Ice and Supercooled Liquid Distributions Based on in Situ Observations and Climate Model Simulations

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ICE AND SUPERCOOLED LIQUID DISTRIBUTIONS BASED ON IN SITU OBSERVATIONS AND CLIMATE MODEL SIMULATIONS

A Thesis

Presented to

The Faculty of the Department of Meteorology and Climate Science

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In Partial Fulfillment

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Master of Science

by

Ching An Yang

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The Designated Thesis Committee Approves the Thesis Titled

ICE AND SUPERCOOLED LIQUID DISTRIBUTIONS BASED ON IN SITU OBSERVATIONS AND CLIMATE MODEL SIMULATIONS

by

Ching An Yang

APPROVED FOR THE DEPARTMENT OF METEOROLOGY AND CLIMATE SCIENCE

SAN JOSÉ STATE UNIVERSITY

July 2022

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ABSTRACT

ICE AND SUPERCOOLED LIQUID DISTRIBUTIONS BASED ON IN SITU OBSERVATIONS AND CLIMATE MODEL SIMULATIONS

by Ching An Yang

Three climate models are evaluated using *in situ* airborne observations from the Southern Ocean Clouds, Radiation, Aerosol Transport Experimental Study (SOCRATES) campaign. The evaluation targets cloud phases, microphysical properties, thermodynamic conditions, and aerosol indirect effects from -40°C to 0°C. Compared with 580-s averaged observations (i.e., 100 km horizontal scale), the Community Atmosphere Model version 6 (CAM6) shows the most similar result for cloud phase frequency distribution and allows more liquid-containing clouds below -10°C compared with its predecessor—CAM5. The Energy Exascale Earth System Model (E3SM) underestimates (overestimates) ice phase frequencies below (above) -20°C. CAM6 and E3SM show liquid and ice water contents (i.e., LWC and IWC) similar to observations from -25°C to 0°C, but higher LWC and lower IWC than observations at lower temperatures. Simulated in-cloud RH shows higher minimum values than observations, possibly restricting ice growth during sedimentation. As number concentrations of aerosols larger than 500 nm (Na_{500}) increase, observations show increases of liquid and ice. Number concentrations of aerosols larger than 100 nm (Na_{100}) only show positive correlations with liquid. CAM6 shows small increases of liquid with Na_{500} and Na_{100}. E3SM shows small increases of. Overall, CAM6 and E3SM underestimate aerosol indirect effects on ice crystals and supercooled liquid droplets over the Southern Ocean.
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LIST OF ABBREVIATIONS

2DS – Two-Dimensional Stereo Probe
CALIPSO – Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation
CAM5 – Community Atmosphere Model version 5
CCN – cloud condensation nuclei
CDP – Cloud Droplet Probe
CESM1 – Community Earth System Model version 1
CESM2 – Community Earth System Model version 2
CLUBB – Cloud Layers Unified By Binormals
cm$^3$ – per cubic centimeter
$D_{\text{max}}$ – maximum particle diameter
DOE – U.S. Department of Energy
e – ambient water vapor partial pressure
E3SM – Energy Exascale Earth System Model
EAMv1 – Earth Atmosphere Model version 1
e$_{\text{e,ice}}$ – saturation vapor pressure with respect to ice
e$_{\text{e,liq}}$ – saturation vapor pressure with respect to liquid
g m$^3$ – gram per cubic meter
GC – generating cells
GCM – global climate model
GV – Gulfstream-V
Hz – hertz
INPs – ice nucleating particles
IWC – ice water contents
K – kelvin
kg m$^3$ – kilogram per cubic meter
km – kilometer
L$^{-1}$ – per liter
LWC – liquid water contents
m s$^{-1}$ – meter per second
MAM3 – modal aerosol module with three modes
Mc – particle mass concentration
m-D – mass-dimension relationships
MERRA2 – Modern-Era Retrospective Analysis for Research and Applications
M-PACE – Mixed-Phase Arctic Cloud Experiment
Na – aerosol number concentration
Na$_{100}$ – number concentrations of aerosols larger than 100 nm
Na$_{500}$ – number concentrations of aerosols larger than 500 nm
Nc – particle number concentration
NCAR – the National Center for Atmospheric Research
N$_{\text{ice}}$ – ice number concentrations
N$_{\text{liq}}$ – liquid number concentrations
nm – nanometer
NSF – the U.S. National Science Foundation
OAP – Optical Array Probes
PDF – probability density function
RF – research flight
RH_{ice} – relative humidity with respect to ice
RH_{liq} – relative humidity with respect to liquid
RICE – Rosemount Icing Detector
SOCRATES – Southern Ocean Clouds, Radiation, Aerosol Transport Experimental Study
CAM6 – Community Atmosphere Model version 6
UHSAS – Ultra-High Sensitivity Aerosol Spectrometer
VCSEL – Vertical Cavity Surface Emitting Laser (version 2)
WBF – Wegener-Bergeron-Findeisen process
\( \mu \)m – micrometer
\( \sigma_D \) – standard deviation of size distribution
Chapter 1

Ice and Supercooled Liquid Water Distributions over the Southern Ocean based on In Situ Observations and Climate Model Simulations

1.1 Introduction

Clouds play a crucial role in influencing Earth’s radiation budget (Liou, 1992). The cloud types, height, the partition of cloud phases, and microphysical properties of liquid droplets and ice crystals are important in determining the cloud radiative effect (T. Chen et al., 2000; Matus & L’Ecuyer, 2017).

Mixed phase clouds, clouds with the coexistence of liquid and ice, have been a focus of cloud microphysics research as many of their properties remain not fully understood (e.g., Korolev et al., 2017; Lohmann et al., 2016). A frequently occurring process in mixed phase clouds, named the Wegener-Bergeron-Findeisen (WBF) process, describes ice crystal growth at the expense of liquid droplets as the liquid droplets evaporate to water vapor that deposits on ice crystals (Bergeron, 1928; Wegener, 1911). This occurs when ambient water vapor partial pressure (e) is lower than the saturation vapor pressure with respect to liquid (e_{s,liq}) but higher than the saturation vapor pressure with respect to ice (e_{s,ice}). The amount of ice and liquid and their mass partition in mixed phase clouds are crucial for determining cloud lifetime, radiative properties, and precipitation (e.g., A. E. Morrison et al., 2010; Mülmenstädt et al., 2015), as well as for developing model parameterizations that represent these properties (e.g., Tan & Storelumo, 2016; M. Zhang et al., 2019).

Supercooled liquid water, i.e., liquid droplets that exist below 0°C in both liquid and mixed phase clouds, was previously found to be underestimated in several global climate model (GCM) simulations, particularly over the Southern Ocean (Bodas-Salcedo et al., 2016;
McCoy et al., 2016; Tan et al., 2016; Williams et al., 2013). Due to the scarcity of in situ observations in remote regions such as over the Southern Ocean, many evaluations of model biases rely on satellite observations (e.g., Kay et al., 2012; Trenberth & Fasullo, 2010). Guo et al. (2020), as an example, used satellite retrieval data from Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) to compare with the Community Atmosphere Model version 5 (CAM5). They concluded that the model misclassifies liquid as ice, leading to an underestimation of liquid cloud occurrence frequencies and an overestimation of ice cloud occurrence frequencies in all vertical levels. The model also shows that the supercooled liquid fraction reaches 50% at -5°C, a much higher temperature than the observed temperature of -20°C where that fraction is reached. When comparing with airborne observations around Punta Arenas, Chile, D’Alessandro et al. (2019) showed that the CAM5 does not allow liquid and mixed phase clouds to exist at temperatures less than -15°C. The model was found to overestimate and underestimate liquid water content (LWC) in liquid and mixed phases, respectively, and underestimate ice water content (IWC) in ice and mixed phases, which demonstrates the importance of analyzing both occurrence frequencies and cloud water content of the three cloud phases. Another study compared ground-based observations of mixed phase clouds over the Arctic with the CAM5, and showed that revising the mixing volumes where supercooled liquid water and ice particles coexist in the model can reduce the effectiveness of the WBF process, which prolongs the lifetime of supercooled liquid water (M. Zhang et al., 2019). Klein et al. (2009) found an underestimation of the median liquid water path by a factor of three in single-column models and cloud-resolving models when comparing with the observations from the Mixed-Phase
Arctic Cloud Experiment (M-PACE). That study emphasized the importance of ice microphysical processes, such as ice initiation and water vapor deposition rate on ice crystals, which contribute to the underestimation of the liquid water path.

Thermodynamic (i.e., temperature and relative humidity) and dynamic (i.e., wind speed and direction) conditions play an important role in the formation of mixed phase clouds. For example, using in-situ aircraft observations over the Southern Ocean, D’Alessandro et al. (2019) showed increasing deviations of RH\textsubscript{liq} from liquid saturation as ice mass fraction increases in the mixture of ice and liquid. Other studies also found that mixed phase clouds are influenced by vertical velocity (e.g., Bühl et al., 2019; Korolev & Field, 2008; Shupe et al., 2008) and horizontal wind direction (e.g., Gierens et al., 2020; Qiu et al., 2018) from the microscale to the mesoscale. Aerosol number concentration and size distribution are also known to influence the formation and evolution of ice particles and supercooled liquid water. Three hypothesized aerosol indirect effects for mixed phase clouds are: (i) the glaciation indirect effect, which describes increases of ice nucleating particles (INPs) that lead to more ice particles and ice phase precipitation (Lohmann, 2002); (ii) the riming indirect effect, which describes increases of cloud condensation nuclei (CCN) concentrations that lead to smaller liquid droplets, less riming and smaller IWC (Borys et al., 2003); and (iii) the thermodynamic indirect effect, which describes increases of CCN concentrations that lead to more liquid droplets, less secondary ice production (SIP) (Hallett & Mossop, 1974) and fewer ice particles (Rangno & Hobbs, 2001). Using airborne observation data, Jackson et al. (2012) found a positive correlation between liquid number concentration inside clouds and aerosol number concentration below clouds. They also found a positive correlation between
ice number concentration and aerosol number concentration above clouds. Storelvmo et al. (2011) conducted a modeling study for aerosol indirect effects on mixed phase clouds and found decreasing cloud lifetime due to increasing INP concentrations. They also found decreasing ice particle sizes and increasing cloud albedo due to increasing INP concentrations, similar to the Twomey effect on liquid clouds. These studies demonstrated the importance of thermodynamic conditions and aerosol indirect effects on cloud microphysical properties in the mixed phase cloud regime.

A recent campaign conducted in 2018 over the Southern Ocean, the Southern Ocean Clouds, Radiation, Aerosol Transport Experimental Study (SOCRATES) (McFarquhar et al., 2021), has provided valuable airborne cloud observations for several scientific studies. For example, Wang et al. (2020) examined the importance of generating cells (GC) for producing mixed phase clouds over the Southern Ocean. They found enhanced IWC and LWC inside the GCs compared with the adjacent environment. D’Alessandro et al. (2021) showed that up to 70% of the supercooled liquid water and most of the spatially heterogeneous mixed-phase segments occurred between -20°C and 0°C. Zaremba et al. (2020) classified cloud top phases as a function of cloud top temperature using radar and lidar and concluded that liquid is the most dominant phase for cloud top. Another study by Gettelman et al. (2020) compared Community Atmosphere Model version 6 (CAM6) simulations against SOCRATES observations and showed that CAM6 provides improved representations of supercooled liquid water and particle size distributions compared with CAM5. However, there is still a question that has not been addressed in these existing studies, that is, “how well do GCMs represent three cloud thermodynamic phases, cloud microphysical properties and aerosol
indirect effects in the mixed-phase cloud regime between -40°C and 0°C over the Southern Ocean?"

This study examines ice particle and supercooled liquid water distributions over the Southern Ocean based on the SOCRATES. In situ observations are compared with simulations of three GCMs: the National Center for Atmospheric Research (NCAR) CAM5 and CAM6, and the Energy Exascale Earth System Model (E3SM) by the U.S. Department of Energy (DOE). CAM5 and CAM6 are the atmospheric component of the NCAR Community Earth System Model version 1 (CESM1) and version 2 (CESM2), respectively. Compared with CAM5, CAM6 has improvements applied to mixed phase cloud parameterization, prognostic precipitation species, and the interaction with aerosol schemes. E3SM uses a similar physics package as CAM6 but includes many differences, such as a different dynamical core, more vertical levels, a more detailed treatment of aerosol variety and properties, etc. The main goals of this work are to advance the understanding of statistical distributions of cloud phase and microphysical properties, the thermodynamic effect, and aerosol indirect effects on cloud characteristics over the Southern Ocean, as well as to provide evaluations on three model simulations.

In Section 2, the observation dataset and the set-up of model simulations are introduced. In Section 3, a case study of three cloud thermodynamic phases are presented. Their thermodynamic conditions (e.g., temperature and RH), cloud phases, and cloud microphysical properties are compared between the observations and model simulations. Statistical distributions of cloud phase occurrence frequencies, mass and number concentrations of cloud hydrometeors, effects of thermodynamic conditions, and aerosol
indirect effects are also analyzed based on a synthesized observation dataset. Lastly, conclusions and implications are given in Section 4.

1.2. Instrumentations and Simulations

1.2.1. In Situ Airborne Observations

The SOCRATES campaign was a flight campaign funded by the U.S. National Science Foundation (NSF) and supported by NCAR (McFarquhar et al., 2021). The campaign was conducted between 62°S – 42°S and 133°E – 164°E over the Australasian section of the Southern Ocean region from January 15 to February 24, 2018. The SOCRATES campaign studied clouds, aerosols, cloud-aerosol interaction, precipitation, and radiation over the remote region of the Southern Ocean, where climate models tend to underestimate the shortwave radiation reflected by the low-level clouds in the Austral summer, especially in the colder sector of low-pressure systems. The research flights (RFs) targeted the cold dry sector of cyclones where the presence of strong westerly and southwesterly flows along with the cold ocean surface temperatures favor the formation of low-level and mid-level clouds such as stratocumulus. For this study, the analysis is restricted to data collected between -40°C and 0°C (also referred to as the mixed phase cloud regime hereafter), which allows for the presence of both supercooled liquid water and ice particles. A total of 15 RFs were conducted, and 111 total flight hours were flown during SOCRATES. Among these observations, 14 and 73 flight hours were at in-cloud and clear-sky conditions at -40°C – 0°C, with average true airspeed at 156 and 178 m s⁻¹, respectively.

The NSF Gulfstream-V (GV) research aircraft was the platform used in the SOCRATES campaign, with scientific instruments installed to measure meteorological conditions, cloud
hydrometeors, aerosol concentrations, and other quantities. A Rosemount temperature probe measured temperature, with an accuracy and precision of ±0.3 K and 0.01 K, respectively. The Two-Dimensional Stereo Probe (2DS) and the cloud droplet probe (CDP) were mounted underneath the aircraft wings to measure particle size distributions. The CDP measures particle sizes from 2 to 50 μm, with large uncertainties in measuring ice crystal size distributions due to potential shattering and their non-spherical nature (Hallett, 2004; McFarquhar et al., 2007). The 2DS uses two orthogonal laser beams that provide a high pixel resolution of 10 μm. It nominally measures particle sizes from 10 to 1280 μm but has an uncertain depth of field for smaller particles (e.g., Baumgardner & Korolev, 1997), giving large uncertainties in calculated size distributions for size less than 50 μm (Jackson et al., 2012). Even though the width of the photodiode array for the 2DS probe is 1280 μm, the sizes of larger particles whose center is in the photodiode array are estimated by fitting a circle around the visible part of the image and then using the diameter of the circle as the dimension. Thus, the range of 40 to 5000 μm is used for 2DS measurement. The Ultra-High Sensitivity Aerosol Spectrometer (UHSAS) measures the number concentrations and size distributions of aerosols in the 60 to 1000 nanometer (nm) size range. The particle sizes measured by UHSAS data may be underestimated compared with the real ambient aerosol sizes due to the sampling process that exposes particles to lower RH than the ambient. Another instrument, the Vertical Cavity Surface Emitting Laser (VCSEL) hygrometer, was mounted on top of the aircraft and reported water vapor molecule number density at 25-Hz resolution with an accuracy of ~6% and a precision of ≤ 1% (Zondlo et al., 2010). Its product of water vapor mixing ratio was reported in 1-Hz resolution, and a PI-calibrated dataset of
water vapor mixing ratio is used in this study based on post-campaign laboratory calibration in summer 2018 (Diao, 2021). Water vapor and temperature data are used to calculate $\text{RH}_{\text{liq}}$ and $\text{RH}_{\text{ice}}$ by using the equations for $e_{s,\text{liq}}$ and $e_{s,\text{ice}}$ in Murphy and Koop (2005), respectively. Combining the uncertainties from water vapor and temperature measurements, the uncertainties for $\text{RH}_{\text{ice}}$ are 6.9% – 6.5%, and the uncertainties for $\text{RH}_{\text{liq}}$ are 6.8% – 6.4% from -40°C to 0°C, respectively. The Rosemount Icing Detector (RICE) measures supercooled water droplets by freezing the droplets collided with the detector and changing its constant vibration frequency. The data are, however, unavailable during the de-icing process and may not detect supercooled droplets for temperatures greater than -5°C. An estimated uncertainty in sensing the presence of LWC is at a limit of about 0.025 g m$^{-3}$ (Mazin et al., 2001). The RICE is used as a supportive instrument in this study for cloud phase verification.

The observed cloud phases are determined based on the cloud phase identification method from Figure 1 of D’Alessandro et al. (2019). This method is optimized for the evaluation of GCMs with a consideration of the coarser grid-box scale and definitions of simulated cloud and aerosol properties. First, this method mainly uses optical array probes (OAP) to derive cloud phases and other microphysical properties (i.e., IWC, LWC, $N_{\text{ice}}$ and $N_{\text{liq}}$), which allows the simulated cloud properties to be truncated to the same size range as the OAP instruments. Second, this method uses a quantifiable threshold – ice mass fraction – to separate ice, mixed, liquid phase, which can also be derived from the model output. These two main aspects differ from another cloud phase identification dataset for SOCRATES at 1-Hz resolution developed by D’Alessandro et al. (2021). That dataset uses LWC measured by
Figure 1. Flight tracks of a total 15 SOCRATES research flights (black) and the grid coordinates simulated by CAM6 (red), CAM5 (red) and E3SM (blue).

King probe in addition to the OAP measurements and defines a cloud segment to be mixed phase when both ice and liquid are observed, while the simulation cannot replicate the sensitivity of King probe and would contain coexisting ice and liquid in most cases at 100-km scale.

Measurements from CDP are categorized into three types (i.e., large aerosols, liquid droplets, and ice particles) based on various thresholds of particle number concentration \(N_{\text{CDP}}\) and mass concentration \(M_{\text{CDP}}\). That is, particles with either \(N_{\text{CDP}} \leq 10^{-1.5} \text{ cm}^{-3}\) or \(M_{\text{CDP}} \leq 10^{-3.4} \text{ g m}^{-3}\) are considered large aerosols; particles with \(10^{-1.5} < N_{\text{CDP}} < 10^{-0.5} \text{ cm}^{-3}\) and \(M_{\text{CDP}} > 10^{-3.4} \text{ g m}^{-3}\) are defined as ice particles; and particles with \(N_{\text{CDP}} \geq 10^{-0.5} \text{ cm}^{-3}\) and \(M_{\text{CDP}} > 10^{-3.4} \text{ g m}^{-3}\) are defined as liquid droplets. For the 2DS, if the ambient temperature \(\geq -30^\circ\text{C}\), particle number concentration \(N_{\text{2DS}}\), the maximum particle diameter \(D_{\text{max,2DS}}\), and the standard deviation of size distribution \(\sigma_{D,2DS}\) are used to categorize
liquid droplets and ice particles. If the ambient temperature < -30°C, a check of CDP reading is also required. After identifying the phase for each cloud probe, LWC is calculated for all probes assuming spherical shape and liquid density of 1000 kg m\(^{-3}\). For the 2DS, IWC is calculated for the range of 40 – 5000 μm following the habit-dependent mass-dimension (m-D) relationships documented in W. Wu and McFarquhar (2016). A crude estimate of spherical shape and ice density (900 kg m\(^{-3}\)) are used to derive IWC if CDP is identified as ice phase, which is subject to large uncertainty. But, the majority of CDP measurements (33232 seconds) are identified as liquid phase, while only a small number of CDP measurements are identified as ice (690 seconds) and typically \(M_{CDP}\) is small for ice phase periods. The final cloud phase identification and cloud microphysical properties are based on a dataset that uses the combined measurements of 2DS and CDP to calculate the total IWC and LWC. Cloud phases are defined by using the mass fraction of ice (hereafter named as the glaciation ratio), i.e., \(\text{IWC/\text{TWC}}\), where TWC stands for total water content and equals the sum of LWC and IWC. Ice, mixed and liquid phases are defined when glaciation ratio > 0.9, 0.1 ≤ glaciation ratio ≤ 0.9, and glaciation ratio < 0.1, respectively (e.g., D’Alessandro et al., 2019; Korolev & Isaac, 2003).

The quality of this cloud phase identification method is further verified by comparing against the RICE detector and manually examining the cloud images from 2DS. In two scenarios, the cloud phases are revised manually after checking the cloud images. One scenario is that at temperatures less than -25°C, there are a few occasions (2386 seconds) where the cloud phase identification method using 2DS and CDP defines the clouds as liquid phase, yet the RICE detector shows no signal of supercooled liquid water, and the cloud
images show ice particles. The other scenario is that at temperatures above 0°C, the method using 2DS and CDP identifies clouds as ice phase, yet the cloud images show drizzles (302 seconds). Besides these rare incidents, the automatic identification of the cloud phase compares well with the RICE detector.

1.2.2. Three GCM Simulations

This study evaluates three model simulations against the in situ observations, including the NCAR CESM1 / CAM5 model, an updated version CESM2 / CAM6 model, and the DOE E3SM / Atmosphere Model version 1 (EAMv1). Figure 1 shows the map of aircraft flight tracks and the collocated model output from three simulations.

Both CAM5 and CAM6 use a finite-volume dynamical core (Lin, 2004). The two models were run with a resolution of 0.9° × 1.25° and 32 vertical levels, a time step of 30 minutes, and were nudged towards MERRA-2 temperature and horizontal wind field reanalysis data. The model output was saved at the closest location to the aircraft flight track for every 1-minute observation, which facilitates a more direct comparison between the simulations and observations. Both CAM5 and CAM6 were run with a spin-up time of one year and a relaxation time of 24 hours when nudged towards the reanalysis data. The CAM5 simulation uses the MG1 cloud microphysics scheme (H. Morrison & Gettelman, 2008) coupled with a modal aerosol module with three modes (MAM3) (X. Liu et al., 2012). A detailed description of CAM5 was previously documented in Neale et al. (2012). The newer version, CAM6, uses the cloud microphysics scheme MG2 with additional improvements of ice nucleation, ice microphysics, prognostic precipitation species, and interaction with aerosol schemes to calculate cloud mass fractions and number concentrations (Gettelman & Morrison, 2015).
The MG2 microphysics scheme is coupled with an updated modal aerosol module with four modes, MAM4 (X. Liu et al., 2016). The MAM4 has an additional aerosol mode named primary carbon compared with MAM3, improving aerosol resuspension, nucleation, scavenging, and sea spray emissions. CAM6 also uses Cloud Layers Unified By Binormals (CLUBB) for turbulence and shallow convection, which replaces the original shallow convection scheme in CAM5 (Park & Bretherton, 2009).

The DOE E3SM / EAM version 1, on the other hand, is a derivative of CAM6 with a spectral element dynamical core and modified physics parameterization schemes (Rasch et al., 2019). The vertical and horizontal resolutions of E3SM are 72 layers and 1° (such gridding resolution is named as ne30), respectively. A nudged simulation towards ERA5 temperature and horizontal wind was performed, with a relaxation time scale of 6 hours (J. Sun et al., 2019). The output closest to flight track location at a 1-minute frequency is used for the comparison. The E3SM nudged simulation started on December 1, 2017 and was initialized with the initial condition output for December from a climatological run (for both atmosphere and land). This allows a relatively short simulation time for the model to spin up compared with using the default initial condition file. Similar to CAM6, E3SM also uses the MG2 microphysics scheme and MAM4, but with more detailed treatments of aerosol categories and processes such as light-absorbing particle deposition.

1.2.3. Approaches to Facilitate Comparisons between Model Simulations and Observations

Due to differences of spatiotemporal resolutions and various definitions of cloud and aerosol variables between in situ observations and model simulations, several approaches are used to select collocated samples and recalculate model output variables for comparisons.
First, for model output in an entire atmospheric column, only the model grid box with the closest location to the vertical location of the aircraft is selected. Second, since simulated cloud hydrometeors in the model cover the size range from zero to infinity, which exceeds the sampling range of CDP and 2DS, a size cutoff is applied to all simulated cloud properties by restricting the particle size to the range of 0 to 5000 μm. As shown in the particle size distribution of simulated hydrometeors by Gettelman et al. (2020), simulated snow partially overlaps with the measurement range of combined 2DS+CDP. Therefore, the sum of simulated ice and snow mass concentrations for a specific size range (i.e., 0 – 5000 μm in this study) is used to compare with the observed IWC derived from measurements of a similar size range. A similar method was previously used in several model-observation comparison studies, such as Fridlind et al. (2007), Eidhammer et al. (2014), and Patnaude et al. (2021). Table S1 shows the contributions of various discrete size ranges of ice and snow to their mass concentrations in the CAM6 simulation. The model output variables being processed for this partial size range include “LWC”, “NUMLIQ”, “IWC”, “NUMICE”, “AQSNOw”, and “ANSNOW”. These variables represent grid-average values for mass and number concentrations of liquid, ice, and snow, respectively. The simulated LWC and liquid number concentration (N_{liq}) are defined by the size-restricted “LWC” and “NUMLIQ”, respectively. We further define the simulated IWC as the sum of size-restricted “IWC” and “AQSNOw” and the simulated ice number concentration (N_{ice}) as the sum of size-restricted “NUMICE” and “ANSNOW”, which means that the simulated ice phase includes both ice crystals and snow since cloud measurements in the observations include both ice crystals and snow. Aerosol number concentrations (Na) from the simulations are also restricted to aerosol
sizes ≤ 1000 nm based on a log-normal distribution, which follows the size range of the UHSAS measurements. The RH_{ice} and RH_{liq} values in the simulations are calculated based on water vapor specific humidity and the saturation vapor pressure equations from Murphy and Koop (2005), which avoids using the RH variable directly reported by the model. The average values of calculated RH_{liq}, RH_{ice} in contrast to the original model output “RELHUM” in every 5-degree temperature bin are shown in supplementary Figure S1. The simulated RH_{liq} shows the maximum values at 101%, 100%, and 105% for CAM6, CAM5, and E3SM, respectively. For the observations, RH_{liq} values greater than 105% are set as NAN values (processed for 2019 seconds) due to the combined uncertainties from water vapor and temperature measurements.

The observations define in-cloud conditions as at least one cloud hydrometeor has been detected by either CDP or 2DS. The maximum and minimum LWC (IWC) values in the observations are 2.41 and 3.06\times10^{-5} \text{ g m}^{-3} (76.5 and 6.05\times10^{-6} \text{ g m}^{-3}), respectively. For simulations, if IWC or LWC is less than 10^{-7} \text{ g m}^{-3}, they are not considered as real hydrometeors and are set to zero. This means that for simulations, in-cloud conditions are defined as either IWC or LWC being greater than 10^{-7} \text{ g m}^{-3}, while the remaining conditions are considered as clear sky. In addition, only N_{liq} and N_{ice} greater than 10^{-7} \text{ cm}^{-3} are used in the analysis of simulations. These in-cloud thresholds are chosen mainly to facilitate the comparison between observations and simulations, since a higher threshold would significantly reduce the number of in-cloud samples from model output. Note that when LWC and IWC are below 0.001 \text{ g m}^{-3}, the observations do not represent real clouds, but rather represent one or a few hydrometeors. For brevity, we call them in-cloud regions, but
they differ from the conventional definition of clouds used in previous studies (e.g., McFarquhar & Heymsfield, 2001; McFarquhar et al., 2007). Table 1 summarizes the maximum and minimum values for thermodynamic conditions (i.e., temperature, pressure, RH_{ice}, and RH_{liq}) and cloud microphysical properties (i.e., LWC, IWC, N_{liq}, and N_{ice}) used for the analysis of observations and simulations.

Table 1. The Maximum and Minimum Values of Thermodynamic Conditions and Cloud Microphysical Properties Used in This Study for Observations and Simulations

<table>
<thead>
<tr>
<th>Variables</th>
<th>1-s observations</th>
<th>200-s observations</th>
<th>580-s observations</th>
<th>CAM6</th>
<th>CAM5</th>
<th>E3SM</th>
</tr>
</thead>
<tbody>
<tr>
<td>T (°C)</td>
<td>-39.7 – 0.0</td>
<td>-39.7 – 0.0</td>
<td>-39.7 – 0.0</td>
<td>-37.6 – 0.0</td>
<td>-37.4 – 0.0</td>
<td>-38.6 – 0.0</td>
</tr>
<tr>
<td>LWC (g m^{-3})</td>
<td>3.06×10^{-5} – 2.41</td>
<td>3.33×10^{-7} – 0.74</td>
<td>1.20×10^{-7} – 0.60</td>
<td>1.00×10^{-7} – 0.36</td>
<td>1.02×10^{-7} – 0.43</td>
<td>2.40×10^{-7} – 0.40</td>
</tr>
<tr>
<td>IWC (g m^{-3})</td>
<td>6.05×10^{-8} – 8.76</td>
<td>3.78×10^{-10} – 0.89</td>
<td>1.30×10^{-10} – 0.67</td>
<td>1.24×10^{-10} – 0.17</td>
<td>1.20×10^{-10} – 0.28</td>
<td>1.03×10^{-10} – 0.18</td>
</tr>
<tr>
<td>N_{liq} (cm^{-3})</td>
<td>5.66×10^{-5} – 565.72</td>
<td>2.96×10^{-7} – 218.19</td>
<td>1.04×10^{-7} – 210.79</td>
<td>4.04×10^{-7} – 111.01</td>
<td>1.93×10^{-7} – 150.09</td>
<td>3.6×10^{-7} – 91.35</td>
</tr>
<tr>
<td>N_{ice} (cm^{-3})</td>
<td>5.66×10^{-5} – 38.89</td>
<td>2.95×10^{-7} – 4.34</td>
<td>1.04×10^{-7} – 1.58</td>
<td>9.49×10^{-8} – 0.09</td>
<td>1.77×10^{-7} – 0.56</td>
<td>9.17×10^{-8} – 2.49</td>
</tr>
<tr>
<td>In-cloud RH_{liq} (%)</td>
<td>2.6 – 105.0</td>
<td>3.9 – 103.1</td>
<td>6.0 – 101.7</td>
<td>18.0 – 101.3</td>
<td>42.3 – 99.5</td>
<td>17.9 – 104.5</td>
</tr>
<tr>
<td>In-cloud RH_{ice} (%)</td>
<td>2.9 – 146.7</td>
<td>4.1 – 140.3</td>
<td>7.3 – 131.7</td>
<td>19.5 – 133.8</td>
<td>52.1 – 115.8</td>
<td>19.1 – 143.8</td>
</tr>
<tr>
<td>Clear-sky RH_{liq} (%)</td>
<td>1.1 – 104.9</td>
<td>1.7 – 93.1</td>
<td>2.0 – 86.9</td>
<td>5.3 – 97.4</td>
<td>5.4 – 97.0</td>
<td>0.0 – 98.8</td>
</tr>
<tr>
<td>Clear-sky RH_{ice} (%)</td>
<td>1.2 – 142.2</td>
<td>2.1 – 117.7</td>
<td>2.3 – 109.2</td>
<td>6.3 – 110.9</td>
<td>6.5 – 103.6</td>
<td>0.0 – 125.0</td>
</tr>
</tbody>
</table>

To examine the effect of spatial scales to the comparison results, 1-Hz observations are averaged by various scales, including 200 seconds (i.e., 34.5 km horizontal resolution, since the average true airspeed of the aircraft at -40°C to 0°C is 172 m s^{-1}) and 580 seconds (i.e., 100 km horizontal resolution) using a moving average method similar to D’Alessandro et al. (2019). A restriction on the averaging process of the observation data is added to reduce the impact of the saw-tooth data collection strategy during the SOCRATES campaign.

Specifically, the observation data used to calculate the moving average surrounding each second must be within the pressure boundaries of one CAM model grid box. In addition, if more than 50% of the averaging data points are outside the pressure boundaries of such CAM model grid box, this averaged datum will be discarded and not used to compare with model
This moving average generally leads to smaller values of average IWC, LWC, \( N_{\text{ice}} \), and \( N_{\text{liq}} \) in coarser scale data than the 1-s data, since the coarser scale data include both clear-sky and in-cloud segments during the averaging process. Since these observation data represent averages over the entire length scale, they are comparable with simulated grid-average cloud quantities.

### 1.3. Results

#### 1.3.1. Cloud Phases, Microphysical Properties, and Thermodynamic Conditions in a Case Study

A segment from the RF 05 was selected to show an example of supercooled liquid water and ice particle distributions using 2DS and CDP (Figure 2). The observed temperature ranges from -9°C to -6°C, and the RH\(_{\text{liq}}\) values are located around or slightly below liquid saturation between UTC 01:53:20 – 01:55:45, favoring the existence of supercooled liquid water at this period (Figure 2a). The magnitude of ice supersaturation (i.e., RH\(_{\text{ice}}\) – 1) is about 5% – 10%, which may partially contribute to the ice formations around UTC 01:56:00 to 01:56:40. The IWC values, derived from the combined 1-Hz CDP and 2DS measurements, remain relatively constant at 0.01 g m\(^{-3}\) for most in-cloud conditions, while the LWC are mostly above 0.1 g m\(^{-3}\) (Figure 2b). The cloud phase identifications are shown in Figure 2c. 2DS cloud imageries are shown in Figure 2d, e, and f for liquid, mixed, and ice phase, respectively. For this specific case, all three models simulate clouds for the entire period. Both CAM6 and E3SM show mixed phase clouds while CAM5 shows ice phase only.
Figure 2. An example time series of clouds with a mixture of ice, liquid, and mixed phases in NSF SOCRATES campaign Research Flight 5 (RF05). (a) 1-Hz observations of temperature (dark green), RH_{ice} (blue), RH_{liq} (red), and RH = 100% (dashed black). (b) 1-Hz observations of log-scale IWC (blue) and LWC (red) using 2DS and CDP probes. (c) Cloud phases identified from the observations (vertical bars) and the models (dots). (d), (e), and (f) illustrate the seconds of liquid, mixed, and ice cloud particle imageries, respectively, captured by the 2DS.

1.3.2. Cloud Phase Occurrence Frequency and Distributions of LWC, IWC and Glaciation Ratio

Cloud phase occurrence frequencies for the entire SOCRATES campaign are compared with model simulations (Figure 3). The number of samples for three cloud phases is shown in the supplementary Figure S2. Figure 3b and c show the cloud phase occurrence frequencies
Figure 3. Cloud phase occurrence frequencies for (a) 1-Hz SOCRATES observations, (b) 200-s averaged observations, and (c) 580-s averaged observations with temperature ranged from -40°C to 0°C, binned by 5°C. Cloud phase occurrence frequencies for CAM6, CAM5, and E3SM are shown for two types of simulated IWC: (d – f) including snow and (g – i) excluding snow. Liquid, mixed, and ice phase are denoted by red, green, and blue colors, respectively.

for 200 s and 580 s spatially averaged observation (horizontal scales of 35 and 100 km, respectively). An increase in spatial scale also increases the occurrence frequencies of mixed phase between -35°C to 0°C by a factor of 2 – 4, i.e., mixed phase frequencies are 0.05 – 0.1 for 1-s observations, compared with 0.1 – 0.2 for 200-s and 0.1 – 0.35 for 580-s observations.

Simulations are further examined with two types of simulated IWC – one contains both ice crystals and snow (Figure 3d–f) which is used as the default definition of simulated IWC,
while the other contains only ice crystals (Figure 3g–i). Excluding snow as part of the simulated IWC increases (decreases) liquid (mixed) phase frequency by 0.1 – 0.2 in three simulations. Compared with the 580-s observations, CAM6 (Figure 3d) shows the most similar cloud phase frequencies for ice, liquid, and mixed phases among the three models. The 580-s averaged observations show an intersection of the liquid (red) and ice (blue) frequency lines with a significant decrease (increase) in ice (liquid) phase frequency between -20°C and -15°C. A similar feature is shown in CAM6 but occurs at a higher temperature range between -15°C and -10°C. The minor issue with CAM6 is the slightly lower mixed phase frequency and higher ice phase frequency by 0.1 at -10°C to -5°C than 580-s averaged observations. CAM6 significantly improves the presence of supercooled liquid water below -10°C compared with CAM5, since CAM5 shows zero frequency of liquid-containing clouds below -10°C. The lack of supercooled liquid water at temperature less than -10°C in CAM5 was also shown in the previous work of D’Alessandro et al. (2019). E3SM (Figure 3f) underestimates (overestimates) the frequency of ice phase clouds below (above) -20°C by 0.1 – 0.3 compared with 580-s averaged observations. E3SM was found to overestimate of liquid cloud fraction between -20°C and -30°C at high latitudes (Y. Zhang et al., 2019). It was also found to underestimate pure ice clouds at most temperatures except for close to -40°C based on a global-scale evaluation (Rasch et al., 2019). A similar result of E3SM overestimating supercooled liquid water below -20°C was also documented in M. Zhang et al. (2020) for an analysis of Arctic clouds. The similarity of E3SM model biases between those previous studies and this study suggests that these model biases of cloud phase frequency are
consistently shown for high latitudinal regions in both hemispheres. Yet more detailed hemispheric comparisons are needed in future work.

Given the importance of temperature nudging on simulation results (e.g., Gettelman et al., 2020), supplementary Figure S3 compares the temperatures of CAM6, CAM5, and E3SM simulations with observations using the nearest model grid to the flight track. In general, both CAM5 and CAM6 show distributions of temperature biases skewed toward low biases, while E3SM shows a more symmetrical temperature bias distribution as well as a smaller mean bias compared with the observations. That is, 65\% of E3SM temperatures are within 1 degree of the observed temperatures (supplementary Figure S3a). This is mainly due to different reanalysis data used for nudging, i.e., E3SM was nudged towards ERA5 data while CAM6 and CAM5 were nudged towards MERRA-2. All three models show similar temperature vertical profiles with respect to pressure compared with the observations (supplementary Figure S3b). This analysis demonstrates that CAM6 having the most similar cloud phase frequency distributions to the observations cannot be explained by a closer temperature match to the observations, since E3SM shows the most similar temperature distribution to the observations.

In addition, the impacts of various in-cloud condition thresholds on cloud phase statistical distributions are also examined in Figure 4. Number of samples for Figures 4 is shown in supplementary Figure S4. Figure 4a–d compare various in-cloud thresholds: $1 \times 10^{-7}$ g m$^{-3}$, $1 \times 10^{-5}$ g m$^{-3}$, 0.001 g m$^{-3}$, and 0.01 g m$^{-3}$. All of these sensitivity tests show very consistent results even when different in-cloud thresholds are used. In addition, using a higher in-cloud threshold (i.e., $1.7 \times 10^{-5}$ g m$^{-3}$,) for the CAM6 simulation (Figure 4e) also shows similar
Figure 4. Sensitivity tests on cloud phase occurrence frequencies using various thresholds for defining in-cloud conditions for SOCRATES observations, including (a) $10^{-7}$ g m$^{-3}$, (b) $10^{-5}$ g m$^{-3}$, (c) 0.001 g m$^{-3}$, (d) 0.01 g m$^{-3}$. (e) CAM6 simulation using in-cloud threshold of $1.7 \times 10^{-5}$ g m$^{-3}$. Using all model output in the entire temperature range of -40°C to 0°C for (f) CAM6, (g) CAM5, and (h) E3SM.

results of cloud phase frequency distribution compared with using the threshold of $1 \times 10^{-7}$ g m$^{-3}$. Another sensitivity test in Figure 4f–h uses the entire model columns between -40°C to 0°C to compare with observations, in contrast to the restriction of comparing observations with the nearest model grid box only in vertical level (as shown in Figures 2, 3 and 5–14).

The result shows that removing the temperature collocation restriction worsens the model-observation comparison results. Nevertheless, the main conclusions are consistently seen. For example, CAM6 still shows the most comparable results to observations, while E3SM still underestimates and overestimates ice phase below -20°C and above -20°C, respectively. This sensitivity test suggests that the model biases seen in this study are unlikely driven solely by the mismatches of time and space for simulated and observed cloud occurrences.
Effects of spatial scales are examined in Figure 5a–c for observations that are spatially averaged by every 10 s, 50 s, 100 s, 200 s, 290 s, and 580 s, which represent horizontal scales of 1.7, 8.6, 17, 35, 50 and 100 km, respectively. The overall trend of an increasing liquid (ice) phase frequency in a warmer (colder) environment remains unchanged. Larger spatial scales consistently show increases in the occurrence frequencies of mixed phase between -35°C to 0°C. Furthermore, length scales of three cloud phases in 1-Hz observations are examined in Figure 5d–f. The number of samples for Figure 5 is shown in supplementary Figure S5. Length scales of individual cloud phase segments are calculated by the consecutive seconds of the same cloud phase in 1-Hz observations. The shorter and longer length scales represent more heterogeneous and homogeneous distributions of cloud phases, respectively. The observations show more mixed phase segments at shorter length scales (1–3 seconds) than longer length scales (> 10 seconds), while the liquid and ice phases dominate the longer length scales (> 10 seconds). This result indicates that the coexistence of ice and liquid occurs more frequently at shorter length scales, likely due to the effective transition from liquid to ice via the WBF process.

Cloud microphysical properties, i.e., LWC, IWC, and glaciation ratios, are examined for various temperatures (binned by 5°C) in Figure 6. The number of samples for this analysis is shown in supplementary Figure S6. 1-s, 200-s, and 580-s averaged observations are compared with model simulations. At temperature greater than -35°C, averaging observations over 200 seconds significantly reduces the average LWC and IWC by 1 – 2 orders of magnitude than the 1-s data, while the 580-s averaged observations show a further reduction of LWC and IWC by up to 0.5 order of magnitude. The moving averages generally lead to
Figure 5. Cloud phase occurrence frequencies for (a) liquid phase, (b) mixed phase, and (c) ice phase are shown for 1-s, 10-s, 50-s, 100-s, 200-s, 290-s, and 580-s averaged observations. (d – f) Occurrence frequency of various length scales of cloud phase, calculated based on 1-Hz observations, including (d) 1-s – 3-s length scale, (e) 4-s – 10-s length scale, and (f) more than 10-s length scale.

smaller values of average IWC, LWC, N_{ice}, and N_{liq} in coarser scale data compared with the 1-s data, since the coarser scale data include both clear-sky and in-cloud segments during the averaging process. Compared with 580-s averaged observations, CAM6 (E3SM) shows similar average LWC to the observations above -25ºC (-20ºC) but higher average LWC at lower temperatures by 1 – 2 orders of magnitude. Consistent with Figure 3, CAM5 lacks LWC at temperatures less than -10ºC. For the average IWC, CAM6 underestimates IWC by 0.5 – 1 order of magnitude between -40ºC and -30ºC compared with 580-s averaged observations. E3SM underestimates IWC by 0.5 – 1 order of magnitude between -40ºC and -20ºC, and overestimates IWC by 0.5 – 1 orders of magnitude between -20ºC and 0ºC.
Figure 6. Averages and standard deviations of (a – c) log-scale LWC, (d – f) log-scale IWC, (g – i) glaciation ratio (i.e., IWC/TWC), and (j – l) glaciation ratio only when ice particles and supercooled liquid water coexist (i.e., IWC/TWC only when both IWC > 0 and LWC > 0). 1-Hz observations (solid black), 200-s observations (dashed black), 580-s observations (dotted black), and model simulations (red) are binned at 5°C interval from -40°C to 0°C.
Two types of glaciation ratios are calculated. One is for all in-cloud conditions (Figure 6g–i), and the other one is for conditions with coexisting ice particles and supercooled liquid water only (j–l). For the former type, the glaciation ratios are controlled by the ratios between ice phase and liquid phase occurrence frequencies, the two dominant phases. For the latter type, the glaciation ratios are controlled by the mass partitioning between ice and liquid when they coexist. For the former type of glaciation ratios, CAM6 shows the most similar results to the 580-s averaged observations (Figure 6g), which is consistent with the cloud phase frequency analysis in Figure 3. The latter glaciation ratios in CAM6 (Figure 6j) are close to the averaged observations above -20°C but are significantly lower by 0.2 – 0.6 below -20°C compared with observations due to the underestimation of IWC. This result is consistent with the LWC and IWC analysis shown in Figure 6a and d. E3SM overestimates the former type of glaciation ratios above -20°C and underestimates both the former and the latter type of glaciation ratios below -20°C. These analyses show that the underestimation of IWC and overestimation of LWC by CAM6 and E3SM at temperatures below -20°C leads to large biases of mass partitioning inside the mixture of ice and liquid.

A similar analysis to Figure 6 is done using simulated IWC containing only ice crystals (supplementary Figure S7). When excluding snow in the simulated IWC, larger model biases of IWC by a factor of 0.5 – 1 are seen compared with including snow in the simulated IWC. Additionally, a sensitivity test is conducted to examine the impacts of model output frequency, by using E3SM output closest to every 1 second, 1 minute, and 10 minutes of observations (supplementary Figure S8). Increasing the temporal resolution also increases the liquid and mixed phase frequencies. Overall, the analysis shows very similar results for
average LWC, IWC, and glaciation ratios at various temperatures regardless of using various model output frequencies.

1.3.3. Thermodynamic Conditions for Clear-Sky, In-Cloud Conditions and Three Cloud Phases

Thermodynamic conditions are crucial for the formation of ice particles and supercooled liquid water, as illustrated in the case study in Section 3.1. Figure 7 shows probability density functions (PDFs) of temperature and RH\textsubscript{ice} categorized by in-cloud and clear-sky conditions (top two rows) and three cloud phases (bottom two rows). The PDF is calculated as the number of samples of a certain condition (such as in-cloud) at each bin divided by the total number of samples of that condition in all bins.

PDFs of temperatures of both clear-sky and in-cloud conditions are comparable between observations and simulations. Specifically, all three models show higher (lower) PDFs of in-cloud condition compared with the PDFs of clear-sky condition at temperatures above (below) -15°C, which is consistent with the distributions seen in the 1-s and 580-s observations. This feature indicates that the general vertical locations of cloud layers are similar between the observations and model simulations.

PDFs of RH\textsubscript{ice} for in-cloud conditions in the simulations show lower maximum values (CAM6 134%, CAM5 116%, E3SM 144%) compared with 1-s observations (147%), but the simulated values of CAM6 and CAM5 are closer to 580-s averaged observations (132%). Similarly, PDFs of RH\textsubscript{ice} for clear-sky conditions in the simulations show lower maximum values (CAM6 111%, CAM5 104%, E3SM 125%) than 1-s observations (142%) but are closer to 580-s averaged observations (109%). The simulations also underestimate the
Figure 7. PDFs of (a – e) temperature and (f – j) RH\textsubscript{ice} for all data (solid black line), clear-sky (dashed black), and in-cloud (dotted black) conditions. PDFs of (k – o) temperature and (p – t) RH\textsubscript{ice} separated into three cloud phases, i.e., ice (blue), mixed (green), and liquid (red) phase. Each PDF is calculated by the number of a certain condition in a bin divided by the total number of samples of that condition of all bins.

frequencies of sub-saturated conditions for in-cloud RH\textsubscript{ice}, since the 1-s and 580-s averaged observations show minimum in-cloud RH\textsubscript{ice} at 3\% and 7\%, respectively, while simulations show minimum values of 19\% – 52\%.
In terms of PDFs of RH_{ice} in three cloud phases, the peak positions of RH_{ice} in 1-s observations are located around 100% – 102% for all three phases. For 580-s observations, the frequencies are similar between the peak position at 102% and the surrounding values from ~85% to 100% due to the inclusion of clear-sky segments in the averaging process. Three simulations show peaks of in-cloud RH_{ice} around 100% but with narrower ranges for all three cloud phases. For both observations and simulations, mixed phase is associated with a narrower RH_{ice} range than ice and liquid phases, consistent with the theoretical condition for WBF process with e_{s,ice} < e < e_{s,liq}. The lack of sub-saturated conditions for ice phase may contribute to the underestimation of ice growth and the riming effect during sedimentation, which possibly leads to lower IWC in the simulations.

1.3.4. Aerosol Indirect Effects on Cloud Microphysical Properties

In this section, aerosol indirect effects on cloud microphysical properties at various temperatures are examined based on the relationships between total aerosol number concentrations (Na) and cloud microphysical properties (Figures 8–11). According to previous studies using in situ measurements to examine aerosol indirect effects (e.g., Chubb et al., 2016; Field et al., 2012; Wood et al., 2018), aerosol number concentrations were often restricted to clear-sky conditions for the analysis of relationships between aerosols and cloud properties in order to reduce the impacts of cloud hydrometeors on aerosol measurements. Thus, average aerosol number concentrations were calculated using clear-sky segments only for 50-second averaged and 200-second averaged observations. Considering that simulated aerosol number concentrations represent the aerosol concentrations in a model grid box, the observed aerosol concentrations are not restricted to only above or below clouds in this
analysis as commonly done for process orientated studies. In addition, since CAM5 significantly underestimates the amount of supercooled liquid water below -10°C, the model evaluation in this section focuses on CAM6 and E3SM only. The analysis is based on Na separated into two groups – aerosols with diameters > 500 nm (hereafter named as Na\textsubscript{500}) and diameters > 100 nm (named as Na\textsubscript{100}). Supplementary Figures S9 and S10 show the number of samples used for the analysis of Na\textsubscript{500} and Na\textsubscript{100}, respectively. Previously, DeMott et al. (2010) showed that at temperatures higher than -36°C, Na\textsubscript{500} is well correlated with the number concentrations of INPs, which can facilitate ice crystal formation during heterogeneous nucleation.

For the impacts of larger aerosols, as \(\log_{10}(Na_{500})\) increases, the 50-s and 200-s averaged observations both show increasing \(\log_{10}(LWC)\) and \(\log_{10}(N_{\text{liq}})\) between -20°C and 0°C and increasing IWC and \(N_\text{ice}\) between -36°C and 0°C (Figure 8). Aerosol indirect effects are further quantified by applying linear regressions to cloud microphysical properties in relation to \(\log_{10}(Na_{500})\) (Figure 9). The linear regression analysis examines the bin-average \(\log_{10}(LWC)\) and \(\log_{10}(N_{\text{liq}})\) between -20°C and 0°C, as well as the bin-average IWC and \(N_\text{ice}\) between -36°C and 0°C, since these two temperature ranges show distinct aerosol indirect effects (Figure 8). The slopes (b) of the linear regressions for logarithmic scale LWC, IWC, \(N_{\text{liq}}\), and \(N_\text{ice}\) in 50-s (200-s) observations are 0.58, 0.57, 0.96, and 0.82 (0.63, 0.47, 0.83, and 0.87), respectively. The similar slopes between LWC and IWC as well as between \(N_{\text{liq}}\) and \(N_\text{ice}\) indicate that Na\textsubscript{500} has similar magnitudes of impacts on liquid droplets and ice particles.
Figure 8. Cloud microphysical properties with respect to logarithmic scale Na\textsubscript{500} for 1-s observations, 580-s averaged observations, CAM6, and E3SM at various temperatures. Bin colors denote the average of (a – d) $\log_{10}(\text{LWC})$, (e – h) $\log_{10}(\text{IWC})$, (i – l) $\log_{10}(\text{N}_{\text{liq}})$, and (m – p) $\log_{10}(\text{N}_{\text{ice}})$.

For the impacts of smaller aerosols (Figures 10 and 11), Na\textsubscript{100} shows similar magnitude of impacts on N\textsubscript{liq} ($b = 0.81, 0.63$) compared with Na\textsubscript{500} ($0.96, 0.83$), but much smaller impacts on LWC ($0.23, 0.18$) compared with Na\textsubscript{500} ($0.58, 0.63$) for 50-s and 200-s observations, respectively. No significant correlation is seen between IWC and Na\textsubscript{100}. These results indicate that both larger and smaller aerosols may serve as CCN to facilitate liquid droplet formation, while larger aerosols more likely exceed the critical radius for spontaneous droplet formation. The lack of impacts from Na\textsubscript{100} on ice phase is consistent with the fact that larger aerosols are more likely to be activated as INPs. Overall, these results
Figure 9. Linear regressions applied (black line) for the bin-average of (a – d) LWC, (e – h) IWC, (i – l) \( N_{\text{liq}} \), and (m – p) \( N_{\text{ice}} \) in relation to \( \log_{10}(N_{a100}) \). Bin sizes follow the respective sub-panels in Figure 8. The slope, intercept, \( R^2 \) values and number of samples are shown in the text box.

indicate Twomey effects from larger aerosols on both liquid droplets and ice particles in the mixed-phase cloud regime, but smaller aerosols only show Twomey effect on liquid droplets. When smaller aerosols act as CCN, the riming indirect effect and the thermodynamic indirect effect can both lead to higher \( N_{\text{liq}} \), which further lead to smaller IWC via less effective riming and less effective SIP, respectively. However, the thermodynamic indirect effect would also lead to smaller \( N_{\text{ice}} \), while the riming indirect effect has smaller impact on \( N_{\text{ice}} \). Since Figure 11 m shows no significant change of \( N_{\text{ice}} \) with respect to increasing \( N_{a100} \), it indicates
that the riming indirect effect is the dominant mechanism seen for indirect effect from small aerosols while the thermodynamic indirect effect is less dominant over the Southern Ocean. On the other hand, the glaciation indirect effect seems to be the dominant indirect effect from larger aerosols, since larger aerosols acting as INPs can lead to higher N_{\text{ice}} and higher IWC, which is consistent with Figure 9e and m. In terms of model simulations, both CAM6 and E3SM capture the decreasing trend of maximum Na_{500} and Na_{100} as temperature decreases. Yet, the maximum values of simulated Na_{500} and Na_{100} are 10 and 100 cm$^{-3}$, respectively, which are 0.5 – 1 order of magnitude smaller than the 200-s observations (Figures 8 and 10).
Figure 11. Similar to Figure 9 but applying linear regressions to cloud microphysical properties in relation to \( \log_{10}(\text{Na}_{100}) \).

For aerosol indirect effects of Na500, CAM6 shows small positive correlations with \( N_{\text{liq}} \) (b = 0.19), negative correlations with IWC (-0.32) and \( N_{\text{ice}} \) (-0.24), and no significant correlations with LWC (Figure 9). E3SM shows small negative correlations with LWC (b = -0.18), \( N_{\text{liq}} \) (-0.087), and small positive correlations with \( N_{\text{ice}} \) (0.112).

For aerosol indirect effects of Na100 (Figure 11), CAM6 shows similar magnitudes of positive correlations with LWC (b = 0.34) and \( N_{\text{liq}} \) (0.46) compared with observations.

However, CAM6 shows negative correlations with IWC (-0.42) and \( N_{\text{ice}} \) (-0.29) while the observations show no correlations. E3SM shows negative correlations with LWC (b = -0.26)
and no correlations with IWC, N_{liq}, and N_{ice}. Overall, these results indicate that CAM6 is able to capture Twomey effect on liquid droplets from Na_{100} but not from Na_{500}, and it shows the opposite impacts on ice crystals from Na_{500} compared with observations. E3SM is able to capture a weak Twomey effect on ice crystals from Na_{500}, but underestimates Twomey effects on liquid droplets from Na_{500} and Na_{100}. In particular, both CAM6 and E3SM underestimate aerosol indirect effects on IWC.

Aerosol indirect effects on phase partitioning and cloud fraction are examined in Figure 12. The number of samples for Figure 12 is shown in supplementary Figure S11. Two types of glaciation ratios are examined – for all in-cloud conditions (Figure 12a–h) and coexisting ice and liquid only (Figure 12i–p). Since aerosols can serve as CCN and INPs as discussed above, the relationships between cloud fraction and aerosol number concentrations are also examined in Figure 12q–x. Cloud fraction is calculated by normalizing the number of in-cloud samples in each bin by the total number of samples in that bin. Note that the cloud fraction for simulations is not based on the model output “cloud fraction” but rather is calculated based on the in-cloud definition described in Section 2.3. 100% cloudiness is seen in 50-s (200-s) observations at Na_{500} > 3 cm^{-3} (> 0.3 cm^{-3}) and Na_{100} > 300 cm^{-3} (>100 cm^{-3}). On the other hand, the temperature effect is much stronger than aerosol indirect effects on cloud fraction in CAM6 and E3SM, with many model bins exceeding 70% cloudiness between -15°C and 0°C.
Figure 12. Relationships of (a – h) glaciation ratio of all clouds, (i – p) glaciation ratio only when ice particles and supercooled liquid water coexist, and (q – x) cloud fraction with respect to (row 1, 3, 5) $\log_{10}(Na_{500})$ and (row 2, 4, 6) $\log_{10}(Na_{100})$ at various temperatures. Columns 1 to 4 represent 50-s observations, 200-s observations, CAM6 and E3SM, respectively.
Two temperature ranges – between -40°C and 0°C and between -20°C and 0°C – show distinctive differences in aerosol indirect effects. Thus, we further examine the correlations of glaciation ratios and cloud fraction in relation to Na_{500} and Na_{100} in these two temperature ranges, separately (Figures 13 and 14). Linear regressions are applied to bin-average glaciation ratios and cloud fraction in relation to \log_{10}(Na_{500}) and \log_{10}(Na_{100}). In the lower temperature range of -40°C to -20°C, observations show positive correlations of two types of glaciation ratios and cloud fraction in relation to Na_{500}, and negative correlations of all three properties in relation to Na_{100}. This result indicates that at lower temperatures, larger aerosols are more effective in increasing IWC than increasing LWC, therefore glaciation ratios increase. Comparatively, smaller aerosols are more effective of increasing LWC and have almost no effects on IWC (Figure 11), therefore glaciation ratios decrease. Since cloud phase is dominated by ice phase at lower temperatures (Figure 3), higher Na_{500} means more larger aerosols that can potentially serve as INPs, therefore cloud fraction also increases. On the other hand, since smaller aerosols are unlikely to serve as INPs, they may experience wet removal through precipitation, which possibly leads to the negative correlations between Na_{100} and cloud fraction. Both CAM6 and E3SM are able to capture increases of glaciation ratios with increasing Na_{500}. However, they both show negative correlations between Na_{500} and cloud fraction at the lower temperatures.
Figure 13. Linear regressions (black line) applied for (a – h) glaciation ratio of all clouds, (i – p) glaciation ratio only when ice particles and supercooled liquid water coexist, and (q – x) cloud fraction in relation to logarithmic scale aerosol number concentrations between -40°C and -20°C. Impacts of log$_{10}$(Na$_{500}$) are shown in rows 1, 3, and 5. Impacts of log$_{10}$(Na$_{100}$) are shown in rows 2, 4, and 6.
Figure 14. Similar to Figure 13 but for temperatures between -20°C and 0°C.

The main difference between the higher temperature range (-20°C to 0°C) and lower temperature range (-40°C to -20°C) is that the higher temperatures show similar impacts
from Na_{500} and Na_{100} on glaciation ratios and cloud fraction, while the lower temperatures show opposite impacts from Na_{500} and Na_{100}. While Na_{500} and Na_{100} increase at the range of -20°C to 0°C, the two types of glaciation ratios decrease and cloud fraction increases. This result indicates that the enhancement of N_{liq} and LWC due to higher aerosol concentrations at the higher temperatures leads to less effective riming and SIP, which results in lower glaciation ratios. Since cloud phase is dominated by liquid phase at this temperature range, the higher N_{liq} and LWC with higher Na contribute to higher cloud fraction, even though ice particle growth and SIP decrease. Both CAM6 and E3SM are able to capture the negative correlations between Na_{500} and glaciation ratios, but only CAM6 captures the negative correlations between Na_{100} and glaciation ratios. E3SM captures the positive correlation between cloud fraction and higher Na_{500}, but shows a negative correlation between cloud fraction and Na_{100}. CAM6, however, does not show significant correlations of cloud fraction in relation to either Na_{500} or Na_{100}.

1.4. Conclusions and Implications to Model Development

This study focuses on examining cloud characteristics at -40°C to 0°C over the Southern Ocean based on in situ aircraft-based observations and three GCM simulations (i.e., CAM6, CAM5, and E3SM). A series of cloud characteristics are examined, including cloud phases, mass and number concentrations of cloud hydrometeors, phase partitioning, thermodynamic conditions, and aerosol indirect effects. Several approaches are used to facilitate the comparison between in situ observations and GCM simulations, including using nudged simulations toward reanalysis data, recalculating cloud properties based on instrument measurement ranges, and examining the impacts of spatial scales on the comparison results.
Spatially averaging observation data from 1 s to 580 s (i.e., from ~0.2 – 100 km in horizontal) is found to affect several variables, such as reducing average LWC and IWC by 1 – 2 orders of magnitude due to the inclusion of clear-sky segments in the grid-mean averages, increasing the occurrence frequency of mixed phase clouds since ice particles and supercooled liquid water are more likely to coexist at coarser scales, reducing the maximum RH_{ice} for in-cloud and clear-sky conditions, and decreasing the peak positions of RH_{ice} PDFs for three cloud phases. For other characteristics, spatial averaging has a small impact on the average glaciation ratios of all in-cloud conditions, and the positive correlations of LWC, IWC, N_{liq}, and N_{ice} with respect to aerosol number concentrations.

Evaluation of three model simulations shows that CAM6 has the most similar cloud phase occurrence frequency to observations compared with CAM5 and E3SM. Particularly, CAM6 and E3SM significantly improve the proportion of liquid and mixed phase clouds below -15°C compared with CAM5. This is most likely due to the removal of a temperature-dependent mass partitioning function between ice and liquid in the shallow convection scheme (Park & Bretherton, 2009) that was previously used in CAM5, as discussed in previous studies (Gettelman et al., 2020; Kay et al., 2016). E3SM underestimates (overestimates) ice phase frequencies below (above) -20°C. When comparing simulated LWC with 580-s observations, CAM6 and E3SM overestimate LWC values by 1 – 2 orders of magnitude below -25°C and -20°C, respectively. Another main model bias is the underestimation of IWC for CAM6 and E3SM below -25°C and -20°C, respectively, by 0.5 – 1 orders of magnitude compared with 580-s observations. Even though CAM6 shows small biases of glaciation ratios of all in-cloud conditions (i.e., with biases less than ±0.2), it
significantly underestimates glaciation ratios of coexisting ice and liquid by 0.2 – 0.6 below -20°C due to the overestimation of LWC.

Thermodynamic conditions, specifically RH, were previously found to be well correlated with model biases of cloud occurrences and cloud phases (e.g., C. Wu et al., 2017). In terms of PDFs of in-cloud RH\textsubscript{ice}, 1-s observations show larger variabilities of in-cloud RH\textsubscript{ice} ranging from 3% to 147%, while the simulations show narrower ranges, i.e., 20%–134% for CAM6, 52%–116% for CAM5, and 19%–144% for E3SM. When averaging the observations into every 580 s, the observed in-cloud RH\textsubscript{ice} is seen from 7% to 132%, indicating that the simulations lack of sub-saturation at in-cloud conditions. This may limit the ranges of cloud microphysical properties, such as underestimating IWC by limiting ice growth and riming in sub-saturated conditions. Q. Liu et al. (2018) previously showed that RH biases might be caused by local processes (i.e., boundary-layer turbulence, shallow and deep convection) or even by biases in remote deep convection via water vapor advection. Therefore, future model development is recommended to examine the representations of these processes in the GCMs.

Regarding aerosol indirect effects on cloud microphysical properties, positive correlations are found between cloud microphysical properties (IWC, LWC, N\textsubscript{ice}, and N\textsubscript{liq}) and the number concentration of larger aerosols (Na\textsubscript{500}) while only liquid properties (LWC and N\textsubscript{liq}) are found to be positively correlated with the number concentration of smaller aerosols (Na\textsubscript{100}) at various temperature ranges. This result suggests Twomey effects of larger aerosols on ice particles (i.e., possibly by acting as INPs) and supercooled liquid water (possibly by acting as CCN), while smaller aerosols only show Twomey effect on
supercooled liquid water (possibly by acting as CCN). The increase of LWC and $N_{\text{liq}}$ with increasing Na are stronger at warmer conditions (-20°C to 0°C) than colder conditions (-40°C to -20°C) in Figure 8, possibly due to less activation of ice nucleation at this temperature range and therefore less reduction of LWC and $N_{\text{liq}}$ due to the WBF process. Aerosol indirect effects also show distinct features in the lower and higher temperature ranges, i.e., -40°C to -20°C and -20°C to 0°C, respectively. At lower temperatures, $Na_{500}$ seem to play a dominant role in controlling glaciation ratio via the glaciation indirect effect, that is, higher $Na_{500}$ leads to higher INP concentrations, higher Nice and IWC, and consequently higher glaciation ratios and higher cloud fraction. At higher temperatures, the negative correlations between aerosol concentrations (both $Na_{500}$ and $Na_{100}$) and glaciation ratios indicate that the riming indirect effects (i.e., more CCN, smaller droplets, less riming and smaller IWC) and/or thermodynamic indirect effect (i.e., more CCN, more liquid droplets, less SIP) are possible pathways for aerosol indirect effects. Furthermore, since $N_{\text{ice}}$ does not show strong correlation with $Na_{100}$ (Figure 11m), the riming indirect effect is likely the dominant mechanism compared with the thermodynamic indirect effect at higher temperatures.

Small increases of LWC and $N_{\text{liq}}$ with increasing Na are seen in CAM6 between -15°C and 0°C. Small increases of $N_{\text{ice}}$ are seen in E3SM only at a narrow temperature range (-20°C to -10°C), and no increases of $N_{\text{ice}}$ are seen in CAM6. In contrast to the observations, both models show stronger temperature effects on glaciation ratios and cloud fraction than aerosol indirect effects. These results suggest that stronger aerosol indirect effects on both liquid droplets and ice particles should be considered for future development of cloud microphysics parameterizations, especially since model parameterizations still have limited aerosol types.
acting as INPs. In addition, the maximum Na_{500} and Na_{100} values are underestimated in CAM6 and E3SM by 0.5 – 1 orders of magnitude compared with 200-s observations, suggesting that higher concentrations of INPs and CCN need to be included in the model. In fact, higher CCN number concentration has also been recommended in another model evaluation study on CAM6 by Gettelman et al. (2020).

Several caveats need to be noted for this study. First, for in situ observations, higher in-cloud thresholds (e.g., 0.01 g m^{-3} in McFarquhar et al., 2007 and 0.001 g m^{-3} in D’Alessandro et al., 2021) have been used to define clouds in previous studies, consistent with the definition of clouds as a visible collection of particles (Sassen & Campbell, 2001). But, due to the significant reduction of model sample sizes when these higher in-cloud thresholds are applied to simulated data, a lower threshold of 10^{-7} g m^{-3} is chosen for this study. A sensitivity test to the observations using in-cloud thresholds at 0.01 g m^{-3} (Figure 4d) shows similar cloud phase frequency distributions as the analysis using the threshold of 10^{-7} g m^{-3}.

Second, the analysis of aerosol indirect effects separates the aerosols into larger and smaller sizes by using aerosols greater than 500 nm as a proxy of INPs based on the study of DeMott et al. (2010). However, that study did not include samples over the Southern Ocean, and therefore the validity of the 500 nm threshold over this region still needs further investigation. Third, the measurements of aerosol size can be complicated by the drier sampling condition compared with the ambient condition, which reduces the size of aerosols. Due to the lack of aerosol composition measurements at 1-Hz resolution, such potential biases cannot be accurately corrected. However, it is unlikely that this potential bias of
aerosol size measurements can explain all the relationships seen between Na₅₀₀, Na₁₀₀, and cloud microphysical properties (Figures 8–14).

Overall, this study provides a series of metrics for model evaluation of ice, liquid, and mixed phase clouds at -40°C to 0°C based on high resolution, in situ observations. Both thermodynamic conditions and aerosol number concentrations are found to be important factors in controlling cloud phases, the mass partition of ice and liquid, and cloud hydrometeor mass and number concentrations. Diagnosis of the parameterizations that drive the model biases on cloud phases and cloud microphysical properties shown in this work is still warranted. The model evaluation in this study is restricted to default configurations of three GCMs, while future work is recommended to investigate the impacts of individual parameters in cloud microphysics parameterizations that may lead to improved results compared with observations. The observation-based statistical distributions of cloud phase frequency, microphysical properties, and their correlations with temperature, RH, and aerosol concentrations can be used to guide future model development at various horizontal scales.
Chapter 2

Development of Aircraft Instrument Simulators for Model Evaluation

2.1. Introduction

One of the major challenges in the GCMs is the simulation of cloud properties on the microphysical scale. Biases were found in the simulated cloud cover and cloud properties, affecting the radiation and contributing to one of the largest uncertainties in predicting the future climate using the GCMs (Bodas-Salcedo et al., 2014; Cesana et al., 2012; Kay et al., 2016; Matus & L’Ecuyer, 2017; Stocker et al., 2013).

With its large spatial coverage and long-term data collection, satellite data have been commonly used in model evaluations (e.g., Cesana et al., 2015; W.-T. Chen et al., 2011; Doutriaux-Boucher & Quaas, 2004; Matus & L’Ecuyer, 2017; Y. Zhang et al., 2010). However, satellite data may face challenges in penetrating thick layer clouds, classifying cloud phases in the lower layer, and providing the finer-scale details for analysis of microphysical processes (Mace et al., 2021). Several agencies, such as the NSF, have recently conducted airborne campaigns targeting cloud microphysics or other cloud-related topics. These in-situ airborne observations provide valuable information for scientists to better understand microscale cloud properties and aerosol-cloud interactions. Some previous studies have used the airborne dataset to evaluate climate model performances by conducting a comparison analysis (e.g., Gettelman et al., 2020; X. Liu et al., 2011; Patnaude et al., 2021; Yang et al., 2021).

Unfortunately, relatively lesser model evaluations were conducted using airborne observations by the modeling community compared to satellite observations. Such underuse
is possibly due to the complexity of deployed instruments, interpretation and quality control of the observation dataset, and the spatial differences between the model simulation and the airborne observations. Therefore, an aircraft simulator package that targets the cloud properties evaluation and intercomparison of climate models is needed. A similar idea has been implemented to the satellite data, such as the International Satellite Cloud Climatology Project (Webb et al., 2001) and the CFMIP Observation Simulator Package that may produce model diagnostics compared to several satellite products (Bodas-Salcedo et al., 2011).

In this project, we aim to develop an aircraft simulator that would bridge the observational instrument and the modeling communities. This simulator will provide a quick and accessible tool for the modeling communities to evaluate their model and simulations on cloud properties and aerosol indirect effects using in-situ observations. This will also encourage the community to take advantage of the airborne observation data.

2.2. Approaches

The aircraft instrument simulator in this project is designed to be offline, which require the user to have a completed simulation output to execute the software. To create a user-friendly software that may provide the user with a quick and reliable diagnosis, the simulation output imported by the user must be in the NetCDF format with the cloud and aerosol attributes listed in the readme file. Building upon the work documented in Chapter 1, the simulator in this project is based on the CESM and the NSF airborne campaigns.

The airborne observation data are also needed for the user to conduct a statistical comparison between the model and the observations using the simulator. The user may choose the spatial scale of the observation data, ranging from 1-s, 10-s, 50-s, 100-s, 200-s, to
580-s, for a scale-aware comparison. The airborne data will be packaged together with the software. The current simulator package supports three NSF campaigns, including the SOCRATES, the O₂/N₂ Ratio and CO₂ Airborne Southern Ocean (ORCAS), and HIAPER Pole-to-Pole Observations (HIPPO) of Carbon Cycle and Greenhouse Gases Study.

Three software are included in the simulator package. The first and second software each process two levels of data, while the third software generates figures for analysis (Figure 15):

- **Level 1 (Software 1):** Unit conversions and calculations are applied to the attributes read from the simulation output. Hydrometeor variables are truncated to the same size range as the observational instruments.
- **Level 2 (Software 2):** Model data are synthesized with the aircraft data by selecting the closest location, time stamp, and vertical level. Quality control, thresholds, and
other definitions are applied to the product. Cloud phases are derived. Variables are merged from several simulation output files into one.

- Software 3: Figure generator

2.3. **Software 1 and Level 1 Simulation Output**

Software 1 prepares all the variables that will be used for the later process. Some variables can be directly read in and used from the NetCDF file (e.g., temperature and vertical velocity); some variables may need to be derived or calculated (e.g., pressure, relative humidity, and cloud properties); others may require unit conversion (e.g., ice and snow number concentrations). The application of size restriction to the simulated hydrometeors is also included in this software. This Level 1 data not only pre-process the attributes that are needed for Software 2, but also provide the user some samples for a quick overview of the simulated variables before proceeding with any further analysis. Moreover, Software 1 is able to accommodate both GCM free-run and nudged runs.

This software would require the user to select one or multiple completed simulation outputs. The simulation output must be in NetCDF format, ending in “.nc”. Table 2 shows the required attributes, units, and names used in the CESM. The software will handle any missing attributes that do not exist in the imported file by automatically creating an array filled with NaNs. While not all required attributes are included in the default CESM output attributes, the user may need to specify these attributes in the output list while running the model simulation.
Table 2. Attributes that Are Required by the Software

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<td>#m-3</td>
<td>grid-averaged ice number conc</td>
</tr>
<tr>
<td>AQRAIN</td>
<td>kgm-3</td>
<td>grid-averaged rain mixing ratio</td>
</tr>
<tr>
<td>ANRAIN</td>
<td>m-3</td>
<td>grid-averaged rain number conc</td>
</tr>
<tr>
<td>AQSNOw</td>
<td>kgm-3</td>
<td>grid-averaged snow mixing ratio</td>
</tr>
<tr>
<td>ANSNOW</td>
<td>m-3</td>
<td>grid-averaged snow number conc</td>
</tr>
</tbody>
</table>

Three instruments are commonly seen in most NSF airborne campaigns related to cloud particle sampling: CDP, Two-Dimensional Optical Array Cloud Probe, or 2DS. The CDP samples particles with sizes between 2 and 50 microns, while 2DC samples 62.5 – 3200 microns and 2DS samples 40 – 5000 microns. While the simulated cloud properties range from zero to infinity, the same size range of 2 – 50, 62.5 – 3200, and 40 – 5000 microns are applied to the simulated hydrometeor variables in the Level 1 data to collocate with CDP,
2DC, and 2DS, respectively. A similar concept is applied to the aerosol variables for aerosol sizes greater than 100 and 500 microns.

A NetCDF file and a mat file will be created for each imported simulation file. The exported output will include all the initial attributes from the imported files, derived variables, and the mass and number concentrations of liquid, ice, rain, and snow for CDP, 2DC, and 2DS sampling size ranges. This exported output file from Software 1 will be needed to execute Software 2.

2.4. Software 2 and Level 2 Simulation Output

After the particle size range restrictions are applied in Level 1 data, Software 2 derives cloud properties such as LWC and IWC and number concentrations ($N_{\text{liq}}$ and $N_{\text{ice}}$). The definition of simulated LWC and IWC in this simulator is similar to Chapter 1. An in-cloud condition threshold of $1 \times 10^{-7}$ g m$^{-3}$ is applied to the mass concentrations. The IWC is redefined as the simulated variables of ice + snow, which is different from the “ice” variables that are directly provided by the simulation output. Cloud phases are also derived and determined by the mass fraction, that is, the ratio of ice mass concentration to the total mass concentrations, or IWC divided by the sum of LWC and IWC (Korolev et al., 1998). If the ratio is greater than 0.9, ice is identified; if it is less than 0.1, liquid is identified; mixed phase is defined with everything in-between. For the cloud phase variable, clear sky is denoted as 0; liquid phase is denoted as 1; mixed phase as 2; and ice phase as 3. Note that if any cloud variables (e.g., LWC or IWC) is missing, the cloud phase would not be generated and would be shown as NaN.
Software 2 also synchronizes the simulation data with the observations. The software selects the data from the simulation output, where the date and time are correlated with the observations. Based on the closest temperature, a vertical grid box will also be selected from the entire simulated atmospheric column to represent the aircraft's location at the vertical level. This step will provide a control on the spatial and temporal scales between the simulation and observation data for intercomparison.

The NetCDF output from Software 1 is required to execute Software 2. One file will be generated for each RF. The user may use a sub-script to merge all the RFs. The user may also use this Level 2 data output for their own analysis, or may proceed to run Software 3, which generates some figures for a quick analysis.

2.5. **Software 3: Figure Generator**

The Level 2 data generated by Software 2 is required to execute Software 3. Software 3 generates several figures that may provide the user with a quick inspection of a comparison between the observations and the simulated data. The figures include basic scatter plots of the cloud variables, thermodynamic analysis of temperatures and pressures, and the analysis of the cloud properties as well as the cloud-aerosol interaction, as listed below:

1. Scatter plot of LWC and IWC vs. temperature for observation and simulation for CDP and 2DS/2DC size range (Figure 16c-f)
2. Scatter plot of total LWC and IWC (combining CDP and 2DS/2DC) vs. temperature (Figure 16a-b)
3. PDF of temperature
4. PDF of temperature differences between the simulation and observations
5. Temperature vs. Pressure for the simulation and observations
6. Cloud phase occurrence frequencies
7. Geometric means of LWCs and IWCs, and glaciation ratio
8. Aerosol number concentration vs. Temperature, colored by the cloud properties
9. Linear regression of aerosol number concentration and the cloud properties

2.6. Conclusion and Future Works

The aircraft simulator aims to provide the modeling community with a quick, easy, reliable tool to evaluate the models using airborne observations. The simulator approaches to
overcome some difficulties of using in-situ observations, including the complexity of the instrument measurements, quality control of the observational data, and the differences in spatial and temporal scales between the model and the observations.

Limitations and restrictions are applied to the simulator, including the required attribute fields that must be included to initial the run, the variable assumptions based solely on the CESM, and the number of available campaigns in the current version.

The idea of this offline aircraft simulator lays a foundation for the future development of an online simulator, which will be integrated as part of the model run. The simulator may also incorporate more research campaigns – including other NSF campaigns and flight campaigns funded by other agencies such as NASA – as well as other GCMs. This not only utilizes the observation data from million-funded campaigns but also help with the model development and parametrization.
3.1. Introduction

Mixed phase clouds, clouds consisting of liquid droplets and ice crystals, exist at temperatures between -40°C and -0°C, where water may remain in liquid phase and ice may also be formed. The radiative properties of mixed phase clouds are significantly influenced by the phase partition of liquid and ice (Z. Sun & Shine, 1994). At temperatures -40°C – 0°C, ice crystals are primarily formed through heterogeneous nucleation, a process of cloud particle formation that involves a foreign substance, such as aerosol, that can be activated as the INP for the water to grow on its surface. The heterogeneous nucleation, however, is not the only explanation for the growth of ice number crystals. It is known that the observed ice number concentrations ($N_{ice}$) are often higher than the INP concentration by several orders of magnitude (e.g., Hobbs & Rangno, 1985, 1998; Korolev et al., 2020; Mossop, 1985). Such a discrepancy is possibly due to the SIP, a formation of ice crystals that involves pre-existing ice crystals (Korolev & Leisner, 2020).

Around 1960s and 1970s, different SIP mechanisms have been proposed, including (i) fragmentation of freezing droplets, a possible crack in the outer ice shell to release the pressure building inside the freezing liquid droplet (e.g., Langham & Mason, 1958; Mason & Maybank, 1960); (ii) splintering during riming, or the Hallett-Mossop process, which refers to the splinters of small ice produced when liquid droplets collide with ice crystals (e.g., Bader et al., 1974; Hallet & Mossop, 1974); (iii) fragmentation an ice crystal collides with another (e.g., Hobbs & Farber, 1972; Vardiman, 1978); (iv) fragmentation due to the latent
heat released when the liquid droplet freezes as it rimes on an ice crystal (e.g., Koenig, 1963); (v) fragmentation when an ice particle sublimates under a subsaturated condition (e.g., Bacon et al., 1998; Oraltay & Hallett, 1989); and (vi) and the water vapor diffusion due to the temperature differences when INP is activated near freezing drops under a high transient supersaturation condition (e.g., Dye & Hobbs, 1968; Gagin, 1972).

While many early SIP studies are based on laboratory experiments (e.g., Bader et al., 1974; Dye & Hobbs, 1968; Hallet & Mossop, 1974; Oraltay & Hallett, 1989), airborne observations have also provided valuable information regarding the thermodynamic and dynamical conditions when SIP occurs in the real atmosphere (e.g., Hobbs & Farber, 1972; Korolev & Isaac, 2004; Ladino et al., 2017). Although some earlier studies and analyses of SIP using aircraft instruments have been challenged due to the possibility of ice crystals shattered into smaller pieces or bouncing off the instruments as the result of aircraft passage in a high speed (Korolev & Isaac, 2005; Schwarzenboeck et al., 2009; Woodley et al., 2003), the results of higher $N_{\text{ice}}$ compared to the INP concentrations remain consistent (Crawford et al., 2012; Ladino et al., 2017) with anti-shattering tips equipped and algorithm applied. Korolev and Isaac (2005) also suggested the exclusion of the first two to three bins from the measurements by OAP to eliminate the contamination of artifacts.

However, challenges and uncertainties remain in the current stages, including measurements of INPs covering a certain spatial range. The INPs are typically determined on a crystal-by-crystal basis (Cziczo & Froyd, 2014; Mertes et al., 2007; Mignani et al., 2019), sometimes with the use of cloud probe imagine (e.g., Stith et al., 2011). One approach is using aerosol number concentrations for aerosols with diameters greater than 500 nanometers
(Na$_{500}$), which can be an indicator of INPs for mixed phase clouds with temperatures warmer than -36°C (DeMott et al., 2010). Some studies have used aerosol number concentrations (Na) for aerosol indirect effects and aerosol-cloud interaction analysis (e.g., Patnaude et al., 2021; Yang et al., 2021).

Although the study focuses on cirrus clouds, Patnaude et al. (2021) used a comprehensive airborne in-situ dataset to conduct a comparative analysis between the observations in the tropical, mid-latitude, and polar regions. Another hemispheric comparison that focuses on the cirrus evolution was conducted by Diao et al. (2014). For mixed phase clouds, hemispheric analyses are mainly based on satellite observations (e.g., Villanueva et al., 2021) or ground-based observations (e.g., Radenz et al., 2021; D. Zhang et al., 2019). No hemispheric analysis has been done on mixed phase clouds using comprehensive airborne observations with a broad spatial coverage.

Previously, studies have shown the differences in cloud microphysical and aerosol properties between the Northern Hemisphere (NH) and Southern Hemisphere (SH). Influenced by anthropogenic aerosol emissions, the effective radius of droplets was found to be smaller in the NH (Feng & Ramanathan, 2017; Han et al., 1994). Aerosol number concentrations in the NH have also been found to be higher than the SH by 2 – 3 orders of magnitude (Minikin et al., 2003). On the other hand, the Southern Ocean is a pristine region with less land mass. Small amounts of dust that may act as ice nuclei for ice formation (Choi et al., 2010) may lead to the higher supercooled liquid observed in the SH (e.g., Bodas-Salcedo et al., 2016; Tan et al., 2014).
In this study, we aim to investigate aerosol and cloud microphysical characteristics and SIP distributions for mixed phase clouds in the NH and SH using airborne observation. This analysis will target the following questions: (1) How does global SIP distribution look like? (2) How do the characteristics of each hemisphere impact aerosol, cloud properties, and SIP? Moreover, (3) Is the difference in SIP caused by more SIP mechanisms in one hemisphere compared to another?

3.2. Data and Methodology

This study uses a comprehensive observation dataset from eleven NSF airborne campaigns, including START08 (L. L. Pan et al., 2010), HIPPO (Wofsy, 2011), PREDICT (Montgomery et al., 2012), TORERO (Volkamer et al., 2015), DC3 (Barth et al., 2015), CONTRAST (C. Pan et al., 2016), WINTER (Lee et al., 2018), CSET (Albrecht et al., 2019), ORCAS (Stephens et al., 2018), SOCRATES (McFarquhar et al., 2021), OTREC (Fuchs-Stone et al., 2020). Table 3 shows the detailed information of campaign names, the number of fights, time, coordinates, total flight hours at all temperatures, hours for in-cloud, clear sky, and all conditions at temperatures between -40°C and 0°C. A total of 504 hours were flown at -40°C – 0°C, with 440 and 64 hours at clear-sky and in-cloud conditions, respectively. Figure 17 shows the number of samples for each campaign, the NH and SH, and the entire dataset. The number of in-cloud samples in the HIPPO campaign contributes almost 50% (68216 samples) of the NH data (140077 samples), with 23 in-cloud hours, as shown in Table 3. Due to the lack of cloud measurements, deployment #1 from HIPPO and ORCAS RF 12 are not included in this study. Collectively, the eleven airborne campaigns


Table 3. Summary of the Eleven NSF Campaigns

<table>
<thead>
<tr>
<th>Acronym</th>
<th>START08</th>
<th>HIPPO</th>
<th>PREDICT</th>
<th>DC3</th>
<th>TORERO</th>
<th>CONTRAST</th>
</tr>
</thead>
<tbody>
<tr>
<td>Field Campaign</td>
<td>Stratosphere-Troposphere Analyses of Regional Transport</td>
<td>HIAPER Pole-to-pole Observation - deployments 2 – 5</td>
<td>PRE-Depression Investigation of Cloud Systems in the Tropics</td>
<td>Deep Convective Clouds and Chemistry Project</td>
<td>Tropical Ocean Roposphere Exchange of Reactive halogen species and Oxygenated voc</td>
<td>CONvective TRansport of Active Species in the Tropics</td>
</tr>
<tr>
<td>Number of Flights</td>
<td>18</td>
<td>46</td>
<td>26</td>
<td>22</td>
<td>17</td>
<td></td>
</tr>
<tr>
<td>Flight Hours</td>
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<td>333</td>
<td>175</td>
<td>136</td>
<td>134</td>
<td></td>
</tr>
<tr>
<td>Clear Sky Hours</td>
<td>28</td>
<td>140</td>
<td>11</td>
<td>37</td>
<td></td>
<td></td>
</tr>
<tr>
<td>In-cloud Hours</td>
<td>23</td>
<td>117</td>
<td>10</td>
<td>31</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Acronym</td>
<td>WINTER</td>
<td>CSET</td>
<td>ORCAS</td>
<td>SOCRATES</td>
<td>OTREC</td>
<td></td>
</tr>
<tr>
<td>Field Campaign</td>
<td>Wintertime Investigation of Transport, Emissions, and Reactivity</td>
<td>Cloud Systems Evolution in the Trades</td>
<td>The O2/N2 Ratio and CO2 Airborne Southern Ocean Study</td>
<td>Southern Ocean Clouds, Radiation, Aerosol Transport Experimental Study</td>
<td>Organization of Tropical East Pacific Convection</td>
<td></td>
</tr>
<tr>
<td>Number of Flights</td>
<td>13</td>
<td>16</td>
<td>18</td>
<td>15</td>
<td>22</td>
<td></td>
</tr>
<tr>
<td>Flight Hours</td>
<td>95</td>
<td>144</td>
<td>95</td>
<td>112</td>
<td>123</td>
<td></td>
</tr>
<tr>
<td>Clear Sky Hours</td>
<td>57</td>
<td>43</td>
<td>40</td>
<td>87</td>
<td></td>
<td></td>
</tr>
<tr>
<td>In-cloud Hours</td>
<td>54</td>
<td>42</td>
<td>33</td>
<td>74</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total Hours</td>
<td>1566</td>
<td>504</td>
<td>440</td>
<td>2</td>
<td>64</td>
<td></td>
</tr>
</tbody>
</table>
Figure 17. Number of samples for all sky (solid), clear sky (dashed), and in-cloud (dotted) conditions for (a–k) each campaign and for (l–m) the merged dataset of all eleven campaigns, separating to Northern and Southern Hemispheres.

conducted between April 2008 and September 2019 cover a spatial range from 75°S to 87°N and from 38°W to 128°E (Figure 18). For this analysis, the observations are further separated into NH and SH, defined to be North or South to the equator at 0° latitude, respectively.

The 1-Hz observations from all eleven campaigns are collected by the NSF GV research aircraft. The temperatures were measured by a Rosemount temperature probe that has an accuracy of \( \pm 0.3 \) K and a precision of 0.01 K. All analyses in this study are restricted to temperatures between -40°C and 0°C, which is the mixed phase cloud regime that both liquid and ice may co-exist. The VCSEL hygrometer provides water vapor readings at 25 Hz resolution with an accuracy of \( \sim 6\% \) and a precision of \( \leq 1\% \) (Zondlo et al., 2010). A calibrated and quality-controlled 1-Hz water vapor dataset is used to derive the relative
humidity with respect to liquid and ice (RH\textsubscript{liq} and RH\textsubscript{ice}) (Murphy & Koop, 2005). The CDP was deployed during every campaign, measuring particles ranging from 2 to 50 μm. The Fast Two-Dimensional Optical Array Cloud probe (Fast-2DC) measures particle sizes from 62.5 to 1600 μm and can be reconstructed up to 3200 μm. The Fast-2DC mass concentrations are derived based on Brown and Francis's (1995) mass-dimension (m-D) relationships, with the small-d equation applied to particles sizes between 62.5 μm and 100 μm and the large-D equation applied to particle sizes greater than 100 μm. The Fast-2DC data are used across all

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{flight_tracks.png}
\caption{Flight tracks of the eleven NSF flight campaigns.}
\end{figure}
the campaigns except for SOCRATES due to the concern of data quality. Instead, the 2DS is used for SOCRATES, providing information for particle sizes from 40 to 5000 μm. The 2DS mass concentrations are calculated and quality-controlled by W. Wu and McFarquhar (2016).

Combining CDP measurements with 2DC or 2DS, the cloud phases are identified using a similar method documented by D’Alessandro et al. (2019) and Yang et al. (2021).

3.3. Findings

3.3.1. Cloud Phase Occurrence Frequencies

Figure 19 shows the cloud phase occurrence frequencies (top) and the number of samples (bottom) for liquid, mixed, and ice phase clouds in NH and SH. The occurrence frequencies are calculated by the number of a cloud phase divided by the number of samples in-cloud for every 5° temperature bin. Liquid phase clouds frequently occur in the SH at temperatures -20°C – 0°C. The frequencies of liquid phase are 0.4 – 0.7 in the SH compared from temperatures -20°C to 0°C, doubling the frequencies observed in the NH (0.1 – 0.35). Such a critical increase (decrease) in liquid (ice) phase in the SH appears at a temperature around -20°C, where the liquid (ice) frequency increases (decreases) from 0.1 to 0.4 (0.85 to 0.55). The mixed phase frequencies are similar between NH and SH, except for the warmest temperature bin -5°C – 0°C, where the mixed phase frequency is 0.2 in the NH and 0.1 in the SH. The number of samples shown in Figure 19 d has confirmed a similar result for liquid and ice phases. However, the number of mixed phase clouds in the SH appears to be decreased as the temperature decreases while it remains more consistent for the NH between -40°C and -20°C.
Figure 19. (a – b) Cloud phase occurrence frequencies and (c – d) number of samples for 1-Hz airborne observations in NH and SH with temperature ranged from -40°C to 0°C, binned by 5°C. Liquid, mixed, and ice phase are denoted by red, green, and blue colors, respectively.

3.3.2. Secondary Ice Production

The discrepancies between the number concentration of ice and INPs would be an indication of the SIP. Aerosol number concentrations are used without a direct measurement of INP concentration. For every 5-degree bin between temperatures -40°C and 0°C, ice with a number concentration exceeding the 75th percentile is used to represent SIP.
In Figure 20, $N_{\text{ice}}$ is sub-sampled into four groups based on the $N_{\text{ice}}$ distribution for its corresponding temperature in every 5-degree: $N_{\text{ice}}$ between 0 and 25\textsuperscript{th} percentile, 25\textsuperscript{th} and 50\textsuperscript{th} percentile, 50\textsuperscript{th} and 75\textsuperscript{th} percentile, and 75\textsuperscript{th} – 100\textsuperscript{th} percentile. A linear fit is applied to each group. We hypothesized that there would be no correlation or weak correlation between the $N_{\text{ice}}$ and Na since the ice does not form through ice nuclei in the SIP. For the NH, weak positive correlations are seen between Na\textsubscript{500} and the three $N_{\text{ice}}$ groups lower than the 75\textsuperscript{th} percentile with slopes around 0.1, but no obvious correlation is seen with Na\textsubscript{100}. Negative correlations are shown between the $N_{\text{ice}}$ exceeding the third quartile and Na\textsubscript{500} and Na\textsubscript{100} with slopes -0.156 and -0.066, respectively, which might indicate the SIP. On the other hand, the maximum of observed $N_{\text{ice}}$ in the SH reaches about 30 cm\textsuperscript{3}, two orders of magnitude higher than the NH. A similar result is seen in the SH between Na\textsubscript{500} and $N_{\text{ice}}$ as for NH, with a weaker negative correlation (b = -0.047) for $N_{\text{ice}}$ exceeding the 75\textsuperscript{th} percentile. Positive correlations are seen in all four $N_{\text{ice}}$ groups and Na\textsubscript{100}.

3.4. Conclusion

This study conducts a hemispheric comparison of clouds, aerosol properties, aerosol-cloud interactions, and SIP in mixed phase clouds using airborne observations from eleven NSF flight campaigns. A comprehensive dataset provides extensive spatial coverage and samples for hemispheric analysis.

Differences between the NH and SH are observed in cloud phases, $N_{\text{ice}}$, and the relationship between $N_{\text{ice}}$ exceeding the 75\textsuperscript{th} percentile and Na. The SH is found to have a higher liquid cloud fraction, as shown previously in other studies (e.g., Badas-Salcedo et al., 2016; Tan et al., 2014), especially for temperatures warmer than -20°C. $N_{\text{ice}}$ that exceeds the
Figure 20. Relationship of Nice with respect to Na500 and Na100 for NH (a – b) and SH (c – d). Linear regression applied for the Nice between 0 and 25\textsuperscript{th} percentile (red), 25\textsuperscript{th} and 50\textsuperscript{th} percentile (blue), 50\textsuperscript{th} and 75\textsuperscript{th} percentile (green), 75\textsuperscript{th} and 100\textsuperscript{th} percentile (magenta) in each in every 5\degree temperature bin.
third quartile is used to represent the SIP, and the aerosol number concentration is used to represent the INPs due to the lack of measurement. No correlation or slightly negative correlations are seen between the $N_{\text{ice}}$ that exceeds the third quartile and the Na, while only $N_{\text{ice}}$ and Na$_{100}$ in SH show a slightly positive correlation.

While justification needs to be made for the representation of INP using aerosol concentrations, a further investigation would need to be done on the characteristics of the SIP mechanism in each hemisphere. The differences in SIP between NH and SH are still unclear, and the leading cause of such differences, if any, would be the next step in our analysis.
References


(CAM5) with in-situ observations. *Atmospheric Chemistry and Physics*, **14**, 10103–10118. https://doi.org/10.5194/acp-14-10103-2014


Appendix

Supplementary Figures

Figure S1. Comparisons among direct model output named “RELHUM” (relative humidity, RH) and the calculated RH\textsubscript{liq} and RH\textsubscript{ice} for simulations based on the saturation vapor pressure equations from Murphy and Koop (2005).
Figure S2. Logarithmic scale of number of samples for Figure 3.

Figure S3. (a) The distributions temperature differences between the models and observations and (b) the averaged pressure values for each 2-degree bin.
Figure S4. Logarithmic scale of number of samples for Figure 4.

Figure S5. Logarithmic scale of number of samples for Figure 5.
Figure S6. Logarithmic scale of number of samples for Figure 6.
Figure S7. Similar to Figure 6, but without adding snow to IWC for the model.
Figure S8. Sensitivity test for E3SM 1-s, 1-min, and 10-min simulations shown with (a – c) cloud phase occurrence frequencies (d – f) log-scale LWC, (g – i) log-scale IWC, (j – l) glaciation ratio (i.e., linear averages of IWC/TWC), and (m – o) glaciation ratio only when ice particles and supercooled liquid water coexist.
Figure S9. Temperature distribution with respect to $\log_{10}(Na_{500})$ for (a – d) liquid containing clouds, (e – h) ice containing clouds, and (i – l) all clouds, colored by $\log_{10}$ (number of samples).

Figure S10. Similar to Figure S9 but for $\log_{10}(Na_{100})$.  

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Figure S11. Logarithmic scale of number of samples for Figure 12.
Table S1. The Contributions of Ice and Snow Mass Concentrations between Size Ranges 5000 – inf, 3200 – inf, 62.5 – inf, 50 – inf, 40 – inf, 2 – inf, and 0 – inf for CAM6, CAM5, and E3SM. Each Percentage is Calculated by the Mass Concentration in Each Size Range Divided by the Total Size Range from 0 to Infinity

<table>
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<tr>
<th></th>
<th>Contribution of Partial Mass Concentration (%)</th>
<th>5000-Inf</th>
<th>3200-Inf</th>
<th>62.5-Inf</th>
<th>50-Inf</th>
<th>40-Inf</th>
<th>2-Inf</th>
<th>0-Inf</th>
</tr>
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<td><strong>CAM6</strong></td>
<td>Ice</td>
<td>2.19</td>
<td>2.21</td>
<td>99.3</td>
<td>99.7</td>
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<td>Snow</td>
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<td>100</td>
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