge University Press.

Low, T. B., and R. List, 1982: Collision, coalescence and breakup of raindrops. Part I: experimentally established coalescence efficiencies and fragment size distributions in breakup. J. Atmos. Sci., 39, 1591–1606, https://doi.org/10.1175/1520-0469(1982)039<1591:CCABOR>2.0.CO;2.

Mason, B. J., 1996: The rapid glaciation of slightly supercooled cumulus clouds. Quart. J. Roy. Meteor. Soc., 122, 357–365, doi:10.1002/qj.49712253003.

Mossop, S. C., 1985: Secondary ice particle production during rime growth: The effect of drop size distribution and rimer velocity. Q. J. R. Meteorol. Soc., 111, 1113–1124, https://doi.org/10.1002/qj.49711146612.

Mülmenstädt, J., Sourdeval, O., Delanoë, J. and Quaas, J., 2015.: Frequency of occurrence of rain from liquid-, mixed-, and ice-phase clouds derived from A-Train satellite retrievals. Geophysical Research Letters, 42(15), pp.6502-6509.

Oraltay, R. G., and J. Hallett, 1989: Evaporation and melting of ice crystals: A laboratory study. Atmos. Res., 24, 169–189, https://doi.org/10.1016/0169-8095(89)90044-6.

Rogers, R. R., and M. K. Yau, 1989: A Short Course in Cloud Physics. Pergamon Press, 290 pp.

Phillips, V. T. J., A. M. Blyth, P. R. A. Brown, T. W. Choularton, and J. Latham, 2001: The glaciation of a cumulus cloud over New Mexico. Q. J. R. Meteorol. Soc., 127, 1513–1534, https://doi.org/10.1002/qj.49712757503.

Phillips, V.T., Choularton, T.W., Blyth, A.M. and Latham, J., 2002: The influence of aerosol concentrations on the glaciation and precipitation of a cumulus cloud. Quarterly Journal of the Royal Meteorological Society: A journal of the atmospheric sciences, applied meteorology and physical oceanography, 128(581), pp.951-971.

Phillips, V. T. J., L. J. Donner, and S. T. Garner, 2007: Nucleation processes in deep convection simulated by a cloud-system-resolving model with double-moment bulk microphysics. J. Atmos. Sci., 64, 738–761, doi:10.1175/JAS3869.1.

Phillips, V.T., Andronache, C., Christner, B., Morris, C.E., Sands, D.C., Bansemer, A., Lauer, A., McNaughton, C. and Seman, C., 2009: Potential impacts from biological aerosols on ensembles of continental clouds simulated numerically. Biogeosciences, 6(6), pp.987-1014.

Phillips, V. T. J., P. J. Demott, C. Andronache, K. A. Pratt, K. A. Prather, R. Subramanian, and C. Twohy, 2013: Improvements to an empirical parameterization of heterogeneous ice nucleation and its comparison with observations. J. Atmos. Sci., 70, 378–409, https://doi.org/10.1175/JAS-D-12-080.1.

Phillips, V. T. J., A. Khain, N. Benmoshe, and E. Ilotoviz, 2014: Theory of Time-Dependent Freezing. Part I: Description of Scheme for Wet Growth of Hail. J. Atmos. Sci., 71, 4527–4557, https://doi.org/10.1175/JAS-D-13-0375.1.

Phillips, V. T. J., M. Formenton, A. Bansemer, I. Kudzotsa, and B. Lienert, 2015: A parameterization of sticking efficiency for collisions of snow and graupel with ice crystals: Theory and comparison with observations. J. Atmos. Sci., 72, 4885–4902, https://doi.org/10.1175/JAS-D-14-0096.1.

Phillips, V. T. J., J. I. Yano, and A. Khain, 2017a: Ice multiplication by breakup in ice-ice collisions. Part I: Theoretical formulation. J. Atmos. Sci., 74, 1705–1719, https://doi.org/10.1175/JAS-D-16-0224.1.

Phillips, V., Yano, J., Formenton, M., Ilotoviz, E., Kanawade, V., Kudzotsa, I., Sun, J., Bansemer, A., Detwiler, A., Khain, A. and Tessendorf, S., 2017: Ice Multiplication by Breakup in Ice–Ice Collisions. Part II: Numerical Simulations. J. Atmos. Sci., 74, 2789–2811, https://doi.org/10.1175/JAS-D-16-0223.1.

Phillips, V. T. J., S. Patade, J. Gutierrez, and A. Bansemer, 2018: Secondary ice production by fragmentation of freezing drops: Formulation and theory. J. Atmos. Sci., 75, 3031–3070, https://doi.org/10.1175/JAS-D-17-0190.1.

Phillips, V.T., Formenton, M., Kanawade, V.P., Karlsson, L.R., Patade, S., Sun, J., Barthe, C., Pinty, J.P., Detwiler, A.G., Lyu, W. and Tessendorf, S.A., 2020: Multiple environmental influences on the lightning of cold-based continental cumulonimbus clouds. Part I: Description and validation of model. Journal of the Atmospheric Sciences, 77(12), pp.3999-4024.

Pruppacher, H. R., and J. D. Klett, 1978: Microphysics of Clouds and Precipitation.

Rangno, A., and P. Hobbs, 2001: Ice particles in stratiform clouds in the Arctic and possible mechanisms for the production of high ice concentrations. J. Geophys. Res., 106, 15 065–15 075, doi:10.1029/2000JD900286.

Rehbein, A., T. Ambrizzi, C. R. Mechoso, S. A. I. Espinosa, and T. A. Myers, 2019: Mesoscale convective systems over the Amazon basin: The GoAmazon2014/5 program. Int. J. Climatol., 39, 5599–5618, https://doi.org/10.1002/joc.6173.

Rehbein, A., T. Ambrizzi, and C. R. Mechoso, 2018: Mesoscale convective systems over the Amazon basin. Part I: climatological aspects. Int. J. Climatol., 38, 215–229, https://doi.org/10.1002/joc.5171.

Romatschke, U., and R. A. Houze, 2013: Characteristics of precipitating convective systems accounting for the summer rainfall of tropical and subtropical South America. J. Hydrometeorol., 14, 25–46, https://doi.org/10.1175/JHM-D-12-060.1.

Schaefer, V. J., and R. J. Cheng, 1971: The production of ice crystal fragments by sublimation and electrification. J. Rech. Atmos., 5, 5–10.

Schaller, R. C., and N. Fukuta, 1979: Ice Nucleation by Aerosol Particles: Experimental Studies Using a Wedge-Shaped Ice Thermal Diffusion Chamber. J. Atmos. Sci., 36, 1788–1802, https://doi.org/10.1175/1520-0469(1979)036<1788:INBAPE>2.0.CO;2.

Scott, B. C., and P. V. Hobbs, 1977: A theoretical study of the evolution of mixed-phase cumulus clouds. J. Atmos. Sci., 34, 812–826, doi:10.1175/1520-0469(1977)034<0812:ATSOTE>2.0.CO;2.

Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K., Tignor, M. and Miller, H., 2007: IPCC fourth assessment report (AR4). Climate change, 374.

Sotiropoulou, G., Sullivan, S., Savre, J., Lloyd, G., Lachlan-Cope, T., Ekman, A.M. and Nenes, A., 2020: The impact of secondary ice production on Arctic stratocumulus. Atmospheric Chemistry and Physics, 20(3), pp.1301-1316.

Sotiropoulou, G., Vignon, É., Young, G., Morrison, H., O'Shea, S.J., Lachlan-Cope, T., Berne, A. and Nenes, A., 2021: Secondary ice production in summer clouds over the Antarctic coast: an underappreciated process in atmospheric models. Atmospheric Chemistry and Physics, 21(2), pp.755-771.

Sullivan, S. C., C. Hoose, and A. Nenes, 2017: Investigating the contribution of secondary ice production to in-cloud ice crystal numbers. J. Geophys. Res. Atmos., 122, 9391–9412, https://doi.org/10.1002/2017JD026546.

Sullivan, S. C., C. Hoose, A. Kiselev, T. Leisner, and A. Nenes, 2018a: Initiation of secondary ice production in clouds. Atmos. Chem. Phys., 18, 1593–1610, https://doi.org/10.5194/acp-18-1593-2018.

Sullivan, S.C., Barthlott, C., Crosier, J., Zhukov, I., Nenes, A. and Hoose, C., 2018b: The effect of secondary ice production parameterization on the simulation of a cold frontal rainband. Atmospheric chemistry and physics, 18(22), pp.16461-16480.

Takahashi, C., and A. Yamashita, 1977: Production of ice splinters by the freezing of water drops in free fall. J. Meteor. Soc. Japan, 55, 139–141, https://doi.org/10.2151/jmsj1965.55.1_139.

Takahashi, T., Y. Nagao, and Y. Kushiyama, 1995: Possible high ice particle production during graupel–graupel collisions. J. Atmos. Sci., 52, 4523–4527, doi:10.1175/1520-0469(1995)052<4523:PHIPPD>2.0.CO;2.

Taylor, J.W., Choularton, T.W., Blyth, A.M., Liu, Z., Bower, K.N., Crosier, J., Gallagher, M.W., Williams, P.I., Dorsey, J.R., Flynn, M.J. and Bennett, L.J., 2016: Observations of cloud microphysics and ice formation during COPE. Atmospheric Chemistry and Physics, 16(2), pp.799-826.

Vardiman, L., 1978: The generation of secondary ice particles in clouds by crystal–crystal collisions. J. Atmos. Sci., 35, 2168–2180, doi:10.1175/1520-0469(1978)035<2168:TGOSIP>2.0.CO;2.

Waman, D., S. Patade, A. Jadav, A. Deshmukh, A. K. Gupta, V. T. J. Phillips, A. Bansemer, and P. J. DeMott, 2022: Dependencies of Four Mechanisms of Secondary Ice Production on Cloud-Top Temperature in a Continental Convective Storm. J. Atmos. Sci., 79, 3375–3404, https://doi.org/10.1175/jas-d-21-0278.1.

Yano, J. I., and V. T. J. Phillips, 2011: Ice-Ice collisions: An Ice multiplication process in atmospheric clouds. J. Atmos. Sci., 68, 322–333, https://doi.org/10.1175/2010JAS3607.1.