# **Observations of clouds, aerosols, precipitation, and surface** 1 radiation over the Southern Ocean: An overview of CAPRICORN, 2 **MARCUS, MICRE and SOCRATES** 3 Greg M. McFarquhar<sup>1,2</sup>, Chris Bretherton<sup>3</sup>, Roger Marchand<sup>3</sup>, Alain Protat<sup>4,5</sup>, Paul J. DeMott<sup>6</sup>, 4 Simon P. Alexander<sup>7,5</sup>, Greg C. Roberts<sup>8, 8b</sup>, Cynthia H. Twohy<sup>9,8</sup>, Darin Toohey<sup>10</sup>, Steve Siems<sup>11</sup>, Yi Huang<sup>12</sup>, Robert Wood<sup>3</sup>, Robert M. Rauber<sup>13</sup>, Sonia Lasher-Trapp<sup>13</sup>, Jorgen Jensen<sup>14</sup>, Jeff Stith<sup>14</sup>, Jay Mace<sup>15</sup>, Junshik Um<sup>1,16</sup>, Emma Järvinen<sup>14,17</sup>, Martin Schnaiter<sup>17</sup>, Andrew 5 6 7 Gettelman<sup>14</sup>, Kevin J. Sanchez<sup>8</sup>, Christina S. McCluskey<sup>14</sup>, Lynn M. Russell<sup>8</sup>, Isabel L. McCoy<sup>3</sup>, Geueman<sup>--</sup>, Kevin J. Sanchez<sup>o</sup>, Christina S. McCluskey<sup>14</sup>, Lynn M. Russell<sup>8</sup>, Isabel L. McCoy<sup>3</sup>, Rachel Atlas<sup>3</sup>, Charles G. Bardeen<sup>14</sup>, Kathryn A. Moore<sup>6</sup>, Thomas C. J. Hill<sup>6</sup>, Ruhi S. Humphries<sup>18</sup>, Melita D. Keywood<sup>18</sup>, Zoran Ristovski<sup>19</sup> Luke Cravigan<sup>19</sup>, Robyn Schofield<sup>12</sup>, Chris Fairall<sup>20</sup>, Marc D. Mallet<sup>5</sup>, Sonia M. Kreidenweis<sup>6</sup>, Bryan Rainwater<sup>10</sup>, John D'Alessandro<sup>1,2</sup>, Yang Wang<sup>1,21</sup>, Wei Wu<sup>1</sup>, Georges Saliba<sup>8</sup>, Ezra J. T. Levin<sup>6,22</sup>, Saisai Ding<sup>23</sup>, Francisco Lang<sup>11</sup>, Son C.H. Truong<sup>11</sup>, Cory Wolff<sup>14</sup>, Julie Haggerty<sup>14</sup>, Mike J. Harvey<sup>24</sup>, Andrew Klekociuk<sup>7,5</sup> and Adrian McDonald<sup>25,26</sup> 8 9 10 11 12 13 14 15 16 <sup>1</sup>Cooperative Institute for Mesoscale Meteorological Studies, University of Oklahoma, Norman, 17 OK <sup>2</sup>School of Meteorology, University of Oklahoma, Norman, OK 18 19 <sup>3</sup>Department of Atmospheric Sciences, University of Washington, Seattle, WA <sup>4</sup>Australian Bureau of Meteorology, Melbourne, Australia 20 <sup>5</sup>Australian Antarctic Programme Partnership, Institute for Marine and Antarctic Science, 21 22 University of Tasmania, Hobart, Australia 23 <sup>6</sup>Department of Atmospheric Science, Colorado State University, Fort Collins, CO 24 <sup>7</sup>Australian Antarctic Division, Hobart, Australia <sup>8</sup>Scripps Institution of Oceanography, La Jolla, CA 25 <sup>8b</sup>Centre National de Recherches Météorologiques, UMR3589, Toulouse, France 26 27 <sup>9</sup>NorthWest Research Associates, Redmond, WA <sup>10</sup>Department of Atmospheric and Oceanic Sciences, University of Colorado, Boulder, CO 28

1

**Early Online Release**: This preliminary version has been accepted for publication in *Bulletin of the American Meteorological Society*, may be fully cited, and has been assigned DOI 10.1175/BAMS-D-20-0132.1. The final typeset copyedited article will replace the EOR at the above DOI when it is published.

29	<sup>11</sup> School of Earth, Atmosphere and Environment, Monash University, Melbourne, Australia
30	<sup>12</sup> School of Earth Sciences, University of Melbourne, Melbourne, Australia
31	<sup>13</sup> Department of Atmospheric Sciences, University of Illinois, Urbana, IL
32	<sup>14</sup> National Center for Atmospheric Research, Boulder, CO
33	<sup>15</sup> University of Utah, Salt Lake City, UT
34	<sup>16</sup> Department of Atmospheric Sciences, Pusan National University, Busan, South Korea
35	<sup>17</sup> Karlsruhe Institute of Technology, Karlsruhe, Germany
36	<sup>18</sup> Climate Science Centre, Oceans and Atmosphere, CSIRO, Melbourne, Australia
37	<sup>19</sup> School of Earth and Atmospheric Sciences, Queensland University of Technology, Brisbane,
38	Australia
39	<sup>20</sup> NOAA, Boulder, Colorado
40	<sup>21</sup> Beijing Normal University, Beijing, China
41	<sup>22</sup> Handix Scientific, Boulder, Colorado
42	<sup>23</sup> Peking University, Beijing, China
43	<sup>24</sup> National Institute of Water and Atmospheric Research, Wellington, New Zealand
44	<sup>25</sup> Gateway Antarctica, University of Canterbury, Christchurch, New Zealand
45	<sup>26</sup> School of Physical and Chemical Sciences, University of Canterbury, Christchurch, New
46	Zealand
47 48 49 50 51 52 53 54 55	Corresponding Author Greg M. McFarquhar Cooperative Institute for Mesoscale Meteorological Studies University of Oklahoma 120 David L. Boren Blvd., Norman, OK 73072 mcfarq@ou.edu; 405-325-3041
30	

Accepted for publication in Bulletin of the American Meteorological Society. DOI 10.1175/BAWS-D-20-0132.1.

- 57 Submit to the Bulletin of the American Meteorological Studies
- 58 11 September 2020

59 60

### ABSTRACT

61 Weather and climate models are challenged by uncertainties and biases in simulating Southern 62 Ocean (SO) radiative fluxes that trace to a poor understanding of cloud, aerosol, precipitation and 63 radiative processes, and their interactions. Projects between 2016 and 2018 used in-situ probes, 64 radar, lidar and other instruments to make comprehensive measurements of thermodynamics, 65 surface radiation, cloud, precipitation, aerosol, cloud condensation nuclei (CCN) and ice 66 nucleating particles over the SO cold waters, and in ubiquitous liquid and mixed-phase clouds 67 common to this pristine environment. Data including soundings were collected from the 68 NSF/NCAR G-V aircraft flying north-south gradients south of Tasmania, at Macquarie Island, and 69 on the RV Investigator and RSV Aurora Australis. Synergistically these data characterize 70 boundary layer and free troposphere environmental properties, and represent the most 71 comprehensive data of this type available south of the oceanic polar front, in the cold sector of SO 72 cyclones, and across seasons.

73 Results show a largely pristine environments with numerous small and few large aerosols above 74 cloud, suggesting new particle formation and limited long-range transport from continents, high 75 variability in CCN and cloud droplet concentrations, and ubiquitous supercooled water in thin, 76 multi-layered clouds, often with small-scale generating cells near cloud top. These observations 77 demonstrate how cloud properties depend on aerosols while highlighting the importance of 78 dynamics and turbulence that likely drive heterogeneity of cloud phase. Satellite retrievals 79 confirmed low clouds were responsible for radiation biases. The combination of models and 80 observations is examining how aerosols and meteorology couple to control SO water and energy 81 budgets.

4

# 82 CAPSULE

Recent air-, ground-, and ship-based observations of clouds, aerosols and precipitation over the
Southern Ocean are used in concert with models and satellite retrievals to provide insight into the
excessive absorption of solar radiation predicted by climate and numerical weather prediction
models.

87

### 88 **1. Introduction**

89 The Southern Ocean (SO) surrounding Antarctica and consisting of parts of the southern 90 Atlantic, Pacific and Indian Oceans, is one of the cloudiest places on Earth. The fractional cover 91 of low clouds (below 3 km altitude) prevalent in the warm and cold sectors of frequent extratropical 92 cyclones reaches nearly 80% year-round (Mace et al. 2009, IPCC 2013). Relative to more easily 93 sampled locations, there is a dearth of in situ observations of aerosols, clouds and precipitation 94 over the SO, especially south of 60°S. This makes it difficult to evaluate remote sensing retrieval 95 products. General circulation models (GCMs) have difficulty with simulating the present-day 96 aerosol, cloud coverage and cloud phase over the SO, with implications for anthropogenic aerosol 97 impacts and cloud feedbacks on climate (e.g., Trenberth and Fasullo 2010, Tan et al. 2016), two 98 key uncertainties in interpreting the historical climate record and projecting future climate change. 99 Numerical weather prediction (NWP) and GCMs have struggled to correctly simulate the 100 radiative budget over the SO due to low cloud biases. Most Coupled Model Intercomparison 101 Project Phase 5 (CMIP5) models predict too much shortwave (SW) radiation absorbed over the 102 SO region (Bodas-Salcedo et al. 2014, 2016; Naud et al. 2014), with impacts on ocean temperature, 103 the Southern Hemisphere (SH) jet (Ceppi et al. 2014), Antarctic sea ice trends (Flato et al. 2013) 104 and tropical rainfall (Hwang and Frierson 2013). Comparisons with satellite data indicate that 105 model radiative biases are due primarily to a lack of low and mid-level clouds in the cold sectors 106 of cyclones (e.g., Flato et al. 2013, Bodas-Salcedo et al. 2014). It was hypothesized on the basis 107 of limited observations, mainly from satellites, that GCMs might be glaciating what are in reality 108 persistent supercooled liquid clouds. Indeed, GCM simulations in which convective 109 parameterizations have been forced to produce greater amounts of supercooled liquid water (SLW) 110 have reduced SW biases (Kay et al. 2014).

6

Accepted for publication in Bulletin of the American Meteorological Society. DOI110.1175/BAMS-D-20-0132.1.

A related motivating issue is the apparent paucity of ice nucleating particles (INPs) over the SO (Bigg 1973; Burrows et al. 2013), due to it being far removed from any continental air sources; INP parameterizations are based mostly on Northern Hemisphere (NH) observations. Satellite retrievals of cloud-top phase indicate that SLW is more prevalent over the SO than at equivalent latitudes in the NH (Choi et al. 2010; Hu et al. 2010; Morrison et al. 2011). This could be because SO supercooled clouds are starved for INPs, as hypothesized by Kanitz et al. (2011) and Vergara-Temprado et al. (2018).

118 A final overarching question is how droplet concentrations are regulated in SO boundary layer 119 (BL) clouds in a synoptically active environment with high winds over a biologically productive 120 ocean. The SO is a biologically unique marine aerosol environment, its pristine nature is as close 121 to pre-industrial conditions as exists on Earth, and thus represents a natural laboratory to study 122 anthropogenic aerosol indirect radiative forcing (Carslaw et al. 2013; Ghan et al. 2013). Hoose et 123 al. (2009) showed that GCMs with prognostic aerosols tended to simulate SO clouds with too few 124 droplets compared to satellite observations, making them overly susceptible to human aerosol 125 perturbations. One hypothesis is that these models underestimate marine biogenic production of 126 cloud condensation nuclei (CCN). Satellite retrievals and some previous field observations show 127 the SO has a strong summertime maximum in cloud droplet concentration  $N_c$  (Boers et al. 1996, 128 1998), CCN (Ayers and Gras 1991), and aerosol concentrations  $N_a$  (Sciare et al. 2009) correlated 129 with phytoplankton productivity. Quinn et al. (2017) found that except for the high southern 130 latitudes, sea spray contributes less than 30% to the total CCN. However, observations in the ACE-131 1 campaign suggested that copious sulfate aerosols can be produced in the outflow of shallow 132 precipitating cumulus clouds from nucleation of marine biogenic gases (Hudson et al. 1998; Clarke 133 et al. 1998; Russell et al., 1998).

Accepted for publication in Bulletin of the American Meteorological Society. DOI110.1175/BAMS-D-20-0132.1.

134 Thus, there is a clear need for observations to help better model the natural aerosol lifecycle 135 and mixed-phase BL cloud over the SO. Prior to the campaigns described here, cloud and aerosol 136 measurements over the SO included those listed in Table 1. But, further observations on cloud and 137 aerosol concentrations over cold waters poleward of 60°S are critical for understanding cloud 138 processes over the SO. To understand the transition of aerosols to CCN over the remote oceans, it 139 is necessary to quantify particle sources and sinks as well as processes related to their aging, 140 including the role of new particle formation in the free troposphere, generation from breaking 141 waves over the ocean, generation of biogenic particles from gas phase oceanic emissions, the role 142 of precipitation scavenging, and the effects of updrafts and dynamics on clouds.

143 Climate model evaluation, and much current knowledge of SO clouds, aerosols, 144 precipitation, and surface radiation properties is based on satellite retrievals. Satellite studies have 145 found that cloud-top SLW is more frequent over the SO (Hu et al. 2010; Choi et al. 2010; Huang 146 et al. 2012a, b; Kanitz et al. 2011; Morrison et al. 2011; Protat et al. 2014; Huang et al. 2015) and 147 Antarctic (Grosvenor et al. 2012) than over the NH, but there are significant variations between 148 satellite retrieval products in the frequency of cloud-top SLW (Delanoe and Hogan 2010; Huang 149 et al. 2015) and these retrievals tell us little about the phase of condensate below cloud top. 150 However, potential errors in cloud retrievals, particularly those related to large solar zenith angles 151 (Grosvenor and Wood 2014) and three-dimensional effects (Wolters et al. 2010; Zeng et al. 2012; 152 Cho et al. 2015) remain a concern. Additional ground-based and airborne remote sensing, and 153 airborne in-situ measurements, are therefore needed to evaluate satellite retrievals.

A 2014 community workshop at the University of Washington discussed these issues, recognizing the need for a large international multi-agency effort to improve the understanding of clouds, aerosols, precipitation and their interactions over the SO (Marchand et al. 2014). The

157 workshop served as a motivation for the proposals of separate, but integrated, projects to various 158 funding agencies in the United States and Australia. These four collaborative projects were (1) the 159 Clouds Aerosols Precipitation Radiation and atmospheric Composition over the Southern Ocean 160 (CAPRICORN) I and II research voyages of the Research Vessel (RV) Investigator, led by the 161 Australian Bureau of Meteorology (BoM), that made extensive in-situ and remote sensing 162 measurements in 2016 and 2018, respectively, (2) the 2017-2018 Measurements of Aerosols, 163 Radiation and Clouds over the Southern Ocean (MARCUS) project, during which the United 164 States Department of Energy Atmospheric Radiation Measurement Program Mobile Facility-2 165 (DOE AMF2) was deployed on the Australian icebreaker Research Supply Vessel (RSV) Aurora 166 Australis (AA) as it made resupply voyages to Australian Antarctic bases, (3) the 2016-2018 167 Macquarie Island Cloud Radiation Experiment (MICRE) acquiring surface in-situ and remote 168 sensing observations using equipment from DOE ARM, the BoM, and the Australian Antarctic 169 Division (AAD), and (4) the 2018 Southern Ocean Cloud Radiation and Aerosol Transport 170 Experimental Study (SOCRATES) using the NSF/NCAR G-V aircraft to sample clouds, aerosols 171 and precipitation from Hobart, Australia, to within approximately 650 km of the Antarctic coast. 172 Although each project was a separate effort and no formal steering committee coordinated the 173 projects, many investigators served on the advisory board of several of the projects and there was 174 much collaboration between the campaigns. There was one integrated planning workshop (2017 175 Boulder) and two integrated data workshops after the completion of the projects (2018 Boulder, 176 2019 Hobart). Data have been freely exchanged among participants, and a special collection of 177 papers in the Journal of Geophysical Research/Geophysical Research Letters covering all four 178 projects has been established and is expected to grow substantially over the next few years. This 179 collaboration is essential to maximize the projects' impacts. Synergistically these data provide the

180 best available measurements of the BL and free troposphere structure, together with vertical 181 distributions of liquid and mixed-phase clouds and aerosols properties, over cold SO waters where 182 SLW and mixed-phase BL clouds are frequent.

183

# 2. Overview of Field Campaigns

184 In this section, the campaigns are introduced, detailing the scientific objectives, the time 185 period of the observations, the instruments and platforms used to acquire the observations, the 186 manner in which the observations were obtained, and a broad overview of the meteorological 187 conditions sampled. The majority of the observations was obtained in a North-South curtain 188 extending from Hobart, Australia to the Antarctic coast in the Australasian sector of the SO. Figure 189 1 shows the ship tracks from CAPRICORN I, II and MARCUS, as well as the G-V flight tracks 190 during SOCRATES and the location of the ground-observing site at Macquarie Island during 191 MICRE.

192 2.i. MICRE

193 The DOE ARM program, the AAD and the BoM collaborated in deploying ground-194 instrumentation to Macquarie Island between March 2016 and March 2018. Macquarie Island is 195 located at 54.5° S, 158.9° E (north of the oceanic polar front, Figure 1) and has a small research 196 station operated by the AAD that is staffed year-round, in part by BoM. The station supports a 197 variety of research activities and includes a long history of surface weather and radiosonde 198 observations (Hande et al. 2012, Wang et al. 2015).

199 The primary objective of MICRE was to collect surface-based observations of radiation, 200 precipitation, BL clouds, and aerosol properties in order to evaluate satellite datasets and to 201 improve knowledge of diurnal and seasonal variations, especially with regards to the vertical 202 structure of BL clouds. Instrumentation deployed during MICRE is listed in Table S1 (in the 203 supplement), along with time periods for which high quality observations are available for each 204 instrument in Table S2. The data include (i) passive surface radiation (solar, longwave, 205 microwave), (ii) surface precipitation rain rates, types and particle sizes, (iii) cloud radar 206 reflectivity and Doppler velocity profiles, ceilometer and lidar backscatter (including 207 depolarization) measurements (that provide information on cloud occurrence, cloud-base and top 208 height, precipitation particle size and phase, and some vertically-resolved aerosol optical 209 properties in cloud-free conditions), (iv) ground-based number concentrations of total aerosol and 210 CCN, and (v) ground-based INP number concentration and type (via filter sample analyses).

211 2.ii. CAPRICORN

212 CAPRICORN was a sea-based field study using the Australian Marine National Facility 213 (MNF) RV Investigator, designed to better understand interrelated aerosol-cloud-precipitation-214 radiation processes responsible for surface SW radiation biases in global models and discrepancies 215 between satellite rainfall measurements south of 40°S (e.g., Skofronick-Jackson et al. 2017; Protat 216 et al. 2019ab). The objectives were to (i) characterize cloud, aerosol, and precipitation properties, 217 BL structure, biological production and cycling of dimethyl sulfide (DMS) in the upper ocean, 218 atmospheric composition, and surface energy budget, as well as their latitudinal variability, (ii) 219 evaluate and improve satellite products (with a focus on the NASA A-Train and NASA / JAXA 220 Global Precipitation Mission (GPM) cloud and precipitation products, and surface heat flux 221 products), and (iii) evaluate and improve the representation of these properties in the regional and 222 global versions of the Australian Community Climate and Earth-System Simulator (ACCESS) 223 model (Puri et al. 2013). A second voyage, CAPRICORN II, occurred simultaneously within the 224 same overall region (south of Australia) as SOCRATES, and included 4 flights where the NCAR 225 G-V aircraft passed over or near the RV Investigator.

Accepted for publication in Bulletin of the American Meteorological Society. DOI 10.1175/BAWS-D-20-0132.1.

226 CAPRICORN I, held 13 March 2016 to 15 April 2016 south of Tasmania, used the 227 instruments listed in Table S3. All instruments operated near 100% of the time, and characterized 228 the basic atmospheric state (~1 radiosonde per day), vertical cloud structure, including integrated 229 liquid water and water vapor contents, cloud phase, and microphysical properties (based on cloud 230 radar, lidar, and microwave radiometer measurements), and rainfall rates and drop size 231 distributions (from disdrometer and micro rain radar measurements). The aerosol size 232 distributions, morphologies and compositions, size-resolved chemical compositions and 233 hygroscopic growth factors, cloud nuclei, CCN and INP concentrations, and some gaseous 234 atmospheric compositions including DMS and VOCs, were measured. Bio-aerosol size 235 distributions, air-sea bulk and turbulent fluxes and surface energy budgets, and sub-surface oceanic 236 properties were also measured. Three CloudSat-CALIPSO overpasses were successfully 237 intersected by the ship. The on-line supplement contains more details about the CAPRICORN-1 238 voyage, including dates and locations of 5 cases when the RV Investigator was in the cold sector 239 of major cold fronts in Table S4.

240 The main limitation of CAPRICORN I was its latitude span, with no measurements 241 collected south of  $55^{\circ}$ S (Figs. 1 and 2), and the period (late austral summer – early fall, thereby 242 not providing observations in the summer season where the largest surface radiation bias is found 243 in GCMs). This motivated CAPRICORN II, where the same comprehensive set of data as 244 CAPRICORN I was collected south of 55°S during summer. CAPRICORN II was held from 11 245 January 2018 to 21 February 2018, in combination with a major oceanographic project on the RV 246 Investigator aimed at quantifying changes in water properties and circulation of the SO, and 247 measuring distributions of trace metals and isotopes in the SO and the physical, chemical, and 248 biological processes controlling their evolving distributions. The objectives were similar to

249 CAPRICORN I, with the additional aim to collect precipitation measurements within the swath of 250 the GPM dual-frequency radar. The instrumentation was similar (Table S5), with notable additions 251 of the C-band dual-polarization Doppler radar (which did not operate during CAPRICORN I) and 252 the NSF-funded contributions as part of SOCRATES that included radiosonde launches every 6 h, 253 remote sensing instruments, and INP and bio-aerosol measurements. More details about the 254 voyage and cloud types sampled are included in the on-line supplement. Seventeen cases of 255 collocated GPM observations were collected with rain, snow, and mixed-phase precipitation 256 (Table S6). The number of identified cold sectors and cold fronts traversed by the RV Investigator 257 during CAPRICORN II are listed in Table S7.

258 *2.iii. MARCUS* 

259 During MARCUS the DOE AMF2 instrument package, including the Aerosol Observing 260 System (AOS) was installed on the AA as it made routine transits between Hobart, Australia and 261 the Australian Antarctic stations of Mawson, Davis and Casey, as well as Macquarie Island 262 between 21 Oct 2017 and 23 Mar 2018. MARCUS observations enhance the CAPRICORN 263 observations in that they were collected over a 5-month period centered upon the Austral summer, 264 allowing transitions from spring to fall to be observed across the 80 days of the MARCUS voyages. 265 Because the data were collected during resupply voyages, the science team had no control on the 266 timing of the voyages, nor could specific cloud types be targeted. Thus, a range of synoptic settings 267 was sampled, providing knowledge of temperature-dependent distributions of cloud properties 268 under a variety of aerosol and cloud conditions.

Specific objectives proposed for MARCUS were to (1) understand the synopticallyvarying vertical structure of SO BL clouds and aerosols, (2) quantify sources and sinks of SO CCN and INPs, including the role of local biogenic sources over spring, summer and fall, (3) quantify 272 mechanisms controlling SLW and mixed-phase clouds, and (4) advance retrievals of clouds, 273 precipitation and aerosols over the SO. Parameterization development and model evaluation 274 requirements were integrated in MARCUS' design so that systematic confrontation and 275 improvement of GCMs and NWP is possible. Instrumentation deployed during MARCUS, listed 276 in Table S8, included active and passive remote sensing instrumentation, in-situ measurements of 277 aerosols, bioaerosols and INPs, trace gas measurements and meteorological measurements 278 including six-hourly radiosonde launches, rain gauges and disdrometers. The conditions sampled 279 are listed in the on-line supplement including passages through cold fronts (Table S9).

280 2.iv. SOCRATES

SOCRATES used the NCAR/NSF G-V aircraft to sample clouds, aerosols and precipitation along (primarily) north-south transects south of Hobart, Australia reaching as far south as 62°S, from 15 Jan to 26 Feb 2018. The G-V made in-situ measurements within the BL and free troposphere, and included remotely-sensed measurements using a cloud-radar and lidar. The G-V flight tracks, shown in Figure 1, were designed to target the cold sectors of cyclones where models have the most trouble producing SLW, and thus were not the same for each day.

The overarching objectives of SOCRATES were similar to those of MARCUS, MICRE and 287 288 CAPRICORN. In particular, the G-V was tasked to obtain a dataset characterizing the structure of 289 the MBL and free troposphere over the SO, including observations of the vertical distribution and 290 properties of clouds and aerosols, including CCN and INPs, so that possible mechanisms to 291 explain the excessive absorbed SW radiation in models could be tested. The instruments and flight 292 paths were designed to gather statistics on aerosols and clouds as a function of latitude, and 293 included measurements over both the RV Investigator during CAPRICORN-2 and Macquarie 294 Island as explained in the on-line supplement. Table S10 lists the instrumentation installed on the

G-V including in-situ cloud and aerosol probes and remote sensing devices. The on-line supplement also provides information about the sampling strategy that was used to execute flights collecting both in-situ and remote sensing data along with a list of all the Research Flights (RFs) in Table S11.

299 Figure 2 shows the normalized fraction of observations made at each latitude during the four 300 campaigns. Apart from time spent at the Australian Antarctic stations and Macquarie Island for 301 resupplying during MARCUS, there is an even sampling of latitudes during both CAPRICORN II 302 and MARCUS. Both the RV Investigator and RSV Aurora Australis spent a large time south of 303 60°S, providing a very rare and invaluable set of data over cold waters poleward of the oceanic 304 polar front. MARCUS data are unique because they provide observations over the sea ice and 305 cover more of a seasonal cycle (October-March) than CAPRICORN II, whereas CAPRICORN I 306 and II included more thorough aerosol, oceanographic, and surface energy budget measurements 307 to put cloud observations in context, and MICRE provides the longest seasonal cycle at a single 308 location. SOCRATES provides the in-situ observations that are critical for process studies and 309 evaluation of remote sensing retrievals, and they are the only direct observations of aerosols below, 310 inside and above cloud. Thus, the combination of CAPRICORN, MICRE, MARCUS, and 311 SOCRATES data are synergistic in their characterization of the latitudinal and seasonal 312 variabilities of aerosol-cloud-precipitation-radiation processes over the SO.

313 **3. Preliminary Findings** 

Much of the initial effort since completing the projects has focused on evaluation of data quality and development of higher-level data products, as well as characterizing cloud and aerosol conditions over the SO. Some of the more noteworthy findings are discussed here. Integration of the datasets and comparison with model simulations and satellite retrievals is starting, a necessary step to evaluate mechanisms responsible for the excess absorption of solar radiation over the SO,
which is the overarching objective of these projects.

#### 320 *3.i. Latitudinal Dependence/Composition of Surface Aerosols*

321 Information on the composition and latitudinal dependence of aerosols is required to 322 understand the origin of aerosols and the role of biological aerosols and sea salt on droplet 323 nucleation in different locations and seasons. Surface aerosol volatility and hygroscopicity were 324 measured during CAPRICORN I at diameters of 40, 100 and 150 nm to provide information about 325 the composition of the Aitken and accumulation modes. Figure 3 shows that the daily averaged 326 number fraction of low volatility aerosol (persisting at 250°C) in the Aitken mode was  $0.22 \pm 0.2$ 327 (mean  $\pm 1\sigma$ ), which indicates that the Aitken mode was largely composed of secondary non-sea 328 salt sulfates. In the accumulation mode, the mean number fraction of low volatility particles was 329  $0.79 \pm 0.2$ , indicating most particles contained a primary sea spray sourced fraction. Low volatility 330 sea spray particle number fractions, particularly the Aitken mode, increased at higher SO latitudes 331 and were associated with higher wind speeds and generally lower particle number concentrations. 332 The proportion of primary sea spray particles observed from volatility measurements during 333 CAPRICORN I was larger than that observed from BL measurements via Scanning Transmission 334 Electron Microscopy (STEM) during SOCRATES in the summer. Further information about 335 aerosol composition and hygroscopicity measurements is provided in the on-line supplement, 336 which include chemical characterization of single particle composition by scanning transmission 337 X-ray microscopy by near-edge X-ray absorption fine structure (STXM-NEXAFS). These 338 measurements show that particles in-cloud and below-cloud have very similar organic functional 339 group compositions (Fig. S6).

Accepted for publication in Bulletin of the American Meteorological Society. DOI110.1175/BAMS-D-20-0132.1.

The average CN number concentrations (diameter greater than 3 nm) during CAPRICORN I were  $290 \pm 170$  cm<sup>-3</sup>, below typical summertime maxima (Gras and Keywood 2017, McCoy et al. 2015) and hence consistent with the seasonal cycle observed at Cape Grim, with summertime maxima of approximately 500 - 550 cm<sup>-3</sup> and wintertime minima of approximately 150 cm<sup>-3</sup>. The seasonal cycle in SO aerosol number is largely driven by enhanced secondary sulfate production in the summer months (Gras and Keywood 2017, McCoy et al. 2015).

346 Information on surface Fluorescent Biological Aerosol Particles (FBAPs) was provided by 347 the WIBS4. It measures the fluorescence from single aerosol particles in 3 excitation/emission 348 channels for particle sizes between 0.8 and 13 µm (Toprak and Schnaiter, 2013) to deduce fluorescent (i.e., biological) and total aerosol number concentrations and size distributions. During 349 350 MARCUS, the FBAP aerosol number concentration was rather low with a median value of 0.43 351  $L^{-1}$  giving an average FBAP fraction of about 0.3% in the MBL at latitudes from 46°S to 68°S 352 consistent with WIBS4 measurements in other projects. The total number concentration varied 353 strongly with latitude while the FBAP concentration was rather stable with indicated minimum 354 around -56° latitude and increasing concentrations towards the north and south. Implications of 355 these results on the sources and sinks of aerosols over the SO are being examined in several 356 publications under preparation.

357 3.ii. BL Aerosol and CCN Vary According to Origin

Aerosol measurements in the BL but above the surface give more information about sources and sinks of aerosols, and their role in droplet nucleation. Ambient aerosols 150 m above the ocean were collected through a CVI inlet on the G-V, but without the counterflow airstream that excludes small particles. Particles in two dry diameter ranges were impacted onto carboncoated nickel grids or silicon nitride windows and stored frozen for subsequent analysis by analytical STEM and X-ray spectroscopy that produces elemental inorganic composition of 364 individual aerosol particles. The size ranges were about 0.1-0.5  $\mu$ m and 0.5-5  $\mu$ m diameter (50%) cut size) for particle densities of 2 g cm<sup>-3</sup> at 1000 mb. Based on size distributions from the UHSAS, 365 366 the 0.1-0.5 µm size range comprised between 54%-93% of the aerosol accumulation-mode number 367 concentration (above the Hoppel minimum), and aerosol concentrations  $>0.1 \,\mu m$  were similar to 368 nearby cloud droplet concentrations  $N_c$ . Thus, particles in this size range would be expected to be 369 representative of the composition of most CCN for the cases analyzed. For the data presented here, 370 heaters on the titanium inlet and stainless steel sample line were turned off to minimize losses of 371 volatile species.

372 Figure 4 shows STEM results for 6 flights after grouping particles into different types based 373 on elemental composition and morphology (Twohy and Anderson, 2008). Fig. 4a shows results 374 for each flight, while Fig. 4b shows the overall mean composition. Particles 0.1-0.5 µm in diameter 375 were dominated by sulfur-based particles (mean 69% by number). Based on the ionic composition 376 measured on the RV Investigator during CAPRICORN-II these particles were primarily acidic 377 sulfate, likely with a small contribution from methanesulfonic acid (MSA) and other organics 378 (Twohy et al., 2020). The second-most frequent particle type in this size range (mean 28% by number) was salt-based sea-spray. Fig. 4b shows different types of sea-spray, which were 379 380 dominated by unprocessed, sodium chloride-based sea-spray particles. However, about 40% of 381 sea spray particles were enriched in sulfur and depleted in chlorine through uptake and 382 condensation of sulfur gases (McInnes et al. 1994), and a small percentage (3%) were salts 383 enriched in calcium or magnesium. Crustal and metallic particles and externally-mixed organics 384 were also detected in the  $<0.5 \ \mu m$  population in approximately equal proportions, but were together only about 3% by number. Overall these data indicate that 0.1-0.5 µm particles in the BL 385 386 were dominated by biogenic sulfates, with a smaller but significant contribution from sea-spray.

Particles >0.5 µm (not shown) were dominated by sea-spray, with only about 2% other aerosol types. Further, many sea-spray particles in the larger size fraction had detectable carbonaceous coatings, which may be important in ice nucleation in the marine environment (McCluskey et al. 2018a). More information about the chemical composition of the organic compounds is shown in the supplement.

Direct observations of CCN make it possible to understand how aerosols act as CCN. To 392 393 investigate controls of CCN, the variability in CCN spectra in the BL was characterized using a k-394 means clustering to group into 4 clusters associated with the observed bimodality in CN and CCN 395 concentrations. Minima in the bimodal frequency distributions of number concentrations occurred at approximately 750 cm<sup>-3</sup> for CN and was dependent on supersaturation for CCN (Figure 5a). The 396 397 four clusters were characterized as follows: 1) low CN/high CCN - southerlies influenced by 398 Antarctic coastal biological productivity, 2) high CN/low CCN - westerlies over the SO 399 characteristic of recent particle formation (RPF) events with low accumulation mode 400 concentrations due to recent precipitation, 3) high CN/high CCN – similar characteristics as high 401 CN/low CCN but with condensational growth of recently formed particles to CCN sizes, 4) low 402 CN/low CCN – aerosol populations scavenged by precipitation and lack of RPF.

403 The CCN concentrations (at 0.3% supersaturation) correlated well (Fig. 5b) with the 404 overlying  $N_c$  indicating large variations in CCN over the SO exist and have an important influence 405 on cloud microphysics. The large variability in CCN led to larger than expected variability in  $N_c$ , 406 which ranged from 10 to 449 cm<sup>-3</sup>. The variation in CN concentration was also notable, ranging 407 from 115-1153 cm<sup>-3</sup>. To understand this variability, HYSPLIT (Stein et al. 2015, Rolph et al. 2017) 408 back trajectories were performed to identify differences in source location and transport history. 409 The back trajectories for the low CN/high CCN were consistently from the south (Figure 6d) along

410 the Antarctic coast. This source location is associated with upwelling and marine biological 411 productivity that produces biogenic gases such as DMS, which can oxidize and condense to form 412 CCN-active particles (Hegg et al. 1991, Covert et al. 1992, Andreae et al. 1995, Read et al. 2008, 413 Sanchez et al. 2018). The two clusters with westerly back trajectories (Figure 6a and 6b) contained 414 the highest CN. High CN over the pristine SO are likely due to RPF aloft (Section 3.iii) and mixed 415 downward into the MBL (Sanchez et al. 2018). The high CN/low CCN cluster contained low 416 concentrations of accumulation mode particles (and consequently, small total aerosol surface area) 417 making conditions ideal for particle formation leading to high CN concentrations (Warren and 418 Seinfeld 1985, Clarke 1993, Pirjola et al 2000). While the high CN/high CCN cluster did not have 419 low accumulation mode concentrations, the spikes in CCN concentrations at the highest 420 supersaturations (Figure 5a, >0.6%) are consistent with RPF where some particles grow to CCN 421 sizes, typically through condensational growth during long residence times over the ocean (Russell 422 et al., 1998; Bates et al. 2000, Kumala et al 2004, Rinaldi et al. 2010, Zhang et al. 2014). The on-423 line supplement offers more information on how the back trajectories were combined with 424 ECMWF reanalysis to identify relations between BL cloud fraction and particle concentration.

# 425 *3.iii. New Particle Formation in Free Troposphere*

Analysis of free-tropospheric (3-6 km) aerosol measurements from the G-V identified signatures of RPF events occurring frequently across the SO, often in association with synopticuplift. It is hypothesized that air masses rich in precursor gasses (i.e. emissions from phytoplankton at the surface) undergo rapid synoptic uplift, are processed through the associated convection, cleansed of coarse and accumulation mode aerosol, and released into the free-tropospheric, lowaerosol surface area environment where gas-to-particle conversion is favored (McCoy et al., 2020 in prep). This synoptic uplift mechanism is complementary but independent from RPF occurring in the outflow of SO shallow cumulus clouds documented during ACE-1 (Clarke et al., 1998). It
is likely both contribute to the widespread observations of high Aitken aerosol number
concentrations throughout the SO free troposphere.

436 A free-tropospheric sample from RF09 is used to illustrate the synoptic-uplift mechanism 437 (Figure 7). During RPF events, simultaneously low accumulation mode aerosol number concentrations (from the wing-mounted UHSAS, 100 < D < 1000 nm) and high total number 438 439 concentrations (from the CN counter, D > 11 nm) occur, indicating presence of large Aitken mode 440 concentrations. High concentrations and rapid spatial variability in CN suggests sampling of 441 particle formation bursts or air masses at different stages of nucleation (Clement et al., 2002). RPF occurrences were prolific during RF09 due to a warm conveyor belt occurring west of Australia 442 and propagating south-east towards Antarctica. For statistical air-mass evolution analysis, 443 444 HYSPLIT (Stein et al., 2015) 72-hour back trajectories initiated at 10-minute intervals along the flight path are identified by maximum CN into RPF ( $CN_{Max} \ge 2500 \text{ mg}^{-1}$ ) and non-RPF events 445 (CN<sub>Max</sub>< 2500 mg<sup>-1</sup>) (Figure 7b). Standard temperature and pressure corrected units (mg<sup>-1</sup>) are 446 447 used to enable altitude invariant analysis across the campaign. The majority of these RF09 air 448 masses are RPF and have undergone recent synoptic uplift (ascent exceeds characteristic vertical velocity for synoptic events, ~1 cm s<sup>-1</sup> (Hakim, 2013)) in the previous 20-30 hours. In the 72-hours 449 450 before sampling, the majority of these air masses have access to the surface (Z < 1 km) and the 451 precursor gases necessary for generating new particles in a low aerosol-surface area environment. 452 RF09 is characteristic of RPF events during SOCRATES and their connection to synoptic 453 uplift. The two most frequent large-scale uplift mechanisms associated with RPF events are warm

Accepted for publication in Bulletin of the American Meteorological Society. DOI 10.1175/BAWS-D-20-0132.1.

454 conveyor belts and sub-polar vortices. Volatility analysis via comparison of heated to unheated 455 CN concentrations confirms that the particles sampled during RPF events are likely composed 456 mostly of  $H_2SO_4$ , a prominent aerosol precursor gas arising from phytoplankton emissions. It is 457 likely that the high concentrations of Aitken-mode aerosol particles produced above-cloud by these 458 RPF events are brought into the BL (Covert et al., 1996) and influence the sub-cloud CN and CCN 459 concentrations (McCoy et al., 2020 in prep; Sanchez et al., 2018) (section 3.ii). This source of 460 Aitken mode aerosol above-cloud may help to buffer SO clouds against precipitation removal, 461 sustaining higher than expected  $N_c$  (on the order of 80-100 cm<sup>-3</sup> between 45-62°S), and explains 462 the larger contribution of sulfur-based particles to sub-cloud CCN compared to sea-spray (section 3.ii, Twohy et al. 2020; McCoy et al., 2020 in prep; Sanchez et al., in prep). Evidence supporting 463 464 this hypothesis, the broader implications for SO cloud-aerosol interactions, and a more detailed 465 assessment of the synoptic uplift mechanism are presented in McCoy et al. (2020 in prep).

# 466 *3.iv. Low INP Concentrations over SO*

To investigate the processes giving rise to extensive SLW over the SO, not only is information about CN and CCN needed, but also about INPs. INP measurements were conducted during the various SO projects to define the spatial and temporal distributions of INPs over the region for the first time since the comprehensive measurements of Bigg (1973). A summary of campaigns, dates and INP sampling methods are given in the on-line supplement and in Table S12. Wide regions of the surface marine BLwere sampled south of 45°S, while INP measurements on the G-V were tailored to the standard flight patterns.

474 Figure 8 gives a broad overview of the INP data sets by focusing on the IS data collected during475 the four ship campaigns. Key findings are the large variability of, but generally very low, INP

476 concentrations at any particular latitude, a weak overall latitudinal dependence, with highest 477 concentrations near land masses (especially toward Australia), and the large discrepancy with 478 historical measurements over the region, first pointed out in the CAPRICORN I study by 479 McCluskey et al. (2018b). McCluskey et al. (2018b) demonstrated that INP concentrations were 480 up to 100 times lower during CAPRICORN I than measured by Bigg (1973) over some of the same 481 regions, that INPs were (excepting episodic events) often predominately organic in nature with 482 contributions of both heat labile and more stable organics, and that the INP content of Austral 483 summer SO seawater samples were lower than those found in Arctic seawater. These results are 484 consistent with a primary ocean sea spray source of SO BL INPs and also lower derived INP site 485 densities (INPs per aerosol surface area) for immersion freezing in SO air compared to north 486 Atlantic air masses. Using CAM-5 with constrained meteorology, McCluskey et al. (2019) 487 demonstrated that using parameterizations linking the number concentrations of mineral dust and 488 surface area of sea spray aerosols in the global aerosol model could predict the magnitude of INPs 489 observed in CAPRICORN I, and that sea spray organic INPs dominated on average, but that 490 episodic incursions of inorganic mineral dust INPs present in the middle troposphere could occur 491 and then dominate ice nucleation in the MBL. This vertical structure of compositions is 492 demonstrated in analyses of collected aerosol compositions above, below and within clouds during 493 SOCRATES (Twohy et al. 2020).

The INP data sets remain to be fully explored to investigate spatial, temporal and compositional variabilities, through aligning with aerosol data including real-time bioaerosol and Next-Generation DNA sequencing of bacteria. Those bacterial sequencing analyses have been completed for aerosol samples collected on equivalent filters to the INP units during CAPRICORN-2. Results reported in Uetake et al. (2020) indicate the predominance of marine

23

bacteria in the MBL during the ship campaign, confirming the pristine marine source of aerosols,
and thus INPs, under most circumstances in this region. Comprehensive INP data from all SO
studies will ultimately be normalized for use in parameterization development (see, e.g.,
McCluskey et al., 2018c; 2019; Vignon et al. 2020), and will serve as a basis for constraining
primary ice nucleation for comparison with observations of ice formation and numerical model
simulations of SO clouds.

505 *3.v. Clouds: In-situ observations of variability in liquid cloud droplet number concentration* 

506 In-situ G-V observations allow for process studies to investigate aerosol-cloud interactions and 507 processes controlling distributions of SLW. For example, using data obtained during 4 ramped 508 ascents and descents through BL clouds, Figure 9 shows  $N_c$  measured by the CDP as a function 509 of altitude. Although all profiles were collected in a similar geographical area on 2 different days, there is considerable variability in  $N_c$ , ranging from less than 50 cm<sup>-3</sup> near cloud top on RF08 at 510 latitude 55.8°S to greater than 450 cm<sup>-3</sup> near the top and in mid-cloud layer for the same flight 511 further south at 58.7°S. Although some lower  $N_c$ , such as concentrations of about 50 cm<sup>-3</sup> seen on 512 513 RF04, were associated with lower wind speeds averaging 5.5 m s<sup>-1</sup>, and some higher concentrations of 250 cm<sup>-3</sup> on RF08 at 59.9°S and up to 450 cm<sup>-3</sup> on RF08 at 58.7°S were associated with larger 514 wind speeds averaging 20.5 and 22.0 m s<sup>-1</sup> respectively, correlation with wind speed was not 515 516 always the case (e.g., low  $N_c$  of less than 50 cm<sup>-3</sup> on RF08 at 55.8°S occurred when wind speeds 517 were 26.6 m s<sup>-1</sup>) as updrafts, dynamics, turbulence and coupling of the cloud with the surface layer 518 can also affect N<sub>c</sub>. Thus, while generation of sea-salt CCN caused by breaking waves associated 519 with high winds likely contribute to variations in  $N_c$ , other factors also contribute significantly, 520 such as the influence of source regions with different bioactivity on the production of CCN and 521 the degree of coupling between the surface and cloud.

Accepted for publication in Bulletin of the American Meteorological Society. DOI 10.1175/BAMS-D-20-0132.1.

### 522 *3.vi. Variable but prevalent supercooled water observed in-situ*

523 Although SLW dominated many BL clouds observed during SOCRATES, information about 524 ice crystals, when present, is important for understanding SLW persistence and cloud glaciation. 525 During SOCRATES ice particle number concentrations and high-resolution images were acquired 526 over a large range of temperature with optical array probes and the PHIPS probe. Using cloud 527 phase determined with a combination of in-situ cloud probes (D'Alessandro et al. 2020, In 528 preparation), Figure 10 shows the distributions of phases as a function of temperature. In some 529 instances, even the identification of phase is poorly defined (e.g., Korolev et al. 2017) as, for 530 example, there is no consensus on how many ice crystals need to be mixed within a sample volume 531 of water drops to be mixed- rather than a liquid-phase cloud. For analysis of in-situ data, the term 532 mixed-phase refers to the occurrence of a liquid mass fraction between 0.1 and 0.9 in a 1 s time 533 period as calculated using data from a combination of size-resolved and bulk mass in-situ probes. 534 In addition to the frequent presence of clouds made exclusively of SLW at very low temperatures, 535 another notable feature was the frequent observation of glaciated clouds at relatively high temperature corresponding to the Hallett-Mossop (H-M) range of  $-2^{\circ}C < T < -8^{\circ}C$ . Figure 11 536 537 shows a collection of representative ice particles images captured by the PHIPS during RF02 538 between 0 and -5°C. Typical ice particle habits were needles that were frequently rimed, thus 539 acting as possible rime splintering sources in the H-M process. Smaller (D<100  $\mu$ m) pristine 540 hexagonal columns and plates were also observed that possibly grew from ice splinters (Korolev 541 et al., 2020) (Fig. 11, first row). Some of the pristine small particles were observed to have been 542 scavenged by the larger needles and needle aggregates (Fig. 11 third row, second needle from the

25

Accepted for publication in Bulletin of the American Meteorological Society. DOI 10.1175/BAWS-D-20-0132.1.

543 left). Occasionally, frozen drizzle droplets were detected – either as complete or sometimes as
544 fractured particles (Fig. 11, second row).

545 The presence of small horizontal scale generating cells were noted near the tops of BL 546 stratocumulus and higher cloud layers in the free troposphere. Such cells are small regions of high 547 reflectivity that frequently produce precipitation streaks below. Although such cells have been 548 observed in other environments, such as the Arctic (McFarquhar et al. 2011) and mid-latitudes 549 (Plummer et al. 2014), the cells observed over the SO had smaller horizontal scales and thus their 550 structure and properties need to be determined to understand precipitation development and cloud 551 life cycles Wang et al. (2020) provide this characterization using times when the G-V was flying 552 near cloud top. Figure 12 shows an example of their approach whereby the probability distribution 553 functions of liquid water content, total concentration, and ice water content were compared inside 554 and outside of generating cells identified by the HCR. All three parameters are higher to a 555 statistically significant degree inside the cells, but substantial liquid water and numbers of particles 556 also occur outside the cells. This shows that although the cells provide a favorable environment 557 for particle nucleation and growth, turbulent mixing at cloud top reduces the gradients inside and 558 outside of the cells. The on-line supplement gives extra information on how the combination of 559 in-situ and remote sensing measurements can be used to identify and characterize the fine-scale 560 structure of SLW, SLD, drizzle and ice crystal type.

The location of liquid water is of interest not only for understanding cloud microphysical process and radiative properties, but also for assessing the potential hazard posed by aircraft icing when the liquid is supercooled. Icing is a significant hazard for aviation, especially over the SO, and is most concerning as the droplets become large enough to impact on areas not typically protected by anti-icing or de-icing systems. Small cloud droplets (D< 50  $\mu$ m) tend to impact on forward edges of aircraft as seen in Figure 13, while larger drops tend not to freeze on impact and instead flow back farther before freezing or else are heavy enough to be somewhat independent of the airflow and actually impact the aircraft behind the forward edges (Figure 13b) (FAA, 2015; Cober and Isaac, 2012). Known as supercooled large drops (SLD), they can accrete on the wing and other important control areas of the aircraft which are outside the heated surfaces. Freezing drizzle and freezing rain are examples of SLD.

572 3.vii. Observing secondary ice production (Rime Splintering) over SO

573 Two research flights (RF11 and RF15) during SOCRATES were dedicated to the sampling of 574 shallow cumulus clouds in the cold sector of extratropical cyclones to understand the possible maintenance of SLW in those clouds. Mossop (1970) had found ample evidence of secondary ice 575 576 production by rime-splintering in cumuli sampled off the western and eastern coasts of Tasmania. 577 Because of the need to focus sampling at multiple levels in the same cumulus field, there was 578 insufficient time to sample the cumuli using the standard curtain flight pattern to  $60^{\circ}$ S. Thus a 579 population of cells as far south as possible, near 55° S, was identified for sampling. Thereafter the 580 G-V flew a series of constant altitude legs about 15 min long targeting the tops of actively growing 581 cells, and also sampling at and below cloud bases, and above the cloud tops, to measure aerosol, 582 CCN and INPs. These two SOCRATES flights provided clear evidence of rime-splintering, farther 583 away from land sources than documented before. Of the 34 sampled shallow cumuli occupying 584 temperatures where rime-splintering can act (-3 to -9  $^{\circ}$ C), 47% contained regions where ice crystals 585 were orders of magnitude more than the INP observed (Scott 2019). The SOCRATES airborne 586 radar data captured the cloud macrostructure needed to place the in situ microphysical data 587 collected near the cloud tops in context (Fig. 14). A complex, multi-thermal structure was common 588 in clouds exhibiting the features of rime-splintering, and lacking in clouds that only contained

Accepted for publication in Bulletin of the American Meteorological Society. DOI110.1175/BAMS-D-20-0132.1.

589 SLW. These new data are being used to guide and constrain detailed process-level numerical 590 modeling, to understand why some SO cumuli glaciate by this mechanism, while others do not.

## 591 3.viii. Himawari-8 retrievals consistent with field observations

592 Satellite data provide both a large-scale context for interpretation of finer resolution remote 593 sensing data and in-situ measurements. The Himawari-8 satellite, developed and operated by the 594 Japan Meteorological Agency, has provided a significant advance in geostationary satellite 595 capability over the Asia-Oceania region. It provided rapid updates on meteorological conditions 596 and cloud systems throughout the SO campaigns which were especially critical for aircraft 597 operations.

598 Figure 15a shows the flight tracks of the 15 SOCRATES missions and the outermost 599 boundaries of the 15 sectors used for the Himawari-8 analysis. Figure 15b shows the frequency of 600 occurrence of Himawari-8 cloud type (Pavolonis 2010) as a function of cloud-top temperature 601 (Heidinger 2011) for the 15 SOCRATES flights. For the duration of each flight, data are taken 602 from a rectangular area that extends from  $45^{\circ}$ S to  $63^{\circ}$ S and covers the entire width of the flight 603 track. The statistics represent the overall atmospheric and cloud conditions sampled during both 604 the in-situ and remote-sensing sampling legs. Figure 15 highlights the prevalence of SLW cloud 605 tops for  $0^{\circ}C > T > -25^{\circ}C$ . More details of this Himawari-8 cloud classification can be found in 606 Huang et al. (2019), and information about how the in-situ cloud properties are being used to 607 evaluate cloud microphysical properties is included in the on-line supplement.

608 3.ix. Radiative Fluxes Confirm Bias in Climate Models

Many studies involving surface radiative fluxes rely on fluxes retrieved from satellites, primarily from the Clouds and the Earth's Radiant Energy System (CERES) instruments or derived from spaceborne cloud radar and lidar observations (CloudSat-CALIPSO). Based on CERES data, 612 most climate models participating in CMIP5 had excessive SW radiation reaching the surface over 613 the SO (Zhang et al. 2016). An evaluation of CERES Synoptic (SYN) and Energy-Balanced and 614 Filled (EBAF) Edition 4 and CloudSat retrieved surface SW and longwave (LW) downwelling 615 fluxes against surface observations collected during MICRE (Hinkelman and Marchand 2020) 616 finds that the overall biases in the CERES-surface fluxes are modest, but slightly larger at Macquarie Island than at most other locations, approximately +10 Wm<sup>-2</sup> for the SW and -10 Wm<sup>-</sup> 617 618 <sup>2</sup> for the LW in the annual mean. The SW bias is positive meaning that climate model biases in 619 downwelling SW fluxes are, if anything, slightly larger than previous studies suggest because 620 CERES downwelling fluxes may be a bit too large and models fluxes are larger yet. However, 621 both the SW and LW bias have significant seasonal and diurnal variations, with SW biases being near +20 Wm<sup>-2</sup> during the SH summer. Biases in LW fluxes are much larger at night (-16 Wm<sup>-2</sup>) 622 than during the day ( $< 2 \text{ Wm}^{-2}$ ) with significant seasonal variations controlled by the relative ratio 623 624 of daytime vs. nighttime, and consequently are largest during the SH winter. This thus confirms 625 that the climate model biases that motivated the projects are indeed real.

### 626 3.x. Low Clouds Responsible for Much of Climate Model Bias

627 Understanding the contributions of different cloud types to the surface SW radiation bias 628 in models is a major objective of these field campaigns, which complements the analysis of large-629 scale environments most conducive to such biases. Figure 16 shows the observed and modelled 630 surface cloud radiative effect (CRE) during CAPRICORN I for different cloud cover types using 631 the BoM ACCESS-C3 numerical weather prediction system (4 km horizontal resolution, no data 632 assimilation, downscaled from the regional 12-km resolution model), which was run for the 633 campaign period. The observed CRE is the difference between the measured downwelling 634 radiative flux at the surface and the simulated clear-sky downwelling radiative flux, accounting

635 for ocean albedo and the broadband infrared emissivity of seawater (e.g., Protat et al. 2017). Over the CAPRICORN I period, the mean SW CRE was -66.7 Wm<sup>-2</sup>, partially offset by a mean LW 636 CRE of 44.4 Wm<sup>-2</sup>, resulting in a mean net CRE of -22.3 Wm<sup>-2</sup> (Figure 16d). A 1-minute merged 637 638 cloud radar - lidar product from ship-based measurements was used to classify the observed cloud 639 profiles into different cloud cover types (Noh et al., 2019) at 1 h resolution to compare with model 640 outputs. Hours that contained only clear skies were classified as clear. Hours that contained more 641 than 30 minutes of clear skies were classified as "mostly clear". Because the cloud cover types 642 containing clouds overlapping low, mid and high-altitude slabs made up only 5% of all the 643 observations, they were grouped into a "thick" cloud type classification. Hours that contained at 644 least 15 minutes of precipitating clouds were classified as precipitating, even if one of the other 645 conditions was met. Lastly, hours that did not meet any of these conditions were classified as 646 "mixed".

647 During CAPRICORN I, 51% of the 697 observation hours were characterized by low 648 clouds, followed by multilayer (14%), precipitating (10%), mostly clear (10%), mixed (8%), thick 649 (5%), and clear conditions (2%). Large negative SW CREs are observed for precipitating, 650 multilayer and low cloud categories and these correspond to a mean positive SW CRE bias for all 651 three clouds types, meaning too much SW flux is reaching the surface in the model under these 652 conditions (Figure 16a). The negative SW CRE and positive SW CRE bias was partially offset by 653 positive LW CREs and a negative LW CRE bias for these cloud types (Figure 16b). This resulted 654 in a net negative CRE and a positive net CRE bias for precipitating, multilayer and low clouds 655 (Figure 16c). While smaller negative (positive) SW (LW) CREs were observed for the other cloud 656 types, the CRE biases for these had little impact on the overall CRE and CRE bias once weighted 657 by their respective frequency of occurrence (Figure 16d). For the measured SW, LW and net CRE,

low clouds were responsible for just over half of the total contribution during CAPRICORN I, with most of the remaining contributions from multilayer and precipitating clouds. Interestingly, however, low clouds were responsible for nearly all of SW, LW and Net CRE biases in ACCESS, highlighting again the need to focus our attention on better understanding these low clouds. This work is being extended to include the MARCUS, MICRE and SOCRATES observations.

663 *3.xi. Remote Sensing Data also shows prevalence of supercooled water* 

The on-line supplement summarizes previous studies that have used the CAPRICORN and 664 665 MARCUS data to determine the frequency of and sources of SLW over the SO. To further 666 understand processes responsible for the production and maintenance of SLW over the SO, and to 667 understand the seasonal and latitudinal dependence of cloud properties, the MARCUS cloud 668 retrievals were combined with a value added product developed to describe the environmental 669 quantities at the position of the AA at 10-min resolution. Parameters examined include ship 670 navigation parameters, local meteorological conditions, SST, location of the AA relative to the 671 oceanic polar front, lower tropospheric stability, marine cold air outbreak index, inversion height, 672 lifting condensation level, location relative to the center of the nearest cyclone, warm front and 673 cold front and location of air parcels 72 hours prior to their arrival at the ship computed from 674 HYSPLIT. Consistent with prior satellite retrievals, the MARCUS data show that low-level liquid 675 water clouds are ubiquitous over the SO and that much of the water is supercooled. For instance, 676 south of 60°S over 49% of non-precipitating clouds had cloud base T < 0°C and mean liquid water paths greater than 50 g m<sup>-2</sup> as measured by the microwave radiometer. Figure 17 shows an example 677 678 of the analysis illustrating how the properties of single-layer, non-precipitating clouds with bases 679 less than 3 km and greater than 500 km away from the nearest cyclone center varied depending on 680 whether the measurements were made north or south of 60°S. The retrievals show that with average

Accepted for publication in Bulletin of the American Meteorological Society. DOI110.1175/BAMS-D-20-0132.1.

cloud base T of about  $-10^{\circ}$ C south of 60°S and hence the location of the oceanic polar front, SLW must extensively exist even though there is less precipitable water than north of 60°S. Further, CCN concentrations and retrieved  $N_c$  peaked in December, but there were large variations over all seasons. Similar ongoing analysis is quantifying the dependence of cloud properties on environmental and aerosol conditions, from which processes responsible for SLW can be better elucidated.

# 687 3.xii. Precipitation observations

688 Recent evaluation studies of satellite rainfall products have highlighted large statistical 689 discrepancies (up to a factor 2) in zonal precipitation averages derived from GPM, CloudSat, and 690 the Global Precipitation Climatology Project (GPCP) south of 40°S and north of 40°N (Grecu et 691 al. 2016; Skofronick-Jackson et al. 2017). Shipborne disdrometer and active remote sensing 692 observations collected during CAPRICORN have been used along with others from several 693 research vessels as part of the OceanRAIN project (Klepp et al. 2018) to establish whether these 694 differences between satellite rainfall products are driven by latitudinal differences in statistical 695 properties of the drop size distribution (DSD) and associated assumptions in GPM radar rainfall 696 retrievals. Results from these investigations are summarized in Protat et al. (2019ab). A large 697 natural, latitudinal, and convective-stratiform variability of the DSD was clearly found, with a 698 much lower drop concentration for diameters smaller than 3 mm and a very different modal value 699 of the DSD shape parameter distribution ( $\mu$ ) to that assumed in the GPM algorithms in the SH 700 high latitude (south of 45°S) and NH polar latitude (north of 67.5°S) bands (Protat et al. 2019a). 701 From a radar rainfall retrieval perspective, the attenuation – reflectivity, drop diameter – 702 reflectivity, and rainfall rate – reflectivity relationships in the SH high latitude and NH polar 703 latitude bands are found to be fundamentally different from those at other latitude bands, producing

Accepted for publication in Bulletin of the American Meteorological Society. DOI 10.1175/BAMS-D-20-0132.1.

smaller attenuation, much larger drop diameters, and lower rainfall rates for a given reflectivity,
which potentially explains the observed discrepancies between satellite rainfall products (Protat et
al. 2019b). Evaluations of CloudSat and other satellite precipitation datasets using MICRE and
SOCRATES datasets are underway, and will be reported in future publications.

708 3.xiii. Unique view of BL Structure from Soundings

709 Across the four field campaigns, a total of 2,186 soundings were obtained. While a variety 710 of spatial and temporal biases exist in the sampling, the collection provides an unprecedented view 711 of the thermodynamic structure of the lower troposphere across the SO. A simple k-means cluster 712 analysis on the lower thermodynamic variables (T, relative humidity, winds (u and v) at 700, 850 713 and 925 hPa levels and surface pressure, T, and relative humidity) (Lang et al. 2018) produces a 714 cluster along the Antarctic coast (C2), another at high latitudes (C1,  $55-65^{\circ}$ S), where polar meso-715 vortices are commonly present, and multiple clusters at lower latitudes across the SO storm track 716 (40-60°S). Increasing the numbers of clusters effectively isolates different sectors of the mid-717 latitude storm track. For brevity the storm track clusters are merged into a single cluster (M).

718 The composite soundings for M, C1 and C2 (Figure 18a-c) illustrate differences in the 719 thermodynamic structure of the atmosphere across the SO. The M composite features strong 720 westerly winds and a low-level inversion near 900 hPa. BL clouds are commonly observed across 721 this region. C1 covers the region where the greatest bias exists in the regional energy budget 722 (Trenberth and Fasullo 2010) and it is also the region where multi-layer clouds are commonly 723 detected by A-train satellites (Mace et al. 2009). For the C1 composite (Figure 18b), the low-level 724 winds are very weak and the atmosphere is near saturation at all altitudes. A more complete analysis of individual soundings (not shown) confirms that multi-layer clouds are frequent, but 725 726 relatively few inversions are present, which suggests that the polar meso-vortices mix the lower

727 free troposphere and that the weak winds may allow for a radiative equilibrium to weaken 728 inversions. Finally, along the Antarctic coast (C2), the composite reveals a very dry, cold 729 atmosphere, commonly cloud free. An illustrative back trajectory (Figure 18d) illustrates that the 730 low-altitude dynamics may be dominated by strong Antarctic outflows such as katabatic winds 731 draining cold, dry air off the Antarctic plateau onto the SO. Using a cyclone tracking algorithm 732 (Lim and Simmonds 2007) on the ERA5 reanalysis, we plot the location of the soundings, by 733 cluster, to the nearest cyclonic core (Figure 18e). The M soundings typically reside to the north of 734 core, the C1 soundings commonly reside just poleward of the core, while the C2 soundings reside, 735 on average, about  $5^{\circ}$  south of polar meso-vortices (Truong et al. 2020).

### 736 3.xiv. Impact of Biological Particles on CCN/Droplet Concentrations near Antarctica

737 The on-line supplement summarizes the use of remote sensing data to derive cloud 738 microphysical properties (Mace and Protat 2018; Mace et al. 2020). In Figure 19, a time series of 739 daily mean  $N_c$  retrieved from non-precipitating liquid MBL clouds during CAPRICORN II is 740 shown. The  $N_c$  represents the mean value from an entire 24 hour period with the error bars showing 741 the standard deviation of the total number of successful retrievals for that day where each retrieval 742 is valid for a 30-second interval (Figure 19e). The latitude of the ship during the 24 hour period is shown in Figure 19d.  $N_c$  decreases steadily from > 100 cm<sup>-3</sup> as the RV Investigator travelled 743 744 south through the Tasman Sea to about 50 cm<sup>-3</sup> as the ship passed into the latitudes of the Antarctic 745 Circumpolar current.  $N_c$  increased by about a factor of 2 occurs on 29 January as the ship passed 746 poleward of 64°S. N<sub>c</sub> remained elevated while the RV Investigator worked along the Antarctic 747 shelf south of 60°S. Poor weather precluded retrievals until 13 February when  $N_c$  was again found to be in the 50 cm<sup>-3</sup> range with the ship working back north of 60°S.  $N_c$  did not climb as rapidly 748 749 with latitude moving northward towards Tasmania.

Accepted for publication in Bulletin of the American Meteorological Society. DOI 10.1175/BAWS-D-20-0132.1.

750 The daily mean  $N_c$  is correlated (r=0.48) with daily averages of sulfate and particulate 751 methanesulfonic acid (MSA) concentrations, but  $N_c$  is less well correlated with CCN at 0.65% 752 super saturation measured at the surface (r=0.38). It was also found (not shown) that  $N_c$  is 753 negatively correlated (r=-0.51) with chloride concentrations. What is reasonably striking in Figure 754 19 is that  $N_c$ , MSA and CCN all increase substantially poleward of 60 S. Because MSA is a marker 755 of DMS oxidation, it is concluded that the higher CCN concentrations in this region are likely 756 driven by the biologically productive latitudes along the Antarctic shelf. It is plausible that this 757 effect is in line with previous observations by Humphries et al. (2016) where increased secondary 758 aerosol formation was observed south of these latitudes.

759 *3.xv. Models Test Ubiquitous Supercooled Water and Role of Biological Particles over SO* 

760 Global-scale, regional-scale, and process-scale modelers were entrained into the SO 761 projects as they were designed. Scientists using the atmospheric component of NCAR's 762 Community Earth System Model version 2 (CESM2) (Danabasoglu et al., 2020) and GFDL's AM4 763 (Zhao et al. 2018) global models participated. The Australian ACCESS model (Puri et al. 2013) 764 was used for operational forecasting during SOCRATES, and its icing products were evaluated. Australian and U.S. groups ran the Weather Research and Forecast (WRF; Skamarock et al. 2005) 765 766 regional model. A large-eddy simulation (LES) model (Atlas et al. 2020) was run with a very fine 767 grid over small domains for selected cases, as was an idealized cloud-resolving model (CM1; 768 Bryan and Fritsch 2002) for process-level studies. Table S13 lists some modeling groups that 769 participated in the projects, as well as the approximate grid resolution; additional details about the 770 LES simulations are also included in the on-line supplement. Two particular foci of the modeling 771 studies were to test hypotheses that (a) the GCMs/NWP models were too quickly glaciating clouds that are in reality persistent SLW clouds, and (b) marine biogenic processes help sustain the natural
aerosol population over the SO.

774 From the start, the modeling team proposed a nudged-meteorology strategy (e. g. Wu et al. 775 2017) to effectively compare the global model with aircraft or ship data in the synoptically active 776 SO. As implemented, three-dimensional model fields of horizontal wind, temperature and surface 777 pressure were nudged toward a global reanalysis with a 24 hour relaxation timescale. The 778 simulated humidity, cloud and aerosol fields freely evolve and can be usefully compared with in-779 situ observations. Ideally, the temperature and wind fields from the nudged simulations and from 780 the reanalysis to which they are being nudged should closely match corresponding observations. 781 This was found to hold remarkably well. For instance, aircraft-measured temperatures were 782 typically within 1 K of the reanalysis and within 2 K of the nudged GCMs. Fig. 5 of Gettelman et 783 al. (2020) shows the example of RF07, in which CAM6 is nudged to the MERRA2 reanalysis. 784 Both MERRA2 and the ERA5 reanalysis used by the nudged AM4 are fine choices for the nudged-785 meteorology approach. ERA5, which input the G-V dropsonde data, was on average about 20% 786 closer to SOCRATES-observed temperature and winds than MERRA2. In addition, the regular 787 radiosonde observations made during MARCUS have been found to improve the forecast track of 788 a mid-latitude low pressure system (Sato el al., 2018).

Figs. 10 and 14 of Gettelman et al. (2020) show examples of comparisons of nudged versions of CAM6 and its predecessor version CAM5 with aircraft cloud microphysical observations from SOCRATES RF07. These show that CAM6 correctly simulates a BL stratocumulus layer that is observed to be primarily supercooled liquid, while CAM5 incorrectly simulates the same cloud layer to mainly be ice. CAM6 is also able to represent the structure of the hydrometeor size distributions (Figure 20, adapted from Figure 9 of Gettelman et al 2020),
but with biases remaining in the representation of the peak liquid size distribution, and in excessive
warm rain. These detailed comparisons allow a new process understanding of weather and climate
models from the in-situ microphysical to the climate scale (Gettelman et al 2020).

798 Both the NSF G-V used in SOCRATES and the RV Investigator in CAPRICORN II 799 gathered extensive vertically-pointing lidar and 94 GHz cloud radar datasets that sampled entire 800 atmospheric columns. Both the CAM6 and AM4 models include implementations of the COSP 801 simulator (Bodas-Salcedo et al. 2011), which includes a 94 GHz radar simulator. This enables a 802 comparison between these powerful remote sensing datasets and the nudged-meteorology GCMs, 803 discussed at length by Zhou et al. (2020). Their Fig. 12 shows such a comparison with the RV 804 Investigator radar data for 1-15 Feb. 2018. This is a sensitivity test of the model cloud 805 microphysics and representation of precipitation. It shows CAM6 has good skill, while AM4 806 greatly underestimates snow reflectivity because its assumed snow particle size is too small. The 807 ship radar often sampled precipitating clouds which the aircraft did not target (and which were 808 often precluded by icing hazard), so it provides complementary information to the in-situ data.

809 **4. Summary and Future Work** 

Motivated by a pressing issue on the absorption of too much solar radiation over the Southern Ocean (SO) (due to problems simulating low-altitude supercooled liquid clouds) by leading climate and numerical weather prediction models, a coordinated multi-agency effort consisting of four field campaigns was held in the time period of 2016 to 2018. The experimental design, platforms, and instruments from four experiments have been summarized here: the groundbased Macquarie Island Cloud Radiation Experiment (MICRE) collecting information on surface aerosol properties in-situ, and clouds, precipitation and radiation using remote sensors; the Clouds 817 Aerosols Precipitation Radiation and Atmospheric Composition over the SO (CAPRICORN) I and 818 II cruises of the RV Investigator that collected aerosol, in-situ and oceanographic measurements 819 in-situ and remotely; the Measurements of Aerosols, Radiation and Clouds over the Southern 820 Ocean (MARCUS) campaign that collected in-situ aerosol and remote sensing observations using 821 instruments installed on the icebreaker RSV Aurora Australis as it made resupply voyages from 822 Hobart to the Australian Antarctic stations and Macquarie Island; and the Southern Ocean Cloud 823 Radiation Transport Experimental Study (SOCRATES) that collected data with the NCAR/NSF 824 G-V aircraft in a north-south direction south of Hobart, Tasmania to approximately 62 S. These 825 data characterize the synoptically and seasonally-varying vertical structure of the SO BLand free 826 troposphere, including the properties of clouds and the variability and sources and sinks of 827 aerosols, cloud condensation nuclei and ice nucleating particles, to a much greater extent than was 828 previously available.

829 The experiments were designed to be complementary in how they contribute to studies of 830 processes, latitudinal variability, seasonal variability, validation of remote sensing retrievals and 831 model evaluation and improvement. MICRE gives a long seasonal record in a single location, 832 CAPRICORN I and II give the most complete ship-borne oceanographic, aerosol and surface 833 energy budget observations, MARCUS covers a longer seasonal cycle than CAPRICORN, and 834 SOCRATES provides the detailed in-situ observations that are required for process-oriented 835 understanding. Combined these data represent the most comprehensive set of data collected on 836 aerosols, clouds and precipitation over the SO over seasonal cycles, especially over cold sectors 837 of extratropical cyclones and at latitudes below 60°S where climate model biases are largest. The 838 related modeling studies tested hypotheses on the cloud processes that lead to the ubiquity of

supercooled clouds and the marine biogenic processes that sustain the natural aerosol populationover the SO.

841 Some first findings from the field campaigns addressing their overarching objectives have 842 been presented here, and are being elaborated upon in several more focused scientific articles. 843 Initial findings included that low clouds were responsible for nearly all the radiative biases in the 844 Australian forecast model ACCESS and the presence of a pristine environment with numerous 845 small and few large aerosols above cloud, highlighting the role of new particle formation in the 846 troposphere and the long-range transport from continents. There is a dearth of INPs (much lower 847 than suggested by much earlier measurements by Biggs 1973) which is a significant factor leading to the ubiquitous presence of supercooled liquid water over the SO. Most INPs appear to have a 848 849 biological source and better understanding of secondary ice nucleating processes related to these 850 particles is needed. Further, there was a suggestion that the higher CCN concentrations south 60° 851 S were likely driven by biologically productive latitudes along the Antarctic shelf, but that sea 852 spray may have more important roles in other latitudinal bands. In the cold dry sectors of cyclones, 853 supercooled liquid water with contents as high as about 0.8 g m<sup>-3</sup> were observed in very thin layers 854 at temperatures as low as -30°C and was frequently associated with the presence of narrow cloud-855 top generating cells. Evaluation of satellite datasets is ongoing, but early results suggest the 856 CERES shortwave fluxes and imager-based (Himawari and MODIS) retrievals for low cloud 857 microphysical properties for stratocumlus are reasonably good, and pointing towards ways in 858 which retrievals of precipitation and cloud-phase among other may quantities might be refined or 859 improved. Finally, ongoing modeling and observations studies are examining how CCN properties 860 are coupled with aerosol properties and meteorological conditions, in order to provide a process-

861 oriented understanding that can be used to improve the performance of models at a variety of862 spatial and temporal scales.

863 In terms of the motivating goal, namely the overprediction of solar radiation over the SO, 864 the hypothesis that SLW is ubiquitous is confirmed. Although measurements verified that there 865 was a dearth of INPs over the SO, the exact mechanisms by which SLW persists over the SO, and 866 the interplay of aerosols, dynamics and meteorology in this persistence are still somewhat 867 uncertain. Now that all data have been processed and conditions over the SO have been 868 characterized, integration of different data sets and comparison against models and satellite 869 retrievals is proceeding rapidly which should lead to a more integrated view of the abundance of supercooled water and its role in reflecting solar radiation to reduce the observed radiative bias. 870

871 Inevitably, the collected data sets have limitations with their temporal and spatial coverage. 872 Use of the data to evaluate and improve satellite retrieval schemes will extend the impact of these 873 SO datasets. Nonetheless, it will be advantageous to collect future aircraft and ship-based datasets 874 over the SO. In order for future data to have the maximum impact, it could be desirable to use a 875 Lagrangian approach to aircraft data collection rather than the Eulerian approach used during 876 SOCRATES. Although the Eulerian approach was beneficial for characterizing the SO 877 environment, a Largrangian approach would allow for better understanding of how clouds evolve 878 over longer periods of time by tracing their evolution on subsequent days. In addition, a focus on 879 the transition season where there is a greater variability in the strength of phytoplankton blooms, 880 and the winter seasons where biological activity is low would allow for testing on hypotheses 881 related to the impact of biogenic aerosol species and generally provide a more thorough 882 understanding of seasonal differences. More comprehensive measurements of aerosol chemical 883 properties as well as of cloud particles with sizes between 50 and 150 µm, perhaps through holographic probes, would also be beneficial. Nevertheless, the publicly available CAPRICORN,

885 MICRE, MARCUS and SOCRATES data significantly extend the availability of data on cloud,

- precipitation and aerosol properties over the SO, and will offer rich datasets for future studies.
- 887

#### 888 Acknowledgments

889 This work was supported by the National Science Foundation (NSF) through grants AGS-

890 1628674 (GM) and AGS-1762096 (GM) and by the United States Department of Energy through

grant DE-SC0018626. PJD, TCJH, and KAM acknowledge NSF grant AGS-1660486 and DOE

- grant DE- SC0018929. KAM acknowledges support by an NSF Graduate Research Fellowship
- under Grant No. 006784. JU was supported by the National Research Foundation of Korea

894 (NRF) grant funded by the Korean government (MSIT) (No. 2020R1A2C1013278) and by Basic

- 895 Science Research Program through the NRF funded by the Ministry of Education
- 896 (No. 2020R1A6A1A03044834). CM was supported primarily by the National Center for
- 897 Atmospheric Research and received travel support from NSF AGS-1660486. The material in the

article is based upon work supported by the National Center for Atmospheric Research, which is

a major facility sponsored by the NSF under Cooperative Agreement No. 1852977. The data

900 were collected using NSF's Lower Atmosphere Observing Facilities, which are managed and

901 operated by NCAR's Earth Observing Laboratory. The efforts of the entire SOCRATES,

902 MARCUS, MICRE and CAPRICORN teams in collecting the high quality data sets are

903 appreciated. Technical, logistical and ship support for MARCUS and MICRE were provided by

- the AAD through Australian Antarctic Science projects 4431, 4292 and 4387, and we thank
- 905 Steven Whiteside, Lloyd Symonds, Rick van den Enden, Peter de Vries, Chris Young, Chris
- 906 Richards, Terry Egan, Nick Cartwright and Ken Barrett for assistance. Logistical and financial

41

907 support was provided for CAPRICORN by the Australian Marine National Facility. Anne Marie 908 Rauker is acknowledged for assistance in INP data processing. Anne Perring is acknowledged 909 for the use of her WIBS-4A during CAPRICORN II. Paul Selleck is acknowledged for his work 910 with the ToF-ACSM during CAPRICORN II. Robyn Schofield also acknowledges support from 911 the Australian Research Council's DP160101598, LE150100048 and CE170100023 grants. The 912 SOCRATES Principal Investigators would like to thank the BoM Tasmanian regional Office for 913 the excellent forecast support and weather briefings provided during the field campaign (with 914 special thanks to Scott Carpentier, Michelle Hollister, Matthew Thomas and Robert Schaap). We 915 thank the Atmospheric Radiation Measurement (ARM) Program sponsored by the U.S. DOE, 916 Office of Science, Office of Biological and Environmental Research, Climate and Environmental 917 Science Division for their support. Any opinions, findings, and conclusions or recommendations 918 expressed in this material are those of the author(s) and do not necessarily reflect the views of 919 the funding agencies.

920

#### 921 Data Availability

922 Copies of all Atmospheric Radiation Measurement (ARM) Program instrument-level data 923 collected during MARCUS and MICRE are permanently stored and available via the ARM data 924 archive (https://adc.arm.gov/). Data from AAD, BoM and CSIRO instruments deployed alongside 925 MARCUS and MICRE instrumentation are available from the Australian Antarctic Data Centre 926 following registration. Copies of non-ARM instrument-level data and derived (multi-instrument) 927 fields (such as cloud liquid water path and effective radius, precipitation particle type) will 928 eventually be available through ARM archive as a primary investigator data (PI data). As of the 929 time this article is being written, the processing of these data is not yet complete but are available 930 at https://atmos.uw.edu/~roj/nobackup/MARCUS\_and\_MICRE/Datasets/. The author(s) wish to acknowledge the SOCRATES Project and the SOCRATES Data Archive Center at NCAR's Earth 931

932	Observing	Laboratory,	https://data.eol.ucar.edu/master_lists/generated/socrates/.	The
933	CAPRICORN	N datasets are av	vailable on the CSIRO Data Access Portal https://data.csiro.au/	<u>'dap</u> .
934				

935 Appendix:

- A list of all the abbreviations used in the main text of the manuscript is provided here.
- 937 AA: RSV Aurora Australis
- 938 AAD: Australian Antarctic Division
- 939 AMF2: ARM Marine Facility 2
- 940 AR: Atmospheric River
- 941 ARM: Atmospheric Radiation Measurement
- 942 BL: Boundary Layer
- 943 BoM; Australian Bureau of Meteorology
- 944 CALIPSO: Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation
- 945 CAPRICORN: Clouds Aerosols Precipitation Radiation and Atmospheric Composition over the
- 946 Southern Ocean
- 947 CCN: Cloud Condensation Nucleus
- 948 CDP: Cloud Droplet Probe
- 949 CERES: Clouds and the Earth's Radiant Energy System
- 950 CFAD: Contour Frequency by Altitude Diagram
- 951 CFDC: Continuous Flow Diffusion Chamber
- 952 CMIP5: Coupled Model Intercomparison Project Phase 5
- 953 CSU: Colorado State University
- 954 DMS: Dimethyl Sulfide
- 955 DOE: Department of Energy

43

956	DSD:	Drop Size Distribution
957	ECMWF:	European Center for Weather Forecasting
958	ERA-5	ECMWF Re-Analysis 5
959	FBAP:	Fluorescent Biological Aerosol Particle
960	FT:	Free Troposphere
961	GCM:	General Circulation Model
962	GPCP	Global Precipitation Climatology Project
963	GPM:	Global Precipitation Measurement
964	G-V:	Gulfstream-V Aircraft
965	HCR:	HIAPER W-band Cloud Radar
966	HGF:	Hygroscopic Growth Factor
967	HIAPER:	High-performance Instrumented Airborne Platform for Environmental Research
968	HSRL:	High Spectral Resolution Lidar
969	HYSPLIT:	Hybrid Single Particle Lagrangian Integrated Trajectory Model
970	INP:	Ice Nucleating Particle
971	IS:	Ice Spectrometer
972	ITCZ:	Intertropical Convergence Zone
973	JAXA:	Japan Aerospace Exploration Agency
974	LW	Longwave
975	MARCUS	Measurements of Aerosols, Radiation and CLouds over the Southern Ocean
976	MBL:	Marine Boundary Layer
977	MICRE:	Macquarie Island Cloud Radiation Experiment
978	MNF :	Australian Marine National Facility

979	MSA:	Methanesulfonic Acid
980	Na:	Aerosol concentration
981	Nc:	Cloud droplet number concentration
982	NASA:	National Aeronautics and Space Administration
983	NCAR:	National Center for Atmospheric Research
984	NH:	Northern Hemisphere
985	NOAA:	National Oceanographic and Atmospheric Administration
986	NSF:	National Science Foundation
987	NWP:	Numerical Weather Prediction
988	PHIPS:	Particle Habit Imaging and Polar Scattering probe
989	PIP:	Precipitation Imaging Probe
990	RF:	Research Flight
991	RPF:	Recent Particle Formation
992	SLW:	Supercooled Liquid Water
993	SO:	Southern Ocean
994	SOCRATES:	Southern Ocean Cloud Radiation Aerosol Transport Experimental Study
995	SST:	Sea Surface Temperature
996	STEM:	Scanning Transmission Electron Microscopy
997	SW:	Shortwave
998	T:	Temperature
999	UHSAS:	Ultra High Sensitivity Aerosol Sampler
1000	VOC:	Volatile Organic Carbon
1001	WIBS4:	Waveband Integrated Bioaerosol Sensor 4

- 1002 WRF: Weather Research and Forecasting Model
- 1003 2DC: Two-Dimensional Cloud Probe
- 1004 2DS: Two-Dimensional Stereo Probe
- 1005  $\lambda$ : Wavelength of Radiation
- 1006
- 1007

#### 1008 **REFERENCES**

- 1009 Ahn, E., Y. Huang, T.H. Chubb, D. Baumgardner, P. Isaac, M. de Hoog, S. T. Siems, M.
- 1010 Manton, 2017: In situ observations of wintertime low-altitude clouds over the Southern
- 1011 Ocean. Quart. J. Roy. Meteor. Soc., 143, 1381–1394, https://doi.org/10.1002/qj.3011
- 1012 Andreae, M. O., W. Elbert and S. J. Demora, 1995: Biogenic sulfur emissions and aerosols over
- 1013 the tropical south-atlantic.3. Atmospheric dimethylsulfide, aerosols and cloud
- 1014 condensation nuclei. J. Geophys. Res 100, 11335-
- 1015 11356, <u>https://doi.org/10.1029/94jd02828</u>.
- 1016 Andreas, A, M Dooraghi, A Habte, M Kutchenreiter, I Reda, and M Sengupta. 2018. Solar
- 1017 Infrared Radiation Station (SIRS), Sky Radiation (SKYRAD), Ground Radiation
- 1018 (GNDRAD), and Broadband Radiometer Station (BRS) Instrument Handbook, U.S.
- 1019 Department of Energy. DOE/SC-ARM-TR-025.
- 1020 https://www.arm.gov/publications/tech\_reports/handbooks/sirs\_handbook.pdf
- 1021 Angulo-Martinez, M., S. Begueria, B. Latorre, and M. Fernandez-Raga, 2018: Comparison of
- 1022 precipitation measurements by OTT Parsivel and Thies LPM optical disdrometers.
- 1023 *Hydrol. Earth Syst. Sci.*, **22**, 2811-2837, <u>https://doi.org/10.5194/hess-22-2811-2018</u>.
- 1024 Atlas R. L., C. S. Bretherton and P. N. Blossey, 2020: How well do high and low resolution
- 1025 models represent observed boundary layer structures and low clouds over the
- 1026 summertime Southern Ocean? J. Geophys. Res. (to be submitted 5/2020).
- Ayers, G. P. and Gras, J. L., 1991: Seasonal relationship between cloud condensation nuclei and
  aerosol methanesulfonate in marine air. *Nature*, **353**, 834–835.

47

1029	Bates, T. S., B. J. Huebert, J. L. Gras, F. B. Griffiths, and P. A. Durkee, 1998a: International
1030	Global Atmospheric Chemistry (IGAC) Project's First Aerosol Characterization
1031	Experiment (ACE 1): Overview. J. Geophys. Res., 103, 16297-16318,
1032	https://doi.org/10.1029/97JD03741.
1033	Bates, T.S., V.N. Kapustin, P.K. Quinn, D.S. Covert, D.J. Coffman, C. Mari, P.A. Durkee, W.
1034	DeBruyn, and E. Saltzman, 1998: Processes controlling the distribution of aerosol
1035	particles in the lower marine boundary layer during the First Aerosol Characterization
1036	Experiment (ACE-1). J. Geophys. Res., 103, 16369-16384,
1037	https://doi.org/10.1029/97JD03720.
1038	Bates, T.S., P.K. Quinn, D.S. Covert, D.J. Coffman, J.E. Johnson, and A. Wiedensohler, 2000:
1039	Aerosol physical properties and processes in the lower marine boundary layer: A

1040 comparison of shipboard sub-micron data from ACE 1 and ACE 2. *Tellus*, **52**, 258- 272,

1041 <u>https://doi.org/10.3402/tellusb.v52i2.16104</u>.

Bigg, E. K., 1973: Ice nucleus concentrations in remote areas, J. Atmos. Sci., 30, 1153–1157,
 https://doi.org/10.1175/1520-0469(1973)030<1153:INCIRA>2.0.CO;2

1044 Bodas-Salcedo, A., M. J. Webb, S. Bony, H. Chepfer, J. L. DuFresne, S. A. Klein, Y. Zhang, R.

1045 Marchand, J. M. Haynes, R. Pincus, and V. O. John, 2011: COSP: Satellite simulation

1046 software for model assessment. *Bull. Amer. Meteor. Soc.*, **92**, 1023-1043,

1047 <u>https://doi.org/10.1175/2011BAMS2856.1</u>

1048 Bodas-Salcedo, A., K.D. Williams, M.A. Ringer, I. Beau, J.N.S. Cole, J.-L. Dufresne, T.

1049 Koshiro, B. Stevens, Z. Wang, and T. Yokohata, 2014: Origins of the solar radiation

48

- 1050 biases over the Southern Ocean in CFMIP2 models. J. Climate, 27, 41–56,
- 1051 https://doi.org/10.1175/JCLI-D-13-00169.1.
- 1052 Bodas-Salcedo, A., T. Andrews, A.V. Karmalkar, and M.A. Ringer, 2016: Cloud liquid water
- 1053 path and radiative feedbacks over the Southern Ocean. *Geophys. Res. Lett.*, **43**, 10938-
- 1054 10946, doi:10.1002/2016GL070770.
- 1055 Boers, R., J. B. Jensen, P. B. Krummel, and H. Gerber, 1996: Microphysical and short-wave
- 1056 radiative structure of wintertime stratocumulus clouds over the Southern Ocean. *Quart. J.*
- 1057 Roy. Meteor. Soc., **122**, 1307–1339, <u>https://doi.org/10.1002/qj.49712253405</u>.
- 1058 Boers, R., J. B. Jensen, and P. B. Krummel, 1998: Microphysical and short-wave radiative
- structure of stratocumulus clouds over the Southern Ocean: Summer results and seasonal
- 1060 differences, Quart. J. R. Meteor. Soc., **124**, 151–168,
- 1061 https://doi.org/10.1002/qj.49712454507.
- 1062 Bryan, G. H. and J.M. Fritsch, 2002: A Benchmark Simulation for Moist Nonhydrostatic
- 1063 Numerical Models. *Monthly Weather Review*, **130**, 2917–2928,
- 1064 <u>https://doi.org/10.1175/1520-0493(2002)130<2917:ABSFMN>2.0.CO;2.</u>
- Burrows, S. M., C. Hoose, U. Pöschl and M.G. Lawrence, 2013: Ice nuclei in marine air:
  biogenic particles or dust? *Atmos. Chem. Phys.*, 13, 245–267. <u>https://doi.org/10.5194/acp-</u>
- 1067 <u>13-245-2013</u>.
- 1068 Carslaw, K, A. Leel, C. L. Reddington, K. J. Pringle, A. Rap, P. M. Forster, G. W. Mann1, D. V.
- 1069 Spracklen, M. T. Woodhouse, L. A. Regayre1 and J. R. Pierce, 2013: Large contribution
- 1070 of natural aerosols to uncertainty in indirect forcing. *Nature*, **503**, 67-71,
- 1071 <u>http://doi:10.1038/nature12674</u>.

- 1072 Carslaw, K. S., O. Boucher, D. V. Spracklen, G. W. Mann, J. G. L. Rae, S. Woodward, and M.
- 1073 Kulmala, 2010: A review of natural aerosol interactions and feedbacks within the Earth 1074 system. *Atmos. Chem. Phys.*, **10**, 1701–1737, https://doi.org/10.5194/acp-10-1701-2010
- 1075 Carslaw, K.S., L. A. Lee, C. L. Reddington, K. J. Pringle, A. Rap, P. M. Forster, G. W. Mann, D.
- 1076 V. Spracklen, M. T. Woodhouse, L. A. Regayre, and J. R. Pierce, 2013: Large
- 1077 contribution of natural aerosols to uncertainty in indirect forcing. *Nature*, **503**, 67-71,
- 1078 <u>https://doi.org/10.1038/nature12674</u>.
- 1079 Ceppi, P., M. D. Zelinka, and D. L. Hartmann, 2014: The response of the Southern Hemispheric
- 1080 eddy-driven jet to future changes in shortwave radiation in CMIP5, *Geophys. Res. Lett.*,
- 1081 **41**, 3244–3250, doi:10.1002/2014GL060043.
- Choi, Y. S., C.H. Ho, S.W. Kim, and R.S. Lindzen, 2010: Observational Diagnosis of Cloud Phase
   in the Winter Antarctic Atmosphere for Parameterizations in Climate Models. *Adv. Atmos.*
- 1084 *Sci.*, **27**, 1233-1245. doi: 10.1007/s00376-010-9175-3.
- 1085 Chubb, T., Y. Huang, J. Jensen, T. Campos, S. Siems, and M. Manton, 2016: Observations of
- 1086 high droplet number concentrations in Southern Ocean boundary layer clouds, *Atmos.*
- 1087 Chem. Phys., 16, 971–987, <u>https://doi.org/10.5194/acp-16-971-2016.</u>
- 1088 Chubb, T. H., J.B. Jensen, S.T. Siems, and M.J. Manton, 2013: In situ observations of
- 1089 supercooled liquid clouds over the Southern Ocean during the HIAPER Pole-to-Pole
- 1090 Observation campaigns, *Geophys. Res. Lett.*, **40**, 5280–5285,
- 1091 <u>https://doi.org/10.1002/grl.50986</u>.
- 1092 Clarke, A. D., 1993: Atmospheric Nuclei in The Pacific Midtroposphere Their Nature,
- 1093 Concentration, and Evolution. J. Geophys. Res., 98, 20633–
- 1094 20647, <u>https://doi.org/10.1029/93jd00797</u>.

- 1095 Clarke, A. D., J.L. Varner, F. Eisele, R.L. Mauldin, D. Tanner and M. Litchy, 1998: Particle
- 1096 production in the remote marine atmosphere: Cloud outflow and subsidence during ACE
- 1097 1. J. Geophys. Res., 103, 16397-16409, <u>https://doi.org/10.1029/97JD02987</u>.
- 1098 Clement, C. F., I. Ford, J., Twohy, C. H., A. Weinheimer, and T. Campos, 2002: Particle
- 1099 production in the outflow of a midlatitude storm. J. Geophys. Res., **107**, AAC 5-1-AAC
- 1100 5-9, https://doi.org/10.1029/2001JD001352.
- 1101 Cober, S.G., and G.A. Isaac, 2012: Characterization of aircraft icing environments with
- supercooled large drops for application to commercial aircraft certification. J. Appl.
- 1103 *Meteor.*, **51**, 265-284, https://doi.org/10.1175/JAMC-D-11-022.1.
- 1104 Covert, D. S., V.N. Kapustin, P.K. Quinn, and T. S. Bates, 2002: New Particle Formation in The
- 1105 Marine Boundary-Layer. J. Geophys. Res., 97, 20581–
- 1106 20589, <u>https://doi.org/10.1029/92jd02074</u>.
- 1107 Covert, D. S., V.N. Kapustin, T.S. Bates, and P.K. Quinn, 1996: Physical properties of marine
- boundary layer aerosol particles of the mid-Pacific in relation to sources and
- 1109 meteorological transport. J. Geophys. Res., 101, 6919-6930,
- 1110 https://doi.org/10.1029/95JD03068.
- 1111 D'Alessandro, J., G. McFarquhar, W. Wu, J. Stith, M. Schnaiter, and E. Jaervinen, 2020: Spatial
- 1112 heterogeneity of liquid, ice and mixed phase low-level clouds over the Southern Ocean
- 1113 derived using in situ observations acquired during SOCRATES. J. Geophys. Res., In
- 1114 preparation.
- 1115 Danabasoglu, G., J.-F. Lamarque, J. Bacmeister, D. A. Bailey, A. K. DuVivier, J. Edwards, L. K.
- 1116 Emmons, J. Fasullo, R. Garcia, A. Gettelman, C. Hannay, M.M. Holland, W.G. Large,
- 1117 P.H. Lauritzen, D.M. Lawrence, J.T.M. Lenaerts, K. Lindsay, W.H. Lipscomb, M.J.
- 1118 Mills, R. Neale, K.W. Oleson, B. Otto-Bliesner, A.S. Phillips, W. Sacks, S. Times, L. van

1119	Kampenhout, M. Vertenstein, A. Bertini, J. Dennis, C. Deser, C. Fischer, B. Fox-
1120	Kemper, J.E. Kay, D. Kinnison, P.J. Kushner, V.E. Larson, M.C. Long, S. Mickelson,
1121	J.K. Moore, E. Nienhouse, L. Polvani, P.J. Rasch, W.G. Strand, 2020: "The Community
1122	Earth System Model Version 2 (CESM2). Journal of Advances in Modeling Earth
1123	Systems 12, e2019MS001916, https://doi.org/10.1029/2019MS001916.
1124	Delanoë J and R. Hogan, 2010: Combined CloudSat-CALIPSO-MODIS retrievals of the
1125	properties of ice clouds. J. Geophys. Res., 115, D00H29, doi:10.1029/2009JD012346.
1126	DeMott, P.J., Hill, T.C., McCluskey, C.S., Prather, K.A., Collins, D.B., Sullivan, R.C., Ruppel,
1127	M.J., Mason, R.H., Irish, V.E., Lee, T. and Hwang, C.Y., 2015. Sea spray aerosol as a
1128	unique source of ice nucleating particles. Proc. Nat. Acad. Sci. USA,
1129	doi:10.1073/pnas.1514034112.
1130	FAA, 2015: Airplane and engine certification requirements in supercooled large drop, mixed
1131	phase and ice crystal icing conditions; Final rule. Parts 25 and 33, Aeronautics and Space,
1132	Title 14, U.S. Code of Federal Regulations, National Archives and Records
1133	Administration, 34 pp.
1134	Flato, G., J. Marotzke, B. Abiodun, P. Braconnot, S.C. Chou, W. Collins, P. Cox, F. Driouech, S.
1135	Emori, V. Eyring, C. Forest, P. Gleckler, E. Guilyardi, C. Jakob, V. Kattsov, C. Reason,
1136	M. Rummukainen, 2013: Evaluation of climate models. In Climate change 2013: The
1137	physical science basis. Contribution of working group I to the fifth assessment report of
1138	the intergovernmental panel on climate change. Cambridge University Press. 741-882,
1139	https://doi.org/10.1017/CBO9781107415324.020

1140	Gettelman, A., C.G. Bardeen, C.S. McCluskey, E. Jävinen, J. Stith, C.G. Bretherton, G.
1141	McFarquhar, C. Twohy, J. D'Alessandro, W. Wu, 2020: Simulating Observations of
1142	Southern Ocean Clouds and Implications for Climate. J. Geophys. Res. (accepted)
1143	Ghan, S. J., S. J. Smith, M. Wang, K. Zhang, K. Pringle, K. Carslaw, J. Pierce, S. Bauer, and P.
1144	Adams, 2013: A simple model of global aerosol indirect effects. J. Geophys. Res., 118,
1145	6688–6707, https://doi.org/10.1002/jgrd.50567.
1146	Gras, J. L., and M. Keywood, 2017: Cloud condensation nuclei over the Southern Ocean: wind
1147	dependence and seasonal cycles, Atmos. Chem. Phys., 17, 4419-4432,
1148	https://doi.org/10.5194/acp-17-4419-2017.
1149	Grecu, M., W.S. Olson, S.J. Munchak, S. Ringerud, L. Liao, Z. Haddad, B.L. Kelley, S.F.
1150	McLaughlin, 2016: The GPM combined algorithm. J. Atmos. Ocean. Tech., 33, 2225-
1151	2245, doi:10.1175/JTECH-D-16-0019.1.

1152 Grosvenor, D.P., and R. Wood, 2014: The effect of solar zenith angle on MODIS cloud optical

and microphysical retrievals within marine liquid water clouds. Atmos. Chem. Phys., 14,
7291-7321, doi:10.5194/acp-14-7291-2014.

1155 Grosvenor, D. P., Choularton, T. W., Lachlan-Cope, T., Gallagher, M. W., Crosier, J., Bower, K.

1156 N., Ladkin, R. S., and Dorsey, J. R.: In-situ aircraft observations of ice concentrations

- 1157 within clouds over the Antarctic Peninsula and Larsen Ice Shelf, Atmos. Chem. Phys., 12,
- 1158 11275–11294, doi:10.5194/acp12-11275-2012, 2012.

53

- Hande, L. B., S.T. Siems, M.J. Manton and D. Belusic, 2012: Observations of wind shear over
  the Southern Ocean. J. Geophys. Res., 117, D12206.
- 1161 https://doi.org/10.1029/2012JD017488.
- 1162 Hakim, J. R. H. G. J. (2013). An Introduction to Dynamic Meteorology (Fifth ed.). Academic
- 1163 Press: Elseveir, 552 pp.
- 1164 Hartery, S., D.W. Toohey, L. Revell, K. Sellegri, P. Kuma, M. Harvey and A.J.
- 1165 McDonald, 2020: Constraining the surface flux of sea spray particles from the Southern
- 1166 Ocean. J. Geophys. Res., **125**, e2019JD032026. <u>https://doi.org/10.1029/2019JD032026</u>.
- 1167 Hegg, D. A., R.J. Ferek, P.V. Hobbs and L.F. Radke, 1991: Dimethyl Sulfide and Cloud
- 1168 Condensation Nucleus Correlations in The Northeast Pacific-Ocean. J. Geophys. Res., 96,
  1169 13189–13191, https://doi.org/10.1029/91jd01309.
- 1170 Heidinger, A. K., 2011: ABI cloud height. NOAA NESDIS Center for Satellite Applications and
- 1171 Research Algorithm Theoretical Basis Doc Ver. 3., 77 pp,
- 1172 https://www.star.nesdis.noaa.gov/goesr/docs/ATBD/Cloud\_Height.pdf
- 1173 Hinkelman, L. and R. Marchand, 2020: Evaluation of CERES and CloudSat surface radiative
- fluxes over the Southern Ocean, *Earth and Space Science*, **7**, e2020EA001224,
- 1175 <u>https://doi.org/10.1029/2020EA001224.</u>
- 1176 Holben, B. N., T.F. Eck, I. Slutsker, D. Tanré J.P. Buis, A. Setzer, E. Vermote, J.A. Reagan, J.
- 1177 Kaufman, T. Nakajima, F. Lavenu, I. Jankowiak, and A. Smirnov, 1998: AERONET A
- 1178 federated instrument network and data archive for aerosol characterization, *Remote Sens*.
- 1179 *Environ.*, **66**, 1–16,

1180	Hoose, C., J.E. Kristjánsson, T. Iversen, A. Kirkevåg, Ø. Seland, and A. Gettelman, 2009:
1181	Constraining cloud droplet number concentration in GCMs suppresses the aerosol indirect
1182	effect, Geophys. Res. Lett., 36, L12807, https://doi.org/10.1029/2009GL038568
1183	Hou, A. Y., K.K. Ramesh, S. Neeck, A.A. Azarbarzin, C.D. Kummerow, M. Kojima, R. Oki, K.
1184	Nakamura, and T. Iguchi, 2014: The global precipitation measurement mission. Bull. Amer.
1185	Meteor. Soc., 95, 701–722, https://doi.org/10.1175/BAMS-D-13-00164.1.
1186	Hu, Y., S. Rodier, K. Xu, W. Sun, J. Huang, B. Lin, P. Zhai, and D. Josset, 2010: Occurrence,
1187	liquid water content, and fraction of supercooled water clouds from combined
1188	CALIOP/IIR/MODIS measurements. J. Geophys. Res., 115, D00H34,
1189	doi:10.1029/2009JD012384
1190	Huang, Y., C.N. Franklin, S.T. Siems, M.J. Manton, T. Chubb, A. Lock, S. Alexander. and A.
1191	Klekociuk, 2015: Evaluation of boundary-layer cloud forecasts over the Southern Ocean
1192	in a limited-area numerical weather prediction system using in situ, space-borne and
1193	ground-based observations. Q.J.R. Meteorol. Soc., 141, 2259-2276.
1194	https://doi:10.1002/qj.2519
1195	Huang, Y., S.T. Siems, M.J. Manton, L.B. Hande and J.M. Haynes, 2012a: The structure of low-
1196	altitude clouds over the Southern Ocean as seen by CloudSat. Journal of Climate, 25,

- 1197 2535–2546. <u>https://doi.org/10.1175/JCLI-D-11-00131.1</u>
- 1198 Huang, Y., S.T. Siems, M.J. Manton, A. Protat and J. Delanoë, 2012b: A study on the low-
- altitude clouds over the Southern Ocean using the DARDAR-MASK. J. Geophys. Res.,
- 1200 **117**, D18204. <u>https://doi.org/10.1029/2012JD017800</u>.
- 1201 Huang, Y., M. Manton, S. Siems, A. Protat, L. Majewskic, and H. Nguyen (2019). Evaluating
- 1202 Himawari-8 Cloud Products Using Shipborne and CALIPSO Observations: Cloud-top

- Height and Cloud-top Temperature. J. Atmos. Ocean. Tech. doi: 10.1175/JTECH-D-180231.1.
- 1205 Hudson, J.G., Y. Xie, and S.S. Yum, 1998: Vertical distribution of cloud condensation nuclei
- 1206 spectra over the summertime Southern Ocean. J. Geophys. Res., 103, 16609-16624,
- 1207 doi:10.1029/97JD03438.
- 1208 Humphries, R. S., A. R. Klekociuk, R. Schofield, M. D. Keywood, J. Ward, and S. R. Wilson,
- 1209 2016: Unexpectedly high ultrafine aerosol concentrations above East Antarctic sea ice.
- 1210 Atmos. Chem. Phys. 16, 2185–2206. <u>https://doi.org/10.5194/acp-16-2185-2016</u>
- 1211 Humphries, R. S., R. Schofield, M. D. Keywood, J. Ward, J. R. Pierce, C. M. Gionfriddo, M. T.
- 1212 Tate, D. P. Krabbenhoft, I. E. E. Galbally, S. B. B. Molloy, A. R. Klekociuk, P. V.
- 1213 Johnston, K. Kreher, A. J. Thomas, A. D. D. Robinson, N. R. P. Harris, R. Johnson, and
- 1214 S. R. Wilson, 2015: Boundary layer new particle formation over East Antarctic sea ice -
- 1215 possible Hg driven nucleation? *Atmos. Chem. Phys.*, **15**, 13339–13364.
- 1216 https://doi.org/10.5194/acp-15-13339-2015
- 1217 Hwang, Y.-T., and D.M.M. Frierson, 2013: Link between the double-Intertropical Convergence
- 1218 Zone problem and cloud biases over the Southern Ocean. *Proc. Natl. Acad. Sci.*, **110**, 49351219 4940.
- 1220 IPCC, 2013: Climate Change 2013: The Physical Science Basis. Contribution of Working Group
- 1221 I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change
- 1222 [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y.
- 1223 Xia, V. Bex and P.M. Midgley (eds.)]. *Cambridge University Press*, Cambridge, United
- 1224 Kingdom and New York, NY, USA, 1535 pp.

- 1225 Kanitz, T., P. Seifert, A. Ansmann, R. Engelmann, D. Althausen, C. Casiccia, and E.G. Rohwer,
  1226 2011: Contrasting the impact of aerosols at northern and southern midlatitudes on
  1227 heterogeneous ice formation. *Geophy. Res. Lett.*, 38, 5. doi: L17802
- Kay, J.E., C. Wall, V. Yettella, B. Medeiros, C. Hannay, P. Caldwell, and C. Bitz, 2016: Global
  climate impacts of fixing the Southern Ocean shortwave radiation bias in the Community
  Earth System Model (CESM). *J. Climate*, **29**, 4617-4636, DOI: 10.1175/JCLI-D-150358.1
- 1232 Klekociuk, A. R., W.J.R. French, S.P. Alexander, P. Kuma and A.J. McDonald, 2020: The state
- of the atmosphere in the 2016 southern Kerguelen Axis campaign region. *Deep Sea Research II*, **174**. https://doi.org/10.1016/j.dsr2.2019.02.001.
- Klekociuk, A.R., D.J. Ottaway, A.D. MacKinnon, I.M. Reid, L.V. Twigger and S.P. Alexander,
  2020: Australian lidar measurements of aerosol layers associated with the 2015 Calbuco

1237 eruption. *Atmosphere*, **11**, 124. <u>https://doi.org/10.3390/atmos11020124</u>

- 1238 Klepp, C., S. Michel, A. Protat, J. Burdanowitz, N. Albern, A. Dahl, M. Kähnert, V. Louf, S.
- 1239 Bakan, and S. A. Buehler, 2018: OceanRAIN, a new in-situ shipboard global ocean
- 1240 surface-reference dataset of all water cycle components. *Sci Data* **5**,180122.
- 1241 https://doi.org/10.1038/sdata.2018.122
- 1242 Korhonen, H., K. S. Carslaw, D. V. Spracklen, G. W. Mann, and M. T. Woodhouse, 2008:
- 1243 Influence of oceanic dimethyl sulfide emissions on cloud condensation nuclei
- 1244 concentrations and seasonality over the remote Southern Hemisphere oceans: A global
- 1245 model study. J. Geophys. Res., 113, <u>https://doi.org/10.1029/2007JD009718.</u>

57

- 1246 Korolev, A., G. McFarquhar, P. Field, C. Franklin, P. Lawson, Z. Wang, E. Williams, S. Abel,
- 1247 D. Axisa, S. Borrmann, J. Crosier, J. Fugal, M. Krämer, U. Lohmann, O. Schlenczaek,
- 1248 and M. Wendisch, 2017: Mixed-phase clouds: progress and challenges. *Meteor. Monogr.*

1249 doi:10.1175/AMSMONOGRAPHS-D-17-0001.1, **58**, 5.1-5.50.

- 1250 Kulmala, M., H. Vehkamäki, T, Petäjä, M. Dal Maso, A. Lauri, V.-M. Kerminen, W. Birmili and
- 1251 P.H. McMurry, 2004: Formation and growth rates of ultrafine atmospheric particles: a
- 1252 review of observations. J. Aerosol Sci., 35, 143–
- 1253 176, <u>https://doi.org/10.1016/j.jaerosci.2003.10.003</u>
- 1254 Kuma, P., A.J. McDonald, O. Morgenstern, S.P. Alexander, J.J. Cassano, S. Garrett, J. Halla, S.
- 1255 Hartery, M.J. Harvey, S. Parsons, G. Plank, V. Varma and J. Williams, 2020: Evaluation
- 1256 of Southern Ocean cloud in the HadGEM3 general circulation model and MERRA-2
- 1257 reanalysis using ship-based observations, Atmos. Chem. Phys. Discuss., 20 (11), 6607-
- 1258 6630, <u>https://doi.org/10.5194/acp-20-6607-2020</u>.
- Lang, F., Y. Huang, S.T. Siems and M.J. Manton, 2018: Characteristics of the marine
- 1260 atmospheric boundary layer over the Southern Ocean in response to the synoptic forcing.
- 1261 J. Geophys. Res. Atmo., **123**, 7799—7820. <u>https://doi.org/10.1029/2018JD028700.</u>
- 1262 Lim, E.-P., and I. Simmonds, 2007: Southern Hemisphere winter extratropical cyclone
- 1263 characteristics and vertical organization observed with the ERA-40 reanalysis data in
- 1264 1979-2001. Journal of Climate, **20**, 2675-2690. <u>https://doi.org/10.1175/JCLI4135.1</u>
- 1265 Mace, G. G., and A. Protat, 2018: Clouds over the Southern Ocean as observed from the RV
- 1266 Investigator during CAPRICORN. Part 2: The properties of non-precipitating
- 1267 stratocumulus. J. Appl. Meteor. Clim., 57, 1805-1823. <u>https://doi.org/10.1175/JAMC-D-</u>
- 1268 17-0195.1

1269	Mace. G.	G.,	O. Zhang, M.	Vaughan.	. R. Marchand.	G. Stepher	ns. C. Trept	e. and D. Winker.
120/	111400, 0.	· •.,	$\chi$ · $\Sigma$ inaing, $i$ · $i$ ·	, aagiiaii	,	C. Stepher	10, 0.10	io, and D millor,

- 1270 2009: A description of hydrometeor layer occurrence statistics derived from the first year
- 1271 of merged Cloudsat and CALIPSO data. J. Geophys. Res., 114, D00A26.
- 1272 https://doi.org/10.1029/2007JD009755
- 1273 Mace, G. G., and Q. Zhang, 2014: The CloudSat radar-lidar geometrical profile product (RL-
- 1274 GeoProf ): Updates, improvements, and selected results. J. Geophys. Res. Atmos., **119**,

1275 9441–9462. <u>https://doi:10.1002/2013JD021374.</u>

- 1276 Marchand, R., R. Wood, C. Bretherton, G. McFarquhar, A. Protat, P. Quinn, S. Siems, C. Jakob,
- 1277 S. Alexander, and B. Weller, 2014: The Southern Ocean Clouds, Radiation Aerosol
- 1278Transport Experimental Study (SOCRATES). whitepaper available from1279https://atmos.uw.edu/~roj/nobackup/Southern\_Ocean\_Workshop\_2014/Southern\_Ocean\_
- 1280 Workshop\_2014\_White\_Paper.pdf.
- 1281 McCoy, D. T., S. M. Burrows, R. Wood, D. P. Grosvenor, S. M. Elliott, P. L. Ma, P. J. Rasch, and
- 1282 D. L. Hartmann, 2015a: Natural aerosols explain seasonal and spatial patterns of Southern
- 1283 Ocean cloud albedo. *Sci. Adv.*, **1**, p.e1500157. <u>https://doi.org/10.1126/sciadv.1500157</u>
- 1284 McCluskey, C. S., T.C.J. Hill, C. Sultana, O. Laskina, J. Trueblood, M.V. Santander, C.M.
- 1285 Beall, J.M. Michaud, S.M. Kreidenweis, K.A. Prather, V. Grassian, P.J. DeMott, 2018a:
- 1286 A Mesocosm Double Feature: Insights into the Chemical Makeup of Marine Ice
- 1287 Nucleating Particles, *J Atmos Sci*, **75**, 2405-2423. <u>https://doi.org/10.1175/JAS-D-17-</u>
- 1288 <u>0155.1</u>
- 1289 McCluskey, C. S., T. C. J. Hill, R. S. Humphries, A. M. Rauker, A. M., S. Moreau, S., P. G.
- 1290 Strutton, S. D. Chambers, A. G. Williams, I. McRobert, J. Ward, M. D. Keywood, J.
- 1291 Harnwell, W. Ponsonby, Z.M. Loh, P. B. Krummel, A. Protat, S.M. Kreidenweis, and

P. J. DeMott, 2018b: Observations of ice nucleating particles over Southern Ocean
waters. *Geophysical Research Letters*, 45, 11,989–11,997. <u>https://doi.</u>

1294 <u>org/10.1029/2018GL079981.</u>

- 1295 McCluskey, C. S., J. Ovadnevaite, M. Rinaldi, J. Atkinson, F. Belosi, D. Ceburnis, S. Marullo, T.
- 1296 C. J. Hill, U. Lohmann, Z. A. Kanji, C. O'Dowd, S. M. Kreidenweis<sup>1</sup>, P. J. DeMott,
- 1297 2018c: Marine and terrestrial organic ice nucleating particles in pristine marine to
- 1298 continentally-influenced northeast Atlantic air masses. J. Geophys. Res., 123, 6196–
- 1299 6212. <u>https://doi.org/10.1029/2017JD028033.</u>
- 1300 McCluskey, C. S., P. J. DeMott, P.-L. Ma, and S. M. Burrows, 2019: Numerical representations
- 1301 of marine ice-nucleating particles in remote marine environments evaluated against
- 1302 observations. *Geophysical Research Letters*, **46**, 7838–7847. <u>https://doi.org/</u>
- 1303 <u>10.1029/2018GL081861.</u>
- 1304 McCoy, D. T., S. M. Burrows, R. Wood, D. P. Grosvenor, S. M. Elliott, P. L. Ma, P. J. Rasch, and
- D. L. Hartmann, 2015a: Natural aerosols explain seasonal and spatial patterns of Southern
  Ocean cloud albedo. *Sci. Adv.*, 1(6), p.e1500157.
- 1307 McCoy, D. T., D. L. Hartmann, M. D. Zelinka, P. Ceppi, and D. P. Grosvenor, 2015b: Mixed-
- phase cloud physics and Southern Ocean cloud feedback in climate models. J. Geophys. *Res.*, **120**(18), 9539-9554.
- 1310 McCoy, I. L., Bretherton, C. S., Wood, R., Twohy, C. H., Gettleman, A. Bardeen, C., 2020:
- 1311 Recent particle formation and aerosol variability near Southern Ocean low clouds. *in*1312 *prep*
- 1313 McFarquhar, G.M., S. Ghan, J. Verlinde, A. Korolev, J. W. Strapp, B. Schmid, J. M. Tomlinson,
- 1314 M. Wolde, S. D. Brooks, D. Cziczo, M. K. Dubey, J. Fan, C. Flynn, I. Gultepe, J. Hubbe,

- 1315 M. K. Gilles, A. Laskin, P. Lawson, W. R. Leaitch, P. Liu, X. Liu, D. Lubin, C.
- 1316 Mazzoleni, A.-M. Macdonald, R. C. Moffet, H. Morrison, M. Ovchinnikov, M. D. Shupe,
- 1317 D. D. Turner, S. Xie, A. Zelenyuk, K. Bae, M. Freer, and A. Glen, 2011: Indirect and
- 1318 Semi-Direct Aerosol Campaign (ISDAC): The impact of arctic aerosols on clouds, *Bull*.
- 1319 *Amer. Meteor. Soc.*, **92**, 183-201.
- 1320 McFarquhar, G. M., J.A. Finlon, D.M. Stechman, W. Wu, R.C. Jackson and M. Freer, 2018:
- 1321 University of Illinois/Oklahoma Optical Array Probe (OAP) Processing Software,
  1322 https://doi.org/10.5281/zenodo.1285969, 2018.
- 1323 McInnes, L. M., D. Covert, P. K. Quinn, and M. S. Germani, 1994: Measurements of chloride
- 1324depletion and sulfur enrichment in individual sea-salt particles collected from the remote
- 1325 marine boundary layer, J. Geophys. Res., 99, 8257-8268.
- 1326 <u>https://doi.org/10.1029/93JD03453</u>
- 1327 Morrison, A. E., S. T. Siems, and M. J. Manton, 2010: A modeling case study of mixed phase
- 1328 clouds over the Southern Ocean and Tasmania. *Mon. Wea. Rev.*, **138**, 839–862.
- 1329 <u>https://doi.org/10.1175/2009MWR3011.1.</u>
- 1330 Morrison, A. E., S. T. Siems, and M. J. Manton, 2011: A three-year climatology of cloud-top phase
- 1331 over the Southern Ocean and North Pacific. J. Climate, 24, 2405–2418.
- 1332 Morrison, H., J. A. Curry, and V. I. Khvorostyanov, 2005: A new double-moment microphysics
- 1333 parameterization for application in cloud and climate models. Part I: Description. J.
- 1334 Atmos. Sci., **62**, 1665-1677. <u>https://doi.org/10.1175/JAS3446.1</u>.
- 1335 Noh, Y.-J., Miller, S. D., Heidinger, A. K., Mace, G. G., Protat, A., & Alexander, S. P. (2019).
- 1336 Satellite-based detection of daytime supercooled liquid-topped mixed-phase clouds over

- 1337 the Southern Ocean using the Advanced Himawari Imager. Journal of Geophysical
- 1338 Research: Atmospheres, 124, 2677–2701. <u>https://doi.org/10.1029/2018JD029524</u>.

1339 Pavolonis, M. J., 2010: GOES-R Advanced Baseline Imager (ABI) algorithm theoretical basis

- 1340 document for cloud type and cloud phase, version 2.0. NOAA NESDIS Center for
- 1341 Satellite Applications and Research Algorithm Theoretical Basis Doc., 86 pp.,

1342 https://www.star.nesdis.noaa.gov/goesr/docs/ATBD/Cloud\_Phase.pdf.

- 1343 Pirjola, L., C.D. O'Dowd, I.M Brooks and M. Kulmala, 2000: Can new particle formation occur
- in the clean marine boundary layer? J. Geophys. Res., 105, 26531–
- 1345 26546. <u>https://doi.org/10.1029/2000jd900310</u>
- 1346 Platnick, S., and S. Twomey, 1994: Determining the susceptibility of cloud albedo to changes in
- 1347 droplet concentration with the advanced very high resolution radiometer, *J. Appl.*

1348 Meteorol., **33**, 334-347. <u>https://doi.org/10.1175/1520-</u>

- 1349 <u>0450(1994)033<0334:DTSOCA>2.0.CO;2</u>
- 1350 Plummer, D.M., G.M. McFarquhar, R.M. Rauber, B.F. Jewett, and D. Leon, 2014: Structure and
- statistical analysis of the microphysical properties of generating cells in the comma-head
  region of continental winter cyclones. *J. Atmos. Sci.*, **71**, 4181-4203.
- 1353 Protat, A., C. Klepp, V. Louf, W. Petersen, S. P. Alexander, A. Barros, and G. G. Mace, 2019:

1354 The latitudinal variability of oceanic rainfall properties and its implication for satellite

- 1355 retrievals. Part 1: The latitudinal variability of drop size distribution properties. J.
- 1356 *Geophys. Res. Atmos.*, **124**, 13291-13311. <u>https://doi.org/10.1029/2019JD031010</u>.
- 1357 Protat, A., C. Klepp, V. Louf, W. Petersen, S. P. Alexander, A. Barros, and G. G. Mace, 2019:
- 1358 The latitudinal variability of oceanic rainfall properties and its implication for satellite
- 1359 retrievals. Part 2: The relationships between radar observables and drop size distribution

- 1360 parameters. J. Geophys. Res., **124**, 13312-13324.
- 1361 https://doi.org/10.1029/2019JD031011.
- 1362 Protat, A., E. Schulz, L. Rikus, Z. Sun, and Y. Xiao, 2017: Shipborne observations of the
- radiative effect of Southern Ocean clouds. J. Geophys. Res., **122**, 318-328.
- 1364 https://doi.org/10.1002/2016JD026061
- 1365 Protat, A., S. A. Young, L. Rikus, and M. Whimpey, 2014: Evaluation of the hydrometeor
- 1366 frequency of occurrence in a limited-area numerical weather prediction system using near
- real-time CloudSat-CALIPSO observations. *Quart. J. Roy. Meteor. Soc.*, **140**, 2430-2443.
- 1368 <u>https://doi.org/10.1002/qj.2308</u>
- 1369 Puri, K., G. Dietachmayer, P. Steinle, M. Dix, L. Rikus, I. Logan, M. Naughton, C. Tingwell, Y.
- 1370 Xiao, V. Barras, I. Bermous, R. Bowen, L. Deschamps, C. Franklin, J. Fraser, T.
- 1371 Glowacki, B. Harris, J. Lee, T. Le, G. Roff, A. Sulaiman, H. Sims, X. Sun, Z Sun, H.
- 1372 Zhu, M. Chattopadhyay, and C. Engel, 2013: Implementation of the initial ACCESS
- 1373 numerical weather prediction system. *Aust. Meteorol. Oceanogr. J.*, **63**, 265–284.
- Quinn, P.K. and T.S. Bates, 2011: The case against climate regulation via oceanic phytoplankton
  sulfur emissions, *Nature*, 480, 51 56. <u>https://doi.org/10.1038/nature10580</u>
- 1376 Quinn, P. K., T.S. Bates, K. Schulz, D. Coffman, A.A. Frossard, L.M. Russell, W.C. Keene, D.
- 1377 Kieber, 2014: Contribution of sea surface carbon pool to organic matter enrichment in
- 1378 sea spray aerosol. *Nat. Geosci.* **7**, 228–232. <u>https://doi.org/10.1038/ngeo2092.</u>
- 1379 Quinn, P., D.J. Coffman, J.E. Johnson, L.M. Upchurch and T.S. Bates, 2017: Small fraction of
- 1380 marine cloud condensation nuclei made up of sea spray aerosol, *Nature Geoscience*, **10**,
- 1381 674-679. <u>https://doi.org/10.1038/ngeo3003</u>

- 1382 Read, K. A., A.C. Lewis, S. Bauguitte, A.M. Rankin, R.A. Salmon, E.W. Wolff, A. Saiz-Lopez,
- 1383 W.J. Bloss, D.E. Heard, J.D. Lee and J.M.C. Plane, DMS and MSA measurements in the
- 1384 Antarctic Boundary Layer: impact of BrO on MSA production, *Atmos. Chem. Phys.*, 8,
- 1385 2985-2997. <u>https://doi.org/10.5194/acp-8-2985-2008</u>
- 1386 Rinaldi, M., S. Decesari, E. Finessi, L. Giulianelli, C. Carbone, S. Fuzzi, C. O'Dowd, D.
- 1387 Ceburnis, and M.C. Facchini, Primary and secondary organic marine aerosol and oceanic
- 1388 biological activity: Recent results and new perspectives for future studies. Advances in
- 1389 *Meteorology*, <u>https://doi.org/10.1155/2010/310682</u>
- 1390 Rolph, G., A. Stein, and B. Stunder, 2017: Real-time Environmental Applications and Display
- 1391 sYstem: READY. Environmental modelling and software, **95**, 210-228.
- 1392 https://doi.org/10.1016/j.envsoft.2017.06.025
- 1393 Russell L.M., D.H. Lenschow, K.K. Laursen, P.B. Krummel, S.T. Siems, A.R. Bandy, D.C.
- 1394 Thornton, and T.S. Bates, 1998: Bidirectional mixing in an ACE 1 marine boundary
- layer overlain by a second turbulent layer. J. Geophy. Res., 103, 16,411-16,432.
- 1396 https://doi.org/10.1029/97JD03437
- 1397 Sanchez, K. J., C.-L. Chen, L.M. Russell, R. Betha, J. Liu, D.J. Price, P. Massoli, L.D. Ziemba,
- 1398 E.C. Crosbie, R.H. Moore, M. Müller, S.A. Schiller, A. Wisthaler, A.K.Y. Lee, P.K.
- 1399 Quinn, T.S. Bates, J. Porter, T.G. Bell, E.S. Saltzman, R.D. Vaillancourt and M.J.
- 1400 Behrenfeld, 2018: Substantial seasonal contribution of observed biogenic sulfate
- 1401 particles to cloud condensation nuclei. *Scientific Reports*, **8**, 3235.
- 1402 https://doi.org/10.1038/s41598-018-21590-9
- 1403 Sanchez, K. J., G.C. Roberts, G. Saliba, L.M. Russell, C. Twohy, M.J. Reeves, R.S. Humphries,
- 1404 M.D. Keywood, J.P. Ward, and I.M. McRobert, 2020: Cloud processes and the transport

1405	of biological emissions regulate Southern Ocean particle and cloud condensation nuclei
1406	concentrations, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-731.
1407	Sato, K., J. Inoue, S.P. Alexander, G. McFarquhar, Y. Yamazaki, 2018: Improved reanalysis and
1408	prediction of atmospheric fields over the Southern Ocean using campaign-based
1409	radiosonde observations, Geophys. Res. Lett., 45, 11406 – 11413,
1410	https://doi.org/10.1029/2018GL079037.
1411	Schmale, J., A. Baccarini, I. Thurnherr, S. Henning, A. Efraim, L. Regayre, C. Bolas, M.
1412	Hartmann, A. Welti, K. Lehtipalo, F. Aemisegger, C. Tatzelt, S. Landwehr, R.L. Modini,
1413	F. Tummon, J.S. Johnson, N. Harris, M. Schnaiter, A. Toffoli, M. Derkani, N.
1414	Bukowiecki, F. Stratmann, J. Dommen, U. Baltensperger, H. Wernli, D. Rosenfeld, M.
1415	Gysel-Beer, and K.S. Carslaw, 2019: Overview of the Antarctic circumnavigation
1416	expedition: Study of preindustrial-like aerosols and their climate effects (ACE-SPACE).
1417	Bull. Amer. Meteor. Soc., 100, 2261-2283. <u>https://doi.org/10.1175/BAMS-D-18-0187.1</u>
1418	Sciare, J., O. Favez, R. Sarda-Este`ve, K. Oikonomou, H. Cachier, and V. Kazan, 2009: Long-
1419	term observations of carbonaceous aerosols in the Austral Ocean atmosphere: Evidence
1420	of a biogenic marine organic source, J. Geophys. Res., 114, D15302.
1421	https://doi.org/10.1029/2009JD011998
1422	Scott, E. L., 2019: The Influence of Primary Nucleation and Rime Splintering on Ice Number
1423	Concentrations in Southern Ocean Cumuli. M.S. Thesis, University of Illinois, 104 pp.

1424 Shupe, M. D., 2007: A ground-based multisensor cloud phase classifier. J. Geophys. Res , 34,

1425 L22809, <u>https://doi.org/10.1029/2007GL031008</u>

- Skamarock, W. C., J. B. Klemp, J. Dudhia, D. O. Gill, D. M. Barker, W. Wang, and J. G. Powers,
  2005: A description of the Advanced Research WRF version 2. NCAR Tech. Note
  NCAR/TN-468+STR, 88 pp.
- 1429 Skofronick-Jackson G, W.A. Petersen, W. Berg, C. Kidd, E.F. Stocker, D.B. Kirschbaum, R.
- 1430 Kakar, S.A. Braun, G.J. Huffman, T. Iguchi, P.E. Kirstetter, C. Kummerow, R. Meneghini,
- 1431 R. Oki, W.S. Olson, Y.N. Takayabu, K. Kurukawa, and T. Wilheit, 2017: The Global
- Precipitation Measurement (GPM) Mission for Science and Society. *Bull. Amer. Meteor. Soc.*, DOI: 10.1175/BAMS-D-15-00306.1
- 1434 Stein, A.F., Draxler, R.R, Rolph, G.D., Stunder, B.J.B., Cohen, M.D., and Ngan, F., 2015: NOAA's
- HYSPLIT atmospheric transport and dispersion modeling system, *Bull. Amer. Meteor.*Soc., 96, 2059-2077, http://dx.doi.org/10.1175/BAMS-D-14-00110.1 □
- 1437 Stephens, B., M. Long, R. Keeling, E. Kort, C. Sweeney, E. Apel, E. Atlas, S. Beaton, J. Bent, N. Blake, J.
- 1438 Bresch, J. Casey, B. Daube, M. Diao, E. Diaz, H. Dierssen, V. Donets, B. Gao, M. Gierach, R.
- 1439 Green, J. Haag, M. Hayman, A. Hills, H. Hoecker-Martinez, S. Honomichl, R. Hornbrook, J.
- 1440 Jensen, R. Li, I. McCubbin, K. McKain, E. Morgan, S. Nolte, J. Powers, B. Rainwater, K.
- 1441 Randolph, M. Reeves, S. Schauffler, M. Smith, K. Smith, J. Stith, G. Stossmeister, D. Toohey, and
- A. Watt, 2018: The O2/N2 ratio and CO2 Airborne Southern Ocean (ORCAS) Study. *Bull. Amer. Meteor. Soc.*, doi:10.1175/BAMS-D-16-0206.1, 99, 381-402.
- 1444 Stevens, B., F. Ament, S. Bony, S. Crewell, F. Ewald, S. Gross, A. Hansen, L. Hirsch, M. Jacob,
- 1445 T. Kölling, H. Konow, B. Mayer, M. Wendisch, M. Wirth, K. Wolf, S. Bakan, M. Bauer-
- 1446 Pfundstein, M. Brueck, J. Delanoë, A. Ehrlich, D. Farrell, M. Forde, F. Gödde, H. Grob,
- 1447 M. Hagen, E. Jäkel, F. Jansen, C. Klepp, M. Klingebiel, M. Mech, G. Peters, M. Rapp,
- 1448 A.A. Wing, and T. Zinner, 2019: A high-altitude long-range aircraft configured as a

- 1449 cloud observatory: The NARVAL Expeditions. *Bull. Amer. Meteor. Soc.*, 100, 1061–
   1450 1077, https://doi.org/10.1175/BAMS-D-18-0198.1
- 1451 Tan, I., T. Storelvmo, and M. D. Zelinka, 2016: Observational constraints on mixed-phase
- 1452 clouds imply higher climate sensitivity, *Science*, **352**, 224-227.
- 1453 https://doi.org/10.1126/science.aad5300
- 1454 Trenberth, K. E., and J.T. Fasullo, 2010: Simulation of present-day and twenty-first-century
  1455 energy budgets of the southern oceans. *Journal of Climate*, 23, 440–454.
- 1456 https://doi.org/10.1175/2009JCLI3152.1
- 1457 Truong, S.C.H., Y. Huang, F. Lang, M. Messmer, I. Simmonds, S.T. Siems, and M.J. Manton,
- 1458 2020: A climatology of the marine atmospheric boundary layer over the Southern Ocean
  1459 from four field campaigns. *J. Geophys. Res. Atmo.*, Submitted.
- 1460 Toprak, E. and M. Schnaiter, 2013: Fluorescent biological aerosol particles measured with the
- Waveband Integrated Bioaerosol Sensor WIBS-4: Laboratory tests combined with a one
  year field study, *Atmos. Chem. Phys.*, 13, 225–243. https://doi.org/10.5194/acp-13-225-
- 1463 <u>2013</u>
- 1464Twohy, C., and J. Anderson, 2008: Droplet nuclei in non-precipitating clouds: Composition and1465size matter. *Environmental Research Letters*. 3, 045002. <a href="https://doi.org/10.1088/1748-">https://doi.org/10.1088/1748-</a>
- 1466 9326/3/4/045002
- 1467 Twohy, C. H., P. J. DeMott, L. M. Russell, D. W. Toohey, B. Rainwater, R. Geiss, K. J.
- 1468 Sanchez, S. Lewis, G. Roberts, R.S. Humphries, C. McCluskey, K. Moore, P. W. Selleck,
- 1469 M. D. Keywood, J. P Ward, and I.M. McRobert, 2020: Cloud-nucleating particles over
- 1470 the Southern Ocean in a changing climate. (submitted to ACPD).

- 1471 UCAR/NCAR- Earth Observing Laboratory (EOL), 1995-present: EOL Field Catalog.
   1472 https://doi.org/10.5065/D6SQ8XFB.
- 1473 Uetake, J., T. C. J. Hill, K. A. Moore, P. J. DeMott, A. Protat, and S. M. Kreidenweis, 2020:
- 1474 Airborne bacteria confirm the pristine nature of the Southern Ocean boundary layer,
- 1475 *Proceedings of the National Academy of Sciences*, doi:10.1073/pnas2000134117.
- 1476 Vallina, S. M., R. Simó, and S. Gassó, 2006: What controls CCN seasonality in the Southern
- 1477 Ocean? A statistical analysis based on satellite-derived chlorophyll and CCN and model-
- 1478 estimated OH radical and rainfall, *Global Biogeochemical Cycles*, **20**,
- 1479 <u>https://doi.org/10.1029/2005GB002597</u>.
- 1480 Veli-Matti, K, Chen X., Vakkari V., Petaja T., Kumala M., and Bianchi F., 2018: Atmospheric
- 1481 new particle formation and growth: review of field observations. *Environ. Res. Lett.* 13
  1482 103003. https://doi.org/10.1088/1748-9326/aadf3c
- 1483 Vergara-Temprado, J., A. K. Miltenberger, K. Furtado, D. P. Grosvenor, B. J. Shipway, A. A.
- 1484 Hill, J. M. Wilkinson, P. R. Field, B. J. Murray, and K. S. Carslaw, 2018: Strong control
- 1485 of Southern Ocean cloud reflectivity by ice-nucleating particles. *Proceeding of the*
- 1486 *National Academy of Sciences*, **115**, 2687-2692.
- 1487 https://doi.org/10.1073/pnas.1721627115.
- 1488 Vignon, É., Alexander, S. P., DeMott, P. J., Sotiropoulou, G., Gerber, F., Hill, T. C. J.,
- 1489 Marchand, R., Nenes, A., Berne, A. (2020), 'Measured ice nucleating particle
- 1490 concentrations improve the simulation of mid-level mixed-phase clouds over the high-
- 1491 latitude Southern Ocean', Journal of Geophysical Research, submitted
- 1492 Waitz, F., M. Schnaiter, T. Leisner, and E. Järvinen, 2020: PHIPS-HALO: the airborne particle
- habit imaging and polar scattering probe Part 3: Single particle phase discrimination and

- particle size distributions based on angular scattering function. *Atmos. Meas. Tech.*, In
  preparation.
- 1496 Wang, Z., S.T. Siems, D. Belusic, M.J. Manton and Y. Huang, 2015: A climatology of the
- 1497 precipitation over the Southern Ocean as observed at Macquarie Island. *Journal of*
- 1498 Applied Meteorology and Climatology, 54, 2321–2337. <u>https://doi.org/10.1175/JAMC-D-</u>
  1499 14-0211.1.
- 1500 Wang, Y., G.M. McFarquhar, R.M. Rauber, C. Zhao, W. Wu, D.M. Stechman, J.A. Finlon, J. Stith,
- 1501 M. Schnaiter, E. Järvinen, J. Jensen, J. Vivekanandan, and M. Dixon, 2019: Microphysical
- 1502 properties of generating cells over the Southern Ocean: Results from SOCRATES. J.
- 1503 *Geophys. Res.*, **125**, e2019JD032237.
- Warren, D.R., and J.H. Seinfeld, 1985: Prediction of aerosol concentrations from a burst of
  nucleation. J. Colloid Interface Sci., 105, 136-142.
- Weber, R.J., P.H. McMurry, L. Mauldin, D.J. Tanner, F.L. Eisele, F.J. Brehtel, S.M. Kreidenweis, G.L.
  Kok, R.D. Schillawski, and D. Baumgardner, 1998: A study of new particle formation and growth
- 1508 involving biogenic and trace gas species measured during ACE 1. J. Geophys. Res., 103, 163851509 16396.
- 1510 Welti, A., E. K. Bigg, P. J. DeMott, X. Gong, M. Hartmann, M. Harvey, S. Henning, P. Herenz,
- 1511 T. C. J. Hill, B. Hornblow, C. Leck, M. Löffler, C. S. McCluskey, A. M. Rauker, J.
- 1512 Schmale, C. Tatzelt, M. van Pinxteren, and F. Stratmann, 2020: Ship-based
- 1513 measurements of ice nuclei concentrations over the Arctic, Atlantic and Southern Ocean,
- 1514 in preparation for submission to *Atmos. Chem. Phys.*
- 1515 Wofsy, S. C., the HIPPO Science Team, and Cooperating Modeling and Satellite Teams, 2011: HIAPER
- 1516 Pole-to-Pole Observations (HIPPO): fine-grained, global-scale measurements of climatically

- 1517 important atmospheric gases and aerosols. *Phil. Trans. Roy. Soc.* A, **369**, 2073-2086,
  1518 doi:10.1098/rsta.2010.0313.
- Wolters, E.L.A., H.M. Deneke, B.J.J.M. van den Hurk, J.F. Merink, and R.A. Roebeling, 2010: Broken and
  inhomogeneous cloud impact on satellite cloud particle effective radius and cloud-phase retrievals. *J. Geophys. Res.*, 115, D10214, https://doi.org/10.1029/2009JD012205.

Wu, C., X. Liu, M. Diao, K. Zhang, A. Gettelman, Z. Lu, J.E. Penner and Z. Lin, 2017: Direct

comparisons of ice cloud macro- and microphysical properties simulated by the

- 1524 Community Atmosphere Model version 5 with HIPPO aircraft observations, *Atmos*.
- 1525 Chem. Phys., **17**, 4731-4749, <u>https://doi.org/10.5194/acp-17-4731-2017</u>.

1522

1523

- 1526 Zeng, S., C. Cornet, F. Parol J. Riedi, and F. Thieuleux, 2012: A better understanding of cloud optical
- 1527 thickness derived from the passive sensors MODIS/AQUA and POLDER/PARASOL in the A-
- Train constellation. *Atmos. Chem. Phys.*, **12**, 11245-11259., doi:10.5194/acp-12-11245-2012et al.
  2012
- 1530 Zhang, X. L., P. Massoli, P.K. Quinn, T.S. Bates and C.D. Cappa, 2014: Hygroscopic growth of
- submicron and supermicron aerosols in the marine boundary layer. J. Geophys.
- 1532 Res., **119**, 8384-8399. <u>https://doi.org/10.1002/2013jd021213</u>
- 1533 Zhao, M., J.-C. Golaz, I.M. Held, H. Guo, V. Balaji, R. Benson, J.-H. Chen, X. Chen, L.J.
- 1534 Donner, J.P. Dunne, K. Dunne, J. Durachta, S.-M. Fan, S.M. Freidenreich, S.T. Garner,
- 1535 P. Ginoux, L.M. Harris, L.W. Horowitz, J.P. Krasting, A.R. Langenhorst, Z. Liang, P.
- 1536 Lin, S.-J. Lin, S.L. Malyshev, E. Mason, P.C.D. Milly, Y. Ming, V. Naik, F. Paulot, D.
- 1537 Paynter, P. Phillips, A. Radhakrishnan, V. Ramaswamy, T. Robinson, D. Schwarzkopf,
- 1538 C.J. Seman, E. Shevliakova, Z. Shen, H. SHin, L.G. Silvers, J.R. Wilson, M. Winton,
- 1539 A.T. Wittenberg, B. Wyman, B. Xiang, 2018: The GFDL Global Atmosphere and Land

70

1540	Model AM4.0/LM4.0: Simulation characteristics with prescribed SSTs. J. Adv. Modeling
1541	Earth Sys., 10, 691-734, https://doi.org/10.1002/2017MS001208.

- 1542 Zhou, X., R. Atlas, I. McCoy, C. S. Bretherton, C. Bardeen, A. Gettelman, P. Lin, and Y. Ming,
- 1543 2020: Evaluation of cloud and precipitation simulations in CAM6 and AM4 using
- 1544 observations over the Southern Ocean. Earth Space Sci., submitted 4/2020. ESSO
- 1545 preprint: doi:10.1002/essoar.10502913.1

#### **Tables**

### 1557 Table 1: Previous field campaigns and data collection activities over the SO

Campaign	Description	Reference
Aerosol Characterization Experiment-1 (ACE-1)	sea-spray aerosol, vertical aerosol profiles and fluxes	Bates et al. 1998a, b; Clarke et al. 1998; Weber et al. 1998; Russell et al. 1998
HIAPER Pole-to-Pole Observations (HIPPO)	4 transects sampling clouds and aerosols south of Macquarie Island	Wofsy et al. 2011; Chubb et al. 2013, 2016
SIPEX II	aerosol number concentrations across polar front	Humphries 2015, 2016
O <sub>2</sub> /N <sub>2</sub> Ratio and CO <sub>2</sub> Airborne Southern Ocean Study (ORCAS)	limited cloud sampling	Stephens et al. 2018; D'Alessandro et al. 2019
Observations near Tasmania	observations in wintertime low-altitude clouds over open ocean near Tasmania	Ahn et al. 2017; Huang et al. 2017
Cape Grim observations	CCN observations at Cape Grim (41°S, 145°E)	Gras et al. 2017
Southern Ocean Cloud Experiment (SOCEX)	aerosol optical depth and composition, clouds	Sciare et al. 2009; Boers et al. 1996, 1998
Recent ship-based observations (separate from campaigns described here)	limited set of cloud radiation and aerosol properties south of 60°S and circumpolar quantification of aerosol properties	Kuma et al. 2020; Klekociuk et al. 2020; Hartery et al. 2020; Schmale et al. 2020
#### 1561 Figures



1562

1563

Figure 1: Ship tracks from CAPRICORN I (dark blue), II (light blue) and MARCUS voyages (green colors), as well as the SOCRATES G-V flight tracks (orange) and the location of the ground-observing site at Macquarie Island during MICRE (red). The locations of Mawson, Davis, and Casey stations are also shown (grey stars). The lower panel depicts the years and seasons corresponding to each campaign.



- 1570
- 1571
- 1572



1574 MARCUS (green), CAPRICORN-1 (dark blue), CAPRICORN-2 (light blue), and

- 1575 SOCRATES (yellow). These are based on the following total number of minutes that each
- 1576 campaign sampled south of 43S: SOCRATES, 6,319 minutes; MARCUS; 182,470 minutes;
- 1577 CAPRICORN-1, 44,408 minutes; CAPRICORN-2, 60,060 minutes.
- 1578





Figure 3: Daily mean number fraction of low volatility particles measured using the V-TDMA during CAPRICORN-1. Pre-selected particle diameters were 40 nm (blue) 100 nm (yellow) and 

150 nm (red). Error bars represent the standard deviation in the daily mean for each preselected 

- particle size.



1587

1588 Fig. 4. a) Compositional fraction of total particles by number in the 0.1-0.5  $\mu$ m dry size range for six 150 m samples on SOCRATES flights 11-15. (Sample times in UTC: 11-D: 04:41:30-04:46:30; 1589 1590 12-D: 04:53:00-04:55:00; 13-G: 04:26:20-04:31:20; 13-I: 05:26:00-05:30:00; 14-C: 04:22:30-1591 04:26:00; 15-F: 05:59:40-06:04:50.) Categories: S-containing: with S (and sometimes, O) primary 1592 elements. Sea Spray: Salts of Na, Cl, Mg, S, K, Ca, sometimes with organic coatings. Other types: 1593 includes crustal dust (silicates and carbonates), metals (Al, Fe, Cr, Ti, Mn, Co, Zn, Cu, O etc.), 1594 primary organics (C and sometimes O) and combustion particles (high S, C, O with K). b) Average 1595 composition by number for all six samples; smaller inset further subdivides Sea Spray into 1596 different types of salts.







16001601Figure 5. a) CCN spectra measured in the marine boundary layer, clustered by CN and1602CCN concentrations. Error bars represent the standard error. b) Measured below-cloud1603CCN concentrations at 0.3% supersaturation vs the observed in-cloud droplet number1604concentration (CDNC). The black line is a linear fit with r = 0.75. The same color scheme1605used to identify the cluster is used in both figures. The white points in (b) represent1606measurements that were not clustered due to missing data.1607



Figure 6. HYSPLIT 5-day back trajectories for the four clusters shown in Figure 1, a) High
CN/High CCN, b) High CN/Low CCN, c) Low CN/Low CCN and d) Low CN/High CCN.
The magenta circles represent the HIAPER G-V location used to initialize the back
trajectory.



1616 Figure 7 (a) Time series of total CN (D > 11 nm) and accumulation mode (wing mounted UHSAS, 1617 100 < D < 1000 nm) aerosol number concentrations sampled in the free tropospheric survey leg 1618 (~6 km) of RF09 as the GV flew south. (b) HYSPLIT 72-hour back trajectories of air masses 1619 initiated in 10 minute intervals (starred locations in (a) with color-coded matched time in (b)) along 1620 the GV flight path. Trajectories dominated by RPF events are identified by where maximum CN over the corresponding 10 minutes exceeds 2500 mg<sup>-1</sup> (dotted line, (a)). In this case, only one 1621 1622 trajectory does not satisfy the criteria for RPF events (dashed line, b). The majority of trajectory 1623 ascents exhibit synoptic uplift (3-6 cm s<sup>-1</sup>) within 20-30 hours of SOCRATES sampling (star) and 1624 are in proximity to phytoplankton emissions (Z < 1 km) in the prior 72 hours.

1615

1626



1629 1630

1630 Figure 8. INP number concentrations per volume of air at -20°C over the SO region for a selection

1631 of the studies (MARCUS, CAPRICORN I, CAPRICORN II, TAN1502) listed in Table S13. Each 1632 data point represents the mid-point position of a single filter collection. Historical data from Bigg

1633 (1973) are shown at right for context (each is the mean of numerous measures at that latitude); all

1634 are the same color since all were >0.1 sL<sup>-1</sup>.



Figure 9: Cloud droplet concentration measured as a function of altitude for four rampedascents/descents through boundary layer cloud for the days indicated in the legend. The time

period, average latitude, and average wind speed of each ascent or descent are indicated. Each
 circle represents 2 sec average.



1643 Figure 10: Relative occurrence frequency of different phases derived from suite of in-situ probes

1644 as a function of temperature (Adapted from D'Alessandro et al. 2020). Black line indicates

1645 number of samples, giving some information about statistical significance of results.



Figure 11: Collection of PHIPS images of ice particles from RF02 sampled in the temperature range 0 to -1650 5°C.



1652



1654 UTC on 8 Feb. 2018. The black line shows the flight level of the G-V aircraft and the shadows

represent the location of the generating cells identified by the method of Wang et al. (2020).

1656 Other plots show statistical analysis of data collected over this time as shown by Kernel

1657 probability distribution functions of properties inside (pink) and outside (blue) of generating cells

- 1658 for b)  $N_t$ , c) LWC and d) IWC. Black box plots show 5<sup>th</sup>, 25<sup>th</sup>, 50<sup>th</sup> (red line), 75<sup>th</sup> and 95<sup>th</sup>
- 1659 percentiles of data. White point indicates mean value. Width of red and blue shaded area
- 1660 represents portion of data located at particular value.







Figure 13: a) The nose cone of the NSF/NCAR GV aircraft large wing pod during an encounter
with small supercooled cloud droplets during SOCRATES. Ice has accreted on the tip of the pod.
b) The nose cone of the GV wing pod under conditions where larger supercooled droplets were
encountered. Notice the drops have impacted and run back before freezing further back on the
pod.



1672 1673

1674 Figure 14: Aircraft pass on 17 Feb 2018, through the tops of two closely-spaced cumuli at a temperature of -9 °C, as shown by the downward-pointing airborne Doppler radar on the G-V 1675 aircraft (top two panels: radar reflectivity and vertical velocity; positive values denote upward 1676 1677 motion of particles). Black horizontal line at top shows aircraft location as it passed through the 1678 cloud tops. Shaded gray boxes demarcate the two clouds, with shaded blue boxes identifying 10-1679 second regions with the highest maximum ice number concentrations (corrected for possible 1680 shattering artifacts) as labeled. Snapshot from forward video (bottom panel) also shows the 1681 multiple thermal structure of these cumuli as the aircraft approached. Particle images (inset) from 1682 PHIPS probe indicated rimed ice and liquid drops were present, necessary for rime-splintering to 1683 be active, as well as the expected products of rime-splintering, pristine and rimed columns.



Figure 15. (a) Flight tracks for the 15 SOCRATES missions and the outermost boundaries of the 15 sectors used for Himawari-8 analysis in (b). Frequency of occurrence of Himawari-8 cloud type as a function of cloud-top temperature for the 15 SOCRATES missions (includes times for both outbound and inbound legs).



1695

Figure 16: The mean CRE biases ((a): short-wave; (b): long-wave and; (c): net) in ACCESS-C3
relative to the mean measured CRE for different cloud types over the Southern Ocean during
CAPRICORN I. Vertical error bars represent the standard deviation of each CRE bias for each
cloud type. The size of the points is proportional to the number of sampling hours of each cloud
type. (d) represents the decomposed CRE and CRE biases, weighted by the relative frequency of
occurrence for each cloud type. The black horizontal bars represent the total CRE and CRE
biases for the whole campaign.



Figure 17: Statistical distribution from MARCUS cruises of how cloud and environmental properties varied depending upon whether measurements were north or south of 60S for time periods with single-layer, non-precipitating clouds with bases less than 3 km and greater than 500 km away from nearest cyclone center. Box and whisker plots show quartiles of the distribution, red line indicates mode, black bars are defined as  $q_3 + 1.5 \times (q_3 - q_1)$  and  $q_1 - 1.5 \times (q_3 - q_1)$ , where  $q_1$  and  $q_3$  are the 25% and 75% percentiles, and red pluses all points outside the black bars.

- 1711
- 1712
- 1713



Figure 18. (a-c) Mean profiles of temperature (red line), dew point temperature (blue line) and vector wind for the clusters M, C1, and C2, respectively, displayed as a skew-T logP diagram, shaded region indicating one standard deviation. (d) 72h HYSPLIT Back trajectory for the nearest soundings to the centroid in each cluster M, C1, and C2, separately. The solid, dash-dotted, and dotted lines represent the tracks at 500, 1500, and 4000 m, respectively. (e) The frequency of occurrence composites of the sounding locations relative to the nearest cyclone centers, concentric circles indicate distances of 5°, 10°, 15° and 20° from the cyclone center.



1720

1721 Figure 19. Daily mean derived and observed properties from the RV Investigator during 1722 Capricorn II. a) Cloud droplet number concentrations (N<sub>c</sub>) derived for non precipitating liquid 1723 clouds using combined radar reflectivity, microwave brightness temperature, and lidar attenuated 1724 backscatter. Error bars shown standard deviation of N<sub>c</sub> during that 24 hour period (only days 1725 with at least 200 30-second retrievals are shown), b) particulate methanesulfonic (MSA) 1726 concentrations (ug m-3), c) CCN measured at 0.25% super saturation, d) latitude of the ship on 1727 that day, e) number of N<sub>c</sub> retrievals used in the N<sub>c</sub> means and standard deviation in panel a. 1728



Figure 20. Size distributions from observations (thin lines) and reconstructed model hydrometeor
size distributions (thick colored lines) for low level clouds (P>750mb) as indicated in the
legend. Cloud probe data shown as 2DS for all particles (black dotted), 2DS round particles
(black dash), CDP (black solid), 2DC all (gray dash), 2DC round (gray dot-das), PIP (gray
dotted), PHIPS all (gray long dash) and PHIPS drop (gray solid). (A) All clouds, (B) Cold
clouds, (C) Warm clouds. Model is sampled along the flight track at aircraft altitude.

# **University Library**



### A gateway to Melbourne's research publications

Minerva Access is the Institutional Repository of The University of Melbourne

#### Author/s:

McFarquhar, G; Bretherton, C; Marchand, R; Protat, A; DeMott, P; Alexander, S; Roberts, G; Twohy, C; Toohey, D; Siems, S; Huang, Y; Wood, R; Rauber, R; Lasher-Trapp, S; Jensen, J; Stith, J; Mace, J; Järvinen, E; Schnaiter, M; Gettelman, A; Sanchez, K; McCluskey, C; Russell, L; McCoy, I; Atlas, R; Bardeen, C; Moore, K; Hill, T; Humphries, R; Keywood, M; Ristovski, Z; Cravigan, L; Schofield, R; Fairall, C; Mallet, M; Kreidenweis, S; Rainwater, B; D'Alessandro, J; Wang, Y; Wu, W; Saliba, G; Levin, E; Ding, S; Lang, F; Truong, S; Wolff, C; Haggerty, J; Harvey, M; Klekociuk, A; McDonald, A

#### Title:

Observations of clouds, aerosols, precipitation, and surface radiation over the Southern Ocean: An overview of CAPRICORN, MARCUS, MICRE and SOCRATES

#### Date:

2020-11-24

#### Citation:

McFarquhar, G., Bretherton, C., Marchand, R., Protat, A., DeMott, P., Alexander, S., Roberts, G., Twohy, C., Toohey, D., Siems, S., Huang, Y., Wood, R., Rauber, R., Lasher-Trapp, S., Jensen, J., Stith, J., Mace, J., Järvinen, E., Schnaiter, M., ... McDonald, A. (2020). Observations of clouds, aerosols, precipitation, and surface radiation over the Southern Ocean: An overview of CAPRICORN, MARCUS, MICRE and SOCRATES. Bulletin of the American Meteorological Society, 102 (4), https://doi.org/10.1175/BAMS-D-20-0132.1.

#### **Persistent Link:**

http://hdl.handle.net/11343/269267

File Description: Published version