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Cirrus Cloud Microphysical Properties and Aerosol Indirect Effects using Airborne Observations and a Global Climate Model

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CIRRUS CLOUD MICROPHYSICAL PROPERTIES AND AEROSOL INDIRECT
EFFECTS USING AIRBORNE OBSERVATIONS AND A GLOBAL CLIMATE
MODEL

A Thesis

Presented to

The Faculty of the Department of Meteorology and Climate Science

San José State University

In Partial Fulfillment

of the Requirements for the Degree

Master of Science

by

Ryan Patnaude

May 2020

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The Designated Thesis Committee Approves the Thesis Titled

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MODEL

by

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SAN JOSÉ STATE UNIVERSITY

MAY 2020

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ABSTRACT

CIRRUS CLOUD MICROPHYSICAL PROPERTIES AND AEROSOL INDIRECT EFFECTS USING AIRBORNE OBSERVATIONS AND A GLOBAL CLIMATE MODEL

by Ryan Patnaude

Cirrus clouds are strong controllers of the global radiation budget, but significant uncertainties remain regarding the thermodynamic, dynamic, and aerosol indirect effects on their microphysical properties. The lack of observational analysis on cirrus cloud controlling factors, i.e., temperature, relative humidity, vertical velocity, and aerosol number concentrations (N_a), limits our understanding of their impact on cirrus microphysical properties, making quantification and parameterization of cirrus clouds in climate models difficult. Using seven flight campaigns funded by the National Science Foundation (NSF), impacts of cirrus controlling factors and their regional distributions are individually quantified, and used to evaluate the National Center for Atmospheric Research (NCAR) Community Atmosphere Model 6 (CAM6). N_a is found to produce positive correlations with respect to cirrus microphysical properties (i.e., ice water content, ice crystal number concentrations, and mean diameter) when N_a is 3 – 10 times higher than average values. The magnitudes of the correlations depend on the size of the aerosols (i.e., > 500 nm or > 100 nm) and environmental conditions, indicating different roles of heterogeneous and homogeneous nucleation, respectively. Compared with observations, CAM6 simulations show large biases in terms of aerosol indirect effects and regional variations of cirrus microphysical properties, underscoring the importance of evaluating and improving cirrus cloud parameterizations in climate models.

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LIST OF ABBREVIATIONS

2DS – 2D-Stereo
CAM6 – Community Atmosphere Model version 6
CLUBB – Clouds Layers Unified by Binormals
 cm^{-3} – per cubic centimeter
CONTRAST – Convective Transport of Active Species in the Tropics
DC3 – Deep Convective Clouds and Chemistry
 D_i – number-weighted mean diameter of ice crystals
Fast-2DC – Fast Two-Dimensional Cloud Probe
GCM – global climate model
 gm^{-3} – grams per cubic meter
G-V – Gulfstream V
HIAPER – High-Performance Instrumented Airborne Platform for Environmental Research
HIPPO – High-Performance Instrumented Airborne Platform for Environmental Research (HIAPER) Pole-to-Pole Observations
Hz – hertz
IN – ice nuclei
INP – ice nucleating particle
IWC – ice water content
ISS – ice supersaturation
K – kelvin
 L^{-1} – per liter
MERRA2 – Modern-Era Retrospective Analysis for Research and Applications version 2
Na – aerosol number concentration
 Na_{100} – aerosol number concentration > 100 nanometers
 Na_{500} – aerosol number concentration > 500 nanometers
NCAR – National Center for Atmospheric Research
 N_i – ice crystal number concentration
NH – northern hemisphere
nm – nanometer
NM – northern hemisphere midlatitude
NP – northern hemisphere polar
NT – northern hemisphere tropical
NSF – National Science Foundation
ORCAS – O_2/N_2 Ratio and CO_2 Airborne Southern Ocean Study
PDF – probability density function
PREDICT – PRE-Depression Investigation of Cloud-systems in the Tropics
PSD – particle size distribution
 r_e – ice crystal effective radius
RH_i – relative humidity with respect to ice
SH – southern hemisphere

SID-2H – Small Ice Detector
SM – southern hemisphere midlatitude
SP – southern hemisphere polar
ST – southern hemisphere tropical
START08 – Stratosphere-Troposphere Analyses of Regional Transport 2008
TKE – turbulent kinetic energy
TOA – top of atmosphere
TORERO – Tropical Ocean Troposphere Exchange of Reactive Halogen Species
and Oxygenated VOC
UHSAS – Ultra-High Sensitivity Aerosol Spectrometer
 μm – micrometer
UT – upper troposphere
VCSEL – Vertical Cavity Surface Emitting Laser
w – vertical velocity

Chapter 1. Aerosol Indirect Effects on Cirrus Clouds based on Global Aircraft Observations

Introduction

Cirrus clouds play an important role in balancing the global radiation budget (Chen, Rossow, & Zhang, 2000). When quantifying the magnitudes of shortwave and longwave radiation, the microphysical properties of ice crystals are important for determining whether the net radiative forcing produces a warming or cooling effect on Earth's surface (Liou, 1986). Ice water content (IWC), number concentration (N_i) and mean diameter (D_i) of ice crystals significantly affect cloud albedo and optical depth. Therefore, quantifying these microphysical properties at various temperatures is crucial for evaluating and improving ice microphysics parameterizations in global climate models (GCMs).

Ice nucleation and ice microphysical properties are affected by various factors, including thermodynamic conditions (i.e., relative humidity and temperature), dynamical conditions (e.g., vertical velocity), and aerosol number concentrations and compositions. Two formation mechanisms contribute to ice nucleation, homogeneous and heterogeneous nucleation (Pruppacher and Klett, 1997). Heterogeneous nucleation requires the presence of ice nucleating particles (INPs) but can occur at relatively lower ice supersaturations (ISS, where relative humidity with respect to ice (RH_i) $> 100\%$). In addition, heterogeneous nucleation generally produces a smaller ice crystal number concentration compared with homogeneous nucleation (Jensen et al., 2013), which subsequently affects the net radiative forcing of cirrus clouds (Lohmann and Gasparini, 2017). Previously, a series of studies based on in situ observations showed that

thermodynamic and dynamical conditions have major impacts on ice microphysical properties (D'Alessandro et al., 2017; Diao, Zondlo, Heymsfield, Beaton, & Rogers, 2013b; Diao et al., 2014a, Diao, Zondlo, Heymsfield, & Beaton, 2014b; Diao et al., 2015; Diao, Bryan, Morrison, & Jensen, 2017; Heymsfield, 1975; Heymsfield, Schmitt, & Bansemer, 2013; Heymsfield et al., 2017; Krämer et al., 2009, 2016; Luebke et al., 2013, 2016; Mcfarquhar and Heymsfield, 1996; Schiller, Krämer, Afchine, Spelten, & Sitnikov, 2008). Other laboratory experiments (Hiranuma et al., 2014) and field observations (DeMott et al., 2015) have showed that thermodynamic conditions can also affect the types and concentrations of aerosols that can be activated as INPs.

Due to the complexity of different ice nucleation mechanisms, the varying effects of ambient conditions and availability of INPs, there are large uncertainties associated with the understanding of aerosol indirect effects on ice clouds as well as the estimates of their climate impacts (Fan, Wang, Rosenfeld, & Liu, 2016; Ghan et al., 2016; Lohmann and Feichter, 2005). Several previous GCM modeling studies evaluated aerosol indirect effects on ice crystal number concentrations via homogeneous nucleation and found a relatively weak relationship between aerosol number concentration and N_i , with an increase of N_i by a factor of 1.3 – 3 when N_a has a tenfold increase (Kärcher and Lohmann, 2002; Kay and Wood, 2008; Liu and Shi, 2018).

Aerosol indirect effects are also found to be affected by other conditions in GCM simulations. For example, the assumption of whether INPs activate under a constant or variable RH_i threshold can modify ice microphysical properties (Kärcher and Lohmann, 2003). The inclusion of sub-grid scale vertical velocity (Penner, Zhou, Garnier, &

Mitchell, 2018; Shi and Liu, 2016; Wang, Liu, Zhang, & Comstock, 2014), aerosol size distribution (Liu and Shi, 2018), and pre-existing ice (Kuebbeler, Lohmann, Hendricks, & Kärcher, 2014; Shi, Liu, & Zhang, 2015) in model parameterizations affects the magnitude of aerosol indirect effects.

To improve the understanding of aerosol indirect effects, several observation-based analyses have been conducted. Using aircraft observations, Cziczo et al. (2013) showed that mineral dust and metallic particles frequently reside in ice residuals, which are the nuclei particles found after removing water from ice particles. Field measurements of INPs show that total aerosol concentrations with diameters greater than 500 nanometers (nm) can be used as a good indicator of the INP number concentrations based on observations of clouds warmer than -36°C (DeMott et al., 2010). A satellite-based analysis demonstrated the importance of separating stronger and weaker convection when examining aerosol indirect effects on convective and anvil clouds (Zhao et al., 2019). The authors showed that, while using aerosol optical depth as a proxy for aerosol loading, ice cloud effective radius (r_e) decreases with polluted continental aerosols near cloud top in strong convection but increases with aerosols in moderate convection. Up to today, there is a lack of in situ observations that studies aerosol indirect effects by simultaneously quantifying the impacts of thermodynamic and dynamical conditions. Ideally, such analysis would not only require high-resolution, accurate measurements of a series of variables such as water vapor, temperature, vertical velocity, aerosol number concentrations, and cloud hydrometeors, but it also relies on a significant number of samples to provide statistically robust results.

This study aims to address this knowledge gap by examining aerosol indirect effects under controlled thermodynamic and dynamical conditions, specifically temperature, RH_i, and vertical velocity. A major advancement of this work is to develop a comprehensive in situ dataset from several flight campaigns, covering from 87°N to 75°S, providing ample number of samples to study various ambient conditions and geographical locations. Seven flight campaigns funded by National Science Foundation (NSF) onboard the NSF/National Center for Atmospheric Research (NCAR) Gulfstream-V (G-V) research aircraft are used in this study. The analyses provide an unprecedented geographical coverage based on research aircraft observations, which can serve as a valuable resource for evaluating model simulations, as well as complementing other measurement techniques.

In Situ Datasets and Instrumentation

For this study, 1-Hz aircraft-based observations are obtained from the NSF G-V research aircraft. A comprehensive dataset is developed based on quality-controlled measurements from seven NSF flight campaigns listed in chronological order: (1) the Stratosphere-Troposphere Analyses of Regional Transport 2008 (START08) campaign (Pan et al., 2010), (2) the High-Performance Instrumented Airborne Platform for Environmental Research (HIAPER) Pole-to-Pole Observations (HIPPO) global campaign (Wofsy, 2011), (3) PRE-Depression Investigation of Cloud-systems in the Tropics (PREDICT) (Montgomery et al., 2012), (4) Tropical Ocean Troposphere Exchange of Reactive Halogen Species and Oxygenated VOC (TORERO) (Volkamer et al., 2015), (5) Deep Convective Clouds and Chemistry (DC3) project (Barth et al., 2015), (6)

CONvective TRansport of Active Species in the Tropics (CONTRAST) (Pan et al., 2016), and (7) the O₂/N₂ Ratio and CO₂ Airborne Southern Ocean Study (ORCAS) (Stephens et al., 2018). Due to the lack of ice particle measurements in HIPPO deployment #1, only HIPPO deployments #2 – 5 are included. Our dataset provides a total of ~1000 flight hours from April 2008 (START08) to February 2016 (ORCAS), including 84 hours and 423 hours of in-cloud and clear-sky conditions at temperatures $\leq -40^{\circ}\text{C}$, respectively. These campaigns provide an extensive spatial coverage from 87°N to 75°S and 128°E to 38°W, from the surface to the upper troposphere and lower stratosphere. The flight tracks for all seven flight campaigns used in this study are shown in Figure 1 a. In-cloud frequency distributions of temperature and vertical velocity for each campaign are shown in Figure 1 b and c, respectively.

Temperature measurements were collected at 1 Hz by the Rosemount temperature probe with an accuracy and precision of $\sim \pm 0.3$ K and 0.01 K, respectively. For this study, observations are restricted to temperatures $\leq -40^{\circ}\text{C}$ to eliminate the presence of supercooled liquid water droplets. Water vapor measurements are from the Vertical Cavity Surface Emitting Laser (VCSEL) hygrometer (Zondlo, Paige, Massick, & Silver, 2010), measuring at 25 Hz with an accuracy and precision of $\sim 6\%$ and $\leq 1\%$, respectively. The data are downsampled to 1 Hz to match temperatures. Relative humidity is derived from temperature and water vapor measurements with a combined uncertainty of 6.9% to 7.8% from -40°C to -78°C , respectively. RH_i is calculated from water vapor mixing ratio, pressure and temperature, following the equation of saturation vapor pressure with respect to ice in Murphy and Koop (2005). The Ultra-High Sensitivity

Aerosol Spectrometer (UHSAS) uses 100 logarithmically spaced bins to measure aerosols from $0.06 - 1 \mu\text{m}$. Ice particle measurements for all campaigns were collected using the NCAR Fast Two-Dimensional Cloud Probe (Fast-2DC), and it is the only cloud probe consistently onboard the G-V aircraft for all seven flight campaigns.

Figure 1. (a) Aircraft flight tracks of seven flight campaigns. Number of samples for (b) temperature and (c) vertical velocity at in-cloud conditions, with colors corresponding to various flight campaigns.

The Fast-2DC has a measuring range of 25 – 1600 μm , and larger particles can be mathematically reconstructed up to 3200 μm . To alleviate the shattering effect and

optical uncertainties, we exclude ice particles $< 62.5 \mu\text{m}$ (omitting the first two bins of $12.5\text{--}37.5 \mu\text{m}$ and $37.5\text{--}62.5 \mu\text{m}$ in Fast-2DC measurements). A previous study showed that various cirrus cloud evolution phases with evolving Ni and Di can still be observed when using a size cutoff of $100 \mu\text{m}$ (Diao et al., 2014b). For definition of in-cloud conditions, we follow several previous studies that analyzed the G-V aircraft observations for clear-sky and in-cloud conditions (D'Alessandro et al., 2017; Diao et al., 2013b, 2014a, 2015, 2017) whereas in-cloud is considered when the 1-second sample of the Fast-2DC probe detects at least one ice particle (i.e., $> 0 \text{ L}^{-1}$), while all the remaining data are considered clear-sky conditions.

Individual Impacts of Thermodynamics, Dynamics and Aerosol Number Concentrations

Cirrus microphysical properties are examined here in relation to four fundamental factors: temperature, RH_i, vertical velocity (w), and Na for in-cloud conditions (Figure 2). Previously, using the IWC–T space, Krämer et al. (2016) classified two general types of cirrus – in situ origin and liquid origin cirrus, and showed their correlations with vertical velocity during evolution. The average IWC increases with increasing temperature, consistent with previous observational studies (Heymsfield et al., 2013; Schiller et al., 2008). The average Ni does not show a clear trend with temperature. At -70°C to -60°C , the average and standard deviation of Di are $100 \mu\text{m}$ and $\sim 100 \mu\text{m}$, respectively, which are smaller than those at -50°C to -40°C (i.e., $200 \mu\text{m}$ and $\sim 200 \mu\text{m}$, respectively). The larger particles at warmer temperature are likely affected by several factors, such as heterogeneous nucleation that produces fewer yet larger ice particles,

higher ice particle growth rate at higher water vapor partial pressure, and sedimentation of ice particles from above the sampling altitude of the G-V.

The relationships between ice microphysical properties and RHi are further examined (Figure 2 d–f). A key step to isolating temperature effect is to define a delta value, such as $dRHi$, which is the deviation of each 1-second RHi datum from the average RHi within each 1°C bin from -40°C to -70°C (equation 1a). Similarly, $d\log_{10}(IWC)$, $d\log_{10}(Ni)$, $d\log_{10}(Di)$, $d\log_{10}(Na_{500})$ and $d\log_{10}(Na_{100})$ are defined as the differences between each 1-second datum and their respective 1°C bin average (1c – 1g), written as:

$$dRHi = RHi_T - \overline{RHi_T}, (1a)$$

$$dw = w_T - \overline{w_T}, (1b)$$

$$d\log_{10}(IWC) = \log_{10}(IWC)_T - \overline{\log_{10}(IWC)_T}, (1c)$$

$$d\log_{10}(Ni) = \log_{10}(Ni)_T - \overline{\log_{10}(Ni)_T}, (1d)$$

$$d\log_{10}(Di) = \log_{10}(Di)_T - \overline{\log_{10}(Di)_T}, (1e)$$

$$d\log_{10}(Na_{500}) = \log_{10}(Na_{500})_T - \overline{\log_{10}(Na_{500})_T}, (1f)$$

$$d\log_{10}(Na_{100}) = \log_{10}(Na_{100})_T - \overline{\log_{10}(Na_{100})_T}, (1g)$$

Here, the temperature effect is mitigated by subtracting the temperature-based averages.

All delta values and correlation analyses are restricted to in-cloud conditions.

Figure 2. Distributions of IWC, Ni and Di in logarithmic scale with respect to (a–c) temperature, (d–f) dRH i, (g–i) dw , (j–l) $d\log_{10}(Na_{500})$ and (m–o) $d\log_{10}(Na_{100})$. Black lines and whiskers denote averages and standard deviations, respectively.

The majority of the higher IWC and Ni values (i.e., $d\log_{10}(\text{IWC}) > 0$ and $d\log_{10}(\text{Ni}) > 0$, respectively) are associated with higher RHi values ($d\text{RHi} > 0$). When average IWC, Ni and Di reach maximum, $d\text{RHi}$ is at 10%, 10%, and 30%–40%, respectively. Here $d\text{RHi}$ has a similar magnitude to that of ISS because the average in-cloud condition is around ice saturation. The fact that the maximum IWC, Ni, and Di do not occur at the highest RHi is likely due to formation and growth of ice particles at the expense of excess water vapor over ice saturation. From ice supersaturated to sub-saturated conditions, the average IWC and Ni decrease 10 – 30 times. The increase of Di at dryer conditions (i.e., from -20% to -70% $d\text{RHi}$) is most likely contributed to by large ice particles sedimenting into sub-saturated layers, which was also observed in a previous analysis of cirrus cloud evolution phases (Diao et al., 2013b). The large variabilities of IWC, Ni, and Di at various RHi have an important implication. That is, if aerosol indirect effects were examined without accounting for the RHi effect, the results would vary significantly depending only on the RHi conditions being sampled.

Similarly, the effect of vertical velocity is examined (Figure 2 g – i). Maximum IWC, Ni, and Di occur at higher updrafts ($dw > 3$ m/s). The minimum values occur at weak downdrafts ($dw = -0.5$ m/s). According to D'Alessandro et al. (2017), higher updrafts increase the occurrence frequency of ISS, particularly for larger ISS (>20%), which likely explains the higher Ni and Di due to higher probability of ice nucleation and faster particle growth. Higher IWC, Ni, and Di are also seen in larger downdrafts with a local maximum at -2.5 m/s. This is most likely due to the proximity between updrafts and downdrafts, consistent with model simulations in D'Alessandro et al. (2017). Comparing

the thermodynamic effect (RHi) and dynamic effect (w), the average IWC and Ni increase one order of magnitude when $d\text{RHi}$ increases from -15% to 10%, comparable to the effect of dw increasing from 0 to 1 m/s.

Lastly, the correlations between cirrus microphysical properties and aerosol number concentrations are examined, specifically for aerosols > 500 nm (Na_{500}) and > 100 nm (Na_{100}) (Figure 2 last two rows). IWC, Ni, and Di are positively correlated with Na_{500} , particularly when Na_{500} is relatively higher than the average condition (i.e., $d\log_{10}(\text{Na}_{500}) > 0.5$). Similar positive correlations are also seen with respect to Na_{100} , but only when Na_{100} is 10 times higher than its average value (i.e., $d\log_{10}(\text{Na}_{100}) > 1$). Na_{500} values are generally considered a good indicator of number concentrations for ice nuclei (IN) facilitating heterogeneous nucleation, although those observations were limited to temperatures warmer than -36°C (DeMott et al., 2010). The Na_{100} values are dominated by smaller aerosols, which more likely contribute to homogeneous nucleation. The fact that both Na_{500} and Na_{100} affect IWC, Ni, and Di suggest that the aerosol indirect effects in the atmosphere are effective in both pathways – heterogeneous and homogeneous nucleation. Minimizing the impacts of temperature by using the delta values is found to be an important step for analyzing aerosol indirect effects. When directly analyzing IWC, Ni, and Di (instead of their delta values) in relation to Na_{500} and Na_{100} , the positive correlations still exist but show more variabilities (Figure 3)

Figure 3. Similar to Figure 2, but without using the delta values. Distributions of IWC, Ni and Di in logarithmic scale with respect to (a–c) RHi, (d–f) w , (g–i) $\log_{10}(\text{Na}_{500})$ and (j–l) $\log_{10}(\text{Na}_{100})$. Black lines and whiskers denote averages and standard deviations, respectively.

Aerosol Indirect Effects on Cirrus Clouds under Controlled Thermodynamic and Dynamical Conditions

Aerosol indirect effects are further quantified for various ranges of temperature, dRH_i , and dw (Figure 4). Specifically, geometric means of IWC, N_i , and D_i in each Na bin are calculated and their linear regressions with respect to Na_{500} and Na_{100} are quantified (Table 1). Using the average ice microphysical properties inside each Na bin ensures that equal weight is assigned for each Na bin, regardless of the distribution of Na (Figure 4 last column).

Figure 4. Linear regressions of $d\log_{10}(\text{IWC})$, $d\log_{10}(\text{Ni})$, and $d\log_{10}(\text{Di})$ with respect to $d\log_{10}(\text{Na}_{500})$ and $d\log_{10}(\text{Na}_{100})$ under controlled temperature (a – c, m – o), $d\text{RH}_i$ (e – g, q – s), and dw (i – k, u – w). The last column shows number of samples for $d\log_{10}(\text{Na}_{500})$ and $d\log_{10}(\text{Na}_{100})$. Markers denote the geometric means of ice microphysical properties in each Na bin.

Linear regressions of IWC, Ni and Di with respect to Na_{500} show higher positive slopes of 0.743, 0.294, and 0.123 at warmer temperatures, while the slopes at colder temperatures are lower, i.e., 0.453, 0.177, and 0.07, respectively. However, this trend is reversed for correlations with Na_{100} , where the highest slopes occur at the coldest temperature range. The positive correlations of IWC, Ni, and Di with respect to Na_{500} and Na_{100} are more pronounced when Na is higher, specifically when $d\log_{10}(Na_{500}) > 0.5$ and $d\log_{10}(Na_{100}) > 1$. A sensitivity test shows similar temperature effects on slope values regardless of restricting linear fits to higher Na (Figure 5). These results indicate that temperature affects the role of various aerosols in cirrus formation, with larger aerosols being more effective at warmer temperatures (possibly via heterogeneous nucleation) and smaller aerosols being more effective at colder temperatures (possibly via homogeneous nucleation). For the entire range of Na, $d\log_{10}(IWC)$ increases by a factor of 2.8 – 5.5 and 2.5 – 4.8 with a tenfold increase of $d\log_{10}(Na_{500})$ and $d\log_{10}(Na_{100})$, respectively (Table 1). At much higher Na, IWC increases by a factor of 10 (100) compared with temperature-based averages as Na_{500} (Na_{100}) increases from 10 to 100 times higher than its average conditions. The smaller increase of IWC with respect to Na_{500} is consistent with heterogeneous nucleation, since only a small fraction of the total Na population can be activated as IN (Rogers, DeMott, Kreidenweis, & Chen, 1998), depending on aerosol composition and morphology, etc. The larger increase in IWC with respect to smaller aerosols is consistent with homogeneous nucleation which freezes a larger fraction of the aerosol population compared to heterogeneous nucleation compared with heterogeneous nucleation (Koop, Luo, Tsias, & Peter, 2000).

Figure 5. Similar to Figure 4 a – d and m – p, but with linear regressions applied to higher Na ranges, i.e., (a – d) $d\log_{10}(\text{Na}_{500}) > 0.5 \text{ \# cm}^{-3}$ and (e – h) $d\log_{10}(\text{Na}_{100}) > 1 \text{ \# cm}^{-3}$.

For the effect of $d\text{RH}_i$, the sub-saturated condition of $d\text{RH}_i \leq -10\%$ shows the lowest intercepts for IWC and Ni regressions, suggesting that the average IWC and Ni are smaller by a factor of 2 – 3 likely due to sublimation. The aerosol indirect effect of small aerosols is more sensitive to $d\text{RH}_i$ (i.e., higher slopes at higher $d\text{RH}_i$) than large aerosols, possibly due to a higher RH_i threshold required for homogeneous nucleation. Lastly, the correlations of IWC, Ni, and Di with respect to Na_{500} and Na_{100} are investigated under various ranges of $d\omega$ (Figure 4 i – l and u – x, respectively). Higher intercepts for IWC, Ni, and Di are seen associated with higher $d\omega$, indicating an increase of 10 – 30 times in strong updrafts. The slope values of IWC, Ni, and Di with respect to Na_{500} decrease with increasing $d\omega$, indicating that under higher updrafts the aerosol indirect effect of large aerosols is reduced. Such dampening effect from higher updrafts is slightly weaker on the aerosol indirect effect of smaller aerosols. From downdrafts to

updrafts, the slopes of $Ni - Na_{500}$ decrease most significantly from 0.345 to 0.066, while its counterpart of $Ni - Na_{100}$ only decreases from 0.368 to 0.247. The large dampening effect on the aerosol indirect effect from larger aerosols indicates that when sufficient cooling is available in updrafts, homogeneous nucleation becomes more viable and plays a more dominant role than heterogeneous nucleation. The decreasing slopes of $Di - Na_{500}$ and $Di - Na_{100}$ with increasing dw is consistent with a previous satellite-based analysis, which showed that impacts of aerosol loading on r_e are modulated by water vapor amount (Zhao et al., 2018)

Table 1. Linear Regressions of IWC, Ni and Di with respect to Na_{500} and Na_{100} under Controlled Temperature, dRH_i , and dw .

	$d\log_{10}(IWC) =$ $a + b \times$ $d\log_{10}(Na_{500})$	$d\log_{10}(Ni) =$ $a + b \times$ $d\log_{10}(Na_{500})$	$d\log_{10}(Di) =$ $a + b \times$ $d\log_{10}(Na_{500})$	$d\log_{10}(IWC) =$ $a + b \times$ $d\log_{10}(Na_{100})$	$d\log_{10}(Ni) =$ $a + b \times$ $d\log_{10}(Na_{100})$	$d\log_{10}(Di) =$ $a + b \times$ $d\log_{10}(Na_{100})$
T (°C)	All T	0.107 ± 0.06	0.064 ± 0.034	0.006 ± 0.008	0.255 ± 0.079	0.158 ± 0.046
	≤ -40	0.6 ± 0.06	0.256 ± 0.034	0.086 ± 0.008	0.532 ± 0.06	0.307 ± 0.035
	-50 – -40	-0.106 ± 0.061	-0.04 ± 0.035	-0.017 ± 0.011	0.171 ± 0.053	0.107 ± 0.027
		0.743 ± 0.061	0.294 ± 0.035	0.123 ± 0.011	0.492 ± 0.041	0.281 ± 0.021
	-60 – -50	0.314 ± 0.116	0.193 ± 0.072	0.027 ± 0.014	0.176 ± 0.101	0.118 ± 0.059
		0.66 ± 0.119	0.332 ± 0.073	0.06 ± 0.014	0.391 ± 0.078	0.231 ± 0.046
dRH_i (%)	-70 – -60	0.184 ± 0.06	0.94 ± 0.039	0.017 ± 0.005	0.216 ± 0.123	0.097 ± 0.072
		0.453 ± 0.061	0.177 ± 0.04	0.07 ± 0.005	0.682 ± 0.099	0.426 ± 0.059
	≤ 10	-0.33 ± 0.089	-0.205 ± 0.051	-0.027 ± 0.014	-0.412 ± 0.102	-0.342 ± 0.068
		0.911 ± 0.091	0.466 ± 0.052	0.115 ± 0.014	0.508 ± 0.086	0.297 ± 0.057
	-10 – 0	0.189 ± 0.089	0.162 ± 0.042	0.001 ± 0.011	0.242 ± 0.084	0.192 ± 0.048
		0.523 ± 0.09	0.219 ± 0.043	0.078 ± 0.011	0.388 ± 0.064	0.219 ± 0.036
dw (m/s)	0 – 10	0.238 ± 0.069	0.143 ± 0.043	0.019 ± 0.008	0.592 ± 0.101	0.356 ± 0.064
		0.542 ± 0.07	0.173 ± 0.044	0.098 ± 0.008	0.383 ± 0.078	0.266 ± 0.049
	> 10	0.139 ± 0.083	0.004 ± 0.054	0.041 ± 0.012	0.481 ± 0.079	0.262 ± 0.045
		0.562 ± 0.084	0.266 ± 0.055	0.065 ± 0.012	0.581 ± 0.06	0.329 ± 0.034
	≤ -0.5	-0.035 ± 0.106	0.01 ± 0.072	-0.019 ± 0.014	-0.117 ± 0.085	-0.029 ± 0.052
		0.683 ± 0.112	0.345 ± 0.076	0.077 ± 0.015	0.623 ± 0.065	0.368 ± 0.04
dw (m/s)	-0.5 – 0	-0.175 ± 0.069	-0.133 ± 0.049	-0.008 ± 0.01	-0.18 ± 0.083	-0.101 ± 0.054
		0.744 ± 0.07	0.343 ± 0.05	0.11 ± 0.01	0.506 ± 0.063	0.295 ± 0.041
	0 – 0.5	0.277 ± 0.046	0.161 ± 0.024	0.024 ± 0.008	0.461 ± 0.088	0.268 ± 0.046
		0.462 ± 0.047	0.181 ± 0.025	0.073 ± 0.008	0.38 ± 0.068	0.254 ± 0.036
	> 0.5	0.759 ± 0.091	0.43 ± 0.046	0.074 ± 0.012	1.037 ± 0.064	0.568 ± 0.035
		0.257 ± 0.093	0.066 ± 0.047	0.041 ± 0.013	0.363 ± 0.05	0.247 ± 0.028

Conclusions and Implications on Numerical Simulations

In this study, the relationships of cloud microphysical properties with respect to thermodynamics, dynamics, and aerosol sizes and concentrations are investigated using a composite in situ observation dataset. The observations are based on seven flight campaigns, including samples over the Pacific Ocean, North and South America, the Southern Ocean, and the Caribbean. Using 1-Hz aircraft observations (~ 230 m horizontal resolution), we employ a new method to isolate specific thermodynamic and dynamical effects, prior to quantifications of aerosol indirect effects on ice microphysical properties. Regarding thermodynamic and dynamical effects, the maximum IWC is found to be associated with warmer temperature (-40°C), RH_i slightly above ice saturation (10% $d\text{RH}_i$), and strong updrafts ($dw > 3$ m/s). The impacts on Ni and Di are similar to those seen in IWC, except that Ni is less sensitive to temperature, and the maximum Di occurs at higher RH_i (30% $d\text{RH}_i$). The effects of temperature, RH_i and w are consistent with several previously studies that only analyzed one of these factors, such as Schiller et al. (2008) (temperature effect in their Figure 2), Diao et al. (2013b) (RH_i effect in their Figure 4), and D'Alessandro et al. (2017) (w effect in their Figures 9 and 10). Our results demonstrate the necessity of controlling temperature, RH_i and w before investigating aerosol indirect effects, since these factors could either obscure or amplify the signal of aerosol indirect effects.

For aerosol indirect effects, we found non-monotonic correlations of IWC, Ni, and Di with respect to concentrations of larger and smaller aerosols (Na_{500} and Na_{100} , respectively). That is, when Na is relatively lower (i.e., $d\log_{10}(\text{Na}_{500}) < 0.5$ or

$d\log_{10}(\text{Na}_{100}) < 1$), IWC, Ni and Di are relatively constant or even slightly decrease with Na; when Na is relatively higher (i.e., $d\log_{10}(\text{Na}_{500}) \geq 0.5$ or $d\log_{10}(\text{Na}_{100}) \geq 1$), IWC, Ni and Di increase with Na. One should caution that the higher Na is a relatively small fraction of the total aerosol population. The decrease of Ni and IWC with Na at lower Na is more pronounced when RH_i is restricted to $> 120\%$, separating cirrus clouds in nucleation phase from the latter phases (Figures 6 and 7). The slight decrease of Ni and IWC at lower Na is likely caused by competition between homogeneous and heterogeneous nucleation, which is known as the anti-Twomey effect and has been shown in model simulations (e.g., Spichtinger and Cziczo, 2010; Spichtinger and Gierens, 2009).

The linear regression slopes of $d\log_{10}(\text{Ni})$ with respect to $d\log_{10}(\text{Na}_{500})$ and $d\log_{10}(\text{Na}_{100})$ at various temperatures are $0.18 - 0.33$ and $0.23 - 0.43$, respectively (Table 1). That is, with a tenfold increase of Na_{500} and Na_{100} , Ni increases by a factor of $1.5 - 2.1$ and $1.7 - 2.7$ compared with temperature-based averages, respectively. These values are slightly lower than that observed in Seifert et al., (2004), which showed a slope of 0.58 when $\text{Na} < 100 \text{ cm}^{-3}$, yet they only sampled ice particles smaller than $15 \text{ }\mu\text{m}$. On the other hand, the slope values in this study are slightly higher than those shown in simulations (slopes of $0.1 - 0.3$) (Kärcher and Lohmann, 2002; Kay and Wood, 2008; Liu and Shi, 2018), possibly because those simulations only considered homogeneous nucleation and turned off heterogeneous nucleation. IWC increases faster than Ni with increasing Na due to the slight increase of Di. Previous observations showed increase of both Ni and Di during cirrus evolution from nucleation phase to early growth and later

growth phases, likely due to new ice particle formation and continuous growth (Diao et al., 2013b, 2014b). Such Ni – Di correlations differ from a single nucleation event, where Ni and Di are anti-correlated given that total water content is constant. The increasing Ni during cirrus evolution is also supported by model simulations under continuous cooling or uplifting (Diao et al. 2017; Spichtinger and Gierens 2009).

Figure 6. Relationships of $d\log_{10}(\text{IWC})$, $d\log_{10}(\text{Ni})$ and $d\log_{10}(\text{Di})$ with respect to $d\log_{10}(\text{Na}_{500})$. Different RH ranges are shown: (a – c) $\text{RH} > 120\%$, (d – f) $100\% < \text{RH} \leq 120\%$, and (g – i) $\text{RH} \leq 100\%$. All the analysis in this figure are restricted to temperature $\leq -40^\circ\text{C}$.

Figure 7. Similar to Figure 6, but for $d\log_{10}(\text{Na}_{100})$.

A highlight of this study is to examine the aerosol indirect effects under controlled thermodynamic and dynamical conditions, which showed varying magnitudes of aerosol indirect effects with temperature, RH_i, and w . Such approach takes advantages of synchronized, fast and accurate in-situ measurements, which often can be difficult to achieve by satellite observations due to uncertainties in retrievals, such as uncertainties of

water vapor by 30%–40% and temperature by 1–2 K in the NASA AIRS satellite data (Diao, Jumbam, Sheffield, Wood, & Zondlo, 2013a).

The observations in this study are restricted to temperature $\leq -40^{\circ}\text{C}$, while some ice may have been formed in warmer temperatures (e.g., warm conveyor belts or deep convection) and uplifted into colder temperatures. To address the potential influence from convection, sensitivity tests are conducted on different latitudes (Figures 8 and 9). In addition, different evolutionary phases of cirrus clouds are examined by separating RHi ranges into $\text{RHi} > 120\%$, $100\% < \text{RHi} \leq 120\%$, and $\text{RHi} \leq 100\%$ (Figures 6 and 7). Similar non-monotonic correlations of IWC, N_i , and D_i with respect to $d\log_{10}(\text{Na}_{500})$ and $d\log_{10}(\text{Na}_{100})$ are shown for all latitudinal ranges and RHi ranges. The relatively higher Na_{500} and Na_{100} were observed in various flight campaigns at different geographical locations (Figure 10), and are associated with various temperatures, RHi and w (not shown). To examine whether cirrus cloud microphysical properties are consistently correlated with Na conditions on coarser scales, we applied 100-second moving averaging to Na_{100} and Na_{500} values (Figure 11). Even though spatial averaging reduces the maximum values of $d\log_{10}(\text{Na}_{100})$ and $d\log_{10}(\text{Na}_{500})$, the non-monotonic correlations with cirrus microphysical properties and their turning points are similar, indicating that aerosol indirect effects are consistently seen at both finer and coarser scales (i.e., ~ 0.23 and 23 km horizontal scales, respectively).

Figure 8. Relationships of $d\log_{10}(\text{IWC})$, $d\log_{10}(\text{Ni})$ and $d\log_{10}(\text{Di})$ with respect to $d\log_{10}(\text{Na}_{500})$. Different latitudinal ranges in the northern hemispheres: (a – c) 30°N – 30°S, (d – f) 30°N – 60°N and 30°S – 60°S, and (g – i) 60°N – 90°N and 60°S – 90°S. The delta values are calculated against the temperature-based average values within each latitudinal range. All the analysis in this figure are restricted to temperature $\leq -40^{\circ}\text{C}$.

Figure 9. Similar to Figure 8 but for $d\log_{10}(\text{Na}_{100})$. Higher $d\log_{10}(\text{Na}_{100})$ values were shown in the tropics possibly due to deep convection.

Figure 10. Geographical locations of the relatively higher Na when (a) $d\log_{10}(\text{Na}_{500}) > 0.5$ and (b) $d\log_{10}(\text{Na}_{100}) > 1$, color coded by the magnitudes of $d\log_{10}(\text{Na}_{500})$ and $d\log_{10}(\text{Na}_{100})$, respectively.

Figure 11. Relationships of $d\log_{10}(\text{IWC})$, $d\log_{10}(\text{Ni})$ and $d\log_{10}(\text{Di})$ with respect to (a – c) $d\log_{10}(\text{Na}_{500})$ and (d – f) $d\log_{10}(\text{Na}_{100})$. The $d\log_{10}(\text{Na}_{500})$ and $d\log_{10}(\text{Na}_{100})$ are calculated based on 100-second moving average of Na conditions. The delta values of cirrus microphysical properties are based on 1-Hz data. Analysis in this figure is restricted to temperature $\leq -40^\circ\text{C}$. The goal of this figure is to illustrate whether Na conditions in a larger horizontal scale would show similar correlations with cirrus microphysical properties.

The role of heterogeneous and homogeneous nucleation can be speculated based on the strengths of aerosol indirect effects from larger and smaller aerosols, respectively. Specifically, the slope values from linear regressions of IWC, Ni and Di with respect to Na₅₀₀ and Na₁₀₀ are used to quantify aerosol indirect effects, with the more positive slope values indicating stronger aerosol indirect effects. The effect of larger aerosols is stronger at warmer temperature, lower RH_i, and smaller w, indicating that heterogeneous nucleation is more effective when the ambient conditions do not exceed the thresholds of homogeneous nucleation. On the other hand, the effect of smaller aerosols is stronger at colder temperature and higher RH_i, and produces higher IWC and Ni at strong updrafts, indicating these are more favorable conditions for homogeneous nucleation. One caveat of these speculations is that larger aerosols may not be well correlated with INP number concentrations at all conditions. Without INP measurements $\leq -40^{\circ}\text{C}$ in the field campaigns analyzed here, these speculations warrant more future investigation on the relationships among Na₅₀₀, Na₁₀₀ and INP number concentrations to confirm the role of heterogeneous and homogeneous nucleation. Another caveat is that smaller ice particles ($<62.5\text{ }\mu\text{m}$) are not included in the measurements. Whether those particles would follow similar correlations with Na requires future investigation based on different instrumentations, such as the 2D-Stereo (2DS) or Small Ice Detector (SID-2H) probes.

Using a global-scale composite in-situ dataset, the observation-based statistical distributions of cirrus cloud microphysical characteristics can be used to evaluate model simulations regarding the thermodynamic, dynamical and aerosol indirect effects. Previous studies have illustrated the sensitivity of model simulations to thermodynamic

and dynamical conditions, such as sub-grid variability of water vapor (Wu et al., 2017) and sub-grid vertical velocity fluctuations (Shi and Liu, 2016). This study can be used as a blueprint for future studies to improve model parameterizations based on the observed relationships of ice microphysical properties with respect to temperature, RH_i , w , and aerosol concentrations.

Chapter 2. Evaluation of Cirrus Cloud Controlling Factors and Aerosol Indirect Effects based on a Global Aircraft Dataset and the Community Atmosphere Model version 6

Introduction

Cirrus clouds represent one of the most ubiquitous global cloud types with an estimated global coverage of nearly 20% (Sassen, Wang, & Liu, 2008). According to the IPCC fifth assessment report (Boucher et al., 2013), the largest uncertainty in estimating the future global climate budget stems from clouds and aerosols. Unlike most other cloud types, cirrus clouds may produce a net positive or negative radiative forcing depending on their microphysical properties (Y. Zhang, MacKe, & Albers, 1999). In addition, the radiative effect can be highly variable depending on thermodynamic conditions and aerosol distributions. Tan et al. (2016) showed the radiative effects of ISS changes with increasing or decreasing ISS. Another modeling study found large differences in the net cloud forcing depending on the fraction of IN that can be nucleated and mechanism through which the clouds form (Storelvmo and Herger, 2014). The large uncertainties in cirrus cloud radiative forcing illustrate the need for further study on the distributions of cirrus cloud controlling factors, and their effects on the microphysical properties.

Ideally, a comprehensive quantification of the spatiotemporal distribution of cirrus clouds' microphysical properties globally would mitigate many uncertainties. However, challenges remain in field and laboratory measurements of ice nucleating particles (INPs) to achieve the necessary spatial coverage (Cziczo and Froyd, 2014). With this being the case, efforts are being made to understand cirrus cloud properties based on their geographical locations. Diao et al. (2014b) performed a hemispheric analysis of in situ

cirrus evolution and found little difference in the clear-sky ice supersaturations frequency between northern and southern hemispheres (NH and SH, respectively). Another study investigated the IWC and snowfall rates for tropical, midlatitude, and Arctic cirrus clouds using in situ observations and found a geographical dependence of IWC (Heymsfield et al., 2017). Wolf et al. (2018) used balloon-based in situ observations to analyze microphysical properties of Arctic ice clouds and found differences in particle size distributions (PSDs) depending on the cloud origin. Moving from north to south using lidar-based observations from two research cruises, from Leipzig, Germany to Puenta Arenas, Chile and the other to Stellenbosch, South Africa, Kanitz et al. (2011) observed a decrease in the efficiency of heterogeneous nucleation in the SH, which could be a result of fewer INPs. This hemispheric difference in aerosol indirect effects is consistent with significantly higher aerosol number concentrations in the NH (Minikin et al., 2003). Krämer et al. (2020) developed a cirrus cloud climatology, focusing on tropical and midlatitude cirrus clouds, and showed that the thickness and optical properties were larger at lower altitudes, thus producing a negative radiative forcing. There remains, however, a lack of study that investigates both the regional differences as well as the statistical conditions of cirrus clouds' controlling factors, (i.e., thermodynamics, dynamics and aerosols).

Recently, developments have been made in quantifying certain microphysical properties with respect to various controlling environmental factors using in situ observations. For example, the effects of temperature have been extensively studied from in situ observations (Heymsfield et al., 2017; Luebke et al., 2013, 2016; Schiller et al.,

2008) and indicate IWC increases towards warmer temperatures. A number of studies focused on distributions of RHi have found that in-cloud RHi occur most frequently at or near 100% (Jensen et al., 2001; Krämer et al., 2009). In addition, the spatial regions of ISS can also vary widely from the micro- to mesoscales, with horizontal extents ranging from 100 m to 100 km, respectively (Diao et al., 2014a). Another study by Diao et al. (2017) found that increasing the RHi threshold of ice nucleation to 130% in an idealized simulation produces the closest results to in situ observations. The effects of vertical velocity on cirrus cloud microphysical properties have been investigated (Muhlbauer et al., 2014a; Muhlbauer, Kalesse, & Kollias, 2014b) and strong updrafts are found to be associated with higher occurrence frequency with higher in-cloud ISS (D'Alessandro et al., 2017).

Finally, the effects of aerosols on cirrus microphysical properties has been extensively studied. For example, Cziczo et al. (2013) investigated ice crystal residuals from in situ observations and found that the majority of ice formed via the freezing of mineral dust and metallic particles. Anthropogenic aerosols, such as secondary organic aerosols have also been found to be less likely to act as INPs compared with mineral dust (Prenni et al., 2009). Another field experiment found that aerosols > 500 nm correlate well with the number of INP in cirrus clouds (DeMott et al., 2010). However, this study only focused on clouds at temperatures $> -36^{\circ}\text{C}$ and has yet to be fully tested for the cirrus regime. Based on remote sensing data, Zhao et al. (2018, 2019) showed the correlations between ice crystal sizes and aerosol optical depth can be either positive or negative depending on the meteorological conditions in convective clouds. Chylek et al.

(2006) showed an increase in ice crystal size during the more polluted winter months than cleaner summer months over the eastern Indian Ocean. Using a global-scale dataset of multiple flight campaigns, Patnaude and Diao (2020) isolated individual effects from temperature, RH_i, vertical velocity and number concentrations of larger and smaller aerosols. They found that when aerosol number concentrations are 3 – 10 times higher than their average conditions, there are strong positive correlations between cirrus microphysical properties such as IWC, N_i and D_i. These aerosol indirect effects are also susceptible to whether or not thermodynamic and dynamical conditions are controlled, demonstrating the need for separating all factors when investigating the aerosol indirect effects.

More recently, in situ observations have been used to provide high-frequency measurements as constraints for cirrus cloud parameterizations in GCMs. Two types of simulations have been used for model evaluation, i.e., free-running (Eidhammer, Morrison, Bansemer, Gettelman, & Heymsfield, 2014; Wang & Penner, 2010; K. Zhang et al., 2013) and nudged and collocated (D'Alessandro et al., 2019; Kooperman et al., 2012; Wu et al., 2017). For free-running simulations, a comparison on statistical distributions of ice microphysical properties is often used for model validation (Penner, Chen, Wang, & Liu, 2009). The second method that nudges wind and temperature measurements from reanalysis towards the aircraft flight tracks has been used with success more recently (e.g., D'Alessandro et al., 2019; Wu et al., 2017). These studies provide a method for direct comparison since the simulated meteorology is collocated to the aircraft observations and allows for an improved analysis of model physics. Given the

importance and limited understanding of how aerosols interact with cirrus clouds, much attention has been dedicated to modeling and the parameterization of aerosol indirect effects (Kärcher and Lohmann, 2002, 2003; Kuebbeler et al., 2014; Wang et al., 2014). Furthermore, Shi et al. (2015) added the effects of pre-existing ice into the NCAR Community Atmosphere Model version 5 model and found a decrease in Ni due to the reduction of homogeneous freezing frequency. Other studies also compared the role of homogeneous and heterogeneous nucleation for the contribution to IWC and Ni (Penner et al., 2018; Zhou, Penner, Lin, Liu, & Wang, 2016).

This study aims to bridge the knowledge gap on how cirrus clouds vary depending on latitude and hemisphere by using a comprehensive in situ dataset that includes seven NSF flight campaigns. Observations were collected aboard the NSF/NCAR G-V research aircraft and provides a large geographical coverage. These data are compared to the Community Atmosphere Model version 6 (CAM6) to evaluate the impacts of all controlling factors (i.e., RH_i, temperature, w and N_a) on cirrus cloud IWC, Ni, and Di as well as differences in free-running and nudged simulations. Additional free-running simulations were evaluated under different CAM6 set-up configurations. Descriptions of the seven flight campaigns, instrumentation, CAM6 model set-up and configurations are provided in Section 2.

Data and Methods

In situ Observations and Instrumentation

In this study, we use airborne in situ observations measured at 1-Hz from the NSF HIAPER G-V research aircraft. We use a comprehensive global dataset combining seven

NSF research campaigns: START08 (Pan et al., 2010), HIPPO deployments 2-5 (Wofsy et al., 2011), PREDICT (Montgomery et al., 2012), TORERO (Volkamer et al., 2015), DC3 (Barth et al., 2015), CONTRAST (Pan et al., 2017), ORCAS (Stephens et al., 2018). Table 1 provides a detailed summary of the seven in situ aircraft campaigns used in this study, including location, timing of flights, and flight hours for in-cloud and clear sky conditions.

For this study, ice measurements are provided by the Fast-2DC probe with a 64-diode laser array with a range of $25\text{ }\mu\text{m} - 1600\text{ }\mu\text{m}$, and can be reconstructed up to $3200\text{ }\mu\text{m}$. In order to mitigate the shattering effect, particles $< 62.5\text{ }\mu\text{m}$ are excluded in icing measurements, this includes mass and number concentrations. The Rosemount temperature probe was used for temperature measurements which has an accuracy and precision of $\sim \pm 0.3\text{ K}$ and 0.01 K , respectively. All analyses are restricted to temperatures $\leq -40^\circ\text{C}$, in order to exclude the presence of super-cooled liquid droplets in mixed-phase clouds. Highly calibrated and quality-checked water vapor data are collected using a VCSEL hygrometer (Zondlo et al., 2010), with a measuring frequency of 25 Hz , and an accuracy of $\sim 6\%$ and precision of $\leq 1\%$. Water vapor for this analysis was used at 1 Hz . Aerosol measurements are collected from the UHSAS, which uses 100 logarithmically spaced bins ranging from $0.06\text{ }\mu\text{m} - 1\text{ }\mu\text{m}$. RH_i is calculated using methods from Murphy and Koop (2005). The combined RH_i uncertainties from the temperature and water vapor measurements range from 6.9% at -40°C to 7.8% at -78°C .

Table 2. *Summary of the Seven Research Campaigns from the NSF G-V Aircraft*

Acronym	Field Program	Time	Lat, Lon	Region	Flight hours	In-cloud hours	Clear sky hours
START08	Stratosphere-Troposphere Analyses of Regional Transport	April – June 2008	26°N – 62°N, 117°W – 86°W	NM, NP	84	2	52
HIPPO	HIAPER Pole-to-pole Observation - deployments 2 – 5	Oct-Nov 2009; Mar – Apr 2010; Jun – July 2011; Aug-Sept 2011	87°N – 67°S, 128°E - 90°W	A	333	7	111
PREDICT	PRE-Depression Investigation of Cloud Systems in the Tropics	Aug – Sept 2010	10°N – 28.5°N, 86°W – 37°W	NT	105	25	66
DC3	Deep Convective Clouds and Chemistry Project	May – June 2012	25°N – 42°N, 106°W – 80°W	NM	144	23	54
TORERO	Tropical Ocean Troposphere Exchange of Reactive halogen species and Oxygenated voc	Jan – Feb 2012	42°S – 14°N, 105°W – 70°W	NT, ST, SM	125	2	52
CONTRAST	CONvective TRansport of Active Species in the Tropics	Jan – Feb 2014	20°S – 40°N, 132°E - 105°W	NM, NT, ST	116	23	48
ORCAS	The O ₂ /N ₂ Ratio and CO ₂ Airborne Southern Ocean Study	Jan – Mar 2016	75°S – 18°S, 91°W – 51°W	SM, SP	95	1	40

N, northern hemisphere; S, southern hemisphere; T, tropics; M, midlatitude; P, polar regions; A, all regions. In-cloud and clear sky hours are only for temperatures $\leq -40^\circ\text{C}$

Measurements are separated by cloud condition whereas in-cloud is defined by the presence of at least one ice crystal from the Fast-2DC probe ($N_i > 0 \text{ L}^{-1}$), which has been used by previous analyses (D'Alessandro et al., 2017; Diao et al., 2013b, 2014b, 2015, 2017; Tan et al., 2016), and all other samples are defined as clear sky. For regional comparisons, data are binned by northern and southern hemispheres and latitudinal region (60°N – 90°N and 60°S – 90°S, Polar; 30°N – 60°N and 30°S – 60°S, Midlatitude; 0° – 30°N and 0° – 30°S, Tropical, respectively).

Climate Model Description and Experiment Design

This study uses model simulations conducted using the NCAR Community Atmosphere Model version 6 (CAM6). Of particular interest, CAM6 implemented a new scheme, the Cloud Layers Unified by Binormals (CLUBB) for representation of boundary layer turbulence, shallow convection and cloud macrophysics (Bogenschutz et al., 2013). CLUBB is a higher-order turbulence closure scheme that calculates prognostic higher-moments based on joint probability density functions (PDFs) for vertical velocity, temperature, and moisture (Golaz, Larson, & Cotton, 2002). An improved bulk two-moment cloud microphysics scheme has been implemented (Gettelman and Morrison, 2015) that replaces diagnostic treatment of rain and snow with prognostic treatment of all hydrometeors (i.e., rain, snow, graupel and hail). This is coupled with a 4-mode aerosol model (MAM4) (Liu et al., 2016) for aerosol cloud microphysics. This allows for ice crystals to form via homogeneous freezing of sulfate aerosols and heterogeneous freezing of dust aerosols (Liu, Penner, Ghan, & Wang, 2007; Liu and Penner, 2005). Finally, the deep convection scheme (G. J. Zhang and McFarlane, 1995) has been tuned to include sensitivity to convection inhibition.

Results from in situ observations are compared with two types of CAM6 simulations, free-running and nudged simulations. Simulations are based on a finite-volume dynamical core (Lin, 2004) with a horizontal resolution of $0.9^\circ \times 1.25^\circ$ with 32 vertical levels, using prescribed sea-surface temperature and 20th century industrial emissions. All simulations are conducted using a 6-month spin-up time with the free-running simulations saving instantaneous output every 6-hours (00, 06, 12, 18 UTC). CAM6

simulations are nudged spatially and temporally with meteorological data (3-D wind and temperature) from the Modern-Era Retrospective Analysis for Research and Applications version 2 (MERRA2) (Gelaro et al., 2017), and collocated with aircraft flight tracks in space and time. A nudged simulation was conducted for each of the seven campaigns independently and was combined into one data set (hereafter named “CAM6-nudg”) to be compared with observations. To maintain the variabilities from in-situ observations, 1-Hz observational data are used to compare with model output on the same frequency. One free-running simulation was conducted for the duration of all flight campaigns from July 2008 to February 2016. When comparing with observations, a total of 24 instantaneous outputs from the free-running simulation are combined into one data set (“CAM6-free” hereafter) (i.e., including 000 and 12 UTC for the first day of each month in 2010). Additional sensitivity tests for annual or seasonal variabilities show very minor differences in the statistical distributions of cirrus microphysical properties and the correlations with their controlling factors when selecting difference years, seasons, and days in a month. Definition for simulated in-cloud conditions are restricted for IWC and N_i values $> 1e-5 \text{ g m}^{-3}$ and $> 1e-2 \text{ # L}^{-1}$, respectively, which are the lower limits from observations. In addition, simulated microphysical properties are restricted to pressure $> 123 \text{ hPa}$, to match the pressure limits from observations.

In addition to the nudged and free-running simulations covering the duration of the flight campaigns, three sensitivity experiments are conducted. Two free-running simulations, CAM6-HOM and CAM6-HET are conducted by turning off heterogeneous and homogeneous freezing modes, respectively. The final experiment examines the

effects of excluding pre-existing ice (CAM6-PRE), since their inclusion has become the default set-up for CAM6 (Shi et al., 2015). The sensitivity experiments are run for a shorter period (i.e., February to September in 2010) than CAM6-free, including 26 outputs for each sensitivity experiment. For a more direct comparison between observations and CAM6, simulated ice crystal number concentrations and mixing ratios are also restricted to $> 62.5 \mu\text{m}$, similar to the size restriction method from Eidhammer et al. (2014), which also includes snow in mass and number concentrations. In addition, simulated aerosol number concentrations are restricted to sizes $> 500 \text{ nm}$ and $> 100 \text{ nm}$ by summing the size-restricted concentrations of the aiten, accumulation and coarse aerosol modes.

Table 3. Summary of CAM6 Simulations.

Simulation	Experiment design
CAM6-free	Free-running during the duration of all seven flight campaigns.
CAM6-nudg	Nudged & collocated along all flight tracks merging meteorological wind (U, V) and temperature (T) conditions.
CAM6-HET	Free running with homogeneous nucleation turned off
CAM6-HOM	Free running with heterogeneous nucleation turned off
CAM6-PRE	Free running with pre-existing ice crystals (Shi et al. 2015) turned off

Cirrus Cloud Regional Analysis

Regional Cirrus Cloud Microphysical Properties with respect to Temperature

Three cirrus cloud microphysical properties, IWC, Ni and Di are examined in relation to temperature and further controlled by hemisphere and latitudinal region for in situ observations and CAM6 are shown in Figure 12. NH in situ IWC and Ni (Figure 12 top two rows) indicate clear latitudinal trends with the highest values occurring in the

midlatitudes, tropics, then polar regions for temperatures $> -60^{\circ}\text{C}$, but for colder temperatures the tropical region shows the highest IWC. However, in the SH highest IWC and Ni occur in the tropics, followed by the polar regions then midlatitudes. Interestingly, there is a significant reduction in IWC and Ni from the NH to SH, with nearly an order of magnitude difference at temperatures $< -50^{\circ}\text{C}$. For example, IWC in the NH midlatitude (NM) is $\sim 0.01 \text{ g m}^{-3}$ compared to $\sim 0.001 \text{ g m}^{-3}$ in the SH midlatitude (SM) at -56°C . Di shows little change between hemisphere and latitudinal region apart from high values in the SH polar (SP) region between -50°C and -60°C . In all regions, the average IWC increases with increasing temperature consistent with previous studies (Krämer et al., 2016; Luebke et al., 2013; Schiller et al., 2008). The average Ni shows no clear trend with temperature while the average Di shows a positive trend with increasing temperature. The reason for large ice crystals at warmer temperatures are likely due to sedimenting large ice crystals, a bias towards heterogeneous nucleation, and faster growth rates due to a higher vapor pressure. Similar to in situ observations, model IWC (Figure 12 g and h) increases with increasing temperature for both latitude regions. Differing from observations, CAM6 produces the highest IWC and Ni in the tropical regions, followed by midlatitudes then polar regions for both hemispheres. Simulated Ni remains relatively constant across all temperatures, while average Di shows a positive trend with increasing temperature, agreeing well with observations. The simulated Di also indicates very little difference between hemispheres and latitudes. However, CAM6 produces slightly lower Di compared with observations, especially for $T > -50^{\circ}\text{C}$. For example, the average Di in the NH tropical (NT) region from observations (CAM6) are

~180 μm (~120 μm), respectively at -50°C . CAM6 simulated Di also indicates significantly more variability in the SH, where standard deviations in the NH and SH are ~50 μm and ~100 μm , respectively, at $T > -50^{\circ}\text{C}$.

A hemispheric evaluation for each latitude region of CAM6 microphysical properties with respect to temperature is shown in Figure 13. For all latitude regions and both hemispheres, in situ IWC values are 3 to 100 times higher than CAM6. However, with the exception of in situ NM, there is little difference between hemispheres in observations and CAM6. In contrast, Ni shows no clear dependence on geographical location from both observations and CAM6. For example, in the tropical region, simulated Ni is higher than observations in both hemispheres at $T > -60^{\circ}\text{C}$, but then is lower than observations at $T < -60^{\circ}\text{C}$. In the NM, in situ Ni is higher than the model except at the warmest temperatures ($> -45^{\circ}\text{C}$), and both in situ and model Ni is higher in the NH. Interestingly, simulated Ni is higher than in situ for the SM. In regard to Di, in situ observations are higher than model Di, and there are little differences between hemispheres. Observed Di values in the tropics are nearly identical in the NH as the SH but are lower in the NH for the polar region. These results indicate that the hemispheric and latitudinal location are important to consider when modeling cirrus cloud microphysical properties

Figure 12. Logarithmic means of (a, b, g and h) IWC, (c, d, i and j) Ni, and linear (e, f, k and l) Di at 4°C temperature intervals between -80°C and -40°C for in situ observations (first three rows) and CAM6 (bottom three rows). Whiskers represent $\pm$ one standard deviation.

Figure 13. Logarithmic means of (a – c) IWC, (d – f) Ni, and linear (g – i) Di at 4°C temperature intervals between -80°C and -40°C for in situ observations (black lines) and CAM6 (red lines). Observed and simulated microphysical properties are binned by hemisphere and region similar to Figure 12, where NH is denoted by solid lines, and SH is denoted by dashed lines.

RHi and w Distributions for In-cloud and Clear Sky Conditions

Regional distributions of observed RHi under clear sky and in-cloud conditions are shown in Figures 14 and 15. For both conditions, RHi magnitudes range from < 5% up to ~180% for temperatures $\leq -40^\circ\text{C}$. Under clear sky conditions, the majority of samples are located below 100%, however there are occurrences that reach up to and exceed liquid saturation level. The few samples that reach above the liquid saturation line are within the

instrument measurement uncertainties. In contrast, the majority of in-cloud samples occur most frequently at or within $\sim 20\%$ of saturation, which agrees well with previous studies (D'Alessandro et al., 2017; Krämer et al., 2009). Additionally, the upper limit of ISS for in-cloud condition remains below the homogeneous freezing line at colder temperatures, excluding NT region (Figure 14 k), which was also observed by Krämer et al. (2009) and described as the “peak RHi” in Kärcher and Lohmann (2002) based on the competition between increasing RHi due to continuous cooling and the depletion of water vapor from newly nucleated ice particles.

Figure 14. Number of samples of RHi for given temperature intervals from in situ observations under (left two columns) clear sky and (right two columns) in-cloud conditions. Solid and dashed black line represents the saturation with respect to ice and liquid water, both calculated based on Murphy and Koop (2005). Dash-dotted line denotes the homogeneous freezing line based on Koop et al. (2000).

CAM6 regional distributions of RHi for clear sky and in-cloud conditions are shown in Figure 15. Similar to observations, simulated clear sky and in-cloud RHi show highest occurrence frequency below and at 100%, respectively. Additionally, CAM6 is able to

represent some ISS up to the homogeneous freezing threshold yet occurrences remain below the liquid-water saturation line. Despite a few clear sky ISS occurrences, the vast majority of samples remain $< 100\%$. In-cloud RH_i ranges from $\sim 160\%$ to $< 10\%$ in the polar regions and midlatitudes, agreeing well with observations, while tropical regions show less frequency of simulated RH_i $< 40\%$. CAM6 in-cloud polar regions represent an interesting comparison to observations. On the one hand, RH_i occurrences are slightly biased towards ISS with the most occurrences around $\sim 110\%$ in agreement with aircraft measurements. On the other hand, simulated RH_i is underestimated, producing significantly more RH_i $< 80\%$ compared to observations. In general, the regional distributions of RH_i in the model closely resemble the observed RH_i distributions.

Figure 15. Similar to Figure 14 but for CAM6 nudged simulations. In-cloud condition uses a threshold when IWC and Ni as $1\text{e-}5\text{ g m}^{-3}$ and $> 1\text{e-}2\text{ \# L}^{-1}$, respectively, and all other samples are labeled clear sky.

Regional distributions of w for in situ observations and CAM6 nudged are shown in Figures 16 and 17, respectively. Observed in-cloud and clear sky w values are highest in the midlatitude and tropical regions (Figure 16 e – l), reaching up to ~ 5 m/s, while vertical motions in the polar regions are significantly weaker with the strongest updraft only reaching up to ~ 3 m/s (Figure 16 a – d). Both conditions exhibit similar distributions of w with maximum occurrences centered at 0 m/s. Muhlbauer et al. (2014a) found similar distributions for midlatitude cirrus when controlling for synoptic patterns. However, that study did not include observations of clear sky conditions. Surprisingly, there is not a difference in maximum w values between clear sky and in-cloud condition or temperature. Distributions of w from CAM6-nudg produce significantly different results (Figure 17). Contrary to observations, the model indicates a bias towards stronger w values when in-cloud, with in-cloud maximum w values 2 – 3 times higher than in clear sky. CAM6 also simulates only updrafts for in-cloud and clear sky conditions. This is likely due to how sub-grid vertical velocity is calculated, which comes from the square root of turbulent kinetic energy (TKE) (Gettelman et al., 2010), therefore only updrafts would occur. However, D’Alessandro et al. (2017) in a model study, showed upward and downward vertical motions frequently occur in close proximity to each other. Besides differences in the vertical gradient between observations and CAM6, the model also underestimates the strength of the updrafts. Peak updrafts occur under in-cloud conditions in the midlatitudes and tropical regions, which agrees with observed w . However, do not even reach 1.5 m/s, significantly lower than the ~ 5 m/s seen in observations. The model demonstrates the need for stronger w for the formation of cirrus clouds, however

observations indicate that the strength of the wind is not a factor for determining clear sky or in-cloud conditions, but likely a combination of thermodynamic and dynamic conditions.

Figure 16. Number of samples of w for given temperature intervals from in situ observations under (left two columns) clear sky and (right two columns) in-cloud conditions. Observations are separated by hemisphere and latitudinal region similar to Figure 12.

Figure 17. Similar to Figure 16 but for CAM6 nudged simulations. In-cloud and clear sky conditions are determined using similar thresholds to Figure 15.

Individual Impacts of Cirrus Cloud Controlling Factors

Thermodynamic, Dynamic and Aerosol Distributions

Probability density functions (PDFs) of cirrus cloud controlling factors are shown in Figure 18. Probabilities of clear sky and in-cloud conditions for a given temperature (Figure 18 a – c) remain nearly the same for all temperatures and in-cloud and clear sky conditions. At very cold temperatures $< -70^{\circ}\text{C}$, in-cloud probabilities in observed and CAM6-nudg drop 2-3 orders of magnitude. While CAM6-free does decrease for this same temperature range, the drop is much weaker and only one order of magnitude. In general, CAM6 is able to represent probabilities with respect to temperature adequately. In regard to probabilities of in-cloud and clear sky conditions with respect to RH_i (Figure 18 d – f), there are significant differences between observations and the model. Observed in-cloud RH_i is most probable at 100% with a large decrease in probabilities with

increasing ISS. While observed in-cloud probabilities of RHi increase monotonically from 50% to 100%, both model simulations exhibit a peak around 80%, and then decrease slightly with increasing RHi, before finally spiking up at 100%. Similar to observations, CAM6-nudg probabilities of in-cloud RHi decrease rapidly with increasing ISS. However, CAM6-free in-cloud decreases much more slowly with increasing ISS and reach up to ~200%. The probability of clear sky RHi is highest at very low RHi (< 5%) with a general decrease in probability with increasing RHi, which is also captured well in both model simulations.

Probabilities of w (Figure 18 g – i) are highest for weak vertical motions and decrease with increasing updraft and downdraft strength. When observed w is very weak (~ 0 m/s), clear sky probabilities are highest, but are generally lower than in-cloud probabilities for stronger vertical motions. Additionally, the in-cloud probabilities are slightly higher for updrafts than downdrafts. As mentioned in the previous section, the model is unable to simulate downdrafts for both in-cloud and clear sky conditions. In addition, the model significantly underestimates the strength of updrafts with maximum w for CAM6-nudg and CAM6-free only reaching 1 m/s and ~ 2 m/s, respectively.

Figure 18. Probability density functions (PDFs) for cirrus cloud controlling factors for observed (left column), CAM6-nudg (middle column) and CAM6-free (right column). Controlling factors include temperature (a – c), RHi (d – f), w (g – i), Na_{500} (j – l) and Na_{100} (m – o).

PDFs of Na_{500} and Na_{100} are shown by Figure 18 j – l and m – o, respectively. Observed PDFs of Na_{500} indicate the highest probability occurs for in-cloud condition. In contrast, model PDFs of Na_{500} , show higher probabilities of in-cloud for low Na_{500} ($< 1 \text{ \#/cm}^3$), but overall the highest probabilities are for higher Na_{500} . In regard to observed PDFs of Na_{100} , the probabilities are highest for moderate values of Na_{100} ($10 - 100 \text{ \#/cm}^3$) and does not show an indication of higher probabilities for in-cloud or clear sky conditions. Similar to PDFs of Na_{500} , the highest probability for in-cloud condition occurs for higher Na_{100} ($> 10 \text{ \#/cm}^3$). A significant difference between observations and the model is the model underestimates total aerosol concentrations. The highest Na_{500} and Na_{100} values are 10 and $100 \text{ \#/cm}^3$ from the model, almost two orders of magnitude lower than the highest values from observations. Although the model simulates the highest probabilities for in-cloud condition, it still fails to correctly simulate the correct Na distribution.

Thermodynamic and Dynamic Impacts

The relationship between ice microphysical properties and RH_i is examined in Figure 19. For in situ observations (Figure 19 a – c), higher IWC and values N_i ($> 0.01 \text{ g m}^{-3}$ and $1 \text{ \# L}^{-1}$, respectively) are found under slight ISS conditions ($\sim 100\%$). As RH_i increases from sub-saturated to saturated conditions, ice microphysical properties follow a positive trend with IWC and N_i increasing for 1 – 2 orders of magnitude from 80% to 100% RH_i. However, the highest IWC and N_i do not peak at the highest RH_i, which is likely due to the depletion of water vapor with new ice nucleation and growth. D_i shows a peak at slightly higher ISS ($\sim 140\%$) than IWC and N_i . The high values of D_i at very

low RH_i (< 40%) are likely a result of sedimenting large ice crystals. This phenomenon has been previously observed by (Diao et al., 2013b), who investigated the evolutionary phases of cirrus clouds. Comparing in situ observations to CAM6 free-running (Figure 19 d – f) and nudged (Figure 19 g – i) indicates some significant differences. Both CAM6 simulations produce a smaller peak in IWC and Ni near ~100% RH_i in agreement with observed IWC and Ni. However, maximum values of IWC and Ni are produced at a higher peak almost 20% lower. This bimodal distribution follows closely with the RH_i distribution shown in Figure 18 and may be the reason for the two peaks in cirrus microphysical properties. Additionally, simulated IWC and Ni do not steadily decline at high ISS (> 120%) but rather show an increase between 110% – 130% and 120% – 140% for CAM6-free and CAM6-nudg, respectively. At extremely high ISS (RH_i > 160%), CAM6-free simulated IWC and Ni remain almost the same with a slight decrease in IWC. CAM6-nudg and CAM6-free produce maximum values of Di occurring at slightly higher RH_i ~140%, which is similar to observed Di. The model is also able to simulate sedimenting high Di values at low RH_i (< 40%). The large variability of observed ice microphysical properties is also significantly underestimated in the model for ISS conditions. Standard deviations are 0.5 – 1 order of magnitude lower for IWC and Ni compared with observations. This illustrates an important flaw in how the model accounts for ice crystal variability under ISS conditions.

Figure 19. Occurrences of in-cloud IWC (left column), Ni (middle column) and Di (right column) for binned intervals of RHi from in situ observations (top row), CAM6 nudged (middle row) free-running (bottom row) simulations. Black line and whiskers denote mean and standard deviation.

The effect of w is similarly examined in Figure 20. In agreement with observations, both CAM6-free and CAM6-nudg show maximum IWC and Ni values occurring for strong updrafts. Although CAM6 underestimates the strength of w in the model, comparing IWC values at the highest simulated w (~ 1.5 m/s), all are within 0.05 g m^{-3} . Despite the agreement in simulated IWC and Ni and observations, the relationship between Di and w indicates some critical differences. For both CAM6 simulations,

increasing w results in a decrease in D_i , while the observed D_i increases with w . The positive correlation between D_i and w in observations is likely due to the competition between both homogeneous and heterogeneous nucleation combined with significant fluctuations in vertical velocity and water vapor. This competition was tested by Zhou et al. (2016) and found N_i magnitudes much closer to observations. Observed maximum IWC, N_i and D_i are found with the highest updrafts (> 2.5 m/s), while minimum respective values occur under weak downdrafts ($w = -0.5$ m/s). Surprisingly, higher values of IWC and N_i occur under strong downdrafts ($w < 3$ m/s), however the highest respective values still occur for updrafts. As mentioned previously, downdrafts and updrafts occur frequently in close proximity, therefore updrafts are likely responsible for the elevated IWC and N_i , before encountering strong downdrafts.

Figure 20. Similar to Figure 19 but for binned intervals of vertical velocity.

Aerosol Indirect Effects

The effects of Na_{500} on ice microphysical properties is further examined for observations, CAM6-free and CAM6-nudg (Figure 21). For all observed cirrus microphysical properties (left column) there is a positive correlation with increasing Na_{500} . These positive correlations are apparent for CAM6-nudg IWC and Ni but show differences for Di and CF. CAM6-nudg Di remains relatively neutral for all but very high Na_{500} ($> 1 \text{ \#/cm}^3$) before decreasing and CF shows no apparent correlation with Na_{500} . A

negative correlation between Di and Na has been observed by a few recent satellite studies (Zhao et al., 2018, 2019), however they only included samples in East Asia and also underscored the importance of contributions from other meteorological parameters. Contrary to observations, CAM6-free (right column) shows significant differences. For example, IWC and Di remain neutral and even decrease for very high Na_{500} ($> 1 \text{ \#/cm}^3$). However, for the same range in Na_{500} , Ni increases. Finally, a significant difference between observations and the model is observed cirrus microphysics are nucleating at significantly higher Na_{500} . The highest simulated ice property occurs at Na_{500} equal to $10 \text{ \#/cm}^3$ compared to nearly $\sim 300 \text{ \#/cm}^3$. Based on these two simulations, it is apparent the CAM6-nudg simulations is able to better resemble the aerosol indirect effects compared to observations. This is likely a result of a more accurate representation of the meteorological conditions and demonstrates the need for such representation to simulate the right sign of the aerosol indirect effect in CAM6.

Figure 21. Aerosol indirect effects on in situ observed (left column), CAM6 nudged (middle column) and free running (right column) IWC, Ni, Di, cloud fraction (CF) and number of samples. Na is restricted to aerosols > 500 nm. Both in situ observations and CAM6 simulated ice properties are restricted to ice crystals > 62.5 μm . Cloud fraction is calculated by taking the ratio of number of in-cloud samples to the total number of samples for a given temperature and Na bin.

Figure 22. Similar to Figure 21 but for CAM6-HOM (left column), CAM6-HET (middle column) and CAM6-PRE (right column).

Similarly, the effects of Na_{500} on cirrus microphysical properties are further examined for three CAM6 free-running experimental simulations (Figure 22). Surprisingly, the aerosol indirect effects for all three experiments produces very similar results. Some key differences can be seen in simulated Ni and Di. For example, CAM6-HOM produces the smallest Di ($\sim 100 \mu\text{m}$) and highest Ni ($> 10 \text{ \# L}^{-1}$), especially for $T < -50^\circ\text{C}$ and lower Na_{500} ($< .01 \text{ \#/cm}^3$). Both CAM6-HET and CAM6-PRE produce lower Ni ($< 10 \text{ \# L}^{-1}$)

and higher Di ($> 150 \mu m$) for the same temperature and Na_{500} range. Simulated IWC remains the same for all three experiments with some higher values at colder temperatures for CAM6-HET and CAM6-PRE. Additionally, simulated CF (Figure 22 bottom row) shows no significant differences between experiments. For all CAM6 experiments, increasing Na_{500} is shown to increase Ni and decrease Di following the conventional Twomey effect (Twomey, 1977). However, this contradicts observations which observed positive correlations in both Ni and Di with Na_{500} . These results indicate the aerosol indirect effect in CAM6 may be less dependent upon the nucleation mechanism and more on the meteorological conditions.

Finally, the aerosol indirect effects with respect to Na_{100} are examined in Figures 23 and 24. Observed aerosol indirect effects with respect to Na_{100} (Figure 23 left column) again produce positive correlations for all cirrus microphysical properties. Aerosol indirect effects with respect to Na_{100} for CAM6 simulations look very similar to results for Na_{500} . Generally, CAM6-nudg shows a positive correlation with Di being the only exception, which varies little with changing Na_{100} . Similarly, experimental simulations indicate the same aerosol indirect effects regardless of the size of aerosols.

Figure 23. Similar to Figure 21 but with respect to Na_{100} .

Figure 24. Similar to Figure 22 but with respect to Na_{100} .

Discussion and Conclusions

In this study, we investigate the regional distributions and impacts of cirrus cloud controlling factors (i.e., t , RHi and w) and aerosol indirect effects and evaluate NCAR CAM6 using a comprehensive in situ observational dataset. The observations were collected from seven NSF flight campaigns over the Pacific and Southern Oceans, North and South America and the Caribbean. We examine the regional distributions of cirrus

cloud meteorological controlling factors by separating 1-Hz observations and CAM6 simulated output into six latitudinal locations. Two types of CAM6 simulations are tested including nudged and collocated and free-running. Finally, three free-running experiments are tested by turning off homogeneous (CAM6-HET) and heterogeneous (CAM6-HOM) nucleation and the inclusion of pre-existing ice (CAM6-PRE).

In regard to the regional effects of temperature on cirrus cloud microphysical properties, observations indicate that both IWC and D_i are maximized at warmer temperatures regardless of geographical location; however, N_i remains neutral for all temperatures. In the NH and warmer temperatures, IWC and N_i follow a clear regional trend with the highest values occurring in the midlatitudes followed by the tropics then polar regions. However, in the SH, the highest IWC and N_i values occur in the tropics. Model IWC and D_i are also underestimated for tropical and polar regions in both hemispheres and just the NH for the midlatitudes. Observed D_i shows no effect by the geographical location. In contrast, CAM6-nugd shows a clear geographical effect, with the highest IWC and N_i occurring in the tropical, followed by the midlatitudes and poles regardless of the hemisphere. The model is able to resemble N_i well compared to observations in the polar and tropical regions. However, it is underestimated and overestimated in the NH and SH, respectively. These regional and hemispheric differences between observed and simulated cirrus cloud microphysical properties illustrate a need for geographical considerations in the model.

For the regional distributions of RH_i from observations we found in-cloud conditions RH_i occur most frequently at or near 100% regardless of the geographical region. Clear

sky distributions of RH_i occur most frequently below saturation but can reach up to the liquid saturation line. This is consistent with the findings of Krämer et al. (2009), but we expanded on their work by analyzing the hemispheric and regional distributions. Clear sky conditions exhibit no difference in RH_i depending on their geographical location. However, in-cloud conditions indicate significantly less variability in RH_i in the poles compared to the tropics and midlatitudes. Our CAM6-nudg simulation is able to successfully resemble the RH_i distributions based on the observations. This illustrates a major improvement to the model's representation of water vapor as previously Wu et al. (2017) described flaws in the model's ability to accurately capture large-scale variability in RH_i. That study also used a larger model resolution and it is unclear whether higher resolution is the sole reason for improved RH_i representation in this case.

Observed regional distributions of w are found to occur most frequently under weak vertical motions. In addition, the difference between regions and in-cloud and clear sky conditions appears to be negligible. CAM6-nudg is found to underestimate vertical motions and only account for updrafts when in-cloud, which is likely a result of how sub-grid velocity is calculated. The model also allows for significantly stronger vertical motions in-cloud with 2 – 3 times stronger updrafts in-cloud compared to clear sky, but overall much weaker than observations. Allowance of stronger sub-grid vertical velocities in both vertical directions in the model would lead to significant improvements more comparable to observations.

The impact of RH_i on cirrus cloud microphysical properties is found to maximize IWC, N_i and D_i values slightly above saturation, while maximum D_i values occur at

slightly higher ISS $\sim 140\%$. In contrast, CAM6-free and CAM6-nudg shows a bimodal distribution with a peak of IWC and Ni values below saturation $\sim 80\%$ and a lower peak at 100%. In addition to the bimodal distribution, the model simulations both dramatically underestimate the peak IWC and overall variability in IWC for all RH_i. The model also does not simulate the correct decrease in IWC or Ni for ISS seen in observations. For both simulations, Di is comparable to observations with the highest Di values occurring at higher ISS $\sim 140\%$ but produce significantly less variability. These results illustrate how the model is unable to resolve the impacts of RH_i on cirrus microphysical properties.

Regarding the dynamic on impact cirrus cloud microphysical properties, the highest values of IWC, Ni and Di are found for strongest updrafts and downdrafts, however they are slightly larger for updrafts. Minimum IWC and Ni are observed under weak downdrafts. The model is able to simulate the highest IWC and Ni values for strongest updrafts in both simulations, agreeing well with observations. However, peak updrafts only reach up to 1 and 2 m/s for CAM6-nudg and CAM6-free, respectively. Simulated Di shows highest values for the weakest vertical motions before decreasing with increasing w . The decrease in ice crystal size with increasing vertical motions agrees with the conventional knowledge, where stronger updrafts increase cooling rates and are more likely to freeze homogeneously. However, Shi and Liu, (2016) showed that GCMs generally overestimate the frequency of homogeneous nucleation due to lack in variability in vertical velocity. In an idealized setting, with mass-conserved water vapor, there would be a negative correlation between Di and w . However, observations include a wide range of w and water vapor allowing for high Ni and Di. Although the model

produces IWC and Ni comparable with observations, significant improvements are still needed on the relationship between ice microphysical properties and w and for a more realistic distribution of w that allows for downdrafts in the model.

Aerosol indirect effects on cirrus cloud microphysical properties with respect to Na_{500} are found to have differing results depending on whether it was observed and the type of CAM6 simulation. Observed aerosol indirect effects indicate a positive correlation between Na_{500} and all microphysical properties. However, positive correlations are only found for IWC and Ni in CAM6-nudg simulations. CAM6-free produces a neutral or negative correlation for IWC and Di, respectively and only produces a positive correlation for CF at extremely high Na_{500} . High CF in CAM6-nudg at low Na_{500} are likely a result of very low samples. Since Na_{500} can act as a good indicator to the number of INPs for heterogeneous freezing (DeMott et al., 2010), these relationships can describe whether increased Na_{500} would lead to an increase in ice crystals forming heterogeneously. Regarding the aerosol indirect effect for CAM6-free experiments, we found positive and negative correlations between Na_{500} and Ni and Di, respectively. The differences in aerosol indirect effects between experiments is negligible, but CAM6-HET and CAM6-PRE do produce lower Ni and higher Di than CAM-HOM at colder temperatures and lower Na_{500} . This is an expected result for CAM6-HET but is a more surprising result for CAM6-PRE as the addition of pre-existing ice was found to reduce the occurrence of homogeneous nucleation (Shi et al., 2015), thus by eliminating this effect should produce higher Ni and lower Di. Similarly, the observed aerosol indirect effects with respect to Na_{100} are found to produce correlations on all cirrus microphysical

properties. CAM6-nudg is found to produce results most in agreement with observations. CAM6-free simulations including the three nucleation experiments produce very similar results as Na₅₀₀. Although nudged and collocated simulations produce better results compared to observations, free-running types was chosen for CAM6 experiments because nudged simulations were found to produce too few samples to distinguish a noticeable difference.

This global-scale observational dataset provides a statistically robust distribution of cirrus cloud microphysical properties that has been used to evaluate the spatial distributions and impacts of thermodynamics, dynamics and aerosols in a global climate model. Expanding on previous studies who have investigated climate model sensitivity in regard to a single cirrus cloud controlling factors, i.e., w (Shi and Liu, 2016), water vapor (D'Alessandro et al., 2019; Tan et al., 2016; Wu et al., 2017), and aerosols (Wang et al., 2014), this study provides a robust analysis of all factors. In addition, further attention was given towards understanding how the model treats these factors based on geographical location. This study underscores the importance of not only correctly representing the thermodynamics, dynamics and aerosol distributions in climate model parameterizations, but also that models must account for the geographical location of cirrus clouds. Failing to do so may result in an under- or overestimation of cirrus cloud microphysical characteristics depending on the region.

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