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Abstract

Cloud phase and relative humidity (RH) distributions at -67° – 0°C over the Southern Ocean during austral summer are compared between in situ airborne observations and global climate simulations. A scale-aware comparison is conducted using horizontally averaged observations from 0.1–50 km. Cloud phase frequencies, RH distributions and liquid mass fraction are found less affected by horizontal resolutions than liquid and ice water content (LWC, IWC), liquid and ice number concentrations (N_{liq} , N_{ice}), and ice supersaturation (ISS) frequency. At -10° – 0°C , observations show 27%–34% and 17%–37% of liquid and mixed phases, while simulations show 60%–70% and 3%–4%, respectively. Simulations overestimate (underestimate) LWC and N_{liq} in liquid (mixed) phase, overestimate N_{ice} in mixed phase, underestimate IWC in ice and mixed phases, and underestimate (overestimate) liquid mass fraction below (above) -5°C , indicating that observational constraints are needed for different cloud phases. RH frequently occurs at liquid saturation in liquid and mixed phases for all datasets, yet the observed RH in ice phase can deviate from liquid saturation by up to 20%–40% at -20° – 0°C , indicating that the model assumption of liquid saturation for coexisting ice and liquid is inaccurate for low liquid mass fractions (<0.1). Simulations lack RH variability for partial cloud fractions (0.1–0.9) and underestimate (overestimate) ISS frequency for cloud fraction <0.1 (≥ 0.6), implying that improving RH subgrid-scale parameterizations may be a viable path to account for small-scale processes that affect RH and cloud phase heterogeneities. Two sets of simulations (i.e., nudged, collocated and free-running) show very similar results (except for ISS frequency) regardless of sample sizes, corroborating the statistical robustness of the model-observation comparisons.

1. Introduction

Clouds over the Southern Ocean are an essential component controlling the energy budget over the region and have an annual mean spatial fraction around 80%–90% (e.g., Kay et al. 2012; McCoy et al. 2014a; Matus and L’Ecuyer 2017). Climate models show large deficiencies in simulating radiative fluxes in the Southern Ocean region ($\sim 50^{\circ}$ – 80° S), and often underestimate reflected shortwave radiation on the order of 10 W m^{-2} (e.g., Bodas-Salcedo et al. 2014; Li et al. 2013; Kay et al. 2012). This is in part due to the fact that climate models (e.g., Trenberth and Fasullo 2010; Kay et al. 2016a; Bodas-Salcedo et al. 2016; Kay et al. 2016b; Cesana and Chepfer 2013; Wang et al. 2018) as well as higher-resolution regional models (e.g., Huang et al. 2014, 2015) generally show lower cloud fraction and less supercooled liquid water (SLW, i.e., liquid water existing at temperatures below 0°C) than the observations in the mid- and high southern latitudes. The amount of SLW plays a critical role in determining cloud radiative forcing (e.g., Ceppi et al. 2014; Lawson and Gettelman 2014; Shupe and Intrieri 2004), cloud feedbacks (e.g., Gettelman and Sherwood 2016; Tsushima et al. 2006; McCoy et al. 2014b), and equilibrium climate sensitivity (e.g., Tan et al. 2016; Frey and Kay 2017). The fundamental mechanism of a negative cloud phase feedback due to ice to liquid transition under surface heating was proposed by Mitchell et al. (1989). Additional considerations need to be taken for mixed phase clouds (MPCs) by characterizing the mass fractions of ice and liquid phases as well as their degree of mixing, which can both substantially alter the radiation budget (Sun and Shine 1994).

Commonly referred to as the Wegener-Bergeron-Findeison (WBF) process, when ice particles and SLW droplets coexist, ice particles grow at the expense of neighboring supercooled liquid droplets, when the equilibrium water vapor partial pressure (e) is less than the saturation vapor pressure with respect to liquid (e_{slq}) and greater than the saturation vapor pressure with

respect to ice (e_{Sice}) (Wegener 1911; Bergeron 1928, 1935, Findeisen 1938, 1940). Ambient relative humidity (RH) has large impacts on cloud phase transition and evolution, therefore quantifying RH conditions becomes a key step for understanding the existence of SLW. Generally speaking, when vapor diffusion effectively equilibrates the vapor pressure between liquid and ice phases, the local RH reaches a steady state around liquid saturation. This feature is frequently shown by in situ observations on the horizontal scales of ~ 100 m (Korolev and Isaac 2006) and cloud-resolving model simulations (Fan et al. 2011). Such assumption of liquid saturation for in-cloud RH when ice and liquid coexist is also used in the parameterization of mixed phase stratiform clouds (Rotsteyn et al. 2000), which has been adopted in several global climate models (GCMs), including the Community Atmosphere Model version 5 (CAM5) (Gettelman et al. 2010; Gettelman and Morrison 2015; Morrison and Gettelman 2008), the European Centre for Medium-Range Weather Forecasts (ECMWF) model (Forbes and Ahlgrimm 2014), and the atmospheric component of the Geophysical Fluid Dynamics Laboratory (GFDL) coupled GCMs – CM2 (Anderson et al. 2004) and CM3 (Donner et al. 2011). Previously, Fu et al. (2004) compared the in-cloud water vapor parameterizations with 1-s in-situ observations in the Arctic. However, the RH conditions as well as other cloud microphysical properties in MPCs have not been examined and compared for various scales of observations from sub-kilometer to tens and hundreds of kilometers.

Validating simulated cloud properties and processes within climate models is crucial as many uncertainties still remain in predicting cloud feedbacks on climatic time scales (Gettelman and Sherwood 2016). Adjusting the parameterized phase partitioning of cloud liquid and ice can either increase or decrease the climate sensitivity parameter when introducing surface temperature perturbations (Li and Le Treut 1992). Tan et al. (2016) constrained the mean state of phase

partitioning in the Community Earth System Model (CESM, of which CAM is a component) simulations using satellite observations and found an increase in equilibrium climate sensitivity by up to 1.3 K with a doubling of CO₂, resulting from a weakened negative cloud phase feedback. Frey and Kay (2017) also found a 1.5 K increase in equilibrium climate sensitivity, partly due to reduced negative cloud phase feedback over the Southern Ocean when decreasing the prescribed cloud ice fraction in the total water detrained from shallow convection (Kay et al. 2016a). McCoy et al. (2014b) showed increasing upwelling shortwave radiation due to liquid replacing ice and increasing optical depths as a result of 1 K tropospheric warming. Climate models were also reported to have difficulties capturing low-level clouds in the cold sector of midlatitude cyclones, potentially due to the prevalence of boundary layer clouds being commonly associated with large-scale subsidence in these regions (Govekar et al. 2014; Bodas-Salcedo et al. 2014), which highlights the importance of local processes relative to large scale processes, e.g., cyclonic activity.

Previously, due to the scarcity of in situ observations over the Southern Ocean, only a few studies analyzed in situ observations (e.g., Huang et al. 2015; Ahn et al. 2017; Chubb et al. 2013; Huang et al. 2017; Ovarlez et al. 2002; Morrison et al. 2010; Jensen et al. 2000) and none of them was directly compared with GCM simulations. In fact, most studies relied on spaceborne observations to analyze cloud microphysical properties (e.g., Bodas-Salcedo et al. 2012, 2014, 2016; McCoy et al. 2016; Cesana et al. 2015; Hu et al. 2010; Morrison et al. 2011a; Huang et al. 2012a,b; Matus and L'Ecuyer 2017) and RH distributions (Lamquin et al. 2012; Gettelman et al. 2006; Kahn et al. 2009; Spichtinger et al. 2003) over this remote region. Although the satellite data provide useful information for climatological purposes, their phase determination often includes a large amount of undefined-phase clouds, and various active and passive detection approaches have large differences in their sampling, accuracy, sensitivity, and assumptions (Nasiri and Kahn 2008;

Kahn et al. 2011; Cho et al. 2009; Hu et al. 2009; Chylek et al. 2006; Naud et al. 2006; Riedi et al. 2010). For example, the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) cloud phase identification is mostly affected by the cloud top (Cesana et al. 2016); the Moderate Resolution Imaging Spectroradiometer (MODIS) cloud-top phase product has limited accuracy from -25° to -5°C (Morrison et al. 2011a); the Polarization and Directionality of Earth Reflectances (POLDER) spaceborne instrument uses visible and near-infrared channels, and has difficulties detecting very thin clouds and cloud edges as well as defining phases for multi-layer clouds (Goloub et al. 2000; Riedi et al. 2001). In addition, it is difficult for satellites to retrieve high-resolution RH distributions relevant to cloud-scale microphysical processes. For example, the NASA Atmospheric Infrared Sounder (AIRS)/Advanced Microwave Sounding Unit (AMSU) temperature and water vapor retrievals have vertical and horizontal resolutions of 1–3 km and 45×45 km, respectively, and have differences of $\sim 1\text{--}2.5$ K in temperature and $\sim 20\%\text{--}60\%$ in water vapor compared with in situ observations (Diao et al. 2013). In contrast, 1-Hz in situ measurements obtained from research aircraft have horizontal resolutions ranging from 100–250 m. Previously, in situ measurements of RH and cirrus cloud microphysical properties were analyzed for temperatures $< -40^{\circ}\text{C}$ over Punta Arenas, Chile (Ovarlez et al. 2002; Gayet et al. 2006), yet a comprehensive analysis for $-40^{\circ}\text{--}0^{\circ}\text{C}$ is still missing.

The purpose of this study is to examine the distributions of cloud phases and RH during austral summer over the Southern Ocean based on in situ observations, and compare with CAM5 simulations. Observations from the National Science Foundation (NSF) O_2/N_2 Ratio and CO_2 Airborne Southern Ocean Study (ORCAS) (Stephens et al. 2018) took place during the austral summer, a time at which a deep, circumpolar storm track often occurs (Hoskins and Hodges 2005). Effects of horizontal spatial resolution on the analyses are examined by spatially averaging

observations from sub-kilometer to tens of kilometers. A series of model-observation comparisons have been conducted, including cloud phase distributions at various temperatures, cloud microphysical properties (i.e., liquid and ice water content (LWC and IWC), liquid and ice number concentration (N_{Clq} and N_{Ice})) for three cloud phases, and RH frequency distributions. The prerequisite condition of ice particle formation – ice supersaturation ($\text{ISS}=\text{RH}_i-100\%$) – has also been compared between observations and simulations from -67° to 0°C . These comparisons will help to improve the representations of cloud thermodynamic phases and to reduce bias of the amount of SLW over the Southern Ocean in GCM simulations.

2. Dataset and experimental set-up

2.1 In situ observations

This study uses 1-Hz airborne measurements from the NSF Gulfstream-V (GV) research aircraft during the ORCAS campaign (Stephens 2017). ORCAS took place from January 15 – February 28, 2016, sampling over the Southern Ocean from 30°S to 75°S and 50°W to 92°W . The ORCAS campaign conducted 19 research flights, and 18 of them reported final data with ~95 hours of flight time in total. Details on flight objectives and analyses can be found in Stephens et al. (2018). Observations in this study are restricted to temperatures $<0^\circ\text{C}$ to exclude warm cloud measurements (i.e., clouds with no ice or SLW). The total flight time at -40° – 0°C is 40 hours. In-cloud sampling time for -10° – 0°C , -20° – -10°C , -30° – -20°C , -40° – -30°C and $<-40^\circ\text{C}$ is 3.2, 1.2, 1.2, 0.73, and 1.3 hours, respectively. The flights in the ORCAS campaign often sampled cyclones and frontal systems associated with strong westerly flow around the Drake Passage and nearby regions. These synoptic scale conditions coupled with cool ocean surface led to frequent cloud cover over the ORCAS flight domain, including many cases of low-level and mid-level stratus and

stratocumulus. Multi-layer stratus and single-layer stratocumulus have been frequently observed in several flights.

Temperature was measured using a Rosemount temperature probe, having an accuracy and precision of ± 0.3 K and 0.01 K, respectively. Water vapor measurements are taken from the 25-Hz Vertical Cavity Surface Emitting Laser (VCSEL) hygrometer (Zondlo et al. 2010), which has an accuracy and precision of $\sim 6\%$ and $\leq 1\%$, respectively. Final data of water vapor mixing ratio and temperature were reported at 1 Hz. Two sets of RH data are calculated, i.e., RH with respect to liquid (RH_{liq}) and ice (RH_i), based on Murphy and Koop (2005) that derived e_{Sliq} and e_{Sice} from a synthesis of laboratory results. For temperatures relevant in this study at -69°C to 0°C , the uncertainties in RH_i range from 7.5%–6.5%, and the uncertainties in RH_{liq} range from 10.4%–6.4%, respectively. Due to combined uncertainties from water vapor and temperature measurements, the observed RH_{liq} greater than liquid saturation is set to be equal to 100.01% (processed for 7745 s of observations). Such restriction does not alter the number of occurrences for liquid or ice supersaturation.

Cloud particle measurements were made by the Fast 2-Dimensional Optical Array Cloud probe (Fast-2DC) and the Cloud Droplet Probe (CDP). The CDP measures particles ranging from 2–50 μm . The Fast-2DC detects particles from 62.5–1600 μm in diameter (excluding the first two bins), and larger particles can be mathematically reconstructed up to 3200 μm . The mass concentrations for Fast-2DC are calculated based on Brown and Francis (1995). We define in-cloud conditions as having either of these two conditions: (1) CDP measurements report both number concentration $> 0.03 \text{ cm}^{-3}$ and derived mass concentration $> 3.98 \times 10^{-4} \text{ g m}^{-3}$; (2) Fast-2DC measurements detect at least one particle in a second, and its minimum derived mass concentration was $4.68 \times 10^{-5} \text{ g m}^{-3}$ in the ORCAS campaign. The rest of the measurements are defined as clear-

sky conditions. The CDP threshold was chosen in order to minimize the impacts of giant aerosols, which were determined by the relationships between number and mass concentrations (Figure 1).

2.2 Global climate model simulations

The model used here is CAM5, which is the atmospheric component of NCAR CESM. A detailed description of CAM5 can be found in Neale et al. (2012). Of particular relevance to this study are the representation of cloud macrophysics (Park et al. 2014), cloud microphysics (Morrison and Gettelman 2008) and shallow convection (Park and Bretherton 2009). Cloud microphysics is coupled with a modal aerosol module (MAM) (Liu et al. 2012) for aerosol–cloud interactions. The version of MAM with three modes (MAM3) is adopted here. Cloud droplets can form via the activation of aerosols (Abdul-Razzak and Ghan 2000). Ice crystals can form via the homogeneous nucleation of sulfate aerosol and/or heterogeneous nucleation of dust aerosol (Liu and Penner 2005; Liu et al. 2007). Hallet-Mossop ice multiplication (secondary ice production) due to accretion of drops by snow is included following Cotton et al. (1986).

Results from in situ observations are compared with two types of CAM5 simulations: the nudged simulation and the free-running simulation. Both simulations were run with a finite volume dynamical core (Lin 2004), a horizontal spatial resolution of $0.47^{\circ} \times 0.63^{\circ}$, a pressure top of 30 hPa and a timestep of 1800 s. The number of vertical levels for nudged and free-running simulations is 56 and 30, respectively. The nudged CAM5 simulation (hereafter named as “CAM-collocated”) was forced to represent meteorological conditions (3-D temperature and wind speed) according to the NASA Goddard Earth Observing System Model version 5 (GEOS-5) data (Lamarque et al. 2012), and the output were saved along the aircraft flight track. The detailed nudging method is described in Wu et al. (2017) and Zhang et al. (2014). The free-running CAM5 simulation (hereafter named as “CAM-domain”) was run with prescribed climatological sea surface

temperature and sea ice extent. The simulation covered the duration of the ORCAS campaign with a 1-year spin-up time and 6-hourly output (00, 06, 12 and 18 UTC). Outputs with the nearest time to the mid-point time stamp of each research flight are selected, which forms a composite CAM-domain data with 18 model outputs, restricted to the horizontal regions (50°W to 92°W and 30°S to 75°S) and altitudes (pressure > 176.38 hPa) comparable to the flight domain.

For analyses of cloud microphysical properties, the model output variables used are “LWC”, “IWC”, “NUMLIQ” ($N_{\text{C}_{\text{liq}}}$) and “NUMICE” ($N_{\text{C}_{\text{ice}}}$), all of which are grid box average quantities. When defining in-cloud conditions, either the value of LWC or IWC being greater than $4.68 \times 10^{-5} \text{ g m}^{-3}$ is considered to be in-cloud condition. The rest is defined as clear-sky condition, with IWC and LWC in clear-sky conditions set to be zero. Such definition is consistent with the minimum value of mass concentration detected by in situ observations. The calculation of RH_i and RH_{liq} is based on Goff and Gratch (1946) in the CAM5 simulations. Since the observations calculate saturation vapor pressures based on equations from Murphy and Koop (2005), both RH_i and RH_{liq} values have differences within $\pm 0.3\%$ compared with those calculated from Goff and Gratch (1946).

2.3 Spatial averaging of observations

One of the main challenges for comparing in situ observations and global climate model simulations is their differences in spatial resolution. To address this inherent problem, 1-Hz airborne observations were averaged over various time intervals, i.e., 10 s, 25 s, 50 s, 100 s and 200 s (named as Obs-Xs). The GV aircraft true air speed ranges from ~100 to 250 m s⁻¹ from near surface to the upper troposphere and lower stratosphere (UT/LS), respectively. Thus, spatially averaged observations at 10–200 s intervals have horizontal resolutions of ~1–20 km near surface and ~2.5–50 km at UT/LS (Table S1). Such averaging mostly stands for horizontal averaging since

aircraft true air speed is one to two orders of magnitude higher in the horizontal direction than in the vertical direction. No spatial averaging was applied to CAM-collocated or CAM-domain data, and they have horizontal resolutions of 14–70 km from higher to lower latitudes (75°S–30°S), comparable to the Obs-100s and Obs-200s data at 10–50 km resolutions, respectively.

Cloud fraction for observations is calculated as the number of 1-Hz in-cloud data divided by the total number of 1-Hz data during the averaging time interval (Δtime). Temperature, RH_i, RH_{liq}, LWC, IWC, N_{cliq} and N_{ci} are averaged over the entire time interval, which correspond well with the grid-average variables from CAM5. A “moving average” method is applied for each 1-Hz sample by averaging between $\pm N$ seconds or between $-N$ and $N+1$ seconds, where N equals $(\Delta\text{time}-1)/2$ or $\Delta\text{time}/2$ for odd or even Δtime values, respectively. If more than 10% of a time interval reports missing data, that sample would be discarded.

3. Results

3.1 Definition of three cloud phases

Cloud phase is defined by the mass fraction of LWC with respect to cloud water content (CWC, i.e., $\text{CWC} = \text{IWC} + \text{LWC}$), similar to the definition used in Korolev et al. (2003). A given cloud sample is considered ice phase at $\text{LWC}/\text{CWC} \leq 0.1$, mixed phase at $0.1 < \text{LWC}/\text{CWC} < 0.9$ and liquid phase at $\text{LWC}/\text{CWC} \geq 0.9$. This definition is applied to both observations and simulations.

Figure 1 a shows a diagram of the cloud phase identification method. All cloud hydrometeors below -40°C are considered ice. LWC and IWC are derived from two cloud probes by combining their ice and liquid measurements, respectively. After removing large aerosols (Section I in Figure 1 a), samples detected by CDP are considered liquid if number concentration ($N_{\text{CDP}} > 10^{-0.5} \text{ cm}^{-3}$ (i.e., 0.316 cm^{-3}), otherwise they are considered ice. At -30°–0°C, 2DC

measurements are considered ice when the maximum diameter ($D_{\text{max_2DC}}$) $> 312.5 \mu\text{m}$ within a 1-Hz measurement by 64 diodes. They are considered liquid for $D_{\text{max_2DC}} < 112.5 \mu\text{m}$. For $112.5 \mu\text{m} \leq D_{\text{max_2DC}} \leq 312.5 \mu\text{m}$, the 2DC sample is considered liquid when the 1-Hz standard deviation of particle diameters (σ_{D_2DC}) $\leq 50 \mu\text{m}$, while the rest of the samples at this $D_{\text{max_2DC}}$ range are considered ice. At -40° to -30°C , a 2DC sample can only be identified as liquid if the concurrent CDP measurement is liquid, as well as the aforementioned criteria for liquid phase are satisfied. Otherwise, that 2DC sample is identified as ice. These key thresholds are illustrated in the relationships of number versus mass concentrations (Figure 1 b and c). The fundamental concept of using $D_{\text{max_2DC}}$ and σ_{D_2DC} is similar to the method in McFarquhar et al. (2007), but the thresholds have been further adjusted for the instrumentation on GV aircraft, and verified by examining the 2DC imagery and comparing with measurements from the King and RICE probes. In addition, a slightly different cloud phase identification method without the temperature threshold of -30°C for the 2DC probe was used to analyze an earlier version (April 2018) of the ORCAS data (D'Alessandro 2018). Sensitivity tests show differences mostly within 10% for cloud phase occurrence frequencies, when examining various thresholds of $D_{\text{max_2DC}}$, σ_{D_2DC} , N_{CCDP} , etc. (supplementary Figure S1)

Time series in Research Flight (RF) 17 and 03 illustrate two typical model-observation discrepancies, that is, simulations misidentifying liquid and mixed phases as ice phase (Figure 2) and misidentifying ice phase as liquid and mixed phases (Figure 3). Cloud phases are highlighted by colored markers below the abscissa (d and e). Exemplary Fast-2DC images show good agreement with the cloud phases being identified (g). In addition, when SLW droplets are identified, the King probe often reports $\text{LWC} > 0.1 \text{ g m}^{-3}$ (d). The King probe infers LWC by the power required to evaporate cloud droplets after their impact with the hotwire (King et al. 1978),

which may involve biases in LWC when sampling droplets larger than 100 μm (Biter et al. 1987) and/or ice particles (Baumgardner et al. 2017). An additional verification of the cloud phase identification method is conducted by comparing with the Rosemount Icing detector (RICE) (Figure S2).

RH values are less variable in averaged observation data (a–c), and compare well with simulations for liquid and mixed phases. However, at RF03 UTC 17:45:00–17:51:00, CAM-collocated data show RH_i at or above ice saturation (Figure 3 c) with coexisting liquid and ice (e), as opposed to the observations that are almost exclusively ice phase at ice subsaturation, potentially from sedimentation (b–d). The underestimated occurrences of ice subsaturation are found to be a common problem in CAM5 simulations, and will be discussed further in Section 3.4. The CDP, Fast-2DC probes and the VCSEL hygrometer all provide open-path measurements, and their data recording synchronization is within milliseconds. For occasions where liquid cloud edge does not match with liquid saturation, it could be due to the combined uncertainties in RH_{liq} (i.e., 6.4%–10.4%) and/or distance between the cloud probes and the VCSEL hydrometer.

Four typical cloud phase segments are seen: (i) a homogeneous, liquid dominant condition (e.g., RF03 UTC 17:38:40–17:41:20), (ii) a homogeneous, ice dominant condition potentially from sedimentation (e.g., RF03 UTC 17:44:30–17:52:00), (iii) a relatively well-mixed segment of mixed phase (e.g., RF17 UTC 19:42:35–19:43:20), and (iv) a spatially heterogeneous mixed phase with a mixture of ice and liquid (e.g., RF17 UTC 19:39:15–19:41:05 and RF03 UTC 17:52:00–17:58:40). Separate pockets of pure ice, pure water and a mixture of both were seen on the order of hundreds of meters based on 1-Hz data, similar to those pockets reported in Korolev et al. (2003), also known as the “conditionally” mixed phase clouds (Korolev et al. 2017). Large spatial heterogeneities in mixed phase are consistent with those reported by several previous studies, such

as the global survey of cloud thermodynamic phase on the sub-km scales based on Hyperion imaging spectrometer onboard NASA’s Earth Observer 1 (EO-1) spacecraft (Thompson et al. 2018), 20-m horizontal resolution satellite data of Arctic clouds (Chylek and Borel 2004), as well as 100-m horizontal resolution in situ and ground-based observations of midlatitude MPCs (Field et al. 2004).

3.2 Cloud phase frequencies and characteristics

Cloud phase occurrence frequencies are examined for all in-cloud conditions (Figure 4), as well as $CWC \geq 0.01 \text{ g m}^{-3}$ and cloud fraction ≥ 0.6 (Table 1). More sensitivity tests using various in-cloud thresholds are shown in Figures S3–S5. Spatially averaging 1-Hz observations into 200 s intervals has approximately doubled the occurrence frequency of mixed phase samples from 17% to 37% at -10° – 0°C and lowered the occurrence frequencies of liquid and ice phase samples when restricting $CWC \geq 0.01 \text{ g m}^{-3}$ (Table 1). Coarser resolution observations show more “conditionally” mixed phase samples than higher resolution data due to spatial averaging of individual cloud phase pockets, as discussed in Korolev et al. (2017). In addition, the spatial averaging leads to fewer liquid phase samples that satisfy the condition of cloud fraction ≥ 0.6 (i.e., 41% for Obs-1s and 5% for Obs-200s). This is also partly due to vertical profiling of the research aircraft at low altitudes, which leads to relatively short residence time in boundary layer clouds. Total number of seconds and lengths of samples based on mean true air speed at various temperature ranges is given in Table S2.

Compared with observations, both simulations have significantly lower frequencies of mixed phase samples, regardless of the restrictions on in-cloud conditions (Figures 4, S3–S5 and Table 1). For $CWC \geq 0.01 \text{ g m}^{-3}$, from -20° – 0°C , the Obs-200s data show 17%–37% of mixed phase, while both CAM-collocated and CAM-domain data show only about 1%–4%. Interestingly,

both simulations show more liquid phase samples (60%–70%) compared with the Obs-200s data (27%) from -10°–0°C. From -20°– -10°C, the CAM-collocated data also contain 53% of liquid phase, which is three times of that in the Obs-200s (16%), and underestimate frequencies of ice and mixed phase samples. Previously, CAM5 simulations have been reported to underestimate SLW content and overproduce ice at temperatures relevant for mixed phase conditions compared with satellite observations (e.g., Komurcu et al. 2014; Cesana et al. 2015; Kay et al. 2016b; Wang et al. 2018). Comparing these previous findings with our results on phase frequencies indicates that satellite observations may be biased to detect a layer of SLW at the top of cold clouds (e.g., Rauber and Tokay 1991) and underestimate ice phase occurrence frequency below cloud top, and/or the underestimation of SLW content in simulations is more likely attributed to underestimating mixed phase frequencies than overestimating liquid phase frequencies. More assessment is needed for the vertical distribution and lifetime of SLW in various cloud phases.

For the examination of liquid and ice mass partitioning, normalized frequencies of LWC/CWC ratios are binned by 10°C (Figure 5) and 5°C (Figure S6) intervals from -40°–0°C, separated into two categories, $CWC \geq 0.01 \text{ g m}^{-3}$ and cloud fraction ≥ 0.6 . All of the observations and simulations show decreasing frequencies of liquid phase (i.e., $LWC/CWC \geq 0.9$) and increasing frequencies of ice phase (i.e., $LWC/CWC \leq 0.1$) with decreasing temperature, consistent with more favorable thermodynamic conditions for ice phase at lower temperatures. The threshold of $CWC \geq 0.01 \text{ g m}^{-3}$ is the same as that used in Korolev et al. (2003). Under this condition, the 1-Hz observations reveal similar distributions of cloud phase occurrence frequencies to those shown in Korolev et al. (2017), which recalculated Nevzorov probe measurements initially reported by Korolev et al. (2003) inside clouds in frontal systems. Specifically, 1-Hz observations in the ORCAS campaign show similar frequencies of liquid (34%), mixed (17%), and ice phases (49%)

from -10° – 0°C , comparable to ~37%, 20% and 43% from observations in the high northern latitudes (42°N – 76°N), respectively (Korolev et al. 2017, 2003). At -30° to -10°C , 1-Hz observations from the ORCAS campaign show lower mixed phase frequencies (3% – 5%) than those in previous studies of Arctic clouds (13% – 17%) (Korolev et al. 2017, 2003) (Table S4).

Figure 6 shows the observed geometric means of LWC, IWC, N_{liq} and N_{ice} at 5°C intervals from -40° – 0°C for respective cloud phases. For liquid and mixed phases, Obs-1s data show larger averaged quantities by 1–2 orders of magnitude compared with the coarser resolution data, since the spatial averaged segments may include both in-cloud and clear-sky conditions. For ice phase, Obs-200s data show larger IWC and N_{ice} than Obs-1s data, likely due to the restriction of no more than 10% of missing values in the spatial averaging, which only allows relatively long, continuous in-cloud segments that are often associated with higher IWC and N_{ice} to be used in Obs-200s data. Trends in the averaged quantities with respect to temperature are generally similar among various scales of observations, except for one temperature bin of -30° – -25°C in mixed phase likely due to the smaller sample sizes (Figure S7). For Obs-1s data, LWC and IWC generally decrease with decreasing temperature for all cloud phases. For Obs-200s data, the average IWC and N_{ice} are 1–2 orders of magnitude higher in ice phase compared with mixed phase, whereas the average LWC and N_{liq} are 3–10 times higher in mixed phase than liquid phase at -10° – 0°C but more similar at other temperatures. For both liquid and mixed phases, average N_{liq} slightly increases with decreasing temperature at -25° to 0°C , and it decreases with decreasing temperature below -25°C . In comparison, the trends in average N_{ice} vary between mixed and ice phases. In mixed phase, average N_{ice} decreases (increases) with decreasing temperature above (below) -25°C , while in ice phase, average N_{ice} increases with decreasing temperature.

Figure 7 further compares the geometric means of IWC, LWC, N_{liq} and N_{ice} between simulations and the Obs-200s data. For liquid phase, simulated average LWC values (i.e., 0.01–0.1 g m⁻³) are about one order of magnitude greater than the Obs-200s data, and are more comparable to those in Obs-1s. The average N_{liq} in liquid phase in CAM-domain data is about three times of that in the observations from -20° to 0°C. Previously, shallow convective detrainment was found to be the main source of cloud liquid over the Southern Ocean (Kay et al. 2016a). Since the parameterization of shallow convective detrainment defines cloud ice mass fraction to be larger than 0.1 below -8°C, much fewer liquid phase samples (i.e., ice mass fraction < 0.1) can exist below -8°C in the simulations (Figure S8). Therefore these liquid phase samples are more likely contributed by shallow convection detrainment above -8°C and by other moist physical processes (e.g., microphysics “MPDLIQ” and macrophysics “MACPDLIQ”) below -8°C.

For mixed phase, the simulations consistently show lower LWC, IWC and N_{liq} than the Obs-200s data at -25°–0°C by a factor of 3–10, while the simulated N_{ice} is much higher than the observations at -25°– -5°C by a factor of 3–500. The overestimation of N_{ice} in simulations is consistent with previous findings of the parameterization of Meyers et al. (1992) overestimating ice number concentrations (e.g., Liu et al. 2011; DeMott et al. 2010). Another main difference is seen for ice phase, where simulated IWC is ~2 orders of magnitude lower than Obs-200s data, suggesting that the simulated ice particles are unrealistically smaller than the observations at -40°– -10°C since the N_{ice} values are similar at this temperature range. In contrast, ice-phase N_{ice} in the simulations is lower than observations by a factor of 5–30 from -10°–0°C, respectively, possibly due to insufficient secondary ice production and/or missing marine organic aerosols as ice nucleating particles (INPs) in the simulations, which can be important at that temperature range

over the Southern Oceans. These findings of ice phase are unlikely affected by the sampling volume, since all the datasets have sufficient number of ice phase samples (Figure S8).

The statistical robustness of the comparisons is supported by the fact that all of the average quantities in Figure 7 show very similar results between CAM-collocated and CAM-domain data, which almost never differ by more than one order of magnitude for the same temperature bins. In addition, sensitivity tests show maximum differences of 33% (29%) and 57% (32%) in mass (number) concentrations of cloud liquid and cloud ice, respectively, due to different ranges of particle size distributions in observations and simulations (Table S3), which proves that the differences in the observed and simulated quantities by more than one order of magnitude cannot be all attributed to size range differences.

Average mass ratios of LWC/CWC are shown at various temperatures for two categories: total in-cloud conditions and mixed phase only (Figure 8), and number of samples are shown in Figure S9. Compared with average LWC and IWC, the observed ratios of LWC/CWC only slightly increase with spatial averaging, because the mass ratios have cancelled out the effect of including more clear-sky conditions on larger averaging scales. Compared with Obs-200s data for total in-cloud conditions, simulations show lower (higher) LWC/CWC ratios by 0.1–0.3 (0.2) at -40° – -5°C (-5° – 0°C), due to the combined effects from overestimated LWC in liquid phase, underestimated LWC in mixed phase, and underestimated IWC in both ice and mixed phases in simulations (Figure 7). The underestimated LWC/CWC ratios for total in-cloud conditions in the simulations is consistent with the previous comparisons between CAM5 simulations and the GCM-oriented CALIPSO Cloud Product (Cesana and Chepfer 2013). On the other hand, the simulations show slightly higher LWC/CWC for mixed phase compared with observations at -20° –

0°C. The differences between total in-cloud and mixed phase only conditions suggest that phase partitioning parameterizations may need to be specified for different phases.

3.3 Relative humidity distributions in relation to temperature and cloud fraction

RH distributions have large influences on the evolution of cloud condensates. Thus, it is important to examine RH distributions in conjunction with cloud microphysical and macrophysical properties. In particular, CAM5 calculates ice cloud fraction as a function of RH_i (Gettelman et al. 2010), highlighting the need for validating the RH distributions at various cloud fractions. Probability density functions (PDFs) of RH_{liq} (Figure 9) and RH_{ice} (Figure 10) are shown for all cloud phases at various cloud fractions. As temperature decreases from -20°–0°C to -40°– -20°C, the peak position of RH_{liq} distribution shifts from around liquid saturation to lower values, in addition to a broadening effect on the distributions. As cloud fraction decreases from >0.9 to <0.1, the peak position of RH_{liq} decreases from 100% to ~85% at -20°–0°C, and from ~75% to 15% at -40°– -20°C, respectively.

The simulations agree well with the observations for RH_{liq} between -20°–0°C. However, the simulations show higher frequencies of RH_i between 105% and 115% than Obs-200s data for cloud fraction >0.9 at all temperature ranges. In addition, the simulations show narrower RH_{liq} and RH_i distributions compared with observations at cloud fractions of 0.1–0.9, lacking the large variabilities of RH_{liq} distributions as seen in the observations. When restricting cloud fractions, the spatial averaging has minimal impacts on RH PDFs, and the two sets of simulations agree well with each other, which further demonstrates the robustness of such comparisons on RH PDFs using datasets with different sample volumes.

A layer normalized frequency distribution of RHi at various temperatures and cloud fractions is shown for the Obs-200s and CAM-domain datasets (Figure 11). The RHi frequency is calculated as the number of samples in each $1^{\circ}\text{C}\times 5\%$ bin divided by the total number of samples in each 1°C interval. Previously, layer-normalized RHi frequency was analyzed for -88° – -23°C based on in situ observations from 68°N to 21°S in the UT/LS (Krämer et al. 2009). Here we extend that previous work to -67° – 0°C at higher southern latitudes. The differences of normalized RHi frequency between the simulations and observations are also shown (Figure 11 g–i). For cloud fraction ≥ 0.9 , Obs-200s data show the maximum RHi at 125%. CAM-domain data show higher maximum RHi but only for very few cases (Figure S10). For cloud fractions of 0.1–0.9, the simulations underestimate the frequencies of $\text{RHi} < 90\%$ and $> 110\%$ compared with the observations. This further illustrates that the smaller variabilities of RH PDFs in the simulations (as seen in Figures 9 and 10) are consistently shown at all the temperatures from -67°C to 0°C . At low cloud fraction (< 0.1), the simulations underestimate the frequencies of relatively dry conditions from -60° – -50°C and underestimate the frequencies of moist conditions (near ice saturation) from -8° – 0°C .

3.4 RH distributions for liquid, mixed and ice phase conditions

In this section, we further examine the influences of cloud phases on cumulative frequency distributions (CFDs) of RH_{liq} and RHi in Figures 12 and 13, respectively. This analysis is restricted to cloud fraction ≥ 0.9 , and separated by 10°C intervals from -40° – 0°C . For observations from ~ 0.1 – 50 km scales, similar CFDs of RH_{liq} are seen for liquid and mixed phases, all of which are close to liquid saturation. For example, 98%, 90% and 64% of the Obs-10s mixed phase data show $\text{RH}_{\text{liq}} > 90\%$ at -10° – 0°C , -20° – -10°C , and -30° – -20°C , respectively. This result is consistent with the theoretical understanding that MPCs generally show RH near liquid saturation

at steady-state conditions (Korolev and Mazin 2003). The slight deviation from liquid saturation in liquid and mixed phases is consistent with previously reported inhomogeneities such as cloud holes (Korolev and Isaac 2006). The simulations show nearly identical RH_{liq} CFDs to observations for liquid phase, except for a lack of liquid droplets for colder temperatures at -40°–-30°C (Figure 12 d). For mixed phase, the CAM-domain data show remarkable similarities to observations for RH_{liq} CFDs at -20°–0°C (note that the CAM-collocated data have very few samples at this condition), yet no mixed phase is shown in the simulations below -30°C.

The largest differences between the observations and simulations are seen in ice phase particularly for -30°–0°C (Figure 12 i–k), where both simulations underestimate the occurrence frequencies of RH_{liq}<95% by a fraction of 0.2–0.4. In fact, for ice phase, only 80%, 59% and 11% of the Obs-10s ice phase data show RH_{liq}>90% at -10°–0°C, -20°– -10°C, and -30°– -20°C, respectively. These results indicate that even though RH is centered at liquid saturation in liquid and mixed phase as low as -30°C on the scale of ~0.1–50 km with liquid mass fraction>0.1 and cloud fraction≥0.9, such assumption is not applicable when LWC/CWC≤0.1. A similar deviation from liquid saturation with increasing IWC/CWC ratio was reported previously (Korolev and Isaac 2008), and our results do not show significant impacts from spatial averaging due to the restriction of cloud fraction≥0.9.

The CFDs of RH_i also show large differences between observations and simulations for ice phase (Figure 13), where simulations overestimate the frequency of ISS conditions at -30°–0°C. Such discrepancy is possibly due to the assumption used in the ice microphysics parameterization in CAM5 (Morrison and Gettelman 2008; Gettelman et al. 2010), that is, RH values reach liquid saturation as long as liquid droplets coexist with ice particles, regardless of the small mass fraction of liquid phase being present. Note that this assumption is accurate for most

of the liquid and mixed phase samples (Figure 12 a–c and e–g), but as liquid mass fraction decreases to below 0.1, it becomes unlikely that the in-cloud conditions can reach a steady-state condition close to liquid saturation when few droplets exist. For liquid and mixed phases, all of the observations show increasing fractions of ISS and increasing magnitudes of maximum ISS with decreasing temperature, which agree relatively well with the simulations. Such increasing magnitudes of ISS are a result of the majority of liquid and mixed phases being around liquid saturation, which leads to higher RH_i as temperature decreases based on the Clausius-Clapeyron equation.

In addition, two other model-observation differences are seen for ice phase. First, the simulations underestimate the occurrence frequencies of ice subsaturation. At -10°–0°C, the observations show ~60% of ice phase being subsaturated, whereas CAM-collocated and CAM-domain show ~25% and ~10%, respectively. The fact that simulations are characterized by lower IWC (Figure 7 g) combined with the lower frequencies of subsaturation for ice phase suggests that the simulations may under-represent ice particles in subsaturated layers during sedimentation (as illustrated in Figure 3), likely due to insufficient accretion rates and/or over-amplified sublimation rates. Second, the simulations do not show any ISS greater than 10% for ice phase at -40°– -30°C (Figure 13 L), which accounts for ~15% of the samples in all the observations. The lower frequency of high ISS at colder temperatures is likely affected by the RH_i threshold for immersion nucleation of ice particles in the ice microphysics parameterization (Liu et al. 2007; Liu and Penner 2005).

3.5 Vertical profiles of ice supersaturation

Ice supersaturation is a key factor that directly affects ice particle growth, deposition and sublimation, which is further examined in Figure 14. Three cases are examined at temperatures

below 0°C: ISS at low cloud fraction (<0.1), ISS at high cloud fraction (≥ 0.6), and non-ISS (i.e., $R_{Hi} \leq 100\%$) at high cloud fraction (≥ 0.6). Frequencies are either normalized by all samples (a–c), or normalized by the total number of the aforementioned three cases (d–f). Comparisons are only shown for the CAM-collocated data (different results for CAM-domain data are not shown), since ISS frequency is affected by local conditions such as vertical velocity, temperature, and existing hydrometeors.

Focusing on -40° – 0° C, Obs-200s and CAM-collocated data show similar profiles of ISS at low cloud fraction with a peak frequency between -15° and -10° C. The simulation also shows a decreasing trend in ISS frequency at high cloud fraction with decreasing temperature, consistent with the observations. However, the simulation shows higher frequencies of ISS at high cloud fraction, which is likely due to the model assumption of liquid saturation for the fraction of the grid box where liquid and ice phases coexist, as discussed in Section 3.4.

For temperatures below -40° C, the CAM-collocated data significantly overestimate the frequencies of ISS by 0.05–0.75 compared with the observations for cloud fractions ≥ 0.6 (Figure 14 b). At temperatures below -50° C, the CAM-collocated data underestimate ISS frequency at cloud fractions <0.1 . This result indicates that additional ISS needs to be allowed for clear-sky conditions rather than in-cloud conditions in CAM5 simulations at cirrus cloud regimes. The relative frequencies of these three cases further illustrate the increasing discrepancies between the observations and simulations with decreasing temperatures below -40° C (Figure 14 d–f). Improvements are potentially needed for parameterizations of clear-sky ISS, ice nucleation condition and water vapor deposition in clouds.

4. Conclusions and implications for climate simulations over the Southern Ocean

In this study, cloud microphysical properties over the Southern Ocean (50°W – 92°W and 30°S – 75°S) are examined based on in situ airborne measurements, and are compared with two sets of CAM5 simulations: one is nudged by the reanalysis meteorological conditions and collocated with the aircraft flight track (CAM-collocated), and the other one is free-running (CAM-domain) and contains all the gridded data within the latitudinal, longitudinal and pressure domain during the ORCAS campaign. The two simulations primarily produce comparable results (Figures 4 – 13) (except for ISS frequency due to sensitivity to local thermodynamic and dynamical conditions), suggesting statistical robustness for the analyses conducted in this work when comparing localized samples with a larger set of samples.

Airborne measurements are averaged over various horizontal scales from ~ 0.1 to 50 km, in order to provide a scale-aware comparison with GCM simulations. For the analyses of cloud phase occurrence frequency, RH PDFs and CFDs, and LWC/CWC ratios, the sensitivity to horizontal resolution is very low, which demonstrates the consistency of these results at various horizontal resolutions. Other microphysical properties, i.e., LWC, IWC, N_{cliq} and N_{ice} , show 1–2 orders of magnitude lower values when averaged over the entire scale that includes both in-cloud and clear-sky conditions. These sensitivity tests demonstrate the applicability of using in situ observations to evaluate GCM simulations for cloud characteristics and RH distributions, which also help to guide future comparisons of datasets at various spatial resolutions.

Several main differences between simulations and observations are identified. Liquid and mixed phases are seen in Obs-1s data at low temperatures (-40° – -30°C) with 3% and 4% frequencies, respectively, which are missing in the simulations. SLW droplets are frequently observed from -20° – 0°C , consistent with previous observations that frequently reported SLW in this region (Morrison et al. 2011a; Chubb et al. 2013; Huang et al. 2012b), while the simulations

show higher liquid phase and lower mixed phase frequencies at this temperature range. Simulations also show higher (lower) LWC in liquid (mixed) phase, higher (lower) $N_{c_{liq}}$ in liquid (mixed) phase, lower IWC in both ice and mixed phases, higher (lower) $N_{c_{ice}}$ below (above) -5°C in mixed phase, and lower (higher) LWC/CWC ratios below (above) -5°C for the total in-cloud conditions. Biases of cloud microphysical properties vary with different cloud phases, indicating that future observational constraints may need to be specified for different phases. Processes of interest not only include the temperature function of phase partitioning such as that used in the shallow convection scheme of Park and Bretherton (2009) and tested by Kay et al. (2016a) and Frey and Kay (2017), but also include ice nucleation and vapor deposition rate during the WBF process as tested by Tan and Storelvmo (2016).

A large amount of ice phase is observed at -10° – 0°C , consistent with Huang et al. (2017) which found relatively high $N_{c_{ice}}$ of small ice particles at $>-9^{\circ}\text{C}$. In comparison, at -5° – 0°C , simulations show smaller ice mass fraction (IWC/CWC) for total in-cloud conditions, potentially due to the insufficient growth of ice particles by deposition and/or accretion. For mixed phase, simulations show higher $N_{c_{ice}}$ at -40° – -5°C and lower $N_{c_{ice}}$ at -5° – 0°C compared with the observations, with the former difference possibly due to ice nucleation parameterization and the latter difference possibly due to insufficient secondary ice production or missing INPs at warmer temperatures over the Southern Ocean. The lack of ice subsaturation in simulations is likely due to too fast removal of ice particles during sedimentation into subsaturated conditions.

In-cloud RH is frequently observed at liquid saturation for liquid and mixed phases. This feature can be a result of several processes, such as vapor diffusion from liquid to ice due to the WBF process, and/or updraft vertical velocity on the order of $0.1\text{--}1\text{ m s}^{-1}$ that can potentially sustain simultaneous growth of liquid droplets and ice particles (Korolev and Field 2008; Korolev

2007; Fan et al. 2011; Korolev 2008). The similarity among various scales of averaged observation datasets highlights the consistency of such findings when a sufficient amount of LWC exists (i.e., $LWC/CWC > 0.1$). However, larger deviations from liquid saturation are observed in ice phase (i.e., $LWC/CWC \leq 0.1$) at various scales. These results suggest that the assumption of liquid saturation for wherever liquid droplets exist (Morrison and Gettelman 2008) is not applicable when mass fraction of LWC is low, even for coexisting liquid droplets and ice particles on ~ 100 m scales. In addition, the increasing deviations from liquid saturation with increasing ice mass fraction imply that RH frequency distributions can serve as an indicator for estimating the dominant phase of cloud condensates (i.e., SLW droplets or ice particles).

Comparing RH distributions, simulations show smaller RH variability for partially cloudy conditions (cloud fraction of 0.1–0.9), and overestimate (underestimate) ISS frequencies for cloud fraction ≥ 0.6 (< 0.1) at -67° – 0°C ($< -40^{\circ}\text{C}$). In addition, ice cloud fraction in CAM5 is parameterized by grid average RH_i and other prescribed parameters (e.g., $RH_{i\max}=1.1$ and $RH_{i\min}=0.8$) (Gettelman et al. 2010). In contrast, observations show larger variabilities of RH_i at various cloud fractions, especially with higher frequencies of $RH_i > 110\%$ and $< 90\%$ at cloud fraction ≥ 0.1 compared with simulations (Figure 11), which in part casts doubt on the simplification of parameterizing ice cloud fraction as a function of the grid average RH_i rather than local RH_i . Previously, the lack of RH variability was found to be a main cause of missing clouds in CAM5 simulations (Wu et al. 2017), and the simulations often overlook water vapor spatial heterogeneities that were ubiquitously shown in observations (Diao et al. 2014). Since RH is a key parameter that links dynamical and thermodynamical conditions with cloud macro- and microphysical properties, improving the subgrid parameterization of RH is not only helpful for improving RH distributions, ISS frequencies and ice cloud fraction, but also could be a viable

approach to represent the impacts of subgrid-scale processes (e.g., eddies and turbulence) that are crucial for maintaining SLW (Field et al. 2014; Hill et al. 2014; Korolev and Field 2008) and calculating the lifetime of MPCs (Korolev and Mazin 2003). For future work, dynamical influences from vertical velocity, such as previously shown for Arctic and midlatitude MPCs (Solomon et al. 2011; Morrison et al. 2011b; Korolev and Field 2008; Shupe et al. 2008; Morrison and Pinto 2005; Naud et al. 2006), warrant more detailed investigation over the Southern Ocean.

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Table 1. Number of samples and occurrence frequencies (%) of three cloud phases at various temperature ranges.*

Dataset	Temperature	CWC $\geq$ 0.01 g m ⁻³			Cloud fraction $\geq$ 0.6		
		Liquid	Mixed	Ice	Liquid	Mixed	Ice
Obs-1s	-10 $^{\circ}$ $\leq$ T<0 $^{\circ}$ C	3114 (34%)	1530 (17%)	4414 (49%)	4245 (41%)	1602 (16%)	4465 (43%)
	-20 $^{\circ}$ $\leq$ T<-10 $^{\circ}$ C	620 (18%)	176 (5%)	2687 (77%)	838 (23%)	182 (5%)	2629 (72%)
	-30 $^{\circ}$ $\leq$ T<-20 $^{\circ}$ C	210 (6%)	114 (3%)	3406 (91%)	426 (10%)	117 (3%)	3672 (87%)
	-40 $^{\circ}$ $\leq$ T<-30 $^{\circ}$ C	62 (3%)	81 (4%)	1663 (92%)	76 (3%)	82 (3%)	2209 (93%)
Obs-10s	-10 $^{\circ}$ $\leq$ T<0 $^{\circ}$ C	2928 (28%)	2249 (21%)	5401 (51%)	3237 (31%)	2524 (24%)	4780 (45%)
	-20 $^{\circ}$ $\leq$ T<-10 $^{\circ}$ C	634 (16%)	276 (7%)	3108 (77%)	659 (18%)	362 (10%)	2722 (73%)
	-30 $^{\circ}$ $\leq$ T<-20 $^{\circ}$ C	180 (5%)	171 (4%)	3607 (91%)	233 (5%)	288 (7%)	3738 (88%)
	-40 $^{\circ}$ $\leq$ T<-30 $^{\circ}$ C	28 (2%)	133 (7%)	1647 (91%)	38 (2%)	154 (7%)	2152 (92%)
Obs-100s	-10 $^{\circ}$ $\leq$ T<0 $^{\circ}$ C	4332 (30%)	4492 (31%)	5772 (40%)	1057 (12%)	3722 (41%)	4228 (47%)
	-20 $^{\circ}$ $\leq$ T<-10 $^{\circ}$ C	881 (17%)	850 (16%)	3454 (67%)	70 (2%)	262 (9%)	2507 (88%)
	-30 $^{\circ}$ $\leq$ T<-20 $^{\circ}$ C	223 (5%)	204 (4%)	4261 (91%)	133 (3%)	115 (3%)	3708 (94%)
	-40 $^{\circ}$ $\leq$ T<-30 $^{\circ}$ C	0 (0%)	205 (9%)	2007 (91%)	0 (0%)	142 (7%)	1760 (93%)
Obs-200s *	-10 $^{\circ}$ $\leq$ T<0 $^{\circ}$ C	4653 (27%)	6186 (37%)	6098 (36%)	434 (5%)	3913 (47%)	4013 (48%)
	-20 $^{\circ}$ $\leq$ T<-10 $^{\circ}$ C	969 (16%)	985 (17%)	3966 (67%)	0 (0%)	57 (2%)	2721 (98%)
	-30 $^{\circ}$ $\leq$ T<-20 $^{\circ}$ C	250 (5%)	258 (5%)	4738 (90%)	10 (0%)	177 (5%)	3631 (95%)
	-40 $^{\circ}$ $\leq$ T<-30 $^{\circ}$ C	0 (0%)	262 (10%)	2261 (90%)	0 (0%)	26 (2%)	1707 (98%)
CAM- collocated	-10 $^{\circ}$ $\leq$ T<0 $^{\circ}$ C	6599 (60%)	289 (3%)	4028 (37%)	4042 (59%)	173 (3%)	2689 (39%)
	-20 $^{\circ}$ $\leq$ T<-10 $^{\circ}$ C	705 (53%)	18 (1%)	604 (46%)	700 (18%)	205 (5%)	2995 (77%)
	-30 $^{\circ}$ $\leq$ T<-20 $^{\circ}$ C	0 (0%)	0 (0%)	978 (100%)	0 (0%)	0 (0%)	3315 (100%)
	-40 $^{\circ}$ $\leq$ T<-30 $^{\circ}$ C	0 (0%)	0 (0%)	292 (100%)	0 (0%)	0 (0%)	2890 (100%)
CAM- domain	-10 $^{\circ}$ $\leq$ T<0 $^{\circ}$ C	112293 (70%)	6735 (4%)	40321 (25%)	84882 (63%)	4802 (4%)	45661 (34%)
	-20 $^{\circ}$ $\leq$ T<-10 $^{\circ}$ C	5404 (29%)	195 (1%)	12793 (70%)	5564 (6%)	1236 (1%)	92236 (93%)
	-30 $^{\circ}$ $\leq$ T<-20 $^{\circ}$ C	66 (1%)	3 (<1%)	12459 (99%)	28(<1%)	32 (<1%)	72318 (100%)
	-40 $^{\circ}$ $\leq$ T<-30 $^{\circ}$ C	0 (0%)	0 (0%)	10558 (100%)	0 (0%)	0 (0%)	54960 (100%)

* The number of samples generally increases with increasing averaging scales of observation data for CWC $\geq$ 0.01 g m⁻³, mostly due to the fact that this restriction can be more easily satisfied for averaged datasets. For example, the center second of the Obs-200s data does not need to be in-

1021 cloud to have averaged $CWC \geq 0.01 \text{ g m}^{-3}$, while each second of Obs-1s data has to be in-cloud to
1022 satisfy this criterion. For Obs-1s data, cloud fraction is calculated for -5 to +4 seconds (a total of
1023 10 second) around each second in Table 1, Figures 5, 9, 10, 12 and 13.

Figure 1. Cloud phase identification method used in this work. (a) A diagram for identifying ice and liquid phase samples from two probes separately, and the total IWC and LWC stand for the sum of IWC and LWC from two probes, respectively. (b) Number versus mass concentrations of CDP probe (N_{CDP} and M_{CDP} , respectively) color coded by D_{max_2DC} . Grey dots show $N_{C2DC}=0$. Section I, II, III are defined as large aerosols, liquid droplets and ice crystals, respectively. (c) Relationship of N_{C2DC} and M_{C2DC} , color coded by the standard deviation of 1-Hz particle size

1031 distribution (σ_{D_2DC}). Black markers highlight where the Fast-2DC measurements are identified as
1032 liquid droplets.

Figure 2. An example time series in Research Flight 17 (RF17) of the NSF ORCAS campaign. (a) 1-Hz observations of temperature, RHliq and RHi, with RH=100% (dashed grey) and 0°C (dotted blue) highlighted; (b) RHi and (c) RHliq for various scales of observations and CAM-collocated

1037 data; (d) log-scale M_{CCDP} (red) and M_{c2DC} (blue) and LWC measured by the King probe (purple);
1038 (e) LWC and IWC from CAM-collocated data. In (d) and (e), cloud phases are identified by
1039 markers below the abscissa. (f) MODIS satellite images around the flight track (black), with cloud
1040 phase highlighted in colors and an orange arrow pointing at the time series segment. The MODIS
1041 satellite image is on February 24, 2016, UTC 19:10:00. (g) Cloud particle imageries from the Fast-
1042 2DC probe for the three individual seconds noted by black arrows 1–3, respectively.

Figure 3. Similar to Figure 2, but for a time series in RF03. The MODIS satellite image is on January 21, 2016, UTC 19:15:00. The three examples (1), (2), and (3) of Fast-2DC imageries are similar to those reported as Type 2, 1a and 1b in Costa et al. (2017), respectively.

Figure 4. Cloud phase occurrence frequency for all in-cloud conditions. Temperature is binned by every 5°C. Sensitivity tests to various in-cloud thresholds are shown in Supplementary Figures S3 – S5.

Figure 5: Occurrence frequencies of LWC/CWC ratios compared between Obs-200s (solid line in all sub-panels) and other data, i.e., Obs-1s (a, b), CAM-collocated (c, d) and CAM-domain (e, f). Two in-cloud thresholds are used: $CWC \geq 0.01 \text{ g m}^{-3}$ (left) and $\text{cloud fraction} \geq 0.6$ (right). Obs-1s data calculate cloud fraction in 10 s intervals ($\pm 5 \text{ s}$). The number of samples of liquid, mixed and ice phases are shown in Table 1. Figure S6 and Table S4 show results using 5°C bins.

Figure 6: Geometric means of LWC, IWC, $N_{c_{liq}}$ and $N_{c_{ice}}$ for three cloud phases at 5°C intervals between -40°C–0°C for various scales of observations. Whiskers in Figures 6–8 represent $\pm$ one

1060 standard deviation, and their number of samples are shown in supplementary Figures S7–S9.
1061 Besides the restriction of no more than 10% of the averaged segments reporting missing values,
1062 the analysis is restricted to observation data with at least one second of $CWC > 0.001 \text{ g m}^{-3}$ and at
1063 least one second of either $N_{\text{Clq}} > 100 \text{ L}^{-1}$ or $N_{\text{Cice}} > 0.01 \text{ L}^{-1}$.

Figure 7: Similar to Figure 6, but for Obs-200s, CAM-collocated and CAM-domain datasets. In addition to the restriction of Obs-200s data as mentioned in Figure 6 caption, simulation output

1067 are restricted to those with both “CLDICE” and “CLDLIQ” $\geq 4.68\text{e-}5 \text{ g m}^{-3}$. These restrictions
1068 help to filter out very low values of IWC, LWC, N_{cliq} and N_{ice} . Particularly, restrictions on
1069 simulated CLDICE and CLDLIQ are used since filtering out low IWC and LWC alone does not
1070 exclude all of the low number concentrations reported in NUMICE and NUMLIQ.

Figure 8: Average LWC/CWC for (a, b) total in-cloud conditions and (c, d) mixed phase only. (Left column) Obs-1s, Obs-100s and Obs-200s. (Right column) Obs-200s, CAM-collocated and CAM-domain.

Figure 9: RHliq PDFs for various temperatures and cloud fractions. (a–d) -20° – 0° C; (e–h) -40° – -20° C. Various scales of observations (colored solid lines), CAM-collocated (dotted) and CAM-domain (dashed) data are shown. Results are binned by 5% of RHliq, except for $<2.5\%$. The number of samples generally increases with increasing averaging scales when using less strict cloud fraction restrictions (i.e., cloud fraction < 0.9), since it is more likely for samples at larger scales to be partially in-cloud than samples at smaller scales.

Figure 10: Similar to Figure 9, except for RH_i .

1084

1085 **Figure 11:** RH_i frequency distributions at various temperatures for (a–c) Obs-200s, (d–f) CAM-
 1086 domain and (g–i) their differences (CAM-domain – Obs-200s). Frequency in each 1°C×5% bin is
 1087 normalized by the total number of samples in each 1°C interval. Solid and dashed lines show ice
 1088 and liquid saturation, respectively.

1089

1090 **Figure 12:** RHliq CFDs for three cloud phases, restricted to cloud fraction ≥ 0.9 . Results are

1091 binned by 5% of RHliq, except for $<2.5\%$.

1092

1093 **Figure 13:** Similar to Figure 12, except for RH.

Figure 14: Vertical profiles for the occurrence frequencies of three cases: (first column) ISS at cloud fraction <0.1 , (second column) ISS at cloud fraction ≥ 0.6 , and (third column) non-ISS at cloud fraction ≥ 0.6 for Obs-200s and CAM-collocated data. (a–c) Frequency normalized by the

1098 total number of samples at each 5°C intervals. Blue dashed lines show the relative differences (in
1099 %, upper abscissa): $(\text{CAM-collocated} - \text{Obs-200s})/\text{Obs-200s} \times 100\%$. Horizontal whiskers
1100 represent the corrected sample standard deviation for 18 research flights. (d–e) Frequency
1101 normalized by the number of samples of these three conditions. (e–f) Number of samples for each
1102 specific condition in that column (solid lines with markers), and number of all the samples at each
1103 5°C interval (dashed lines).