The Association Between Cloud Droplet Number Over the Summer Southern Ocean and Air Mass History

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Abstract

The cloud properties and governing processes in Southern Ocean marine boundary layer clouds have emerged as a central issue in understanding the Earth's climate sensitivity. While the simulated cloud feedbacks in Southern Ocean clouds have evolved in the most recent climate model intercomparison, the background properties of simulated summertime clouds in the Southern Ocean are not consistent with measurements due to known biases in simulating cloud condensation nuclei concentrations. This paper presents several case studies collected during the Capricorn 2 and Marcus campaigns held aboard Australian research vessels in the Austral Summer of 2018. Combining the surface-observed cases with MODIS data along forward and backward air mass trajectories, we demonstrate the evolution of cloud properties with time. These cases are consistent with multi-year statistics showing that long trajectories of air masses over the Antarctic ice sheet are critical to creating high droplet number clouds in the high latitude summer Southern Ocean. We speculate that secondary aerosol production via the oxidation of biogenically derived aerosol precursor gasses over the high actinic flux region of the high latitude ice sheets is fundamental to maintaining relatively high droplet numbers in Southern Ocean clouds during Summer.

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Abstract: The cloud properties and governing processes in Southern Ocean marine boundary layer clouds have emerged as a central issue in understanding the Earth's climate sensitivity. While the simulated cloud feedbacks in Southern Ocean clouds have evolved in the most recent climate model intercomparison, the background properties of simulated summertime clouds in the Southern Ocean are not consistent with measurements due to known biases in simulating cloud condensation nuclei concentrations. This paper presents several case studies collected during the Capricorn 2 and Marcus campaigns held aboard Australian research vessels in the Austral Summer of 2018. Combining the surface-observed cases with MODIS data along forward and backward air mass trajectories, we demonstrate the evolution of cloud properties with time. These cases are consistent with multi-year statistics showing that long trajectories of air masses over the Antarctic ice sheet are critical to creating high droplet number clouds in the high latitude summer Southern Ocean. We speculate that secondary aerosol production via the oxidation of biogenically derived aerosol precursor gasses over the high actinic flux region of the high latitude ice sheets is fundamental to maintaining relatively high droplet numbers in Southern Ocean clouds during Summer.

Plain Language Summary: The amount of warming the Earth will experience because of increasing carbon dioxide levels in the atmosphere is sensitive to the properties of clouds that occur over the Southern Ocean. The atmosphere over the circumpolar Southern Ocean is poorly understood and presents significant challenges to climate models. Here we document the properties of the ubiquitous Southern Ocean low-level clouds that exert a strong influence on the albedo of this region. We find that high cloud droplet number concentrations are associated with air masses that have taken paths over the high-altitude ice sheets. The chemistry of the aerosol on which the cloud droplets form suggests that newly formed aerosols that have condensed from gasses emitted by phytoplankton in the highly productive waters near Antarctica are an important component of the cloud properties that must be correctly simulated in models.

Main Point 1: High cloud droplet number concentrations are associated with air masses that have recently passed over continental Antarctica.

Main Point 2: Cloud droplet number concentrations decrease with time as clouds evolve in over-water trajectories due to scavenging by precipitation.

Main Point 3: Katabatic flows bring high concentrations of cloud condensation nuclei to the marine boundary layer where they influence cloud properties.

1 Introduction

As one of the cloudiest regions on Earth, with cloud cover near 90% (Mace et al., 2009; Mace & Zhang, 2014), the high latitude Southern Ocean from roughly poleward of the Antarctic Circumpolar Current to the Antarctic Continent is a critical component of the Earth's climate system. In addition to fundamental oceanographic processes such as the uptake of atmospheric heat and carbon in the formation of the intermediate and deep-water masses (Armour et al., 2016; Morrison et al., 2016), the albedo of the extensive cloud cover, particularly in summer, induces significant controls on the column energy budget of the region and influences the Earth's climate (Zelinka et al., 2020; Gettelman et al., 2019).

The high cloud cover of the Southern Ocean is primarily due to extensive marine boundary layer (MBL) clouds, most of which are liquid phase and subfreezing (Bodas-Salcedo et al., 2016). These clouds, while mostly nonprecipitating (Huang et al., 2016), generate both liquid- and ice-phase precipitation (Mace et al., 2021). Because precipitation exerts controls on cloud cover (Albrecht et al., 1989), precipitation in these clouds continues to be a topic of active research (Kang et al., 2022). However, cloud properties and the associated cloud albedo, as well as the precipitation processes, respond to a significant seasonal cycle in cloud condensation nuclei (CCN) that directly influence cloud droplet number concentration (N_d) (McCoy et al., 2015; Kruger and Grasl, 2011; Mace et al., 2023, 2021). Analyzing cloud properties derived from A-Train data, Mace and Avey (2017) find that the Summer N_d in the SO is approximately a factor of two higher in summer compared to winter. While this seasonal cycle in N_d is thought to be associated with seasonally higher CCN associated with due to secondary aerosol production derived from the oxidation of biogenically-derived precursor gasses such as dimethyl sulfide (DMS) (Lana et al., 2012), aerosol measurements collected during recent campaigns have not directly observed new particle formation events. However, robust circumstantial evidence suggests that new particle formation of sulfur-based particles with a volatility similar to sulfuric acid in the SO free troposphere is widespread and frequent (McCoy et al., 2021).

This hypothesis is consistent with prior studies suggesting that particle concentrations in the free troposphere in marine regions are controlled by sulfuric-acid-driven nucleation of nanoparticles resulting from the oxidation of oceanic dimethylsulfide transported upward. Downward transfer of these nanoparticles then supplies the marine boundary layer with CCN concentration. This mechanism of aerosol production has been observed (Clarke, 1993) and modeled (Russell et al., 1994; Raes, 1995) previously. Most recently, laboratory work on sulfuric acid nucleation and further growth have demonstrated that these processes can survive evaporation and even grow during these transport processes and temperature transitions (Tiszenkel et al., 2019)

Trajectory analyses also point to a source of CCN from airmasses emanating from the Antarctic continent (Humphries et al., 2016; Twohy et al., 2021; Simmons et al., 2021). Specifically, the Antarctic continent in summer has been identified as a potential source of sulfate aerosol in work dating back to the 1980's (Shaw, 1988). Presumably, air masses with high DMS and oxidants are lofted over the high albedo and mostly cloud-free ice sheets in synoptic scale ascent, where most ambient aerosols are removed by precipitation. New sulfate-containing aerosol particles nucleate and grow during the time spent over the ice sheets in the cold free-troposphere. Katabatic flows then cause these newly formed aerosols to emerge into the marine boundary layer in the waters adjacent to the Antarctic continent (Simmons et al., 2021).

Using MODIS data, we recently showed that N_d in the East Antarctic sector of the SO exhibits strong latitudinal gradients during Summer, with the highest N_d in the waters immediately adjacent to the Antarctic Continent (Mace et al., 2023). These results agreed broadly with findings compiled from the Measurements of Aerosols, Radiation, and Clouds over the Southern Ocean (MARCUS) and the Clouds, Aerosols, Precipitation, Radiation, and atmospheric Composition over the Southern Ocean (CAPRICORN II) (McFarquhar et al., 2021) campaigns that collected data in this region during Summer 2018 (Mace et al., 2021). The latitudinal gradient in N_d is a curious feature of the SO region. It seems consistent with the idea that the highest CCN concentration is found immediately adjacent to the Antarctic continent. Cloud and precipitation processing of the aerosol then decrease N_d with distance away from the Antarctic coast. However, the CCN concentration and N_d remain mostly above sea salt aerosol concentration, suggesting a source of new particles mixing into the MBL from the free troposphere (McCoy et al., 2021). The extent to which the background CCN and N_d are maintained against cloud processing by mixing from the free troposphere, as suggested by Twohy et al. (2021) and McCoy et al. (2021), is unknown.

The processes that maintan N_d in the summer SO are crucial to understanding the Earth's climate sensitivity. In the most recent climate model intercomparison project (CMIP6; Eyring et al., 2016), the high latitude SO had significantly less negative cloud feedback than in CMIP5 and earlier model intercomparison studies. While this change in simulated cloud feedback was due partially to an observationally consistent decrease in ice-phase precipitation, the cloud feedback change occurred within a simulated background state in cloud properties that is unrealistic. As shown by McCoy et al. (2021), the CAM6 model consistently underpredicts, by approximately a factor of 2, the N_d in comparisons to collocated measurements during the Southern Ocean Cloud Radiation Aerosol Transport Experimental Study (SOCRATES) campaign (McFarquhar et al., 2021). McCoy et al., (2021) trace this bias to an insufficient DMS oxidation mechanism in the model among other issues. Since N_d and precipitation scale inversely in MBL clouds (Comstock et al., 2004), a correct simulation of N_d is a prerequisite to correctly predicting the precipitation that is driving the negative feedback changes in CAM6 (Gettleman et al., 2019) poleward of about 50°S.

This paper examines three case studies collected in the Australian East Antarctic sector of the SO from the Summer of 2018 during the MARCUS and CAPRICORN II campaigns. Using a combination of surface-based cloud property diagnostics and aerosol in situ measurements, we examine MODIS-derived cloud properties along backward and forward trajectories. We show that clouds forming in air masses recently descended in Katabatic flows from the high-altitude Antarctic ice sheets have significantly higher N_d than the mean for that latitude. The N_d then decreases with time as the air masses pass over the open ocean. We then place these case studies within a larger context of MODIS data collected over 5 Austral Summers to show a consistent relationship between N_d and trajectories emanating from the Antarctic continental ice sheets.

2. Methods

In this study, we combine data from the MARCUS and CAPRICORN II ship-based campaigns with MODIS level 2 data products extracted from overpasses along backward and forward airmass trajectories centered on the ships. This combination of surface and satellite measurements linked by trajectory modeling allows us to exploit the synergy in the surface and satellite measurements. While polar-orbiting satellite measurements provide spatial context, they only provide a single snapshot in time and have only limited information on the cloud field. Surface-based measurements fill in the missing temporal element and add significant detail but are limited spatially. Using airmass trajectories derived from model output, we can combine the spatial information from MODIS with the much more detailed information from the surface to address the role of airmass history in governing cloud properties in the high-latitude Southern Ocean.

a. Ship Data

The CAPRICORN II voyage occurred from 11 January to 21 February 2018 between Hobart, Tasmania, and the East Antarctic coastal region. CAPRICORN II collected oceanographic data along transects between the 130°-150°E meridians by occupying some 88 stations from 6 to 24 hours where oceanographic data were collected, followed by movement to the next station several tens of km distant. This approach was advantageous for atmospheric data collection, allowing for longer periods of measurements at latitudes infrequently sampled by voyages focusing on the highest latitudes. On the other hand, the DOE ARM-funded MARCUS deployed instrumentation on the Aurora Australis that was conducting resupply of the Australian Antarctic stations (McFarquhar et al., 2019). Since resupply was the primary objective of the ship, the Aurora Australis made best time between Hobart and the Australian stations in 5 voyages starting in Hobart between November 2017 and March 2018. For a more thorough description of these campaigns, see Mace et al. (2021) and McFarquhar et al. (2019, 2021). The cloud and aerosol measurement instrumentation are described in detail in the supplemental material of McFarquhar et al. (2021).

Cloud properties from the surface-based measurements are derived using a synergistic approach described in Mace et al. (2021). Combining measurements from collocated and vertically pointing W-Band radars, optical elastic lidars, and microwave radiometers (MWR) on each ship, we derive N_d and cloud-top effective radius (r_e) of shallow cloud layers based in the MBL. Our methodology allows for a liquid water path (LWP) constraint provided by the MWR (Turner et al., 2016), and the radar and lidar are combined to constrain N_d and r_e given the LWP. Constraints on N_d and r_e are possible by assuming that N_d is constant with height and the observed liquid water path is distributed subadiabatically, knowing the cloud geometry. The LWP uncertainty is 20% for LWP more than 100 g m^{-2}, scaling to 100% near the detection limit of 20 g m^{-2} while the r_e and N_d retrievals have uncertainty of 30% and 100%, respectively.

b. Airmass Trajectories

We derive 120-hour airmass back and forward trajectories based on the ship location for specific case studies centered on the time of a MODIS overpass. The trajectories are calculated using the National Center for Environmental Prediction’s (NCEP) Global Data Assimilation System (GDAS) (GDAS; Kanamitsu,1989) with the HYSPLIT program (HYSPLIT; Stein et al., 2015) using the Stochastic Time Inverted Lagrangian Transport Model (STILT) (Fasoli et al., 2018) in concentration mode for 100 particle tracers starting near the top of the marine boundary layer. We use the STILT model in our analysis to gauge the extent to which the center of mass trajectory that we derive from the dispersion of the tracers is a viable description of the likely parcel track. We assume that as the particles disperse, the center of mass trajectory becomes less informative of the likely path of the air mass. We use standard deviational ellipses to determine the extent of the particle dispersion within one standard deviation of the center of mass parcel trajectory. We assume that the trajectory is not informative after the ellipse area reaches 40,000 km^2, equivalent approximately to a circle with a 224 km diameter. However, we continue to use the trajectory for anecdotal information on airmass origin, recognizing the uncertainty. While the cutoff size of the ellipse is arbitrary, it is approximately twice the area of a MODIS analysis scene that we use for averaging. We reason that a trajectory uncertainty much larger than this significantly influences our knowledge of the likely location of the airmass and the cloud properties along the trajectory. We run HYSPLIT/STILT for each case study, storing data in 1-hour intervals with the parcel's endpoint being the central latitude and longitude of the cloud scene.

We use reanalysis data sets to describe the meteorology along trajectories from a combination of MERRA-2 reanalysis data and the GDAS model output. MERRA-2 is based on the MERRA system (GEOS-5.2.0) (Rienecker et al., 2011), with numerous updates described in detail in Bosilovich et al., 2015. GDAS provides analyses four times a day and forecasts using the Global Forecast System (GFS) (Abreu et al., 2012).

c. MODIS Data

The AQUA and TERRA satellites each pass over the high latitude SO region twice per day but only once during daylight, providing potentially two views of the cloud field separated by several hours (typically 4-6 hours). Terra usually passes over around 00 UTC or mid-local morning, while Aqua passes closer to 06 UTC or in the early local afternoon. Moderate Resolution Imaging Spectroradiometer (MODIS) level 2 cloud products (MOD06_L2) from MODIS of the AQUA and TERRA satellites provide effective radius (r_e) and optical depth (τ) during daytime from which LWP is derived (Platnick et al., 2003, 2015). We then use the MODIS Level 2 retrievals of τ and r_e to calculate N_d with the method described by Grosvenor et al. (2018), who assume an adiabatically shaped cloud profile among other assumptions. In Mace et al. (2023), we evaluate and use the method outlined in Grosvenor et al. (2018), where we examine the sources of error in the N_d computation from the MODIS retrievals concurring with the analysis in Grosvenor et al. (2018) that a pixel level uncertainty of approximately 80% is appropriate.

We use the same basic approach to sampling the MODIS data used in Mace et al. (2023), where the MODIS data are filtered by solar and view zenith angles, cloud type, and coverage. Every center-of-mass point along a trajectory is assigned a nearest MODIS scene in space and time where a scene is a set of 1-km MODIS pixels in 1° latitude by 2° longitude regions. Only when the MODIS scene is reliably composed of low-level clouds as described in Mace et al. (2023) and is within 180 km and 4 hours of a trajectory point, do we use the scene in further analysis. In a scene, we store the statistics of cloud properties (LWP, τ, r_e, and N_d). Over a trajectory we tend to find clusters of 1 to 3 MODIS overpasses between 00 and 06 UTC each day allowing for a coarse time series of spatially averaged cloud properties along the trajectory paths. We also directly compare the cloud properties retrieved from MODIS with those derived from the ship-based remote sensors. For this comparison, we typically focus on a 50 km diameter set of pixels centered on the ship.

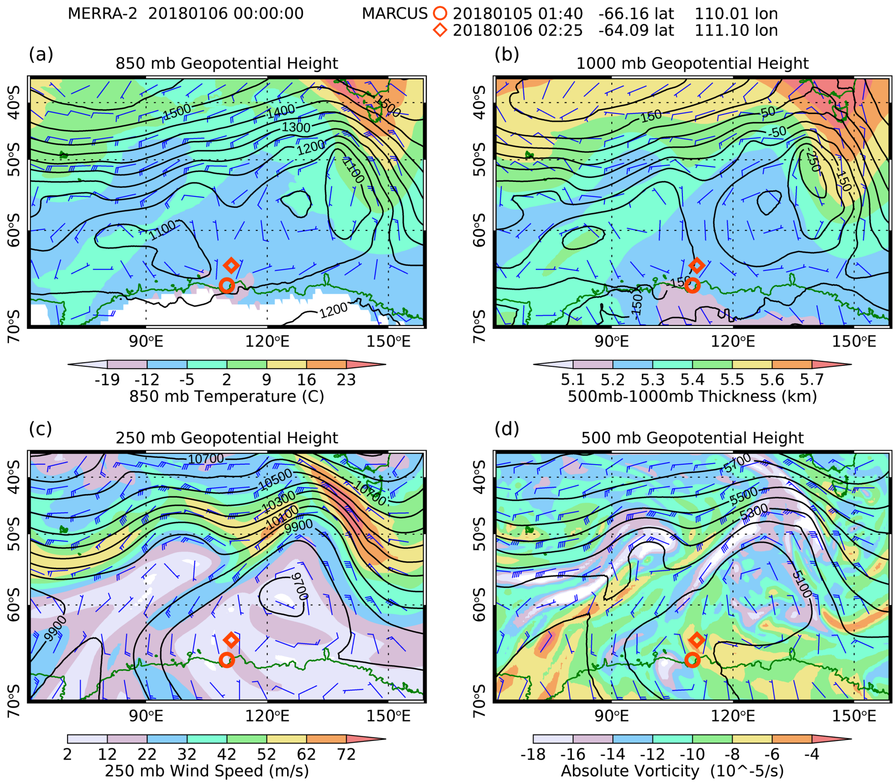

Figure 1. Large-scale meteorology from MERRA-2 during the January 5 and 6 case studies. Location of the ship during the January 5 Terra overpass is marked with a red circle. A red diamond marks the location of the ship on January 6. a) 850 hPa geopotential heights (contours, meters), and 850 hPa temperatures (color shading), b) 1000 hPa geopotential height (contours, m) and 500-1000 hPa thickness (km), c) 250 hPa geopotential height (contours, m) and wind speed, d) 500 hPa Geopotential height (contours, m) and absolute vorticity

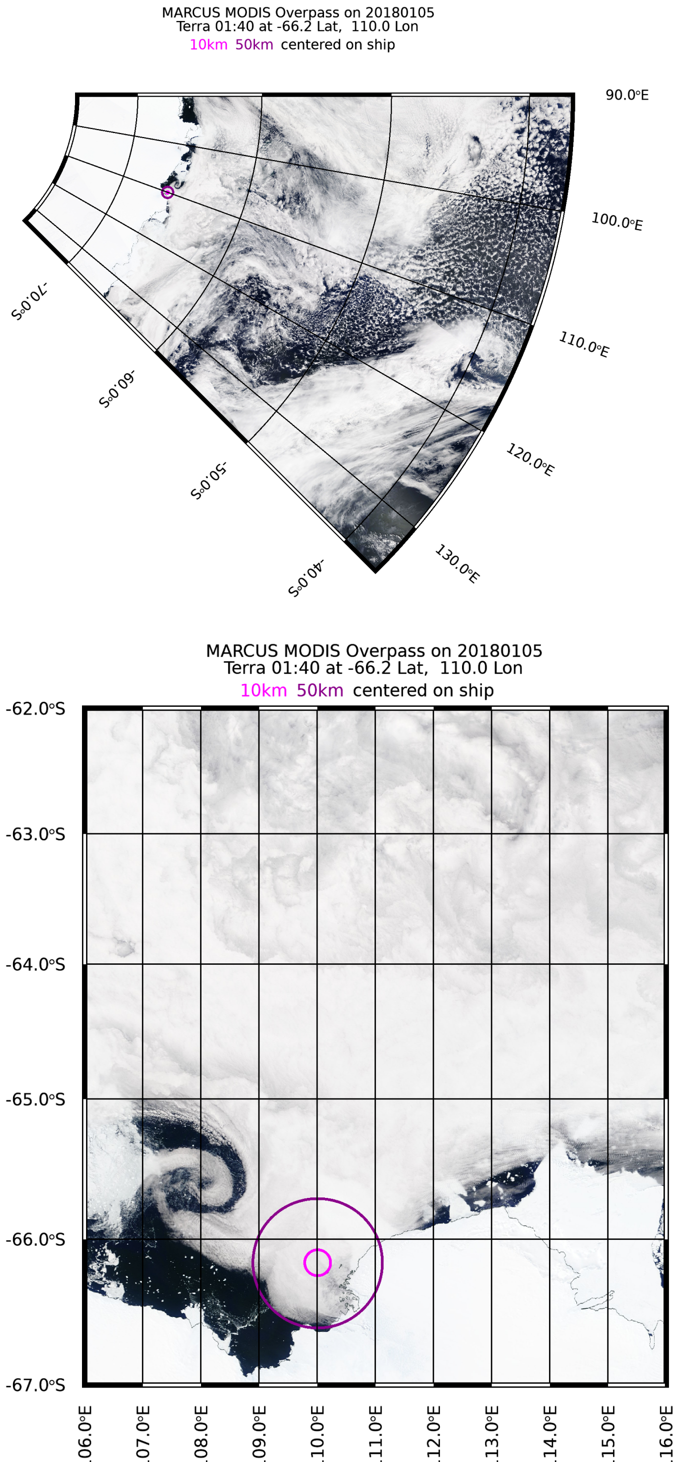

Figure 2. Terra MODIS imagery (3-color visible composite) collected at 0140 UTC January 6, 2018. Ship location shown by concentric 10 and 50 km circles.

We present three case studies - two from the Marcus Campaign and one from Capricorn II. In choosing these case studies, we sought examples that illustrate the statistical findings in Mace et al. (2021 and 2023) and presented in Section 4 of this paper where cloud properties derived from satellite measurements demonstrate a gradient in cloud properties with distance from the Antarctic coast. These case studies had high view-zenith MODIS overpasses of the ship that allows us to establish the validity of derived cloud properties by comparing the MODIS scene statistics with cloud properties derived from the surface-based data. The first two case studies we present are derived from a consecutive 30-hour period during Marcus. We present it as two case studies because the ship, while transiting northeast, sampled two distinctly different air masses that had very different histories and very different cloud and aerosol properties. Case Study 1 is the 12-hour period from 00 UTC until 16 UTC on 1/5 while Case Study 2 begins at ~20 UTC on 1/5 and extends until 05 UTC on 1/6. Case study 3 is from the Capricorn II campaign and extends over a two-day period on the fourth and fifth of February 2018.

3.1 January 5, 2018 - Marcus

The first case study is from data collected on 5 January 2018 (hereafter 1/5) when the Aurora Australis was heading away from Casey Station on a return trip to Hobart. The ship had recently spent nearly a week at Casey and collected atmospheric data as part of Marcus during the port period, as discussed in Mace et al. (2021) where we noted a persistent BL cloud deck with relatively high N_d in an offshore flow. The large-scale atmospheric state at 00 UTC on 1/6 features a jet stream oriented well north along 50°S, and a broad region of weak low pressure with little thermal contrast oriented in a broad trough along 60°S placing the Wilkes Land region of the East Antarctic Coast in a weak easterly flow.

The cloud field observed by MODIS on Terra at 0140 UTC in the region of the Aurora Australis on 1/5 (Fig. 2) shows an extensive BL cloud field with the ship located south-east of a mesoscale cyclonic feature that was also observed in the MODIS Aqua overpass at 07 UTC on this day that gives the appearance of a terrain-induced coastal low. A close examination of the image reveals many small-scale vortices in the near shore closed-cell stratocumulus likely associated with the easterly flow interacting with the complicated coastal geography. Offshore, the closed cellular pattern appears to become more cumuliform with an open cellular structure north of the trough axis along 60°S.

The local meteorology observed by instruments on the ship and 6-hourly soundings (not shown) document an ~5 m/s northeasterly wind with surface temperatures near -2°C. The potential temperature difference between the surface and the 850 hPa layer was 8°C. The boundary layer was well mixed up to an inversion based near 1 km, and the clouds had a temperature near -10°C. Photos and video recorded during this day (not shown) show calm seas with only scattered sea ice and mostly open water under a persistent low overcast.

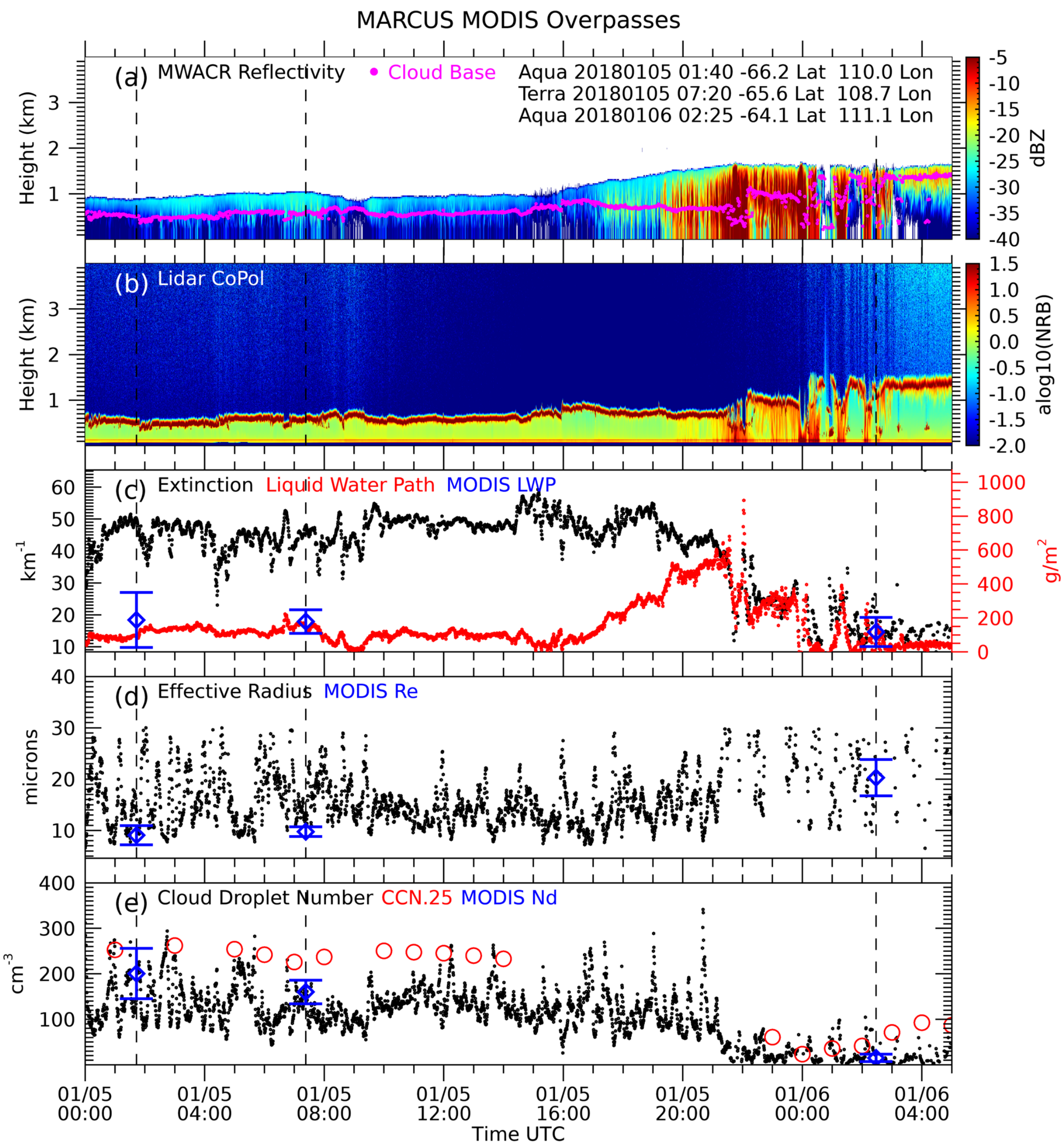

Figure 3. Marcus Surface measurements and derived cloud properties collected from aboard the Aurora Australis on January 5 and 6, 2018. a) w-band radar reflectivity with ceilometer cloud base overlaid (purple), b) lidar copolar attenuated backscatter, c) visible extinction derived from the lidar data using the method of Li et al. (2011), d) effective radius (black) and liquid water path (red), e) cloud droplet number concentration (black) and hourly averaged CCN at 0.25% supersaturation (red circles). MODIS overpass averages and standard deviations shown as blue circles with error bars to indicate standard deviation of 50 km domain centered on the ship.

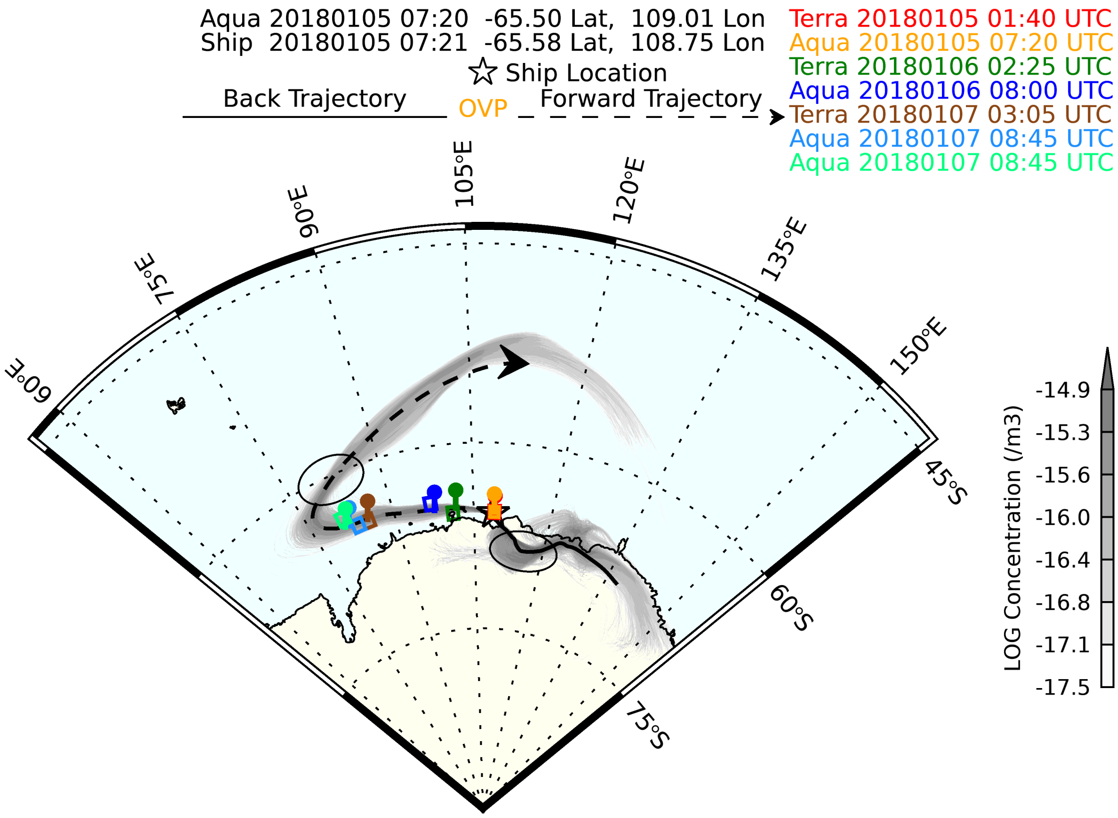

Figure 4. 5-day Hysplit backwards and forwards trajectories from the Marcus ship location (star). The solid and dashed curves show the center-of-mass trajectory from the tracer concentrations (gray). Drop pins denote MODIS overpasses over water with color code corresponding to inset information at the bottom of the figure. The ellipses denote the point in the trajectories where the concentration of tracers denote significant uncertainty in the center of mass trajectory (see text for explanation).

A time series of remote sensing and derived cloud properties in Fig. 3 show an optically thick cloud layer that fully attenuates the lidar. We note that a transition in observed and derived cloud properties begin near 16 UTC on 1/5. This transition period occurs as the ship transits northeastward and encounters an airmass with very different history as discussed below. From

00 to 16 UTC, the layer base is near 600 m, with tops near 1 km. The liquid water path (LWP) derived from the microwave radiometer suggests an approximate 2-hour periodicity in enhanced LWP that correlates with periods of episodic drizzle occurring during the high LWP part of the day. The more extended period oscillations in LWP are also matched in the lidar-derived extinction. On a much more rapid pace, we note oscillations in cloud droplet number Nd, r_e, and lidar-derived extinction. These oscillations are approximately in phase with the drizzle events that had a timescale of 10-15 minutes. Overall, extinction's high (low) values correlate with local maxima (minima) in N_d. The r_e is anticorrelated with N_d, but these quantities are not independent in the retrieval algorithm. We find that the MODIS retrievals are in reasonable agreement with the surface retrievals regarding the mean values and the variability. The depolarization ratio of the lidar is poorly known below values of 0.4 due to a substandard window that was repaired on 13 January, so we infer liquid drizzle based on the Doppler velocity and the low radar reflectivity.

CCN remains in the 250 cm^{-3} range throughout case 1 - well beyond the 75th percentile of CCN at 0.2% supersaturation for this latitude belt compiled from Capricorn II and MARCUS data reported in Humphries et al. (2021). N_d ranges up to the values of the CCN in between

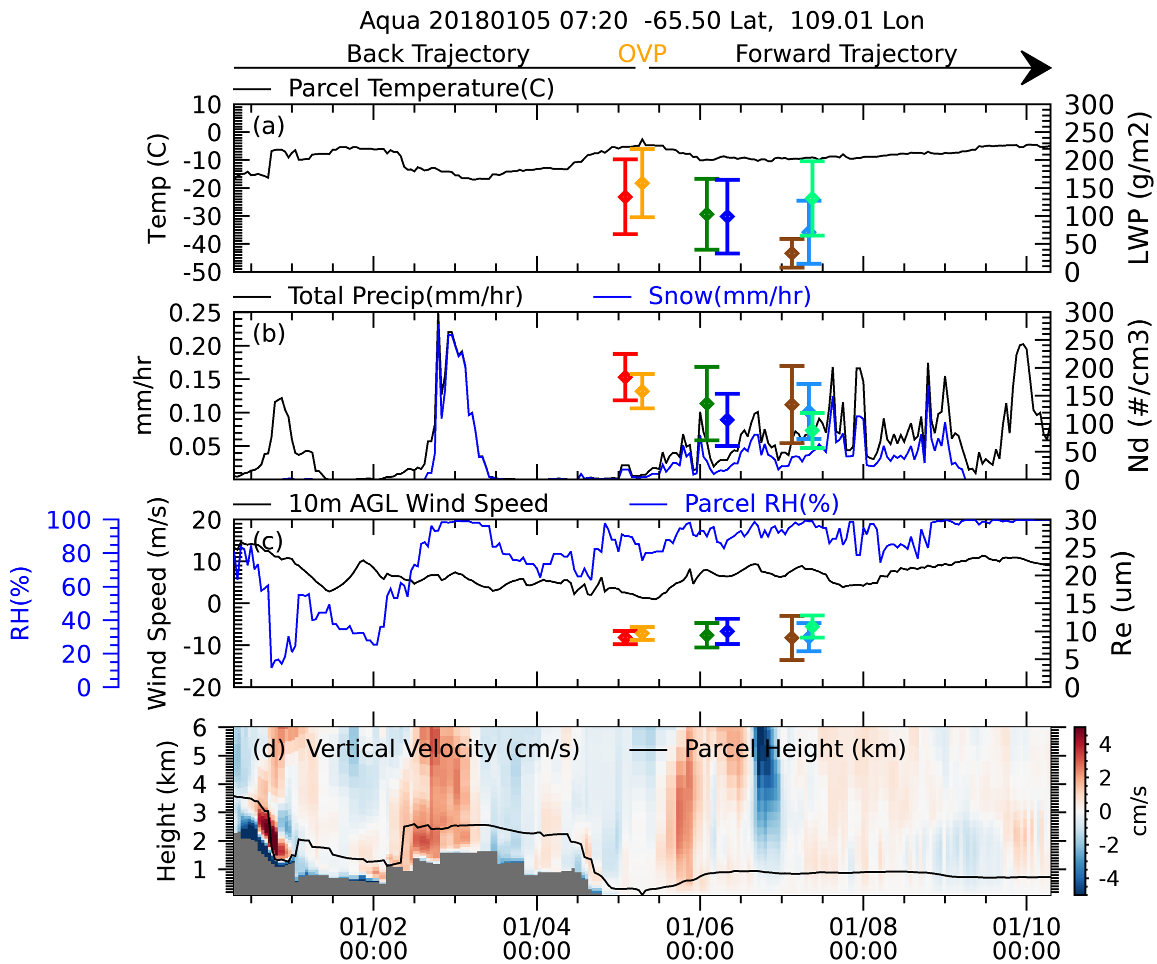

Figure 5. Meteorological quantities and MODIS cloud properties from along the backwards and forwards trajectories depicted in Fig. 4. OVP is the overpass time of the ship by MODIS Aqua on 07:20 UTC. A) Parcel temperature and MODIS LWP, b) Total and snow precipitation rates and MODIS Nd, c) Parcel relative humidity and 10 m wind speed and MODIS r_e, d) large-scale vertical motion and parcel height.

drizzle events to values as low as 80 cm^{-3} in regions near more intense drizzle where the extinction is also a minimum.

Back trajectories (Figs. 4 and 5) show that the air mass had recently descended to near the sea surface, as depicted in downward vertical motions along the parcel trajectory late on 1/4.

Before this, the trajectories suggest that the air mass transited the Law Ice Dome during the previous 48 hours near 2 km altitude and temperatures near -15°C. The MERRA-2 model suggests clear skies during the time over the Law Ice Dome, and this seems reasonable given the quiescent large-scale meteorology. While the back trajectories are quite uncertain during the back trajectory, there is an indication that before being over the Law Ice Dome, the air mass had been at a higher elevation over the ice sheet and then descended to near sea level, passing over the Totten Ice Shelf. Thus, it appears that the air mass had spent little time over the SO in the days before the case study period examined here.

The first cluster of MODIS overpasses of the trajectory are those that passed over the ship on 1/5. After that, two additional sets of overpasses occurred on 1/6 and 1/7. The cloud properties derived along the forward trajectories clustered in time show reasonable consistency in the scene-averaged r_e, N_d, and LWP. During the forward trajectory as the air mass continues roughly westward over the open ocean, the MODIS retrievals show little change in r_e although both LWP and N_d decreased over the ensuing three days. N_d drops from near 200 cm^{-3} during the overpasses of the ship on 1/5 to values near 100 cm^{-3} in the cluster of overpasses on 1/7. The MERRA-2 model suggests that light precipitation in the form of snow in the BL clouds increased after 1/5, consistent with decrease in N_d and LWP.

3.2 January 6, 2018 - Marcus

On the day following the 1/5 case study, January 6, 2018 (1/6), the Aurora Australis had progressed ~240 km to the northeast. They continued northeastward through open water with icebergs and wildely scattered sea ice notable on the video and still photography. The winds had increased and ranged between 10-15 m s^{-1} from the northeast during the early UTC hours of 1/6. The sea state remained unremarkable with no apparent whitecaps that would indicate active sea salt aerosol production locally. Surface temperatures were near and just below freezing, having warmed slightly from the previous day. The sounding at 06 UTC (not shown) shows that the boundary layer had deepened considerably yet remained well mixed with the base of the marine inversion near 1.6 km. The clouds observed by the ARM remote sensors (Fig. 3) show a marked contrast from the previous day with a transition in cloud properties that began at ~16 UTC and was complete by 00 UTC. A persistent layer of BL clouds continues with bases now near 1.4 km

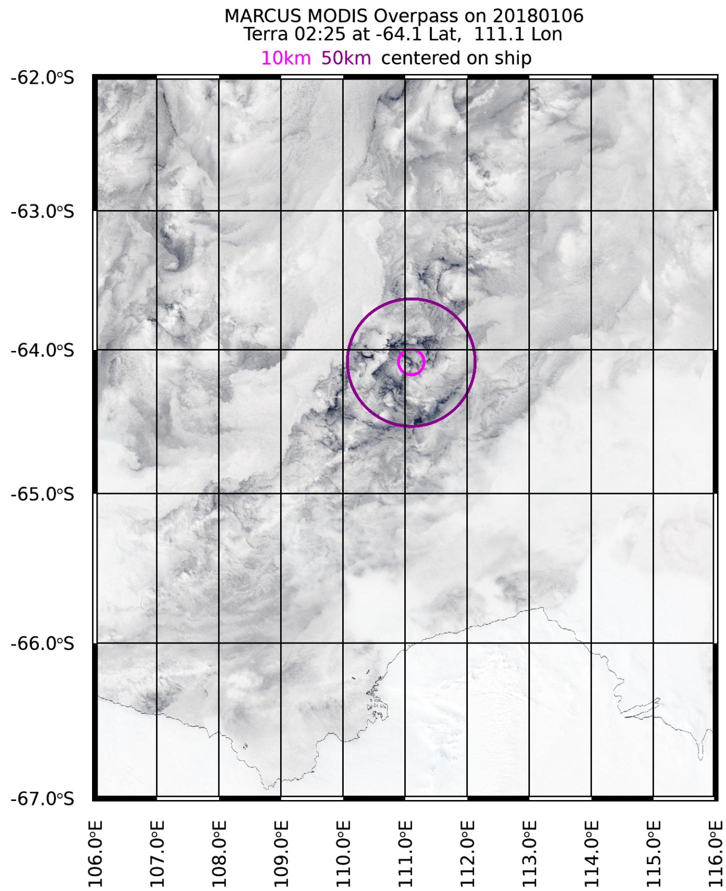

Figure 6. As in Figure 2b but for the Terra MODIS overpass at 02:25 UTC on 1/6.

and with layer top at the bottom of the marine inversion at temperatures near -15ºC. Higher radar reflectivities are noted in precipitation that, according to lidar depolarization above 0.4 during events before 03 UTC, seems to be snow. After 03 UTC, we cannot establish with certainty the phase of the lighter precipitation because of ambiguity in the lidar depolarization noted earlier. The cloud field is variable before 03 UTC, with breaks in the lidar data and lower cloud bases in heavier precipitation. Widely scattered clouds are observed below the bottom of the primary cloud layer.

The variability in the cloud field is consistent with satellite imagery and photography recorded at the ship (Figs. 6 and 7). The MODIS overpass at 0225 UTC places the ship in a region of mostly overcast cloud cover with the sea surface visible in various breaks in the cloud field in the vicinity of the ship. A photograph (Fig. 7) taken at 0226 UTC at the approximate time of the MODIS overpass looks towards the northeast and shows thick boundary layer clouds with breaks in the layer.

Figure 7. Photograph taken from the Aurora Australis at 02:26 UTC on 1/6 near the time of the MODIS overpass. Perspective is looking northeast.

The derived cloud microphysical properties had also changed considerably from 1/5. N_d is overall much lower, ranging between 10 and 50 cm^{-3} with substantial variability in regions of precipitation (Fig. 3). These values of N_d are in broad agreement with MODIS, which records a mean of ~15 cm^{-3}. The effective radius is also larger than on 1/5 and is in the 20-30 um range from the surface, while MODIS retrieves a value near 20. um. LWP is highly variable in and around precipitation, reaching several hundred g m^{-2}. After 03 UTC, the cloud properties become much more steady as the ship moves into the less variable cloud field to the northeast in Fig. 6. However, light precipitation from the cloud continues. The derived cloud properties also continue to show markedly low N_d and high r_e, with the LWP now in the 50 g m^{-2} range, suggesting that the cloud properties feature low N_d and high r_e throughout this area and not just in regions of active precipitation.

The 5-day back trajectories (Fig. 8) suggest that the air mass had exited continental Antarctica near the beginning of the 5-day period on January 1 and likely came from the region of the Amery Ice Shelf in what is known as Prydz Bay. The trajectories are substantially

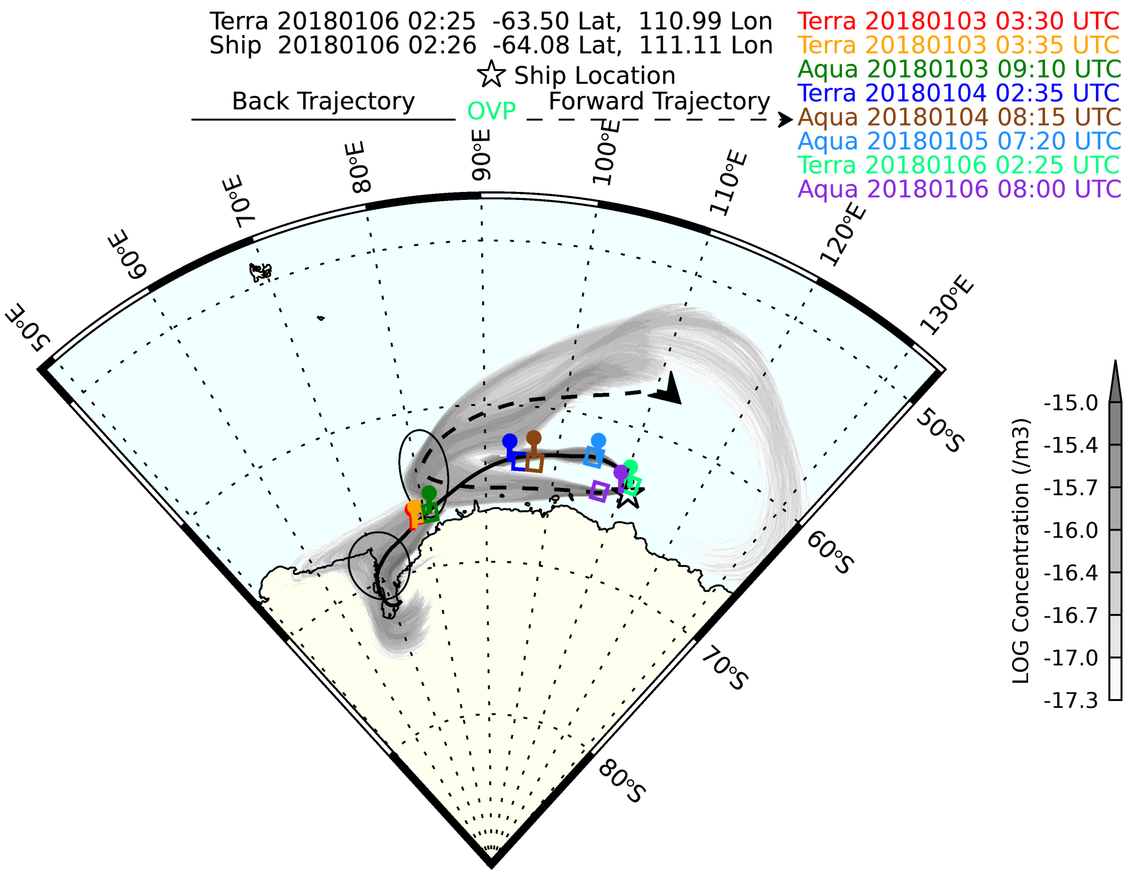

Figure 8. As in Figure 4 except for 1/6.

uncertain. However, the majority of the individual trajectories do show this path. Other parcels follow the coastline near the Amery ice shelf. We can confidently say that the air had been over open water for most of the past five days. There is a well-defined trend in MODIS cloud properties along the trajectory. We pick up the first set of MODIS overpasses of open water early on January 3 and have 4 clusters of overpasses on each succeeding day until 1/6. On 1/3 the Nd was in the 200 cm^{-3} range (Fig. 9) with r_e near 8 um - similar to the values found on 1/5 in the air that had recently descended from the Law Ice Dome. The N_d decreased, and the r_e increased in each successive set of overpasses until 1/6. The LWP, on the other hand, shows a different trend. MODIS records a Maximum LWP near 200 g m^{-2} early on 1/4 while a minimum in LWP occurs 24 hours later. While it borders on speculation, the precipitation (mostly snow) predicted by MERRA-2 seems to lag the LWP by about ½ day.

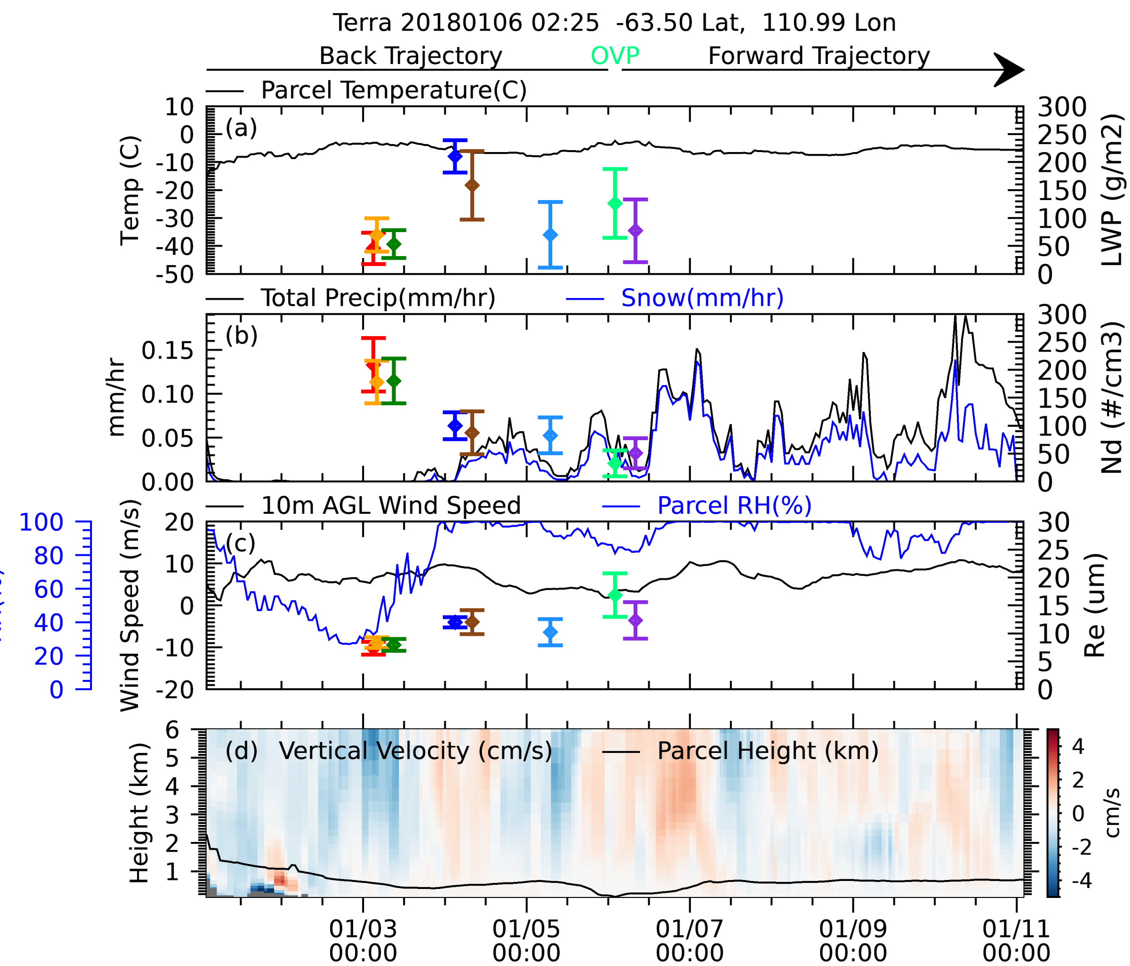

Figure 9. As in Figure 5 except for the 1/6 case study.

It is interesting to note in Fig. 3 that the ship time series of CCN (0.25% supersaturation) during the 1/6 case study period. Recall that 24 hours prior, CCN was in the 250 cm^{-3} range and close to the 90th percentile of CCN for this latitude band. While CCN is missing late on 1/5, the cloud properties transition from the high N_d and small r_e on 1/5 to what is observed on 1/6. It seems clear that the air mass sampled by the Aurora Australis shifted from one that had recently emerged from continental Antarctica to an air mass that had experienced land multiple days prior that now had much lower CCN and N_d.

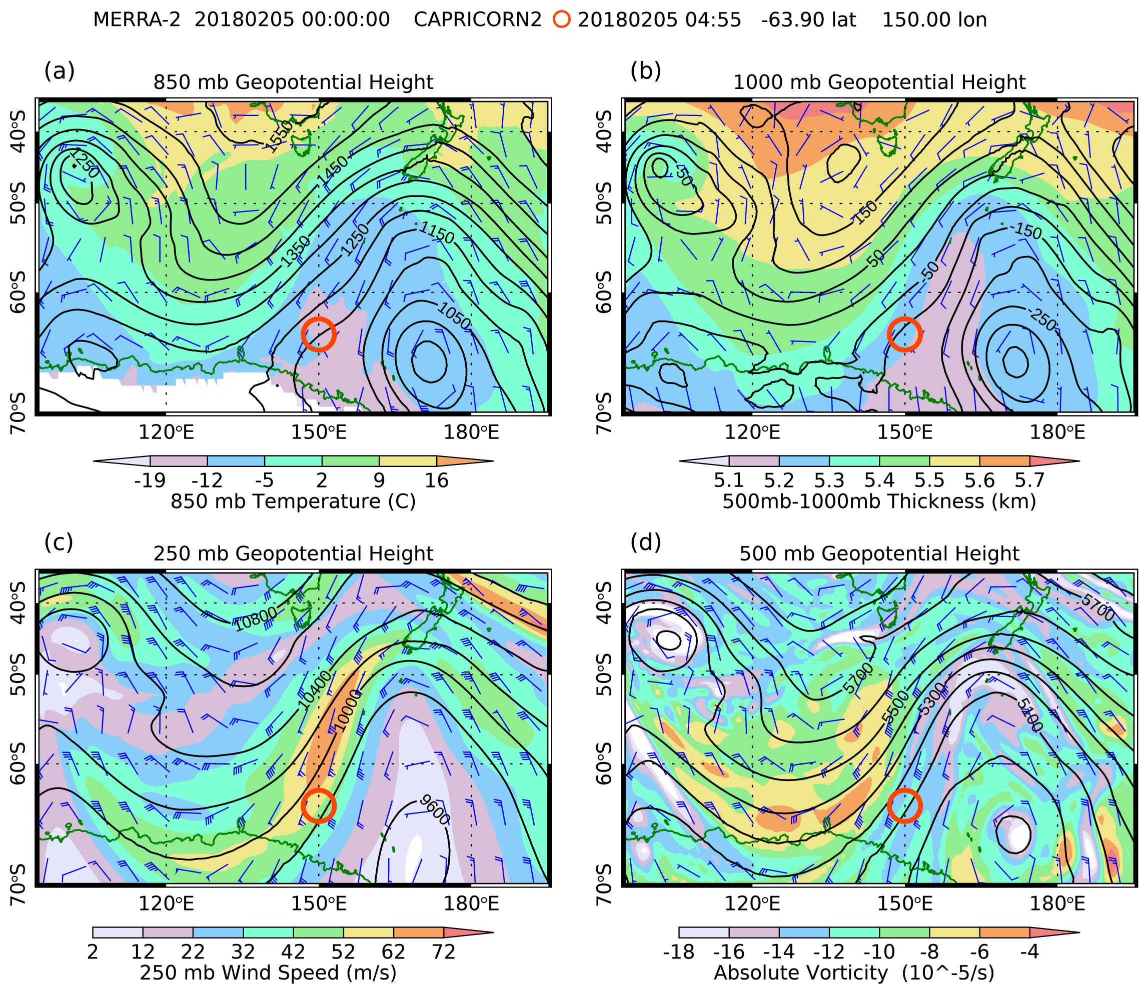

Figure 10. As in Fig. 1 except for the 2/4-2/5 case study. The red circle marks the location of the R/V Investigator on 2/5 at 00 UTC.

3.3 February 4 and 5, 2018 – Capricorn II

We take our final case study from data collected during the Capricorn II campaign on February 4 and 5, 2018 (2/4, 2/5) when the R/V Investigator was conducting oceanographic measurements within 100 km of 65°S, 150°E. The ship began a northward transit at 10 knots during midday on 2/5. The large-scale meteorology (Fig. 10) at 00 UTC on 2/5 shows a highly amplified upper-level pattern with a trough axis along 170°E and a ridge along about 130°E. A frontal system associated with the low pressure centered at 65°S, 170°E at 00 UTC 2/5 had passed over the ship on 2/3 at around 15 UTC, turning the flow out of the south. By 00 UTC on 2/5, this southerly flow extended through the entire troposphere.

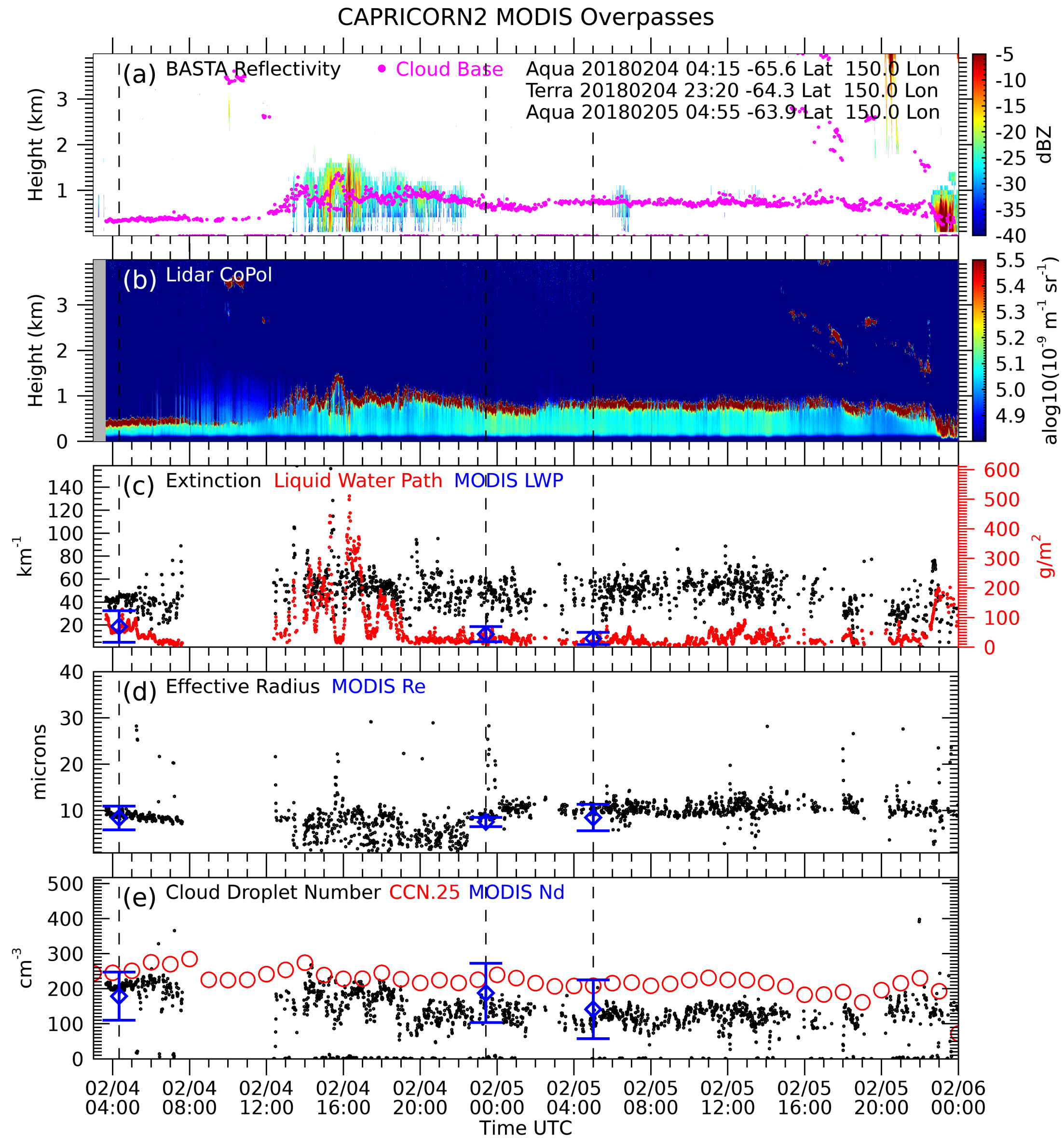

Figure 11. As in Figure 3 except for the 2/4-2/5 Capricorn II Case Study

Weather at the ship evolved somewhat during these two days. Surface temperatures remained between -2° and -4°C, and surface winds were steady out of the south at 10-15 m s^{-1}. Photography collected from the ship shows only modest wind waves, given the ship's proximity to the sea ice edge with very scattered white caps, suggesting the possibility of some local sea salt aerosol generation. A marine inversion was not evident in the 3-hourly soundings until 16 UTC on 2/4 when it was based near 1.5 km with a temperature of -15°C. The inversion strengthened as the pattern progressed, and surface pressure rose through 2/5. By mid-day on 2/5, a strong marine inversion with a 10°C potential temperature difference between the surface and 850 hPa was based near 1 km at a temperature of -10°C.

The BL clouds observed during this case study period (Fig. 11) were based near 500 m and, except for a period between 15 UTC and 23 UTC on 2/4, the clouds were below the detection threshold of the cloud radar on the R/V Investigator (~ -30 dBZe). The lidar shows a mostly overcast cloud layer with some regions of broken clouds and optically thinner clouds where the lidar transmits through the layer. Lidar depolarization ratios (not shown) in the sub-cloud precipitation between 15 and 23 UTC appear to be primarily liquid except for two brief episodes of snow that correlate with maxima in the radar reflectivity of more than -10 dBZe between 15 and 16 UTC. MODIS imagery collected at 04 UTC on 2/4 and 05 UTC on 2/5 (Fig. 12) show the ship's position in relation to the cloud field. Cloud streets that formed as the southerly flow encountered the sea surface and then evolved into closed cellular stratocumulus are evident from the sea ice region to the south of the ship location. The cloud field remained consistent during this southerly flow period.

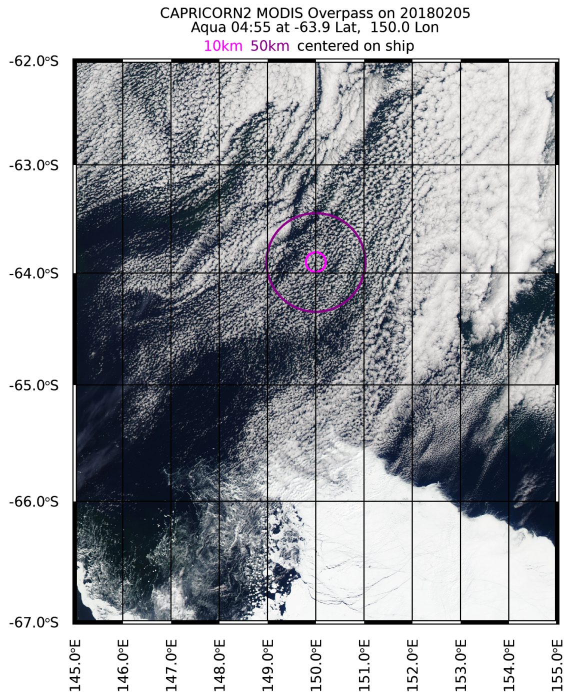

Figure 12. As in Fig. 2b except for a MODIS Aqua overpass of the 2/4-2/5 case study.

Derived cloud properties (Fig. 11) are also remarkably steady during the case study period. N_d decreases from near 200 cm^{-3} in the early part of the period to near 100 cm^{-3} after the light precipitation event at 19 UTC. CCN at 0.25% supersaturation remains steady throughout the case study between 200-300 cm^{-3}. This value of CCN, as the 1/5 case study presented earlier, is beyond the 90th percentile of CCN collected in this latitude belt. r_e follows N_d and remains at or below roughly 10 µm during most of the period. Except in the period with precipitation, the LWP remains in the 50-100 g m^{-2} range. Overall, the MODIS retrievals conducted from the overpasses agree reasonably with the surface-based cloud properties with high N_d and small r_e and LWP less than 50 g m^{-2} at the times of the overpasses. The MODIS N_d is well beyond the upper quartile of N_d recorded by MODIS during a 5-summer period examined in Mace et al. (2023).

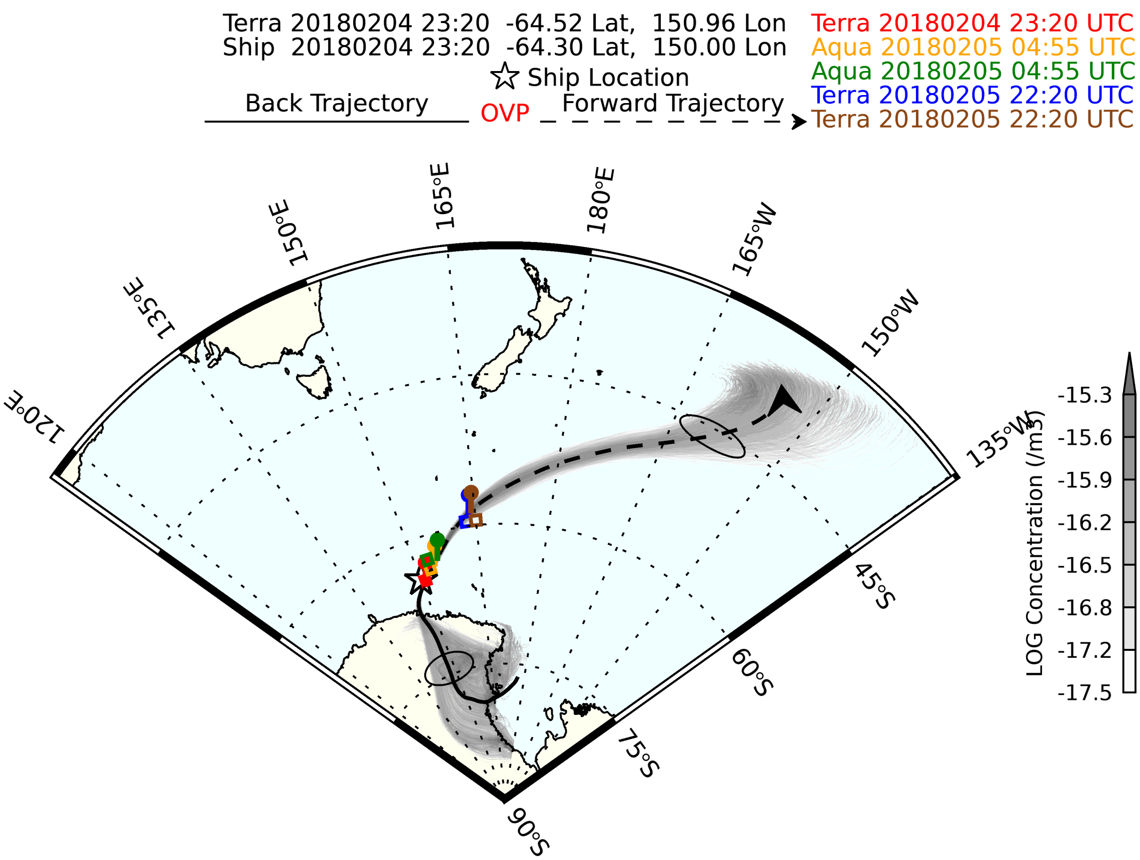

Figure 13. As in Fig. 5 except for 2/5 at 23 UTC.

As shown in Fig. 13, the air sampled during the case study period had spent the previous 48 hours transiting the high-altitude ice dome known as Dome C after it seemingly ascended from the Ross Sea. At least according to the back trajectories, the parcel was well into the free troposphere near 5 km altitude during this transit of Dome C, where predicted temperatures were near -40C. The air sampled at the ship had only been over the sea surface for a very short time after descending in a strong katabatic flow depicted in Fig. 14. After passing over the ship, the forward trajectories show that the air mass continued northeast. In addition to the overpasses near the ship, we find two additional sets of overpasses along the forward trajectory on 2/6 and 2/7. These show that the clouds evolved with increasing LWP and r_e and decreasing N_d to values of 70 cm^{-3} on 2/7.

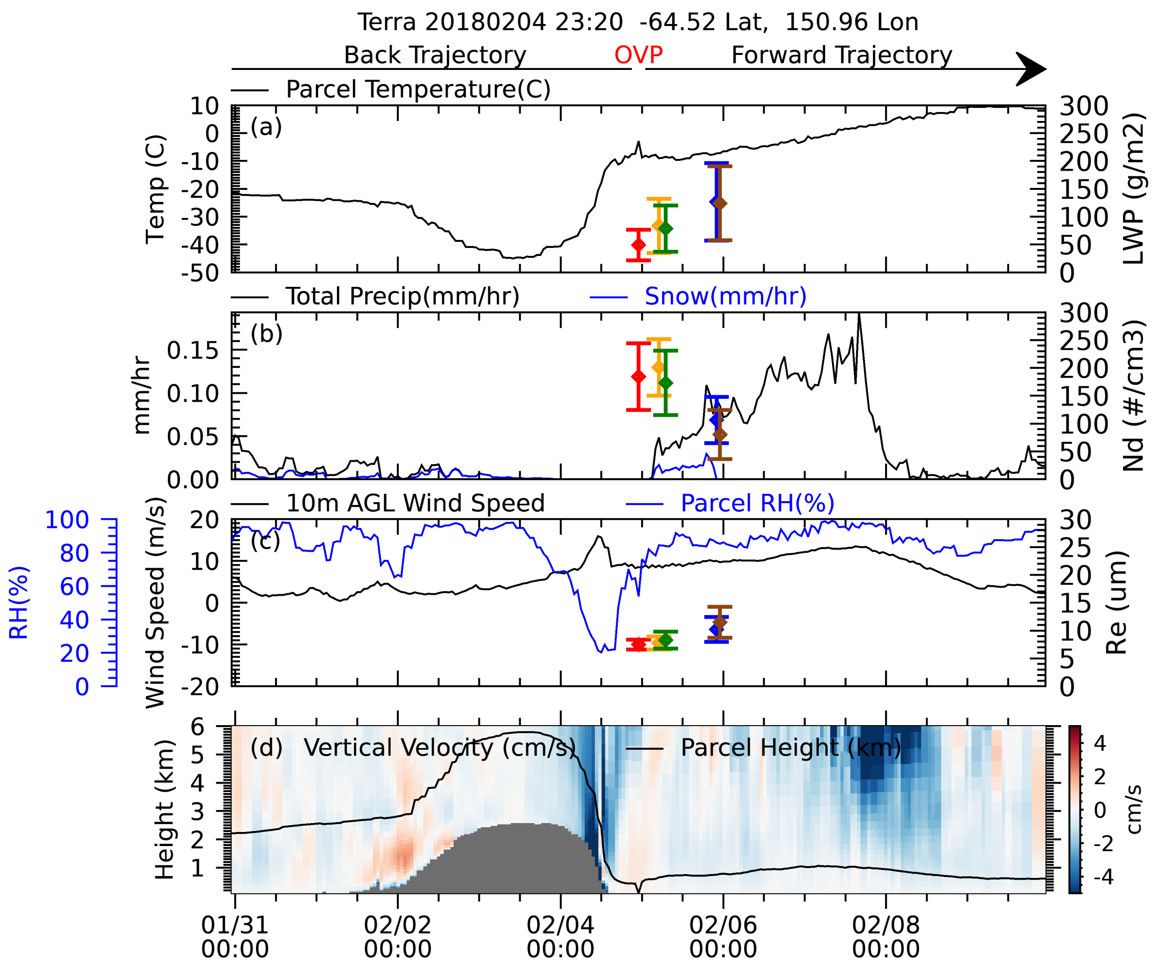

Figure 14. As in Fig. 5 except for 2/4 at 23 UTC.

The Capricorn II data included a Time-of-Flight Aerosol Chemical Speciation Monitor (TOF-ACSM) that allows for inference of aerosol composition. Humphries et al. (2021) present an analysis of the aerosol composition recorded as a function of latitude for recent Southern Ocean voyages. We show the compositional breakdown of the aerosol for the 2/4-2/5 case study in Fig. 15. We find that this aerosol mainly was (90%) sulfate and methanesulfonic acid (MSA). Very low values of other chemical species are indicated, including chloride – a proxy for sea salt. The sea salt and organic fraction of the aerosol composition in the 2/4-2/5 case study was less than half of that compared to the latitudinal average.

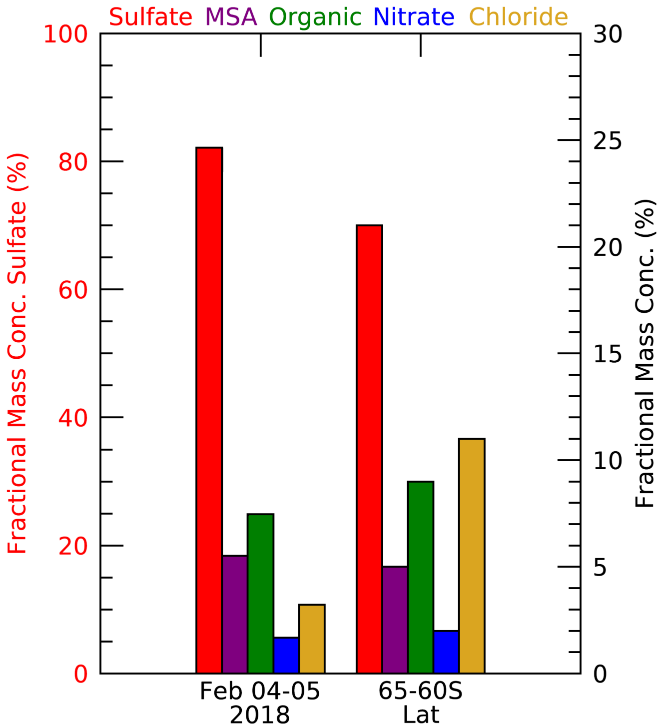

Figure 15. Aerosol compositional fractions from TOF-ACSM for the 2/4-5 case study compared to the latitudinal band average.

4. MODIS statistics

The three case studies we present seem to convey a picture consistent with the statistical findings of Mace et al. (2021 and 2023) and the earlier results of Humphries et al. (2021 and 2016) and going back to Shaw (1988) that air masses that have spent significant time over the Antarctic continental ice sheet emerge over the Southern Ocean with CCN concentrations in the 200-300 cm^{-3} range – significantly higher than the latitudinal average (Humphries et al., 2021). To see how general this association is we examine MODIS data collected from Terra and Aqua during Austral Summers (December through February) between 2014 and 2019 south of 60°S (the same data set used in Mace et al. 2023), except that here we examine the sensitivity of N_d in

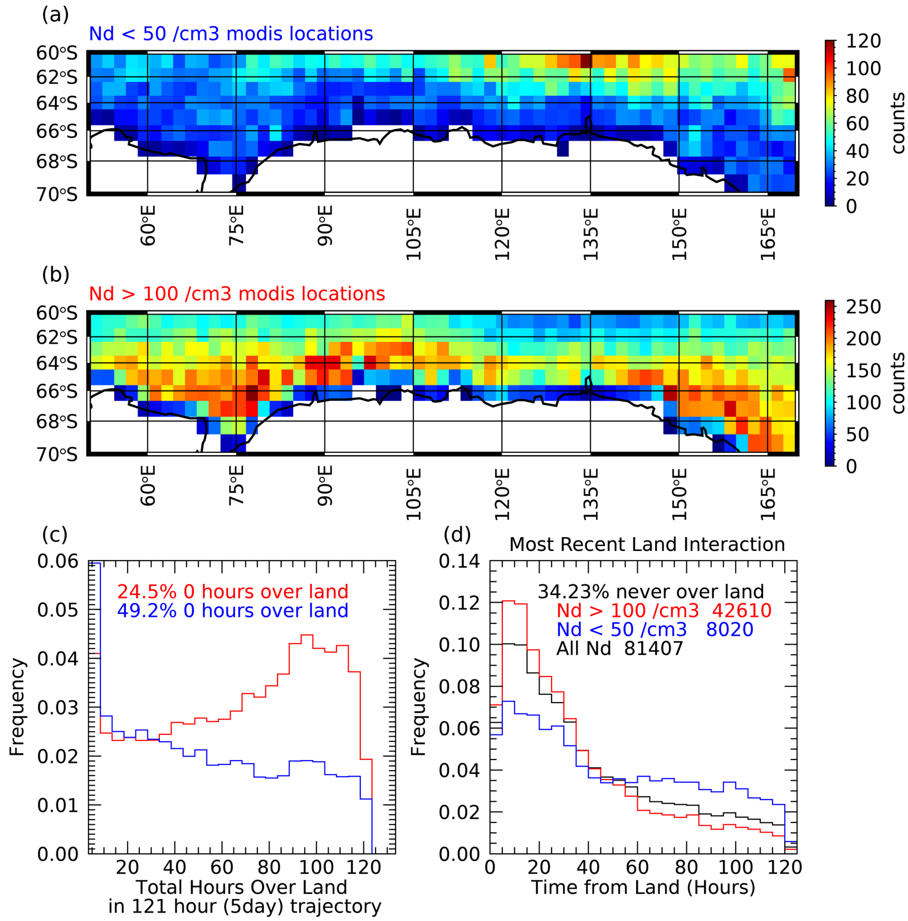

Figure 16. Occurrence of lower (a) and upper (b) Nd quartiles from MODIS data collected during the summers of 2014-2019. c) shows the frequency of occurrence of low (blue) and red (upper) Nd quartiles as a function of total hours spent over continental Antarctica in the 5-day back trajectory. d) shows the frequency of occurrence of the upper (red) and lower (blue) quartiles for over water scenes as a function of time since the back trajectory had encountered the Antarctic land mass.

a MODIS scene to the time since a back trajectory from that scene had encountered the Antarctic Continent. This analysis includes a total of 123784 trajectories. To demonstrate these relationships more clearly, we focus on the upper (100 cm^{-3}) and lower (50 cm^{-3}) N_d quartiles for all MODIS data in the Southern Ocean, as established in Mace et al. (2023).

The occurrence of the upper and lower N_d quartiles is strongly related to latitude (Fig. 16), with the high N_d quartiles tending to be adjacent to coastal Antarctica. Not surprisingly then, we also find that being in one or the other of the N_d quartiles is a strong predictor of the time since a back trajectory had encountered land (Fig. 16d). On the other hand, and perhaps less obviously, the amount of time spent over continental Antarctica seems to be predictive of membership in either the high or low N_d quartile. Nearly half of the low N_d quartile had not encountered land in the 5-day back trajectory, while only 25% of the high N_d quartile had been entirely over open water. Of the back trajectories that had passed over the continent, membership in either quartile depends oppositely on the amount of time spent over land. High N_d seems much more likely as time over the continent increases – rising sharply after 72 hours. On the other hand, low N_d is decreasingly likely as time over the continent increases.

5. Conclusions

While changes to model simulations of cloud feedbacks in the Southern Ocean are primarily responsible for the increased climate sensitivity of CMIP 6 (Zelinka et al., 2020), past work has documented that the cloud properties simulated in models are not consistent with observations of cloud properties Southern Ocean low-level clouds. Because cloud properties are strongly linked to precipitation this lack of consistency between models and observations calls into question the validity of the increased climate sensitivity. Fundamental to this is that the models seem to lack a mechanism to create the observed CCN concentrations (McCoy et al., 2021) over the summertime Southern Ocean. While the primary mechanism behind the high concentrations of CCN is likely related to aerosol nucleation and subsequent growth associated with sulfate precursor gasses emitted by the highly productive ocean biology in the high latitudes, the exact mechanism for the CCN production has not been established and are not simulated in global models of CCN production (Dunne et al., 2016; Lee et al., 2013). In this paper, we build on past work to show that air masses emerging into the MBL over the Southern Ocean after having spent time transiting the continental ice sheets of Antarctica during summer appear predisposed to have anomalously high CCN concentrations and produce clouds that have similarly anomalously high N_d.

Statistics derived from MODIS data demonstrate the relationship between time over Antarctica and high N_d clouds. These statistics are consistent with three case studies collected during ship-based observing campaigns in the Summer of 2018. A case on January 5, 2018, from the Marcus campaign and the 2/4-2/5 case from Capricorn II were of air masses that had recently emerged over the Southern Ocean after spending multiple days over continental Antarctica and had N_d from both MODIS and surface-based retrievals well within the upper quartile of what we find from multi-year MODIS analysis (> 100 cm^{-3}). CCN observed at the surface in the case studies was also anomalously high relative to the mean in the southern high latitudes. In both cases, the N_d along the trajectories decrease with time as the air masses transit over the open ocean and are diagnosed to no longer have N_d that would place it in the upper N_d quartile. On the other hand, the air mass sampled in the 1/6 case from Marcus had not been in contact with land for at least five days, and Nd from both surface and MODIS put it well within the lower N_d quartile. However, MODIS data from along the back trajectories suggest that N_d had decreased from values that placed it within the upper N_d quartile near the start of the 5-day back trajectory after the airmass likely emerged onto the Southern Ocean from the continental landmass.

The relationship between summertime transits of air masses over the continent and the production of high CCN air masses has been described in the literature (Shaw, 1988; Humphries et al., 2016). The highly productive waters of the Antarctic continental shelf produce copious precursor gasses beginning as DMS from phytoplankton along with oxidants such as BrO and IO. These maritime air masses would also contain preexisting primary and secondary aerosols such as sea salt and organics. Under synoptic scale conditions conducive to forced ascent, these air masses ascend the ice domes, where cloud and precipitation processes scavenge most of the ambient aerosol from these air masses. The air masses with high concentrations of aerosol precursor gasses now devoid of ambient aerosol then spend multiple days in the free troposphere at cold temperatures and extremely high actinic flux, enabling new particle formation and growth (Raes, 1995). Growth from the nucleation mode to CCN sizes would take several days, so long trajectories over the ice are favored. The air masses, now with high CCN, descend off the ice sheets in katabatic flows to the Southern Ocean marine boundary layer, forming the high N_d clouds found in observations.

While aspects of this storyline are speculative (the new particle formation process has not been observed in the free troposphere over the ice sheets, for instance), the critical elements of the story are reasonably well documented in past literature or cases and statistics presented in this paper. Additional observations are needed to establish this mechanism's validity more rigorously. It is fortuitous that several upcoming campaigns into the Southern Ocean are planned during this decade (Mallet et al., 2023). What is not planned but needed are airborne campaigns into the high latitudes to document the chemical pathways favored in taking aerosol precursor gasses to new particles that then grow into CCN and eventually seed the high albedo clouds observed by satellite and ground-based sensors.

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Open Research

All data used in this study are available in public archives. MARCUS data are available from the DOE ARM archive at https://adc.arm.gov/armlogin/login.jsp, SOCRATES data are available at https://data.eol.ucar.edu/project/SOCRATES, and CAPRICORN I and II data are available at https://doi.org/10.25919/5f688fcc97166. MODIS cloud products can be found for Terra and Aqua at https://doi.org/10.5067/TERRA/MODIS/L3M/CHL/2018 and https://doi.org/10.5067/MODIS/MYD06_L2.006. GDAS data are obtained from ftp://ftp.arl.noaa.gov/pub/archives/gdas (National Centers for Environmental Prediction, 2022). Computer code for this study including analysis code and graphic generation code is available at https://doi.org/10.7278/S50d-bpx8-gmtt.

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The Association Between Cloud Droplet Number Over the Summer Southern Ocean and Air Mass History.

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Abstract: The cloud properties and governing processes in Southern Ocean marine boundary layer clouds have emerged as a central issue in understanding the Earth's climate sensitivity. While the simulated cloud feedbacks in Southern Ocean clouds have evolved in the most recent climate model intercomparison, the background properties of simulated summertime clouds in the Southern Ocean are not consistent with measurements due to known biases in simulating cloud condensation nuclei concentrations. This paper presents several case studies collected during the Capricorn 2 and Marcus campaigns held aboard Australian research vessels in the Austral Summer of 2018. Combining the surface-observed cases with MODIS data along forward and backward air mass trajectories, we demonstrate the evolution of cloud properties with time. These cases are consistent with multi-year statistics showing that long trajectories of air masses over the Antarctic ice sheet are critical to creating high droplet number clouds in the high latitude summer Southern Ocean. We speculate that secondary aerosol production via the oxidation of biogenically derived aerosol precursor gasses over the high actinic flux region of the high latitude ice sheets is fundamental to maintaining relatively high droplet numbers in Southern Ocean clouds during Summer.

Plain Language Summary: The amount of warming the Earth will experience because of increasing carbon dioxide levels in the atmosphere is sensitive to the properties of clouds that occur over the Southern Ocean. The atmosphere over the circumpolar Southern Ocean is poorly understood and presents significant challenges to climate models. Here we document the properties of the ubiquitous Southern Ocean low-level clouds that exert a strong influence on the albedo of this region. We find that high cloud droplet number concentrations are associated with air masses that have taken paths over the high-altitude ice sheets. The chemistry of the aerosol on which the cloud droplets form suggests that newly formed aerosols that have condensed from gasses emitted by phytoplankton in the highly productive waters near Antarctica are an important component of the cloud properties that must be correctly simulated in models.

Main Point 1: High cloud droplet number concentrations are associated with air masses that have recently passed over continental Antarctica.

Main Point 2: Cloud droplet number concentrations decrease with time as clouds evolve in over-water trajectories due to scavenging by precipitation.

Main Point 3: Katabatic flows bring high concentrations of cloud condensation nuclei to the marine boundary layer where they influence cloud properties.

1 Introduction

As one of the cloudiest regions on Earth, with cloud cover near 90% (Mace et al., 2009; Mace & Zhang, 2014), the high latitude Southern Ocean from roughly poleward of the Antarctic Circumpolar Current to the Antarctic Continent is a critical component of the Earth's climate system. In addition to fundamental oceanographic processes such as the uptake of atmospheric heat and carbon in the formation of the intermediate and deep-water masses (Armour et al., 2016; Morrison et al., 2016), the albedo of the extensive cloud cover, particularly in summer, induces significant controls on the column energy budget of the region and influences the Earth's climate (Zelinka et al., 2020; Gettelman et al., 2019).

The high cloud cover of the Southern Ocean is primarily due to extensive marine boundary layer (MBL) clouds, most of which are liquid phase and subfreezing (Bodas-Salcedo et al., 2016). These clouds, while mostly nonprecipitating (Huang et al., 2016), generate both liquid- and ice-phase precipitation (Mace et al., 2021). Because precipitation exerts controls on cloud cover (Albrecht et al., 1989), precipitation in these clouds continues to be a topic of active research (Kang et al., 2022). However, cloud properties and the associated cloud albedo, as well as the precipitation processes, respond to a significant seasonal cycle in cloud condensation nuclei (CCN) that directly influence cloud droplet number concentration (N_d) (McCoy et al., 2015; Kruger and Grasl, 2011; Mace et al., 2023, 2021). Analyzing cloud properties derived from A-Train data, Mace and Avey (2017) find that the Summer N_d in the SO is approximately a factor of two higher in summer compared to winter. While this seasonal cycle in N_d is thought to be associated with seasonally higher CCN associated with due to secondary aerosol production derived from the oxidation of biogenically-derived precursor gasses such as dimethyl sulfide (DMS) (Lana et al., 2012), aerosol measurements collected during recent campaigns have not directly observed new particle formation events. However, robust circumstantial evidence suggests that new particle formation of sulfur-based particles with a volatility similar to sulfuric acid in the SO free troposphere is widespread and frequent (McCoy et al., 2021).

This hypothesis is consistent with prior studies suggesting that particle concentrations in the free troposphere in marine regions are controlled by sulfuric-acid-driven nucleation of nanoparticles resulting from the oxidation of oceanic dimethylsulfide transported upward. Downward transfer of these nanoparticles then supplies the marine boundary layer with CCN concentration. This mechanism of aerosol production has been observed (Clarke, 1993) and modeled (Russell et al., 1994; Raes, 1995) previously. Most recently, laboratory work on sulfuric acid nucleation and further growth have demonstrated that these processes can survive evaporation and even grow during these transport processes and temperature transitions (Tiszenkel et al., 2019)

Trajectory analyses also point to a source of CCN from airmasses emanating from the Antarctic continent (Humphries et al., 2016; Twohy et al., 2021; Simmons et al., 2021). Specifically, the Antarctic continent in summer has been identified as a potential source of sulfate aerosol in work dating back to the 1980's (Shaw, 1988). Presumably, air masses with high DMS and oxidants are lofted over the high albedo and mostly cloud-free ice sheets in synoptic scale ascent, where most ambient aerosols are removed by precipitation. New sulfate-containing aerosol particles nucleate and grow during the time spent over the ice sheets in the cold free-troposphere. Katabatic flows then cause these newly formed aerosols to emerge into the marine boundary layer in the waters adjacent to the Antarctic continent (Simmons et al., 2021).

Using MODIS data, we recently showed that N_d in the East Antarctic sector of the SO exhibits strong latitudinal gradients during Summer, with the highest N_d in the waters immediately adjacent to the Antarctic Continent (Mace et al., 2023). These results agreed broadly with findings compiled from the Measurements of Aerosols, Radiation, and Clouds over the Southern Ocean (MARCUS) and the Clouds, Aerosols, Precipitation, Radiation, and atmospheric Composition over the Southern Ocean (CAPRICORN II) (McFarquhar et al., 2021) campaigns that collected data in this region during Summer 2018 (Mace et al., 2021). The latitudinal gradient in N_d is a curious feature of the SO region. It seems consistent with the idea that the highest CCN concentration is found immediately adjacent to the Antarctic continent. Cloud and precipitation processing of the aerosol then decrease N_d with distance away from the Antarctic coast. However, the CCN concentration and N_d remain mostly above sea salt aerosol concentration, suggesting a source of new particles mixing into the MBL from the free troposphere (McCoy et al., 2021). The extent to which the background CCN and N_d are maintained against cloud processing by mixing from the free troposphere, as suggested by Twohy et al. (2021) and McCoy et al. (2021), is unknown.

The processes that maintan N_d in the summer SO are crucial to understanding the Earth's climate sensitivity. In the most recent climate model intercomparison project (CMIP6; Eyring et al., 2016), the high latitude SO had significantly less negative cloud feedback than in CMIP5 and earlier model intercomparison studies. While this change in simulated cloud feedback was due partially to an observationally consistent decrease in ice-phase precipitation, the cloud feedback change occurred within a simulated background state in cloud properties that is unrealistic. As shown by McCoy et al. (2021), the CAM6 model consistently underpredicts, by approximately a factor of 2, the N_d in comparisons to collocated measurements during the Southern Ocean Cloud Radiation Aerosol Transport Experimental Study (SOCRATES) campaign (McFarquhar et al., 2021). McCoy et al., (2021) trace this bias to an insufficient DMS oxidation mechanism in the model among other issues. Since N_d and precipitation scale inversely in MBL clouds (Comstock et al., 2004), a correct simulation of N_d is a prerequisite to correctly predicting the precipitation that is driving the negative feedback changes in CAM6 (Gettleman et al., 2019) poleward of about 50°S.

This paper examines three case studies collected in the Australian East Antarctic sector of the SO from the Summer of 2018 during the MARCUS and CAPRICORN II campaigns. Using a combination of surface-based cloud property diagnostics and aerosol in situ measurements, we examine MODIS-derived cloud properties along backward and forward trajectories. We show that clouds forming in air masses recently descended in Katabatic flows from the high-altitude Antarctic ice sheets have significantly higher N_d than the mean for that latitude. The N_d then decreases with time as the air masses pass over the open ocean. We then place these case studies within a larger context of MODIS data collected over 5 Austral Summers to show a consistent relationship between N_d and trajectories emanating from the Antarctic continental ice sheets.

2. Methods

In this study, we combine data from the MARCUS and CAPRICORN II ship-based campaigns with MODIS level 2 data products extracted from overpasses along backward and forward airmass trajectories centered on the ships. This combination of surface and satellite measurements linked by trajectory modeling allows us to exploit the synergy in the surface and satellite measurements. While polar-orbiting satellite measurements provide spatial context, they only provide a single snapshot in time and have only limited information on the cloud field. Surface-based measurements fill in the missing temporal element and add significant detail but are limited spatially. Using airmass trajectories derived from model output, we can combine the spatial information from MODIS with the much more detailed information from the surface to address the role of airmass history in governing cloud properties in the high-latitude Southern Ocean.

a. Ship Data

The CAPRICORN II voyage occurred from 11 January to 21 February 2018 between Hobart, Tasmania, and the East Antarctic coastal region. CAPRICORN II collected oceanographic data along transects between the 130°-150°E meridians by occupying some 88 stations from 6 to 24 hours where oceanographic data were collected, followed by movement to the next station several tens of km distant. This approach was advantageous for atmospheric data collection, allowing for longer periods of measurements at latitudes infrequently sampled by voyages focusing on the highest latitudes. On the other hand, the DOE ARM-funded MARCUS deployed instrumentation on the Aurora Australis that was conducting resupply of the Australian Antarctic stations (McFarquhar et al., 2019). Since resupply was the primary objective of the ship, the Aurora Australis made best time between Hobart and the Australian stations in 5 voyages starting in Hobart between November 2017 and March 2018. For a more thorough description of these campaigns, see Mace et al. (2021) and McFarquhar et al. (2019, 2021). The cloud and aerosol measurement instrumentation are described in detail in the supplemental material of McFarquhar et al. (2021).

Cloud properties from the surface-based measurements are derived using a synergistic approach described in Mace et al. (2021). Combining measurements from collocated and vertically pointing W-Band radars, optical elastic lidars, and microwave radiometers (MWR) on each ship, we derive N_d and cloud-top effective radius (r_e) of shallow cloud layers based in the MBL. Our methodology allows for a liquid water path (LWP) constraint provided by the MWR (Turner et al., 2016), and the radar and lidar are combined to constrain N_d and r_e given the LWP. Constraints on N_d and r_e are possible by assuming that N_d is constant with height and the observed liquid water path is distributed subadiabatically, knowing the cloud geometry. The LWP uncertainty is 20% for LWP more than 100 g m^{-2}, scaling to 100% near the detection limit of 20 g m^{-2} while the r_e and N_d retrievals have uncertainty of 30% and 100%, respectively.

b. Airmass Trajectories

We derive 120-hour airmass back and forward trajectories based on the ship location for specific case studies centered on the time of a MODIS overpass. The trajectories are calculated using the National Center for Environmental Prediction's (NCEP) Global Data Assimilation System (GDAS) (GDAS; Kanamitsu,1989) with the HYSPLIT program (HYSPLIT; Stein et al., 2015) using the Stochastic Time Inverted Lagrangian Transport Model (STILT) (Fasoli et al., 2018) in concentration mode for 100 particle tracers starting near the top of the marine boundary layer. We use the STILT model in our analysis to gauge the extent to which the center of mass trajectory that we derive from the dispersion of the tracers is a viable description of the likely parcel track. We assume that as the particles disperse, the center of mass trajectory becomes less informative of the likely path of the air mass. We use standard deviational ellipses to determine the extent of the particle dispersion within one standard deviation of the center of mass parcel trajectory. We assume that the trajectory is not informative after the ellipse area reaches 40,000 km^2, equivalent approximately to a circle with a 224 km diameter. However, we continue to use the trajectory for anecdotal information on airmass origin, recognizing the uncertainty. While the cutoff size of the ellipse is arbitrary, it is approximately twice the area of a MODIS analysis scene that we use for averaging. We reason that a trajectory uncertainty much larger than this significantly influences our knowledge of the likely location of the airmass and the cloud properties along the trajectory. We run HYSPLIT/STILT for each case study, storing data in 1-hour intervals with the parcel's endpoint being the central latitude and longitude of the cloud scene.

We use reanalysis data sets to describe the meteorology along trajectories from a combination of MERRA-2 reanalysis data and the GDAS model output. MERRA-2 is based on the MERRA system (GEOS-5.2.0) (Rienecker et al., 2011), with numerous updates described in detail in Bosilovich et al., 2015. GDAS provides analyses four times a day and forecasts using the Global Forecast System (GFS) (Abreu et al., 2012).

c. MODIS Data

The AQUA and TERRA satellites each pass over the high latitude SO region twice per day but only once during daylight, providing potentially two views of the cloud field separated by several hours (typically 4-6 hours). Terra usually passes over around 00 UTC or mid-local morning, while Aqua passes closer to 06 UTC or in the early local afternoon. Moderate Resolution Imaging Spectroradiometer (MODIS) level 2 cloud products (MOD06_L2) from MODIS of the AQUA and TERRA satellites provide effective radius (r_e) and optical depth (τ) during daytime from which LWP is derived (Platnick et al., 2003, 2015). We then use the MODIS Level 2 retrievals of τ and r_e to calculate N_d with the method described by Grosvenor et al. (2018), who assume an adiabatically shaped cloud profile among other assumptions. In Mace et al. (2023), we evaluate and use the method outlined in Grosvenor et al. (2018), where we examine the sources of error in the N_d computation from the MODIS retrievals concurring with the analysis in Grosvenor et al. (2018) that a pixel level uncertainty of approximately 80% is appropriate.

We use the same basic approach to sampling the MODIS data used in Mace et al. (2023), where the MODIS data are filtered by solar and view zenith angles, cloud type, and coverage. Every center-of-mass point along a trajectory is assigned a nearest MODIS scene in space and time where a scene is a set of 1-km MODIS pixels in 1° latitude by 2° longitude regions. Only when the MODIS scene is reliably composed of low-level clouds as described in Mace et al. (2023) and is within 180 km and 4 hours of a trajectory point, do we use the scene in further analysis. In a scene, we store the statistics of cloud properties (LWP, τ, r_e, and N_d). Over a trajectory we tend to find clusters of 1 to 3 MODIS overpasses between 00 and 06 UTC each day allowing for a coarse time series of spatially averaged cloud properties along the trajectory paths. We also directly compare the cloud properties retrieved from MODIS with those derived from the ship-based remote sensors. For this comparison, we typically focus on a 50 km diameter set of pixels centered on the ship.

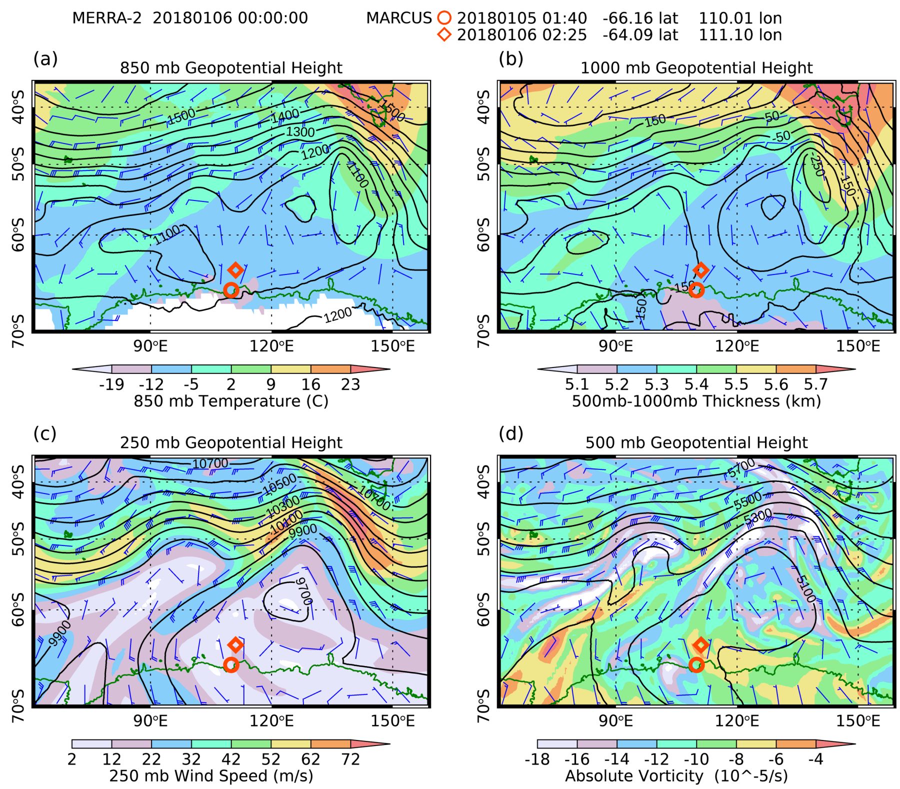

Figure 1. Large-scale meteorology from MERRA-2 during the January 5 and 6 case studies. Location of the ship during the January 5 Terra overpass is marked with a red circle. A red diamond marks the location of the ship on January 6. a) 850 hPa geopotential heights (contours, meters), and 850 hPa temperatures (color shading), b) 1000 hPa geopotential height (contours, m) and 500-1000 hPa thickness (km), c) 250 hPa geopotential height (contours, m) and wind speed, d) 500 hPa Geopotential height (contours, m) and absolute vorticity

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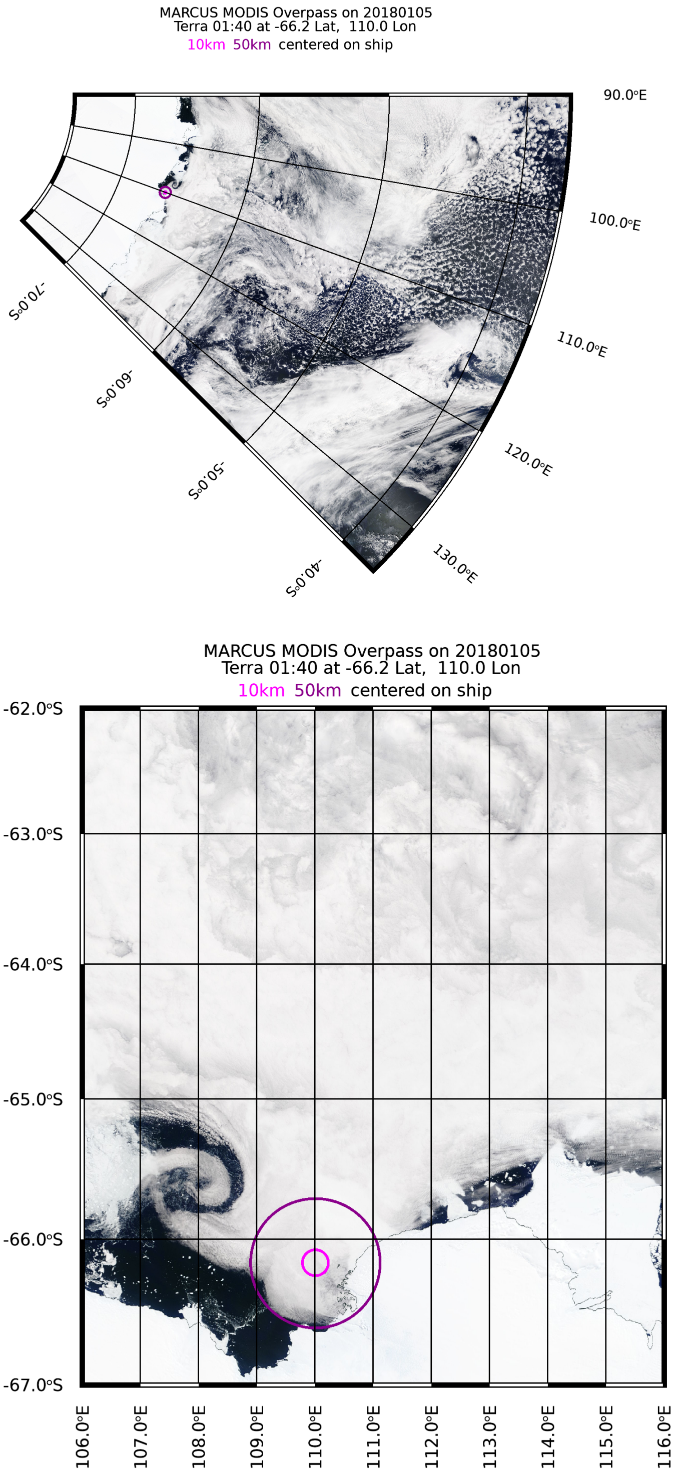

Figure 2. Terra MODIS imagery (3-color visible composite) collected at 0140 UTC January 6, 2018. Ship location shown by concentric 10 and 50 km circles.

We present three case studies - two from the Marcus Campaign and one from Capricorn II. In choosing these case studies, we sought examples that illustrate the statistical findings in Mace et al. (2021 and 2023) and presented in Section 4 of this paper where cloud properties derived from satellite measurements demonstrate a gradient in cloud properties with distance from the Antarctic coast. These case studies had high view-zenith MODIS overpasses of the ship that allows us to establish the validity of derived cloud properties by comparing the MODIS scene statistics with cloud properties derived from the surface-based data. The first two case studies we present are derived from a consecutive 30-hour period during Marcus. We present it as two case studies because the ship, while transiting northeast, sampled two distinctly different air masses that had very different histories and very different cloud and aerosol properties. Case Study 1 is the 12-hour period from 00 UTC until 16 UTC on 1/5 while Case Study 2 begins at ~20 UTC on 1/5 and extends until 05 UTC on 1/6. Case study 3 is from the Capricorn II campaign and extends over a two-day period on the fourth and fifth of February 2018.

3.1 January 5, 2018 - Marcus

The first case study is from data collected on 5 January 2018 (hereafter 1/5) when the Aurora Australis was heading away from Casey Station on a return trip to Hobart. The ship had recently spent nearly a week at Casey and collected atmospheric data as part of Marcus during the port period, as discussed in Mace et al. (2021) where we noted a persistent BL cloud deck with relatively high N_d in an offshore flow. The large-scale atmospheric state at 00 UTC on 1/6 features a jet stream oriented well north along 50°S, and a broad region of weak low pressure with little thermal contrast oriented in a broad trough along 60°S placing the Wilkes Land region of the East Antarctic Coast in a weak easterly flow.

The cloud field observed by MODIS on Terra at 0140 UTC in the region of the Aurora Australis on 1/5 (Fig. 2) shows an extensive BL cloud field with the ship located south-east of a mesoscale cyclonic feature that was also observed in the MODIS Aqua overpass at 07 UTC on this day that gives the appearance of a terrain-induced coastal low. A close examination of the image reveals many small-scale vortices in the near shore closed-cell stratocumulus likely associated with the easterly flow interacting with the complicated coastal geography. Offshore, the closed cellular pattern appears to become more cumuliform with an open cellular structure north of the trough axis along 60°S.

The local meteorology observed by instruments on the ship and 6-hourly soundings (not shown) document an ~5 m/s northeasterly wind with surface temperatures near -2°C. The potential temperature difference between the surface and the 850 hPa layer was 8°C. The boundary layer was well mixed up to an inversion based near 1 km, and the clouds had a temperature near -10°C. Photos and video recorded during this day (not shown) show calm seas with only scattered sea ice and mostly open water under a persistent low overcast.

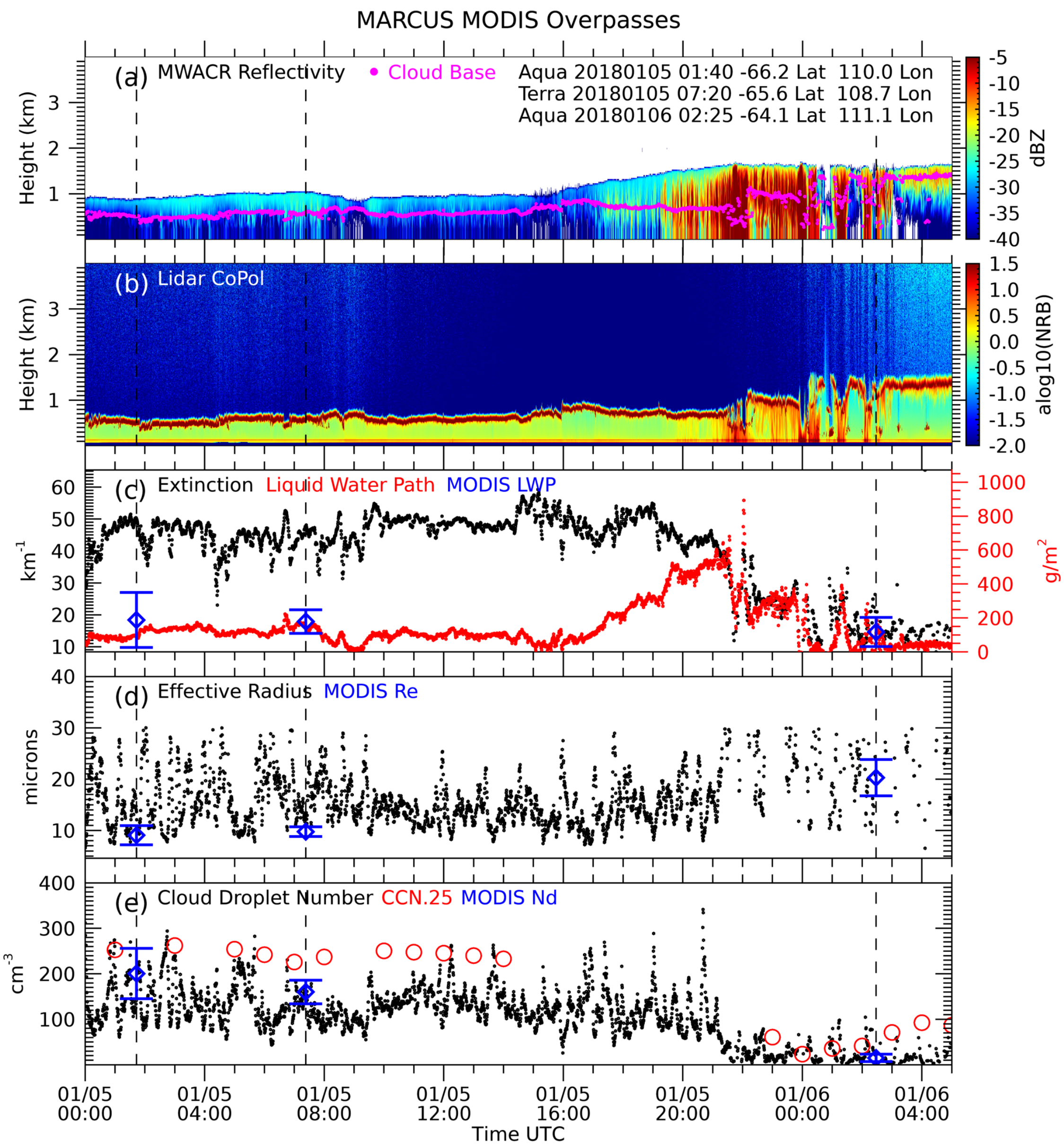

Figure 3. Marcus Surface measurements and derived cloud properties collected from aboard the Aurora Australis on January 5 and 6, 2018. a) w-band radar reflectivity with ceilometer cloud base overlaid (purple), b) lidar copolar attenuated backscatter, c) visible extinction derived from the lidar data using the method of Li et al. (2011), d) effective radius (black) and liquid water path (red), e) cloud droplet number concentration (black) and hourly averaged CCN at 0.25% supersaturation (red circles). MODIS overpass averages and standard deviations shown as blue circles with error bars to indicate standard deviation of 50 km domain centered on the ship.

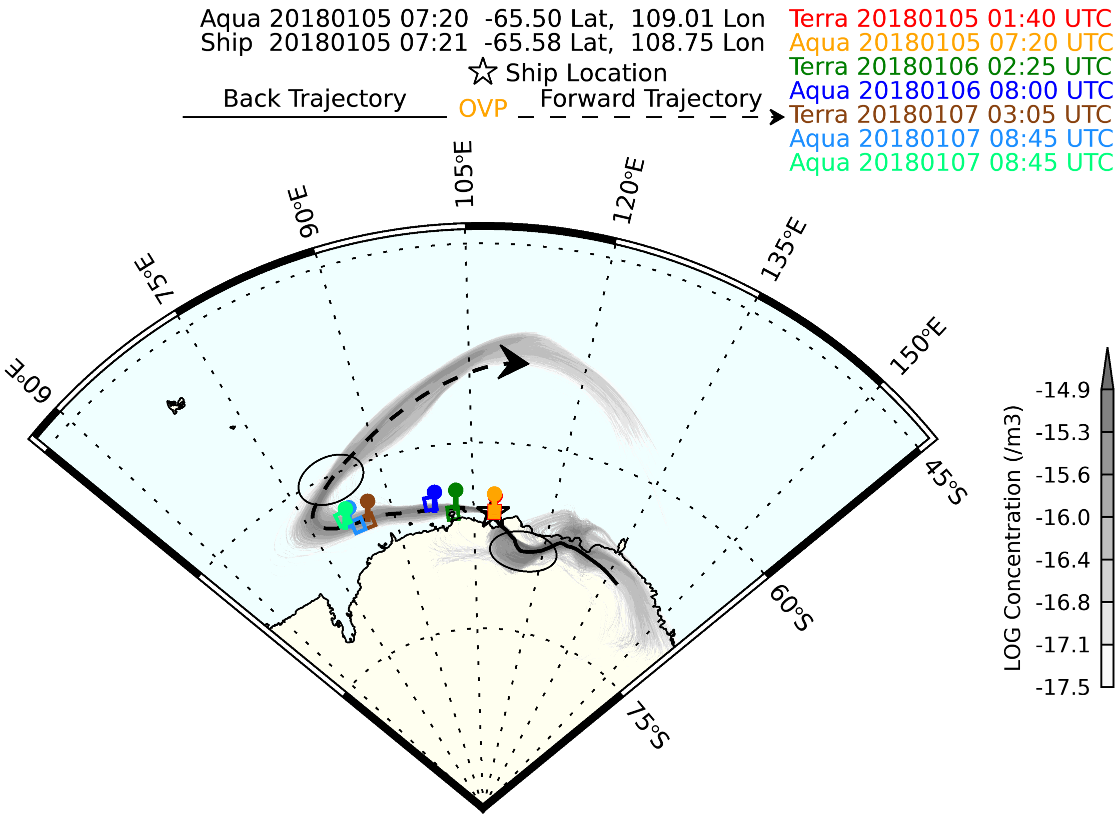

Figure 4. 5-day Hysplit backwards and forwards trajectories from the Marcus ship location (star). The solid and dashed curves show the center-of-mass trajectory from the tracer concentrations (gray). Drop pins denote MODIS overpasses over water with color code corresponding to inset information at the bottom of the figure. The ellipses denote the point in the trajectories where the concentration of tracers denote significant uncertainty in the center of mass trajectory (see text for explanation).

A time series of remote sensing and derived cloud properties in Fig. 3 show an optically thick cloud layer that fully attenuates the lidar. We note that a transition in observed and derived cloud properties begin near 16 UTC on 1/5. This transition period occurs as the ship transits northeastward and encounters an airmass with very different history as discussed below. From

00 to 16 UTC, the layer base is near 600 m, with tops near 1 km. The liquid water path (LWP) derived from the microwave radiometer suggests an approximate 2-hour periodicity in enhanced LWP that correlates with periods of episodic drizzle occurring during the high LWP part of the day. The more extended period oscillations in LWP are also matched in the lidar-derived extinction. On a much more rapid pace, we note oscillations in cloud droplet number Nd, r_e, and lidar-derived extinction. These oscillations are approximately in phase with the drizzle events that had a timescale of 10-15 minutes. Overall, extinction's high (low) values correlate with local maxima (minima) in N_d. The r_e is anticorrelated with N_d, but these quantities are not independent in the retrieval algorithm. We find that the MODIS retrievals are in reasonable agreement with the surface retrievals regarding the mean values and the variability. The depolarization ratio of the lidar is poorly known below values of 0.4 due to a substandard window that was repaired on 13 January, so we infer liquid drizzle based on the Doppler velocity and the low radar reflectivity.

CCN remains in the 250 cm^{-3} range throughout case 1 - well beyond the 75th percentile of CCN at 0.2% supersaturation for this latitude belt compiled from Capricorn II and MARCUS data reported in Humphries et al. (2021). N_d ranges up to the values of the CCN in between

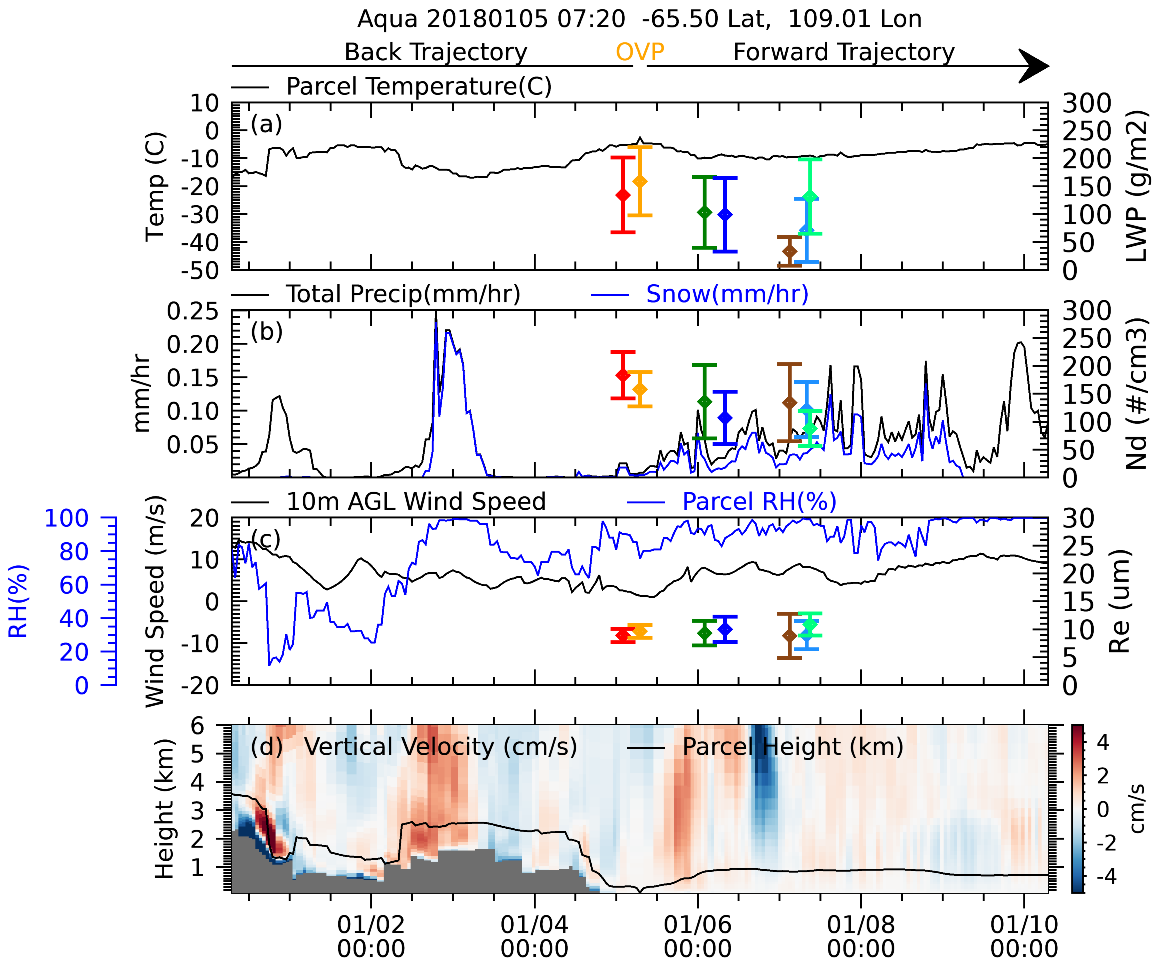

Figure 5. Meteorological quantities and MODIS cloud properties from along the backwards and forwards trajectories depicted in Fig. 4. OVP is the overpass time of the ship by MODIS Aqua on 07:20 UTC. A) Parcel temperature and MODIS LWP, b) Total and snow precipitation rates and MODIS Nd, c) Parcel relative humidity and 10 m wind speed and MODIS r_e, d) large-scale vertical motion and parcel height.

drizzle events to values as low as 80 cm^{-3} in regions near more intense drizzle where the extinction is also a minimum.

Back trajectories (Figs. 4 and 5) show that the air mass had recently descended to near the sea surface, as depicted in downward vertical motions along the parcel trajectory late on 1/4.

Before this, the trajectories suggest that the air mass transited the Law Ice Dome during the previous 48 hours near 2 km altitude and temperatures near -15°C. The MERRA-2 model suggests clear skies during the time over the Law Ice Dome, and this seems reasonable given the quiescent large-scale meteorology. While the back trajectories are quite uncertain during the back trajectory, there is an indication that before being over the Law Ice Dome, the air mass had been at a higher elevation over the ice sheet and then descended to near sea level, passing over the Totten Ice Shelf. Thus, it appears that the air mass had spent little time over the SO in the days before the case study period examined here.

The first cluster of MODIS overpasses of the trajectory are those that passed over the ship on 1/5. After that, two additional sets of overpasses occurred on 1/6 and 1/7. The cloud properties derived along the forward trajectories clustered in time show reasonable consistency in the scene-averaged r_e, N_d, and LWP. During the forward trajectory as the air mass continues roughly westward over the open ocean, the MODIS retrievals show little change in r_e although both LWP and N_d decreased over the ensuing three days. N_d drops from near 200 cm^{-3} during the overpasses of the ship on 1/5 to values near 100 cm^{-3} in the cluster of overpasses on 1/7. The MERRA-2 model suggests that light precipitation in the form of snow in the BL clouds increased after 1/5, consistent with decrease in N_d and LWP.

3.2 January 6, 2018 - Marcus

On the day following the 1/5 case study, January 6, 2018 (1/6), the Aurora Australis had progressed ~240 km to the northeast. They continued northeastward through open water with icebergs and wildely scattered sea ice notable on the video and still photography. The winds had increased and ranged between 10-15 m s^{-1} from the northeast during the early UTC hours of 1/6. The sea state remained unremarkable with no apparent whitecaps that would indicate active sea salt aerosol production locally. Surface temperatures were near and just below freezing, having warmed slightly from the previous day. The sounding at 06 UTC (not shown) shows that the boundary layer had deepened considerably yet remained well mixed with the base of the marine inversion near 1.6 km. The clouds observed by the ARM remote sensors (Fig. 3) show a marked contrast from the previous day with a transition in cloud properties that began at ~16 UTC and was complete by 00 UTC. A persistent layer of BL clouds continues with bases now near 1.4 km

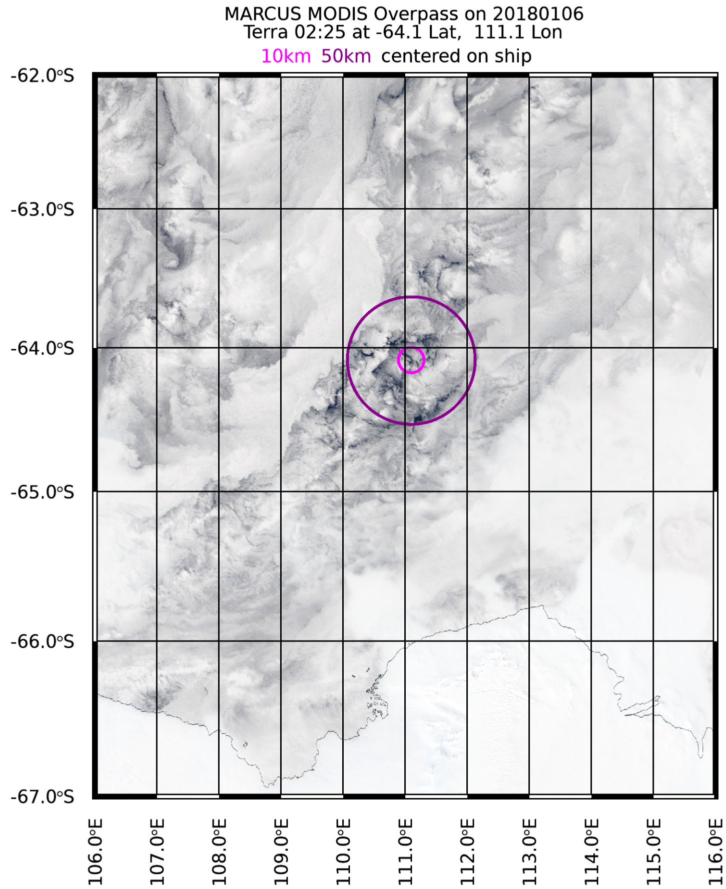

Figure 6. As in Figure 2b but for the Terra MODIS overpass at 02:25 UTC on 1/6.

and with layer top at the bottom of the marine inversion at temperatures near -15ºC. Higher radar reflectivities are noted in precipitation that, according to lidar depolarization above 0.4 during events before 03 UTC, seems to be snow. After 03 UTC, we cannot establish with certainty the phase of the lighter precipitation because of ambiguity in the lidar depolarization noted earlier. The cloud field is variable before 03 UTC, with breaks in the lidar data and lower cloud bases in heavier precipitation. Widely scattered clouds are observed below the bottom of the primary cloud layer.

The variability in the cloud field is consistent with satellite imagery and photography recorded at the ship (Figs. 6 and 7). The MODIS overpass at 0225 UTC places the ship in a region of mostly overcast cloud cover with the sea surface visible in various breaks in the cloud field in the vicinity of the ship. A photograph (Fig. 7) taken at 0226 UTC at the approximate time of the MODIS overpass looks towards the northeast and shows thick boundary layer clouds with breaks in the layer.

Figure 7. Photograph taken from the Aurora Australis at 02:26 UTC on 1/6 near the time of the MODIS overpass. Perspective is looking northeast.

The derived cloud microphysical properties had also changed considerably from 1/5. N_d is overall much lower, ranging between 10 and 50 cm^{-3} with substantial variability in regions of precipitation (Fig. 3). These values of N_d are in broad agreement with MODIS, which records a mean of ~15 cm^{-3}. The effective radius is also larger than on 1/5 and is in the 20-30 um range from the surface, while MODIS retrieves a value near 20. um. LWP is highly variable in and around precipitation, reaching several hundred g m^{-2}. After 03 UTC, the cloud properties become much more steady as the ship moves into the less variable cloud field to the northeast in Fig. 6. However, light precipitation from the cloud continues. The derived cloud properties also continue to show markedly low N_d and high r_e, with the LWP now in the 50 g m^{-2} range, suggesting that the cloud properties feature low N_d and high r_e throughout this area and not just in regions of active precipitation.

The 5-day back trajectories (Fig. 8) suggest that the air mass had exited continental Antarctica near the beginning of the 5-day period on January 1 and likely came from the region of the Amery Ice Shelf in what is known as Prydz Bay. The trajectories are substantially

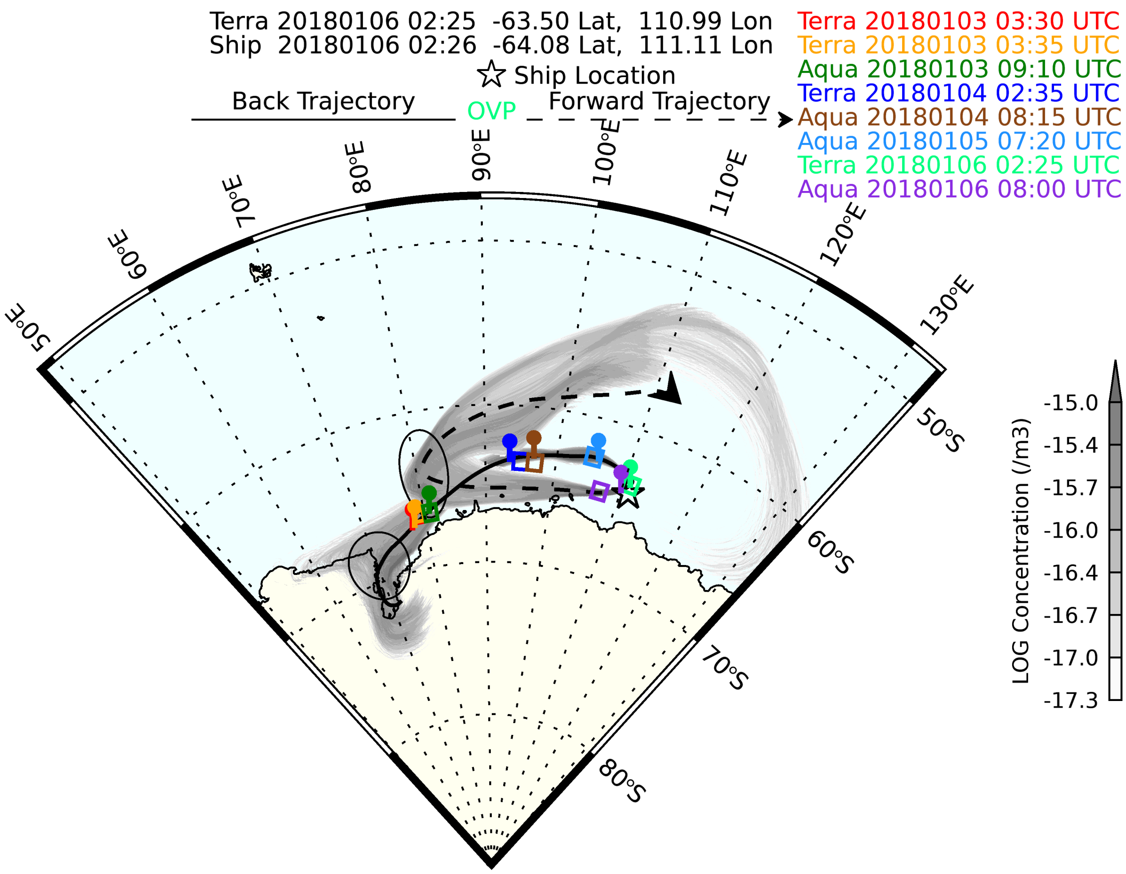

Figure 8. As in Figure 4 except for 1/6.

uncertain. However, the majority of the individual trajectories do show this path. Other parcels follow the coastline near the Amery ice shelf. We can confidently say that the air had been over open water for most of the past five days. There is a well-defined trend in MODIS cloud properties along the trajectory. We pick up the first set of MODIS overpasses of open water early on January 3 and have 4 clusters of overpasses on each succeeding day until 1/6. On 1/3 the Nd was in the 200 cm^{-3} range (Fig. 9) with r_e near 8 um - similar to the values found on 1/5 in the air that had recently descended from the Law Ice Dome. The N_d decreased, and the r_e increased in each successive set of overpasses until 1/6. The LWP, on the other hand, shows a different trend. MODIS records a Maximum LWP near 200 g m^{-2} early on 1/4 while a minimum in LWP occurs 24 hours later. While it borders on speculation, the precipitation (mostly snow) predicted by MERRA-2 seems to lag the LWP by about ½ day.

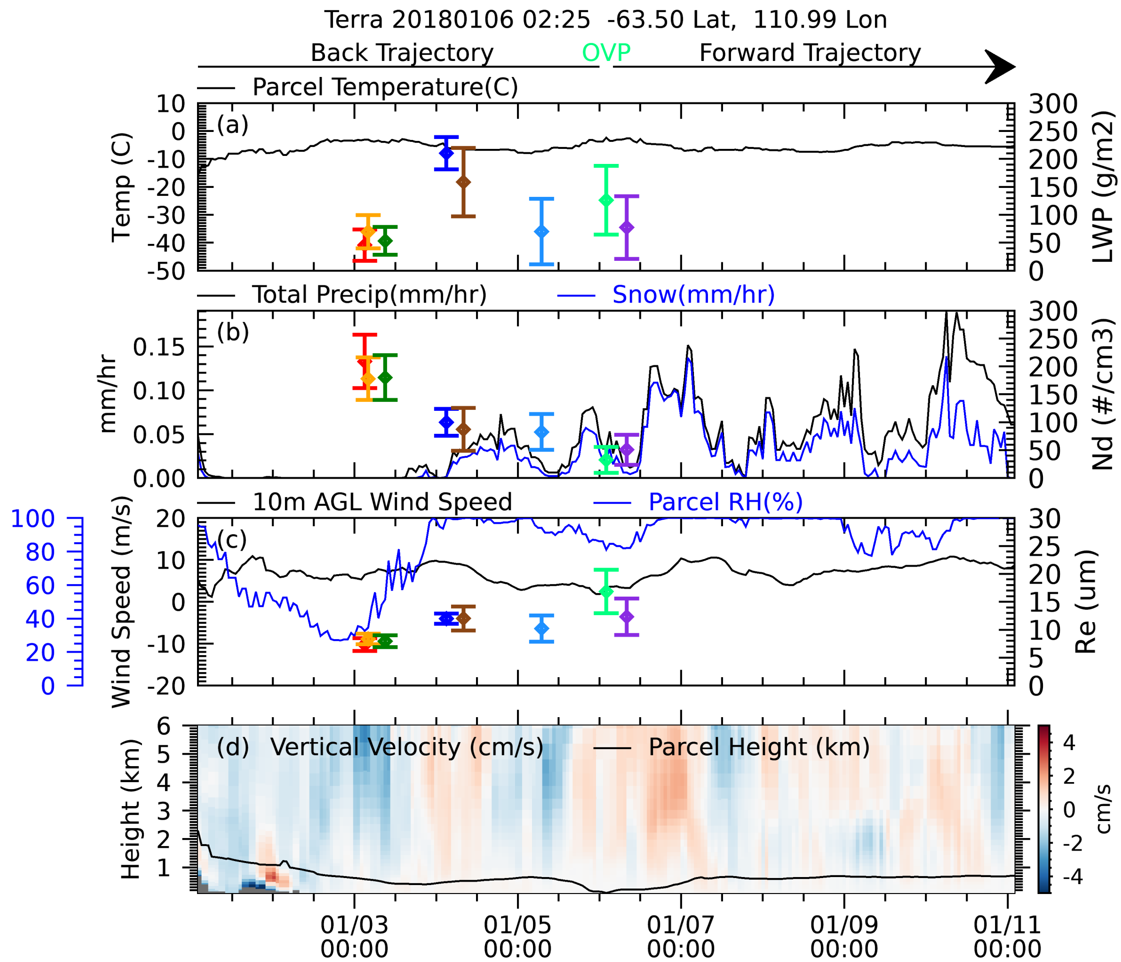

Figure 9. As in Figure 5 except for the 1/6 case study.

It is interesting to note in Fig. 3 that the ship time series of CCN (0.25% supersaturation) during the 1/6 case study period. Recall that 24 hours prior, CCN was in the 250 cm^{-3} range and close to the 90th percentile of CCN for this latitude band. While CCN is missing late on 1/5, the cloud properties transition from the high N_d and small r_e on 1/5 to what is observed on 1/6. It seems clear that the air mass sampled by the Aurora Australis shifted from one that had recently emerged from continental Antarctica to an air mass that had experienced land multiple days prior that now had much lower CCN and N_d.

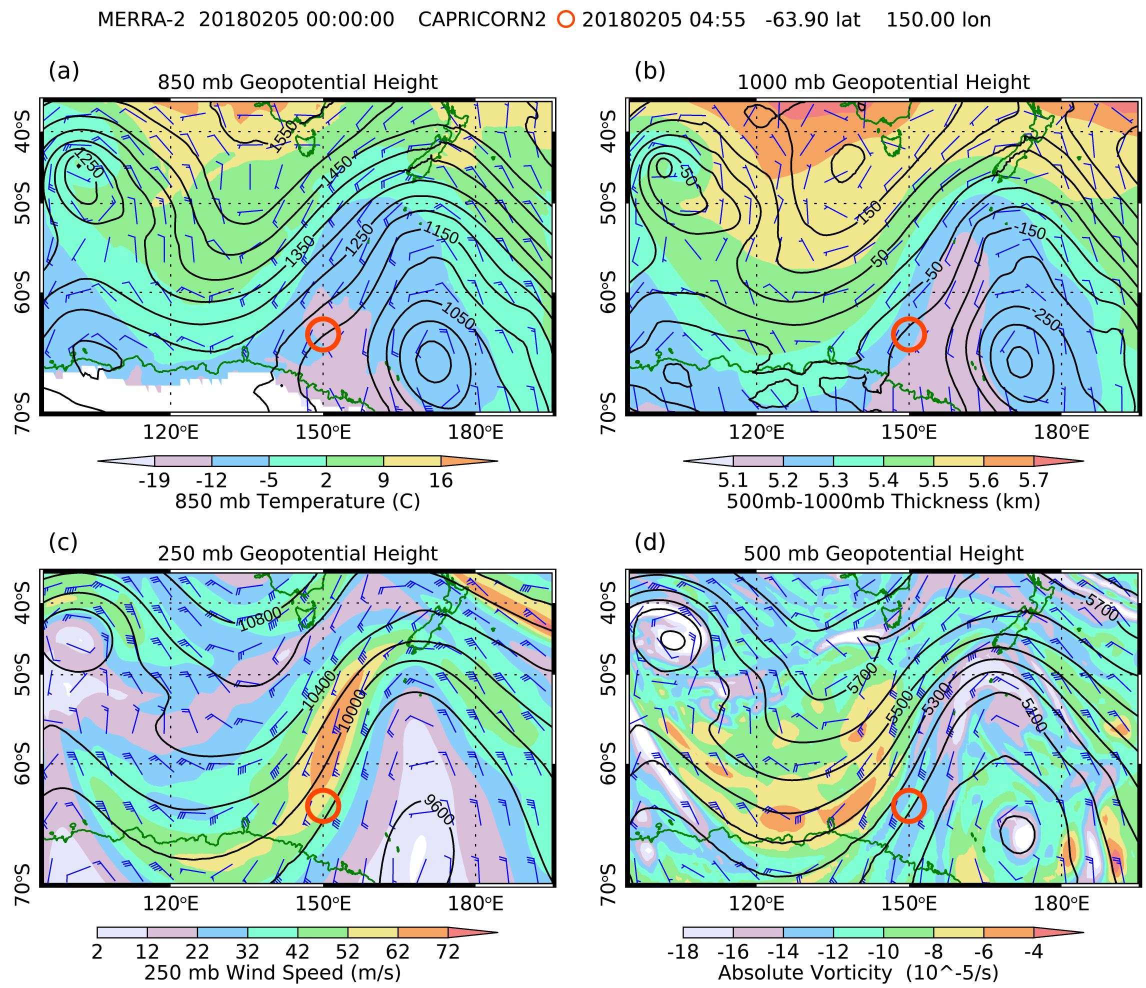

Figure 10. As in Fig. 1 except for the 2/4-2/5 case study. The red circle marks the location of the R/V Investigator on 2/5 at 00 UTC.

3.3 February 4 and 5, 2018 – Capricorn II

We take our final case study from data collected during the Capricorn II campaign on February 4 and 5, 2018 (2/4, 2/5) when the R/V Investigator was conducting oceanographic measurements within 100 km of 65°S, 150°E. The ship began a northward transit at 10 knots during midday on 2/5. The large-scale meteorology (Fig. 10) at 00 UTC on 2/5 shows a highly amplified upper-level pattern with a trough axis along 170°E and a ridge along about 130°E. A frontal system associated with the low pressure centered at 65°S, 170°E at 00 UTC 2/5 had passed over the ship on 2/3 at around 15 UTC, turning the flow out of the south. By 00 UTC on 2/5, this southerly flow extended through the entire troposphere.

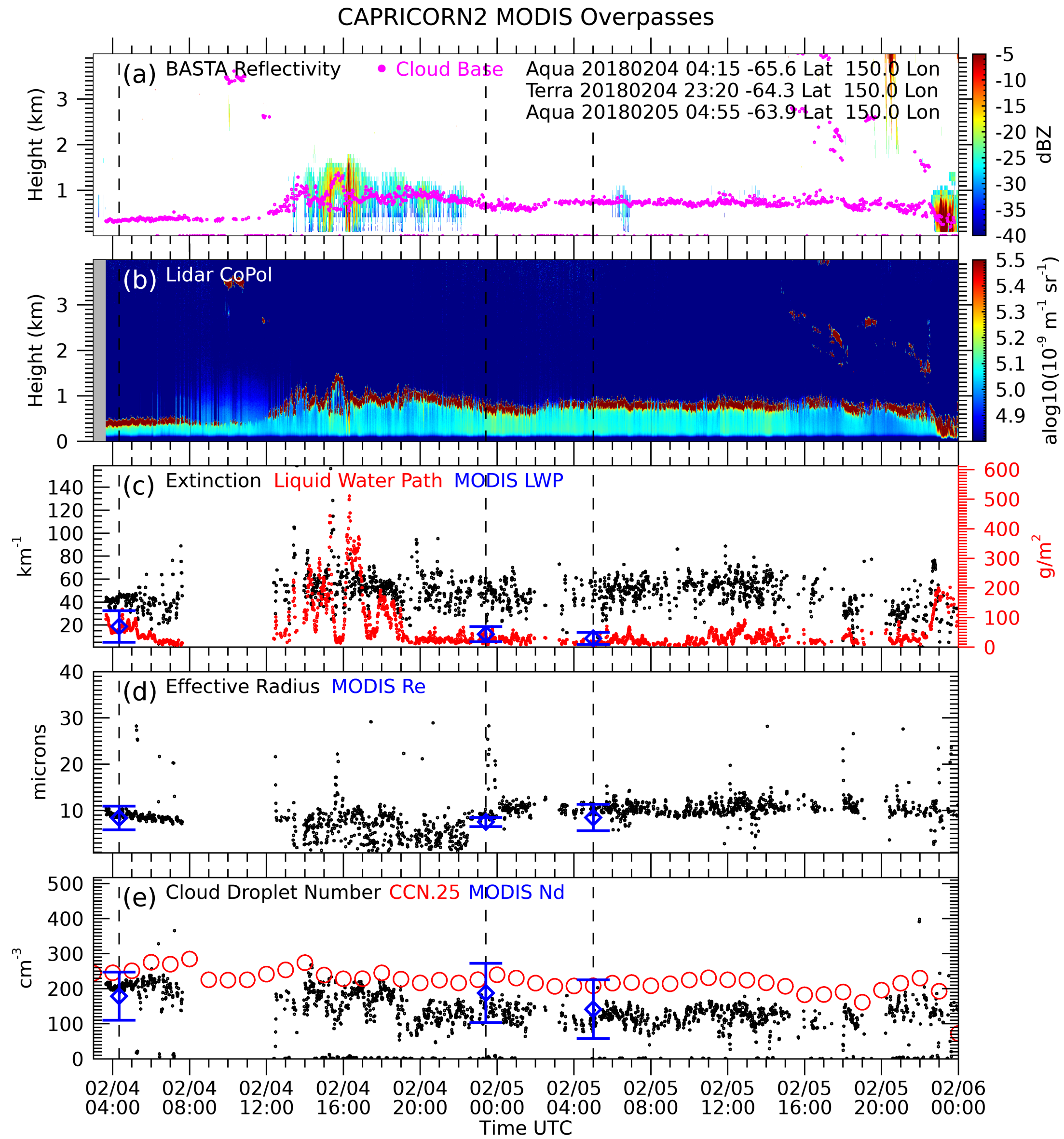

Figure 11. As in Figure 3 except for the 2/4-2/5 Capricorn II Case Study

Weather at the ship evolved somewhat during these two days. Surface temperatures remained between -2° and -4°C, and surface winds were steady out of the south at 10-15 m s^{-1}. Photography collected from the ship shows only modest wind waves, given the ship's proximity to the sea ice edge with very scattered white caps, suggesting the possibility of some local sea salt aerosol generation. A marine inversion was not evident in the 3-hourly soundings until 16 UTC on 2/4 when it was based near 1.5 km with a temperature of -15°C. The inversion strengthened as the pattern progressed, and surface pressure rose through 2/5. By mid-day on 2/5, a strong marine inversion with a 10°C potential temperature difference between the surface and 850 hPa was based near 1 km at a temperature of -10°C.

The BL clouds observed during this case study period (Fig. 11) were based near 500 m and, except for a period between 15 UTC and 23 UTC on 2/4, the clouds were below the detection threshold of the cloud radar on the R/V Investigator (~ -30 dBZe). The lidar shows a mostly overcast cloud layer with some regions of broken clouds and optically thinner clouds where the lidar transmits through the layer. Lidar depolarization ratios (not shown) in the sub-cloud precipitation between 15 and 23 UTC appear to be primarily liquid except for two brief episodes of snow that correlate with maxima in the radar reflectivity of more than -10 dBZe between 15 and 16 UTC. MODIS imagery collected at 04 UTC on 2/4 and 05 UTC on 2/5 (Fig. 12) show the ship's position in relation to the cloud field. Cloud streets that formed as the southerly flow encountered the sea surface and then evolved into closed cellular stratocumulus are evident from the sea ice region to the south of the ship location. The cloud field remained consistent during this southerly flow period.

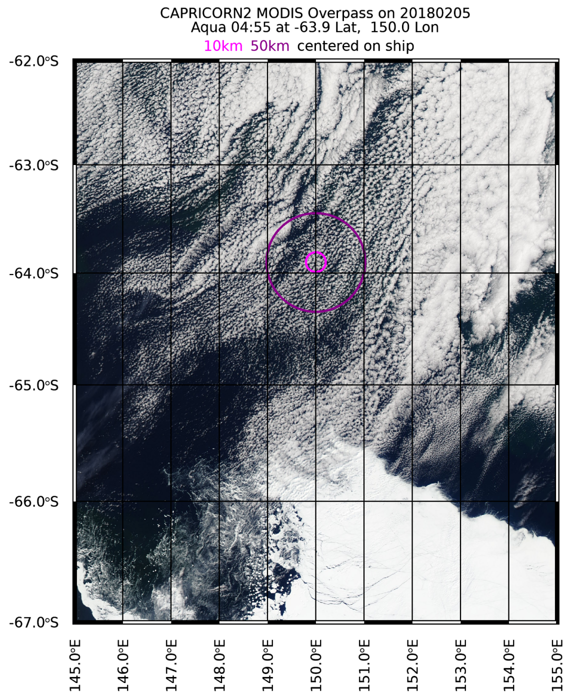

Figure 12. As in Fig. 2b except for a MODIS Aqua overpass of the 2/4-2/5 case study.

Derived cloud properties (Fig. 11) are also remarkably steady during the case study period. N_d decreases from near 200 cm^{-3} in the early part of the period to near 100 cm^{-3} after the light precipitation event at 19 UTC. CCN at 0.25% supersaturation remains steady throughout the case study between 200-300 cm^{-3}. This value of CCN, as the 1/5 case study presented earlier, is beyond the 90th percentile of CCN collected in this latitude belt. r_e follows N_d and remains at or below roughly 10 μm during most of the period. Except in the period with precipitation, the LWP remains in the 50-100 g m^{-2} range. Overall, the MODIS retrievals conducted from the overpasses agree reasonably with the surface-based cloud properties with high N_d and small r_e and LWP less than 50 g m^{-2} at the times of the overpasses. The MODIS N_d is well beyond the upper quartile of N_d recorded by MODIS during a 5-summer period examined in Mace et al. (2023).

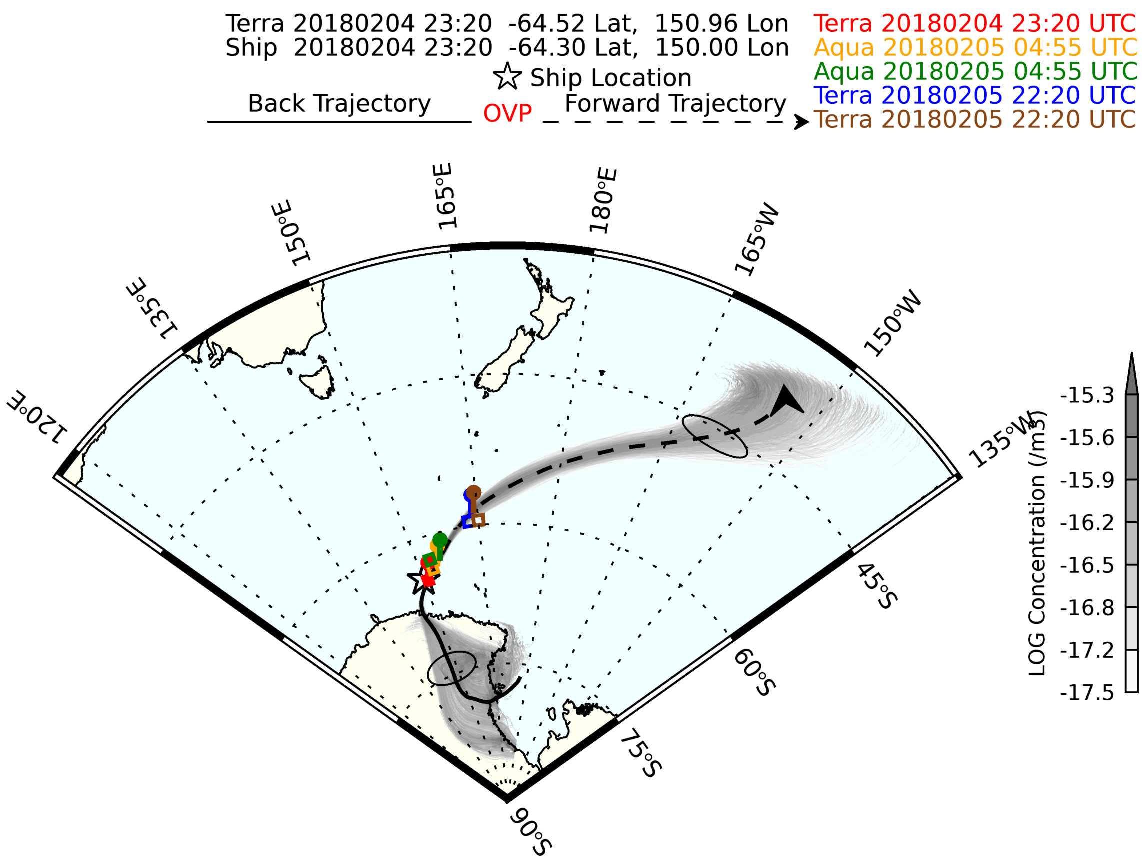

Figure 13. As in Fig. 5 except for 2/5 at 23 UTC.

As shown in Fig. 13, the air sampled during the case study period had spent the previous 48 hours transiting the high-altitude ice dome known as Dome C after it seemingly ascended from the Ross Sea. At least according to the back trajectories, the parcel was well into the free troposphere near 5 km altitude during this transit of Dome C, where predicted temperatures were near -40C. The air sampled at the ship had only been over the sea surface for a very short time after descending in a strong katabatic flow depicted in Fig. 14. After passing over the ship, the forward trajectories show that the air mass continued northeast. In addition to the overpasses near the ship, we find two additional sets of overpasses along the forward trajectory on 2/6 and 2/7. These show that the clouds evolved with increasing LWP and r_e and decreasing N_d to values of 70 cm^{-3} on 2/7.

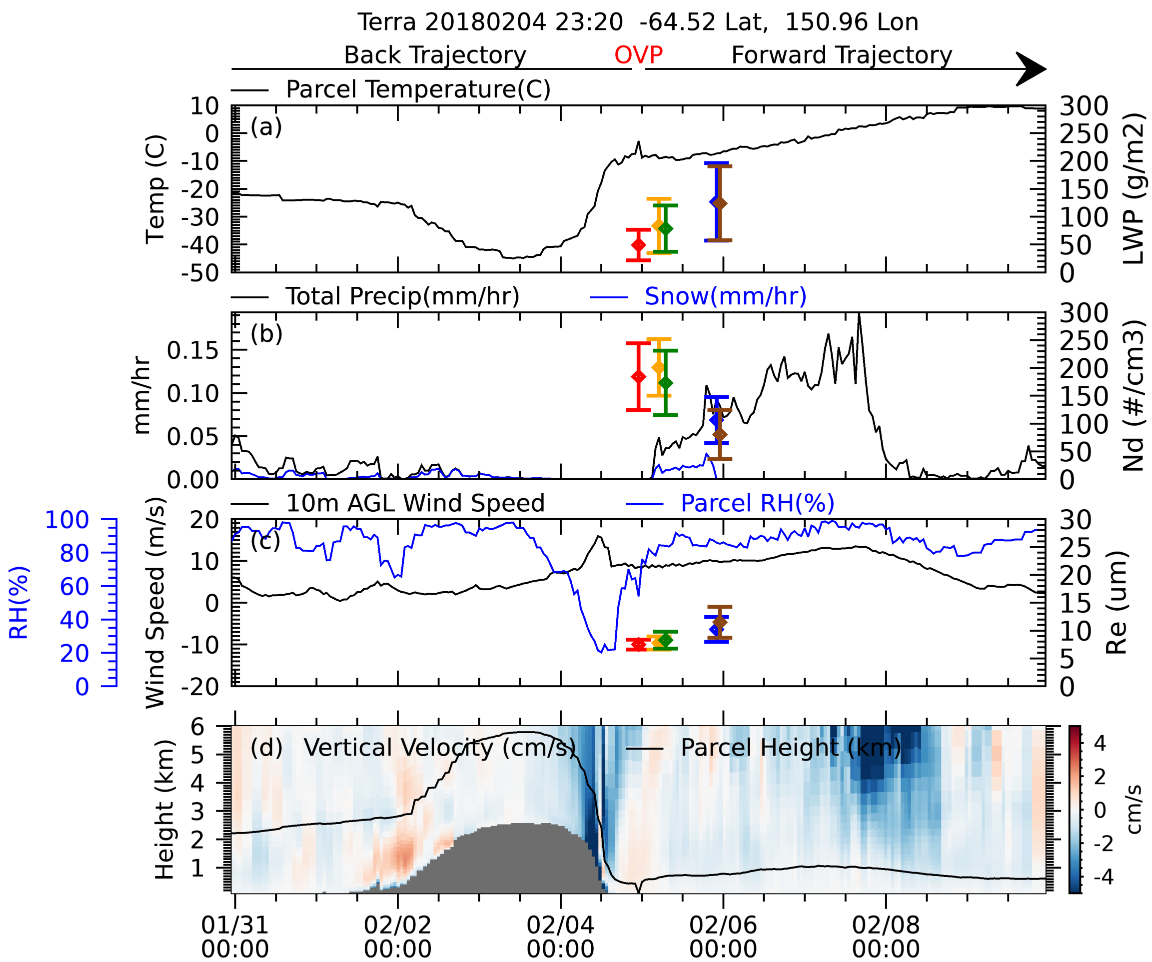

Figure 14. As in Fig. 5 except for 2/4 at 23 UTC.

414 The Capricorn II data included a Time-of-Flight Aerosol Chemical Speciation Monitor
415 (TOF-ACSM) that allows for inference of aerosol composition. Humphries et al. (2021) present
416 an analysis of the aerosol composition recorded as a function of latitude for recent Southern
417 Ocean voyages. We show the compositional breakdown of the aerosol for the 2/4-2/5 case
418 study in Fig. 15. We find that this aerosol mainly was (90%) sulfate and methanesulfonic acid
419 (MSA). Very low values of other chemical species are indicated, including chloride – a proxy
420 for sea salt. The sea salt and organic fraction of the aerosol composition in the 2/4-2/5 case
421 study was less than half of that compared to the latitudinal average.

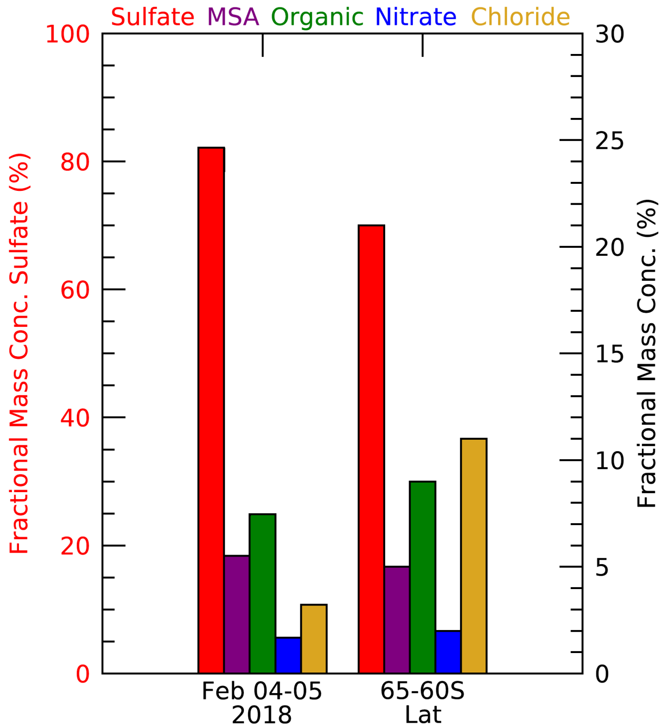

Figure 15. Aerosol compositional fractions from TOF-ACSM for the 2/4-5 case study compared to the latitudinal band average.

4. MODIS statistics

The three case studies we present seem to convey a picture consistent with the statistical findings of Mace et al. (2021 and 2023) and the earlier results of Humphries et al. (2021 and 2016) and going back to Shaw (1988) that air masses that have spent significant time over the Antarctic continental ice sheet emerge over the Southern Ocean with CCN concentrations in the 200-300 cm^{-3} range – significantly higher than the latitudinal average (Humphries et al., 2021). To see how general this association is we examine MODIS data collected from Terra and Aqua during Austral Summers (December through February) between 2014 and 2019 south of 60°S (the same data set used in Mace et al. 2023), except that here we examine the sensitivity of N_d in

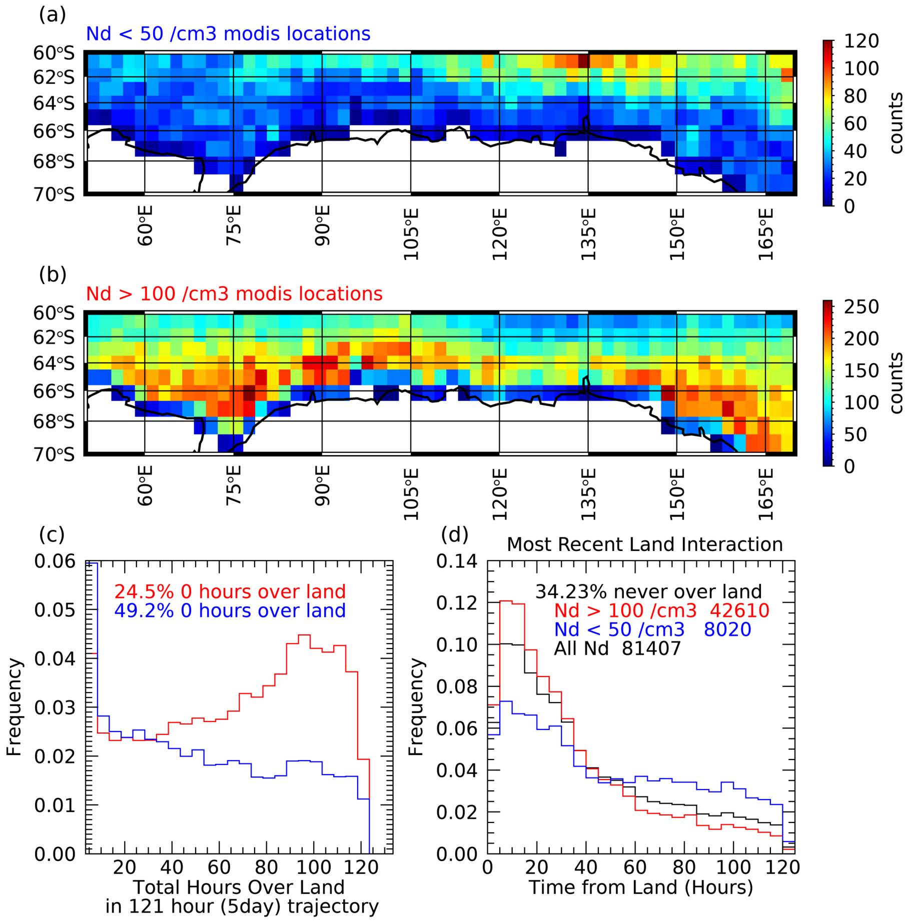

Figure 16. Occurrence of lower (a) and upper (b) Nd quartiles from MODIS data collected during the summers of 2014-2019. c) shows the frequency of occurrence of low (blue) and red (upper) Nd quartiles as a function of total hours spent over continental Antarctica in the 5-day back trajectory. d) shows the frequency of occurrence of the upper (red) and lower (blue) quartiles for over water scenes as a function of time since the back trajectory had encountered the Antarctic land mass.

a MODIS scene to the time since a back trajectory from that scene had encountered the Antarctic Continent. This analysis includes a total of 123784 trajectories. To demonstrate these relationships more clearly, we focus on the upper (100 cm^{-3}) and lower (50 cm^{-3}) N_d quartiles for all MODIS data in the Southern Ocean, as established in Mace et al. (2023).

The occurrence of the upper and lower N_d quartiles is strongly related to latitude (Fig. 16), with the high N_d quartiles tending to be adjacent to coastal Antarctica. Not surprisingly then, we also find that being in one or the other of the N_d quartiles is a strong predictor of the time since a back trajectory had encountered land (Fig. 16d). On the other hand, and perhaps less obviously, the amount of time spent over continental Antarctica seems to be predictive of membership in either the high or low N_d quartile. Nearly half of the low N_d quartile had not encountered land in the 5-day back trajectory, while only 25% of the high N_d quartile had been entirely over open water. Of the back trajectories that had passed over the continent, membership in either quartile depends oppositely on the amount of time spent over land. High N_d seems much more likely as time over the continent increases – rising sharply after 72 hours. On the other hand, low N_d is decreasingly likely as time over the continent increases.

5. Conclusions

While changes to model simulations of cloud feedbacks in the Southern Ocean are primarily responsible for the increased climate sensitivity of CMIP 6 (Zelinka et al., 2020), past work has documented that the cloud properties simulated in models are not consistent with observations of cloud properties Southern Ocean low-level clouds. Because cloud properties are strongly linked to precipitation this lack of consistency between models and observations calls into question the validity of the increased climate sensitivity. Fundamental to this is that the models seem to lack a mechanism to create the observed CCN concentrations (McCoy et al., 2021) over the summertime Southern Ocean. While the primary mechanism behind the high concentrations of CCN is likely related to aerosol nucleation and subsequent growth associated with sulfate precursor gasses emitted by the highly productive ocean biology in the high latitudes, the exact mechanism for the CCN production has not been established and are not simulated in global models of CCN production (Dunne et al., 2016; Lee et al., 2013). In this paper, we build on past work to show that air masses emerging into the MBL over the Southern Ocean after having spent time transiting the continental ice sheets of Antarctica during summer appear predisposed to have anomalously high CCN concentrations and produce clouds that have similarly anomalously high N_d.

Statistics derived from MODIS data demonstrate the relationship between time over Antarctica and high N_d clouds. These statistics are consistent with three case studies collected during ship-based observing campaigns in the Summer of 2018. A case on January 5, 2018, from the Marcus campaign and the 2/4-2/5 case from Capricorn II were of air masses that had recently emerged over the Southern Ocean after spending multiple days over continental Antarctica and had N_d from both MODIS and surface-based retrievals well within the upper quartile of what we find from multi-year MODIS analysis (> 100 cm^{-3}). CCN observed at the surface in the case studies was also anomalously high relative to the mean in the southern high latitudes. In both cases, the N_d along the trajectories decrease with time as the air masses transit over the open ocean and are diagnosed to no longer have N_d that would place it in the upper N_d quartile. On the other hand, the air mass sampled in the 1/6 case from Marcus had not been in contact with land for at least five days, and Nd from both surface and MODIS put it well within the lower N_d quartile. However, MODIS data from along the back trajectories suggest that N_d had decreased from values that placed it within the upper N_d quartile near the start of the 5-day back trajectory after the airmass likely emerged onto the Southern Ocean from the continental landmass.

The relationship between summertime transits of air masses over the continent and the production of high CCN air masses has been described in the literature (Shaw, 1988; Humphries et al., 2016). The highly productive waters of the Antarctic continental shelf produce copious precursor gasses beginning as DMS from phytoplankton along with oxidants such as BrO and IO. These maritime air masses would also contain preexisting primary and secondary aerosols such as sea salt and organics. Under synoptic scale conditions conducive to forced ascent, these air masses ascend the ice domes, where cloud and precipitation processes scavenge most of the ambient aerosol from these air masses. The air masses with high concentrations of aerosol precursor gasses now devoid of ambient aerosol then spend multiple days in the free troposphere at cold temperatures and extremely high actinic flux, enabling new particle formation and growth (Raes, 1995). Growth from the nucleation mode to CCN sizes would take several days, so long trajectories over the ice are favored. The air masses, now with high CCN, descend off the ice sheets in katabatic flows to the Southern Ocean marine boundary layer, forming the high N_d clouds found in observations.

While aspects of this storyline are speculative (the new particle formation process has not been observed in the free troposphere over the ice sheets, for instance), the critical elements of the story are reasonably well documented in past literature or cases and statistics presented in this paper. Additional observations are needed to establish this mechanism's validity more rigorously. It is fortuitous that several upcoming campaigns into the Southern Ocean are planned during this decade (Mallet et al., 2023). What is not planned but needed are airborne campaigns into the high latitudes to document the chemical pathways favored in taking aerosol precursor gasses to new particles that then grow into CCN and eventually seed the high albedo clouds observed by satellite and ground-based sensors.

Acknowledgements

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Open Research

All data used in this study are available in public archives. MARCUS data are available from the DOE ARM archive at https://adc.arm.gov/armlogin/login.jsp, SOCRATES data are available at https://data.eol.ucar.edu/project/SOCRATES, and CAPRICORN I and II data are available at https://doi.org/10.25919/5f688fcc97166. MODIS cloud products can be found for Terra and Aqua at https://doi.org/10.5067/TERRA/MODIS/L3M/CHL/2018 and https://doi.org/10.5067/MODIS/MYD06_L2.006. GDAS data are obtained from ftp://ftp.arl.noaa.gov/pub/archives/gdas (National Centers for Environmental Prediction, 2022). Computer code for this study including analysis code and graphic generation code is available at https://doi.org/10.7278/S50d-bpx8-gmtt.

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