npj | climate and atmospheric science

Article

Published in partnership with CECCR at King Abdulaziz University

https://doi.org/10.1038/s41612-024-00625-1

# Wet deposition in shallow convection over the Southern Ocean

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Southern Ocean (SO) air is amongst the most pristine on Earth, particularly during winter. Historically, there has been a focus on biogenic sources as an explanation for the seasonal cycle in cloud condensation nuclei concentrations ($N_{CCN}$). $N_{CCN}$ is also sensitive to the strength of sink terms, although the magnitude of this term varies considerably. Wet deposition, a process encompassing coalescence scavenging (drizzle formation), is one such process that may be especially relevant over the SO. Using a boundary layer cloud climatology, $N_{CCN}$ and precipitation observations from Kennaook/Cape Grim Observatory (CGO), we find a statistically significant difference in $N_{CCN}$ between when the upwind meteorology is dominated by open mesoscale cellular convection (MCC) and closed MCC. When open MCC is dominant, a lower median $N_{CCN}$ (69 cm$^{-3}$) is found compared to when closed MCC (89 cm$^{-3}$) is dominant. Open MCC is found to precipitate more heavily (1.72 mm day$^{-1}$) and more frequently (16.7% of the time) than closed MCC (0.29 mm day$^{-1}$, 4.5%). These relationships are observed to hold across the seasonal cycle with maximum $N_{CCN}$ and minimum precipitation observed during Austral summer (DJF). Furthermore, the observed MCC morphology strongly depends on meteorological conditions. The relationship between $N_{CCN}$ and precipitation can be further examined across a diurnal cycle during the summer season. Although there was again a negative relationship between precipitation and $N_{CCN}$, the precipitation cycle was out of phase with the $N_{CCN}$ cycle, leading it by ~3 hours, suggesting other factors, specifically the meteorology play a primary role in influencing precipitation.

The atmosphere over the Southern Ocean (SO) is renowned for being the most pristine on Earth[1,2], since it is largely free of anthropogenic and terrestrial emissions. It is further renowned for its high fractional cloud cover[3] and high precipitation frequency[4] throughout the year, an immediate consequence of the strong latent heat flux that arises along the SO storm track. When these elements are combined, the SO can serve as a proxy for a pre-industrial environment, providing a natural testbed for aerosol-cloud-precipitation interactions (ACPI)[5–7]. Yet the limited understanding of SO clouds results in radiation budget biases in both reanalysis products and climate simulations[8–14].

These persistent biases have garnered extensive attention from the scientific community with numerous international field campaigns undertaken[15–17]. Many of these efforts have focused on better categorizing the sources and magnitude of cloud condensation nuclei (CCN) and ice nucleating particles (INP) given their effect on cloud microphysical properties[18].

For many years, dimethylsulfide (DMS), which is formed by planktonic algae in sea water and oxidizes in the atmosphere to make sulfate aerosol, has been thought to be the main source of CCN over the oceans[19–21]. First published more than three decades ago, the 'CLAW' hypothesis, an

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acronym from the first letters of the authors' surnames[19], defines a negative climate feedback loop between incident solar radiation, marine biogenic emissions, CCN and cloud properties, which has sparked sustained scientific interest. Many subsequent studies have confirmed specifics of the connections between oceanic phytoplankton and DMS emission to the atmosphere (e.g.[22]), and the relation of DMS-derived aerosol mass, CCN number concentrations ($N_{CCN}$) and various cloud properties like cloud droplet number and cloud optical properties (e.g.[23–27]).

Although several links in the suggested loop between cloud albedo, CCN, DMS, and phytoplankton have been confirmed, there has not yet been a comprehensive scientific synthesis of it on a worldwide basis[21]. Quinn and Bates[28] examined observations of the individual steps of this loop and concluded that the sources of CCN over the marine atmospheric boundary layer (MABL) are more complex than previously recognized and there should be other sinks and sources than DMS, especially during winter[26,29]. Later research focused less on DMS and more on other CCN sources. Leck and Bigg[30] illustrated that bubble bursting at the ocean surface is a significant source of CCN to the MABL, consistent with previous studies[31–33]. They demonstrated while a marine biological source of reduced sulfur dominates $N_{CCN}$ over the summer months, other components, such as wind-generated coarse-mode sea salt, are important CCN components year-round, especially in winter due to higher winds[22,26,34–37].

Numerous recent research efforts have further underscored the importance of considering regionally varying meteorological factors for understanding ACPI[38–40]. For example, Zhang and Feingold[38], revealed the global distribution of marine low-cloud albedo susceptibility, emphasizing the strong influence of large-scale meteorological conditions on cloud albedo in various regions. They considered crucial factors such as lower-tropospheric stability, free-tropospheric relative humidity, sea surface temperature, and boundary layer depth, highlighting the need to account for these variables when assessing the response of cloud albedo to aerosol perturbations at different scales.

The removal of aerosols from the MABL by their activation into cloud droplets and subsequent development into precipitation-sized particles through collision-coalescence has long been acknowledged as a prominent mechanism addressing probable CCN sinks[7,41–43]. Recent research by Tornow et al.[44] demonstrated that frozen hydrometeors in marine cold air outbreaks can affect cloud liquid water and early consumption of CCN, leading to a reduction in cloud droplet number concentration ($N_d$). According to aircraft observations over the SO, Hudson found that CCN have lower concentrations in the MABL under cloudy conditions than clear conditions[2,45]. Also, the average $N_d$ for wintertime flights with non-drizzling clouds is roughly three times higher than the overall average[45,46]. Bennartz[47] also used observations from the MODerate resolution Imaging Spectroradiometer (MODIS) to study the sensitivity of maritime clouds to precipitation and concluded that $N_d$ was approximately 2.5 times greater in non-drizzling clouds than in drizzling clouds. In their idealized model of $N_d$ for MABL clouds, Wood et al.[42] demonstrated that precipitation was the main sink of $N_d$. Using the same budget model, McCoy et al.[7] showed that coalescence scavenging might reduce the mean $N_d$ to around 30% of the value that would occur otherwise. However, using a modified version of the same budget model, Kang et al.[43] found a reduction in anticipated $N_d$ of up to 90% depending on the rate of precipitation. The impact of wet deposition, which is a direct aerosol removal process, is still poorly understood, especially over the SO, where great uncertainty exists in the amount and nature of precipitation[48–51].

MABL clouds, which are frequently shallow boundary-layer clouds, are the dominant cloud type over the SO[51–55]. Light precipitation from shallow clouds, on the other hand, has been found to be common over the SO as non-frontal precipitation[56–58]. According to satellite observations, MABL clouds frequently display some mesoscale morphological types, each of which is distinguished by certain patterns of cloud organization. Wood and Hartmann[59] categorized these clouds into open mesoscale cellular convection (MCC), closed MCC, no MCC, and cellular but disorganized clouds based on the mesoscale organization. The type of MCC introduces significant mesoscale variability in both the microphysical (e.g., $N_d$, effective radius, precipitation rate) and macrophysical (e.g., cloud albedo, cloud coverage) properties of clouds[59–63]. In-situ observations have found that drizzle/light precipitation is more frequent and intense in open MCCs, while closed cells have very few drizzle drops[43,46,64,65].

Building upon these field observations, we seek to quantify climatological differences in the relationships between cloud morphology, precipitation and their meteorological controls, and to extend this relationship to $N_{CCN}$ as observed at the Kennaook/Cape Grim Observatory (CGO), Tasmania. This will be done by examining the sensitivity of $N_{CCN}$ to precipitation from boundary layer clouds over the SO, considering the nature of the upwind shallow convection. The hypothesis is that "highly pristine conditions/low $N_{CCN}$ over the SO are associated with periods of relatively high precipitation arising mainly from open MCC".

## Results and discussion

### CCN and precipitation relation within cloud morphology

From the 17,470 h of baseline data at CGO, the median $N_{CCN}$ and mean precipitation are determined for samples with a specified upwind domain radius, upwind averaging time and the fraction of coverage for both open and closed MCC ($FC_{MCC}$) threshold. Tables 1, 2 show the results for the 50 and 80% $FC_{MCC}$ threshold for both open and closed MCC, respectively. For each upwind radius and averaging time, the median $N_{CCN}$ is shown along with the 5th and 95th percentiles; the number of samples is also indicated. The tables reveal that the results are robust across different upwind radii, averaging times, and $FC_{MCC}$ thresholds.

The number of samples varies systematically with upwind radius and time. For an upwind averaging time of 3 h, for 80% $FC_{MCC}$ threshold (Table 2), the maximum number of samples for open MCC (3403) occurs at an upwind radius of 200 km while the maximum for closed MCC (1050) occurs at a radius of 100 km. The number of samples depends upon the match between radius and time, accounting for the advection speed of cloud and the persistence of cloud type within the baseline sector. Qualitatively, the results are the same when the domain radius is set at 300 km or less. At larger radii the number of records drops steeply.

We assessed whether the examined properties of open and closed MCC (e.g., median $N_{CCN}$ or mean precipitation rate and frequency) are different for each upwind radius and time, with the null hypothesis that any differences are only due to random variations. The analysis with the Whitney *U* test revealed statistically significant differences, with median $N_{CCN}$ consistently smaller for open than closed MCC ($p < 0.05$). Similarly, the results of the two-tailed Student's *t* test confirmed that differences in precipitation rate and frequency between open and closed MCC are statistically significant ($p < 0.05$). As such for all results hereafter, we only consider the 100 km upwind average, 80% $FC_{MCC}$ threshold (Table 2) for both open and closed and 3 h upwind averaging time, which gives a median $N_{CCN}$ of 69 $cm^{-3}$ from 3285 samples for open MCC and 89 $cm^{-3}$ from 1050 samples for closed MCC. The precipitation rate for open MCC (1.72 mm $day^{-1}$) is 6 times greater than for closed MCC (0.29 mm $day^{-1}$) and the frequency of precipitation is also more frequent during open MCC, occurring 16.7% of the time compared to 4.5% for closed MCC. These results support the hypothesis that open MCC have higher precipitation than closed MCC, climatologically, which is consistent with previous field observations[46,66,67].

Figure 1a compares the probability distribution function (PDF) of $N_{CCN}$ for open and closed MCC, while Fig. 1b compares the PDF of the precipitation. The precipitation from closed MCC is predominantly drizzle (less than 7.2 mm $day^{-1}$ or 0.3 mm $h^{-1}$) rather than rain, with a frequency of 98.8% (orange solid line in Fig. 1b). Conversely, in the case of open MCC, drizzle accounts for precipitation 92.1% of the time (blue solid line in Fig. 1b). Figure 1a shows that very clean air with $N_{CCN}$ less than 50 $cm^{-3}$ is much more common under open MCC than closed MCC, suggesting that CCN could be washed out by wet deposition during the heavier rain under open MCC. Since both MCC types have similar modes within 50 to 100 $cm^{-3}$, the relative frequency of $N_{CCN}$ higher than 100 $cm^{-3}$ is lower for open than closed MCC. The bottom plots depict PDF of Mean Sea Level Pressure

**Table 1 | Median $N_{CCN}$ and mean precipitation rate and frequency**

| Radius (km)-dominated MCC type | $N_{CCN}$ (cm$^{-3}$) (5th, 95th) (No. of cases) 1 h | Precipitation intensity (mm day$^{-1}$), Frequency (%) 1 h | $N_{CCN}$ (cm$^{-3}$) (5th, 95th) (No. of cases) 3 h | Precipitation intensity (mm day$^{-1}$), Frequency (%) 3 h | $N_{CCN}$ (cm$^{-3}$) (5th, 95th) (No. of cases) 6 h | Precipitation intensity (mm day$^{-1}$), Frequency (%) 6 h |
|---|---|---|---|---|---|---|
| 1000-Open MCC | 85 (27, 211) (4273) | 2.88, 19.6% | 85 (27, 209) (4169) | 2.64, 18.9% | 85 (28, 207) (4057) | 2.42, 17.9% |
| 500-Open MCC | 78 (24, 199) (5121) | 2.72, 19.6% | 78 (25, 199) (5045) | 2.34, 18.3% | 77 (24, 199) (4970) | 2.01, 17.2% |
| 300-Open MCC | 74 (24, 193) (5136) | 2.25, 18.3% | 74 (24, 193) (5081) | 1.98, 17.2% | 74 (24, 193) (5013) | 1.82, 16.1% |
| 200-Open MCC | 72 (23, 189) (5074) | 1.98, 17.2% | 72 (23, 189) (4997) | 1.85, 16.7% | 72 (23, 189) (4922) | 1.78, 16.1% |
| 100-Open MCC | 71 (23, 183) (4710) | 1.84, 16.6% | 70 (23, 183) (4637) | 1.79, 16.6% | 70 (22, 183) (4546) | 1.77, 16.3% |
| 50-Open MCC | 70 (23, 180) (4370) | 1.77, 16.6% | 69 (22, 180) (4331) | 1.86, 16.9% | 69 (22, 180) (4211) | 1.88, 17.1% |
| 1000-Closed MCC | 88 (24, 207) (1010) | 1.34, 11.2% | 91 (25, 203) (952) | 1.04, 9.9% | 96 (26, 202) (877) | 0.74, 8.2% |
| 500-Closed MCC | 91 (26, 236) (1855) | 0.49, 6.7% | 93 (25, 230) (1787) | 0.49, 6.7% | 93 (25, 228) (1640) | 0.45, 6.2% |
| 300-Closed MCC | 91 (27, 242) (2047) | 0.50, 6.9% | 92 (29, 240) (1937) | 0.44, 6.2% | 93 (29, 235) (1822) | 0.49, 6.1% |
| 200-Closed MCC | 91 (29, 242) (2039) | 0.51, 7.1% | 91 (30, 234) (1900) | 0.44, 6.3% | 91 (30, 222) (1721) | 0.44, 5.9% |
| 100-Closed MCC | 91 (31, 231) (1894) | 0.45, 6.6% | 91 (32, 225) (1754) | 0.41, 5.9% | 91 (32, 219) (1597) | 0.42, 5.1% |
| 50-Closed MCC | 91 (32, 226) (1726) | 0.44, 6.6% | 90 (32, 224) (1629) | 0.44, 6.1% | 90 (32, 221) (1453) | 0.40, 4.7% |

Results are for different upwind radii of baseline (different rows) and different averaging times (different columns) based on the 50% $FC_{MCC}$ threshold for 2016–2021. The $N_{CCN}$ columns contain the 5th and 95th percentiles and the number of cases in two separate brackets, respectively.

**Table 2 | Median $N_{CCN}$ and mean precipitation rate and frequency**

| Radius (km)-dominated MCC type | $N_{CCN}$ (cm$^{-3}$) (5th, 95th) (No. of cases) 1 h | Precipitation intensity (mm day$^{-1}$), Frequency (%) 1 h | $N_{CCN}$ (cm$^{-3}$) (5th, 95th) (No. of cases) 3 h | Precipitation intensity (mm day$^{-1}$), Frequency (%) 3 h | $N_{CCN}$ (cm$^{-3}$) (5th, 95th) (No. of cases) 6 h | Precipitation intensity (mm day$^{-1}$), Frequency (%) 6 h |
|---|---|---|---|---|---|---|
| 1000-Open MCC | 87 (29, 207) (1335) | 2.65, 19.0% | 88 (30, 206) (1225) | 2.27, 17.6% | 88 (30, 206) (1074) | 1.89, 15.2% |
| 500-Open MCC | 77 (25, 186) (2883) | 2.23, 18.4% | 77 (25, 183) (2677) | 1.83, 17.0% | 77 (25, 186) (2365) | 1.63, 22.5% |
| 300- Open MCC | 74 (24, 184) (3441) | 1.95, 17.4% | 73 (24, 181) (3207) | 1.71, 16.2% | 72 (23, 179) (2867) | 1.61, 15.6% |
| 200-Open MCC | 72 (23, 179) (3734) | 1.82, 16.7% | 71 (23, 178) (3403) | 1.70, 16.3% | 70 (22, 178) (2991) | 1.60, 15.9% |
| 100-Open MCC | 70 (23,177) (3759) | 1.80, 16.9% | 69 (23, 174) (3285) | 1.72, 16.7% | 68 (22, 177) (2861) | 1.64, 16.1% |
| 50-Open MCC | 69 (23, 176) (3679) | 1.78, 16.8% | 68 (23, 173) (3100) | 1.74, 17.1% | 67 (22, 173) (2666) | 1.71, 16.4% |
| 1000-Closed MCC | 97 (30, 173) (93) | 1.03, 9.7% | 98 (36, 173) (79) | 1.09, 11.4% | 101 (41, 168) (58) | 0.50, 6.9% |
| 500-Closed MCC | 88 (35, 238) (599) | 0.39, 5.0% | 89 (35, 228) (538) | 0.32, 4.8% | 91 (37, 224) (424) | 0.18, 3.1% |
| 300-Closed MCC | 89 (35, 236) (953) | 0.37, 5.5% | 93 (36, 226) (824) | 0.25, 3.6% | 93 (37, 214) (672) | 0.21, 3.3% |
| 200-Closed MCC | 91 (34, 229) (1152) | 0.36, 5.3% | 91 (35, 224) (969) | 0.28, 4.1% | 90 (35, 211) (765) | 0.26, 3.7% |
| 100-Closed MCC | 88 (33, 229) (1296) | 0.37, 5.6% | 89 (34, 222) (1050) | 0.29, 4.5% | 86 (33, 211) (768) | 0.28, 4.2% |
| 50-Closed MCC | 90 (33,225) (1312) | 0.38, 6.0% | 88 (34, 214) (976) | 0.31, 4.7% | 86 (32, 205) (690) | 0.34, 5.1% |

Results are for different upwind radii of baseline (different rows) and different averaging times (different columns) based on the 80% $FC_{MCC}$ threshold for 2016–2021. The $N_{CCN}$ columns contain the 5th and 95th percentiles and the number of cases in two separate brackets, respectively.

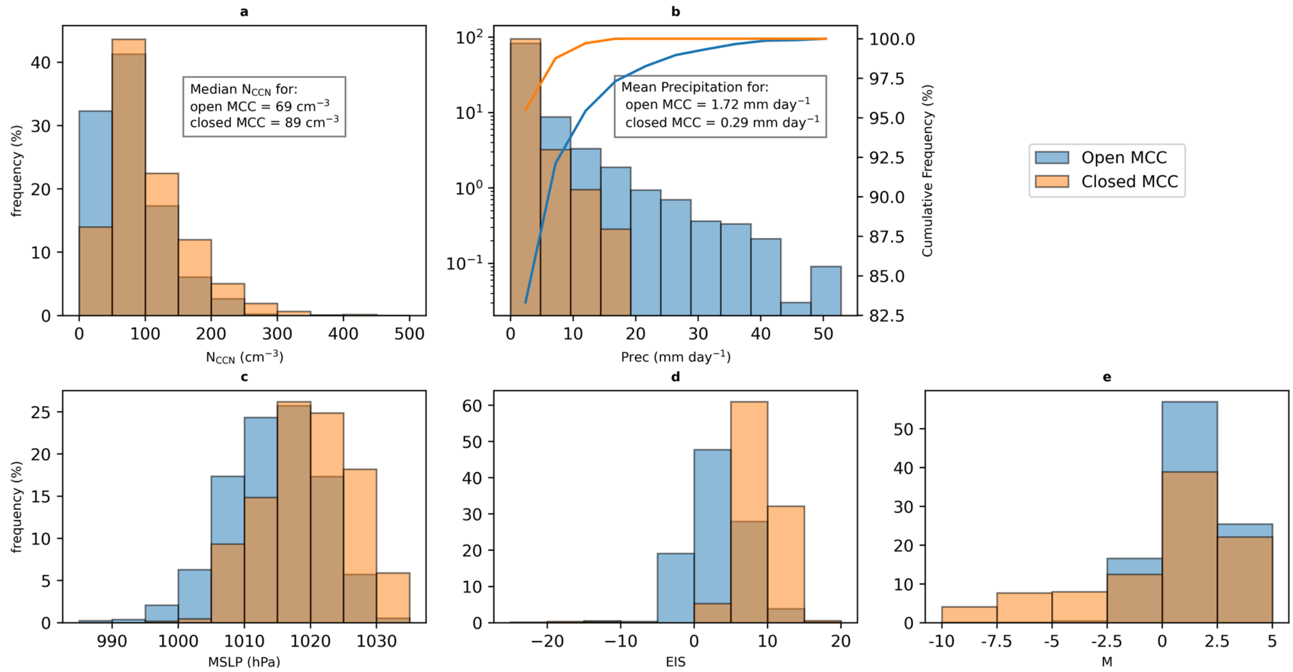


**Fig. 1 | General atmospheric condition under different MCCs.** Overlapping Probability Density Function (PDF) plot for $N_{CCN}$ (**a**), PDF plot (left axis) and the accumulated frequency (solid lines-right axis) for the precipitation (**b**) and the PDF plot for MSLP (**c**), EIS (**d**) and M (**e**) in open (blue) and closed (orange) MCC conditions for the 100 km upwind radius and 3 h upwind averaging time and 80% $FC_{MCC}$ threshold.

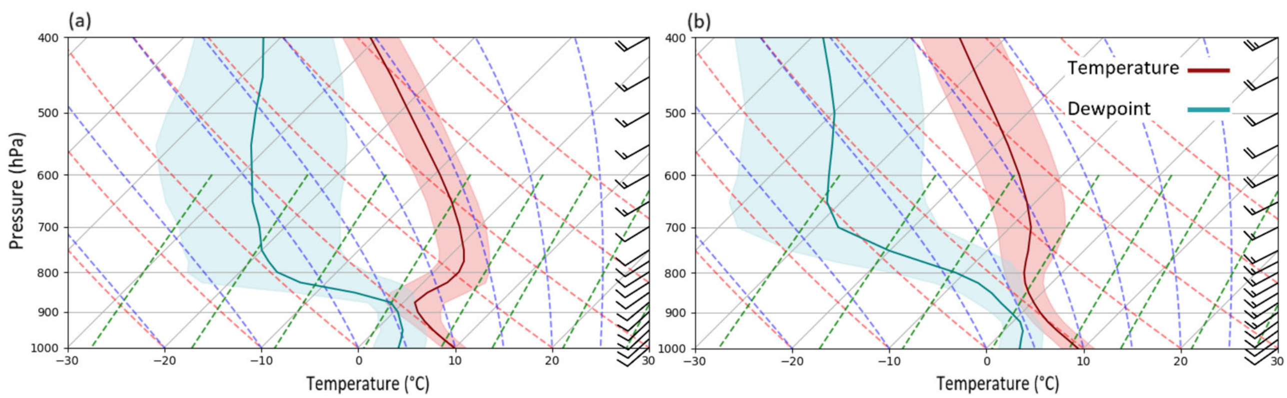


**Fig. 2 | Composite soundings.** Composite sounding profile for (**a**) closed MCC and (**b**) open MCC for the 100 km upwind radius and 3 h upwind averaging time and 80% $FC_{MCC}$ threshold. Mean profiles of temperature (Red lines), dew point temperature (blue lines), and vector winds, shaded region indicates standard deviation.

(MSLP), the Estimated Inversion Strength (EIS) and M of both open and closed MCC from left to right respectively. These parameters offer insights into the meteorological controls operating under each condition. Analysis of these plots reveals a higher incidence of high-pressure systems during closed MCC, whereas open MCC tends to occur in regions with lower MSLP values (Fig. 1c). Lang et al.[68] presents observations from a recent field campaign illustrating the common post-frontal structure of the MCC upwind of CGO. Both stability parameters highlight a more stable condition for the closed MCC with a higher EIS and a lower M range (Fig. 1d, e), consistent with McCoy et al.[61]. Furthermore, statistical analysis, specifically the Kolmogorov–Smirnov (KS) test, was employed to test the null hypothesis that the distributions of these parameters are the same for open and closed MCC. The results confirmed significant differences in these distributions ($p < 0.05$), underscoring the distinct characteristics associated with each MCC type.

To further investigate the meteorological differences under open and closed MCC, we also examined their composite soundings using ERA5. As shown in Fig. 2, closed MCC (Fig. 2a) exhibits a stronger inversion consistent with the higher EIS in Fig. 1d. On the other hand, open MCC (Fig. 2b) displays a higher inversion altitude, which is typically associated with conditions favorable for enhanced precipitation[57].

## Seasonal variations in the CCN-precipitation relationship

Although we have less than six complete years of all observations, it is of interest to examine the seasonal cycle of these records (Table 3), in an effort to investigate the role of precipitation in this cycle. Turning first to the median baseline $N_{CCN}$, we observe a strong seasonal cycle with an Austral summer (DJF) maximum of 157 cm$^{-3}$ and a winter (JJA) minimum of 54 cm$^{-3}$, which is consistent with the long-term CGO records[20,21,28,29,34,37,69,70]. The average baseline precipitation rate and frequency exhibit a pattern opposite to that of median $N_{CCN}$, with a maximum in winter (2.69 mm day$^{-1}$ occurring 20.3% of the time) and a minimum in summer (0.68 mm day$^{-1}$ occurring 6.4% of time). We note that the average baseline precipitation rate (1.69 mm day$^{-1}$) substantially contributes to the overall precipitation rate across this latitude band (2.5–3.2 mm day$^{-1}$)[48], which is found in various precipitation products[48]. Statistical independent sample

**Table 3 | Seasonal variations**

| Season (No. of hours in baseline) | Frequency of coverage (%) | Median $N_{CCN}$ (5th, 95th) | Precipitation rate (mm day$^{-1}$) | Precipitation frequency (%) |
|---|---|---|---|---|
| Annual (17470) All Baseline | – | 87 (23, 248) | 1.69 | 13.6 |
| Summer (4394) All Baseline | – | 157 (52, 327) | 0.68 | 6.4 |
| Autumn (4462) All Baseline | – | 81 (23, 201) | 1.95 | 15.4 |
| Winter (3836) All Baseline | – | 54 (16, 127) | 2.69 | 20.3 |
| Spring (4778) All Baseline | – | 84 (24, 203) | 1.58 | 13.1 |
| Annual Open MCC | 18.8 | 69 (23, 174) | 1.72 | 16.7 |
| Summer Open MCC | 6.2 | 145 (72, 241) | 1.07 | 12.4 |
| Autumn Open MCC | 14.3 | 78 (32, 160) | 1.97 | 17.2 |
| Winter Open MCC | 30.7 | 47 (18, 115) | 1.77 | 18.9 |
| Spring Open MCC | 25.0 | 75 (26, 174) | 1.68 | 15.2 |
| Annual Closed MCC | 6.0 | 89 (34, 222) | 0.29 | 4.5 |
| Summer Closed MCC | 4.8 | 165 (76, 271) | 0.10 | 2.9 |
| Autumn Closed MCC | 8.3 | 90 (36, 189) | 0.44 | 6.5 |
| Winter Closed MCC | 5.2 | 64 (29, 125) | 0.33 | 4.5 |
| Spring Closed MCC | 5.6 | 86 (32,171) | 0.16 | 3.0 |

Median $N_{CCN}$, mean precipitation rate and frequency across all baseline conditions regardless of MCC type and also for the open and closed MCC cases within the 100 km radiuses and 3 h averaging time and the $FC_{MCC}$ threshold of 80% for 2016–2021 along with the frequency of their presence.

tests, including two-tailed Student's *t* test and Whitney *U* test, were conducted to assess the significance of the observed differences in $N_{CCN}$ and precipitation. These tests aimed to test the null hypotheses that there is no difference in $N_{CCN}$ and precipitation across the seasons. The results confirmed that these differences are statistically significant ($p < 0.05$), reinforcing the validity of our findings.

After segregating the records into open and closed MCC cases, we observe an inverse tendency between $N_{CCN}$ and precipitation when we analyze the data across different seasons (except for the closed MCC during the winter, which can be attributed to the limited number of cases with precipitation from closed MCC during this season). Further we find that for all seasons, open MCC has a greater average precipitation rate where it is more frequent as well and lower median $N_{CCN}$ compared to closed MCC (Table 3). The relative difference in median $N_{CCN}$ is least in summer (12%) and greatest in winter (27%). The most pristine air is observed at CGO when open MCC is present upwind during the winter season. Using the statistical tests (including two-tailed Student's *t* test and Whitney *U* test), we tested the null hypotheses that there is no difference in $N_{CCN}$ and precipitation between open and closed MCC across different seasons, which confirm the significance of these differences ($p < 0.05$).

We further note a strong seasonal cycle in the frequency of coverage of open MCC with a wintertime peak of 30.7% of all baseline records and a summertime minimum of 6.2% (Table 3). Such a seasonal cycle is consistent with numerous climatological studies (e.g.[60,68]). Conversely no substantial seasonal cycle is evident in the frequency of occurrence of closed MCC (Table 3).

Using HYSPLIT back trajectories we sought to establish whether differences in the air mass origin could be linked to differences in $N_{CCN}$, and potentially cloud morphology. The 72-h back trajectories (Fig. 3), however, are not conclusive with only weak differences between seasons and between the open and closed MCC. Immediately upwind of Tasmania, back trajectories of closed MCC predominantly come from the west, while open MCC trajectories have mostly a south-westerly origin. During winter, open MCC back trajectories tend to originate at higher latitudes than closed MCC. Also, for closed MCC during winter, there is an interesting air mass origin region in the Australian Bight, near the coast. The average $N_{CCN}$ for these scenes was similar to overall average. In general, the baseline air masses originate from higher latitudes, as previously established. Only rarely do these back trajectories cross over Antarctica. The vertical component of these back trajectories (Fig. 4) suggests that large-scale subsidence prevails for both open and closed MCC, somewhat stronger for closed MCC. Back trajectories of open MCC in winter show considerably more spread through the boundary layer, suggesting a well-mixed boundary layer due possibly to active shallow convection. There is less spread in the vertical history of closed MCC back trajectories, consistent with widespread subsidence.

An alternate hypothesis for the cause of the observed seasonal cycle in open MCC and precipitation at CGO pertains to the annual migration in the subtropical ridge, which reaches its highest latitude (38° S) along Australia during February with an average intensity of 1016 hPa[71]. During winter the subtropical ridge strengthens in intensity but retreats to lower latitudes (28° S) over the Australian continent. Manton et al.[48] found a strong correlation coefficient (~−0.6) between precipitation and MSLP across the SO, highlighting the importance of the general circulation in precipitation processes.

The seasonal cycles of atmospheric stability parameters were analyzed (Table 4) to further assess their relationship to the baseline conditions and cloud morphology. These parameters are important for determining the depth of the MABL and evolution of low clouds[61], which are a major source of precipitation in many regions. Lang et al.[72] also found a connection between EIS and the occurrence of precipitation over Macquarie Island. Both M and EIS show a seasonal cycle with stronger stability (lower M and higher EIS) during summer (DJF), consistent with Lang et al.[72] and McCoy et al.[61]. Also, there is a stronger inversion (61% greater EIS) under closed MCC coverage, consistent with the composite sounding (Fig. 2a). Thus, these meteorological controls may be of further importance in setting the seasonal cycle of precipitation, which may also contribute to the seasonal cycle of $N_{CCN}$.

In order to evaluate the importance of wet deposition in the evolution of the CCN budget from closed to open MCC across the SO, we sought to employ a quantitative approach. Previous studies have noted the importance of precipitation in the transition from closed to open MCC[64,66,73] and the hypothesis is that wet deposition will clean out the MABL by removing particles. Kang et al.[43] utilized the recent summertime Clouds Radiation Aerosol Transport Experimental Study (SOCRATES) campaign over the SO to drive the CCN budget model developed by Wood et al.[42]. This model considers various source and sink terms, including entrainment of CCN from the free troposphere ($N_{FT}$), primary production at the sea surface from sea-spray ($N_s$) and precipitation that is induced by coalescence scavenging ($N_p$)[7,42,43]. Kang et al.[43] assumed the system was at steady state and found that

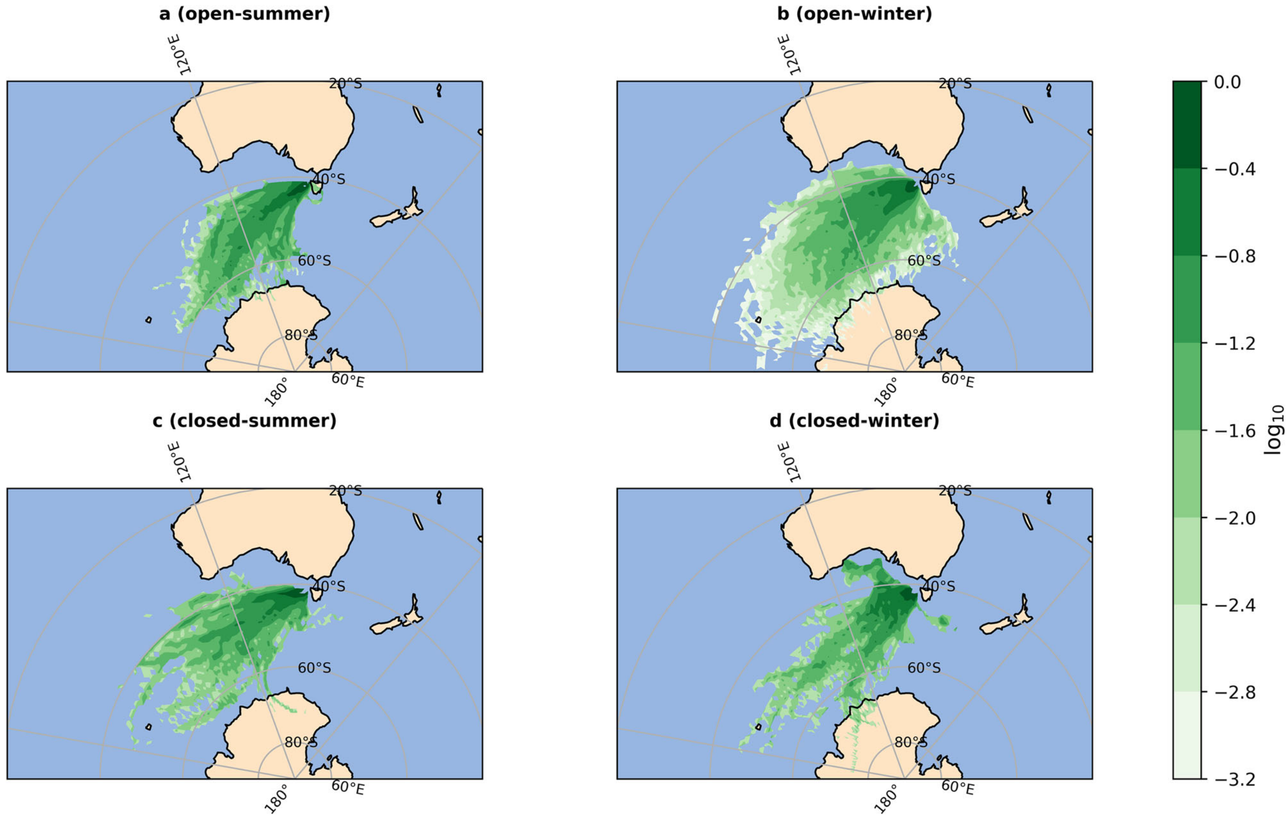


**Fig. 3 | Air masses origin.** Back trajectories of air parcels at a height of 1000 m at Kennaook/Cape Grim for a 72-h period, during Austral summer (left) and winter (right) when more than 80% of the baseline with a radius of 100 km was covered by open MCC (top) and closed MCC (bottom) for 3 h averaging time (2016–2021).

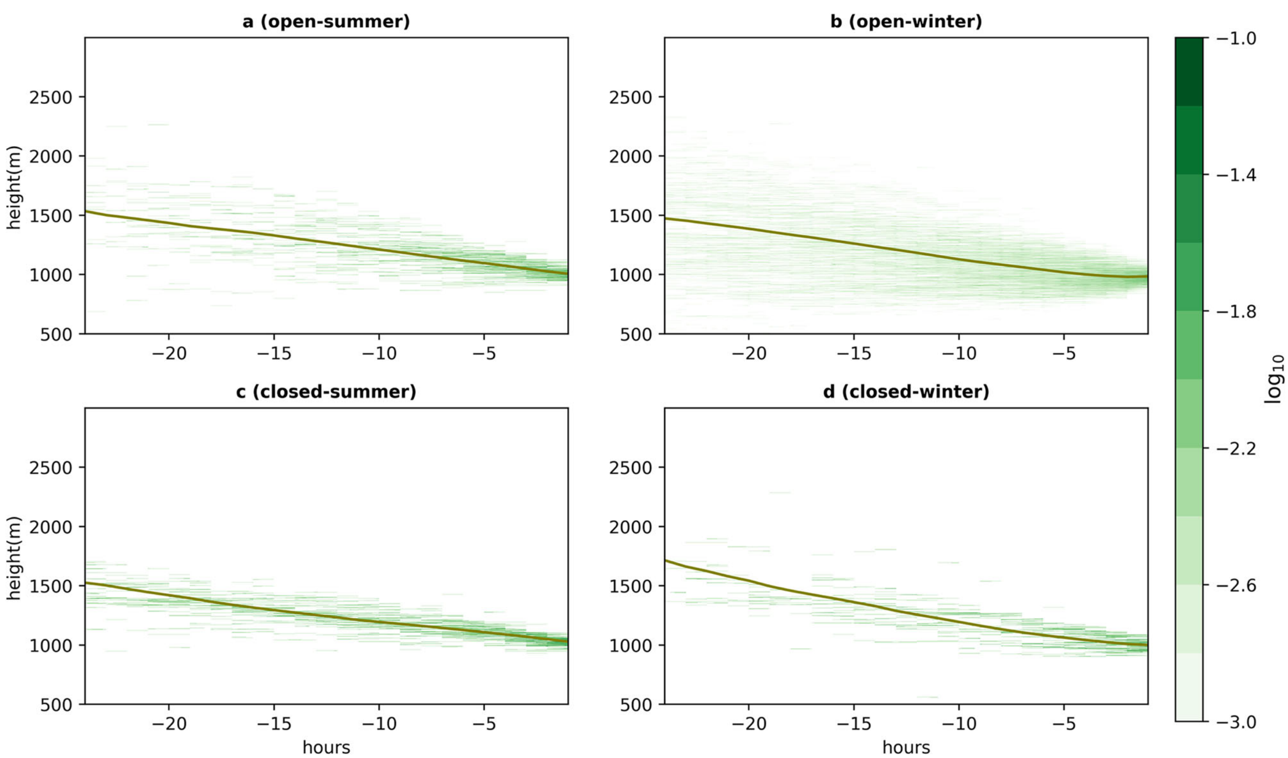


**Fig. 4 | Back trajectory altitude.** Vertical motion along the average back trajectory (solid line) of the 24-h back trajectories of air parcels at a height of 1000 m, for cases where more than 80% of the baseline with a radius of 100 km in 3 h averaging time, was covered by open MCC (top) and closed MCC (bottom) for Austral summer (left) and winter (right) (2016–2021).

**Table 4 | Seasonal variations for meteorological parameters**

| Season (No. of hours in baseline) | M | EIS | MSLP (hPa) |
|---|---|---|---|
| Summer (4394) All Baseline | −3.66 | 6.26 | 1012.9 |
| Autumn (4462) All Baseline | −0.69 | 5.85 | 1016.9 |
| Winter (3836) All Baseline | 0.20 | 4.26 | 1016.6 |
| Spring (4778) All Baseline | −1.48 | 5.29 | 1015.1 |
| Summer Open MCC | 1.94 | 2.73 | 1010.5 |
| Autumn Open MCC | 2.24 | 3.69 | 1014.8 |
| Winter Open MCC | 1.30 | 3.27 | 1015.7 |
| Spring Open MCC | 0.98 | 3.29 | 1014.1 |
| Summer Closed MCC | −3.95 | 8.43 | 1014.8 |
| Autumn Closed MCC | 0.77 | 9.28 | 1019.8 |
| Winter Closed MCC | 0.27 | 7.40 | 1024.0 |
| Spring Closed MCC | −1.38 | 8.66 | 1019.9 |

Average MSLP, M and EIS of the whole baseline followed with the open and closed condition considering the cases for the 100 km radiuses and 3 h averaging time and the $FC_{MCC}$ threshold of 80%.

the CCN budget was most sensitive to the $N_{CCN}$ of the overlying free troposphere compared to other sources. Coalescence scavenging was also found to be an important sink.

Following Kang et al.[43], we also employed the CCN budget model along with available datasets to perform a CCN budget analysis. Considering the relatively constant $N_{FT}$ and $N_s$ during the summer when moving between open and closed MCC (as suggested by Kang et al.[43]), we can assume that

$$\frac{N_{closed} - N_{open}}{\Delta t} = \dot{N}_{pclosed} - \dot{N}_{popen} \quad (1)$$

This equation suggests that the reduction in $N_{CCN}$ from closed to open MCC can be attributed to precipitation along the trajectory. The precipitation sink term depends upon the following terms[74]:

$$\dot{N}_P = K\, N\, P_{CB} h / z_i \quad (2)$$

where $P_{CB}$ represents the precipitation rate at cloud base, $K = 2.25\ m^2\ kg^{-1}$ is a constant that depends on the collection efficiency of cloud droplets by drizzle drops, $h$ is the cloud thickness and $z_i$ is the depth of the MABL[74]. Due to the lack of cloud base precipitation measurements in our datasets, we use surface precipitation data as a proxy for $P_{CB}$. Cloud thickness and boundary layer height are estimated using the composite ERA5 soundings.

Our calculations indicate that it would take approximately 3 h for precipitation during the transition from closed to open MCC to remove the 12% median CCN differences between closed (165 $cm^{-3}$) and open MCC (145 $cm^{-3}$) during summer. It should be noted that this estimate is defined by constraints in our calculation. Extending such quantitative analysis to other seasons is limited by a lack of enough observational information (e.g., the $N_{FT}$ and $N_s$). Nonetheless, our expanded budget analysis for other seasons indicates transition time within the range of 2–6 h. Another limitation of this analysis pertains to the estimation of precipitation that is induced by coalescence scavenging ($N_p$), which relies on the precipitation rate at the cloud base level[74]. Our measurements, however, are only available at the surface. Recent research by Kang et al.[75], found substantial differences between precipitation rates at the cloud base and the surface for MABL clouds over the SO using in situ and airborne cloud radar observations. K, which has been used in estimating $N_p$, is also subject to uncertainty. Nevertheless, these limitations highlight the significance of future coordinated efforts, such as multi-platform measurement campaigns in this region to supply critical datasets needed for cloud-aerosol-precipitation research, including a more comprehensive CCN budget analysis.

## Meteorological influences on diurnal precipitation

Moving beyond the seasonal cycle, we can also explore potential relationships between the median $N_{CCN}$ and precipitation at the diurnal time scale, aiming to understand whether CCN acts as the driver of the precipitation, or vice versa. We limit this analysis to the summer season, when solar forcing is most intense and can readily decouple, thin and even completely burn off MABL clouds[76,77]. The Lang et al.[68] cloud climatology readily found such a diurnal cycle in closed MCC over the latitude band between 40° to 50° south. For this analysis, however, we employ all baseline samples (open, closed and other) to increase the sample size and ensure a sufficient amount of data for analysis and to clearly illustrate the diurnal cycle. Furthermore, in order to adequately assess the diurnal cycle, data for precipitation and $N_{CCN}$ from 2011 to 2015 has been included in this analysis (both data were not available before 2011). Looking first at the precipitation (Fig. 5b, c), a strong diurnal cycle is evident in both mean precipitation rate and frequency with a peak in the early morning (5 AM local time) and a minimum in the afternoon. It should be mentioned that the 25th and 75th percentile of precipitation were not presented as both values were zero during the summer. Lang et al.[72] and Tansey et al.[58] found a similar diurnal cycle in precipitation over Macquarie Island. The median $N_{CCN}$ at CGO (Fig. 5a) also displays a diurnal cycle, contrasting with the precipitation pattern with a peak concentration observed at 8 PM. Ayers and Gillet[20] and Ayers et al.[69] also found a diurnal cycle for $N_{CCN}$ over the CGO. Wider research suggests that the thinning of the marine boundary layer clouds through solar forcing/burn off is likely to be driving the diurnal cycle of the precipitation rate[72,78]. Additionally, we observe a diurnal cycle in MSLP (Fig. 5d) and the two stability parameters (M and EIS) (Fig. 5e, f). These meteorological conditions, particularly the stability of the atmosphere and related dynamics, are likely other drivers of the observed diurnal cycles in precipitation. It is especially intriguing to note that the minimum of MSLP coincided with the peaks of precipitation frequency and intensity. These findings further emphasize the crucial role of meteorological controls on ACPI.

In our effort to examine the meteorological impacts on ACPI behaviors, we employed multilinear regression to examine the relationship between meteorological factors, precipitation and aerosol. Subsequently, we removed the meteorological influences on precipitation and $N_{CCN}$ and examined the residuals. Notably, our findings indicate that, after controlling for the meteorology, the correlation between precipitation and $N_{CCN}$ decreased from −0.045 to −0.034 (see details in the attached supplementary materials), suggesting that a substantial portion of the relationship between precipitation and CCN is explained by meteorological factors. Additionally, we conducted a comprehensive analysis comparing multilinear regression models of precipitation with and without considering CCN, revealing minimal differences in $R^2$ values and correlation coefficients (see details in the attached supplementary materials). This underscores the limited influence of CCN on precipitation, emphasizing the substantial role played by meteorological factors in shaping their relationship. It is important to note that our findings do not imply that the role of CCN is negligible, especially in a changing climate. Even minor influences from the CCN could have important effects on climate given the complexity of the climate system.

Importantly we note that the diurnal cycle in precipitation leads the small diurnal cycle in CCN by roughly 3 h (Fig. 5). To test the significance of this time lag, we applied the Bootstrapping method by which 10000 samples were randomly drawn with a sample size of 8000 (from a total of 8200-time steps). The results indicate that in over 98% of the instances, the minimum $N_{CCN}$ occurred after the maximum precipitation (in just 2% of the instances, the minimum $N_{CCN}$ preceded the maximum precipitation). This suggests that the diurnal cycle in precipitation is unlikely to be led by CCN (with a confidence level of 98%). These analyses emphasize that multiple factors, particularly the meteorology, are likely at play in shaping the diurnal patterns of precipitation, and the influence of CCN on precipitation is not a dominant mechanism over our study area. Again, further investigation is needed to explore how precipitation may be influencing the diurnal cycle of CCN through processes like wet deposition, but it is clear that the relationship between CCN and precipitation is complex.

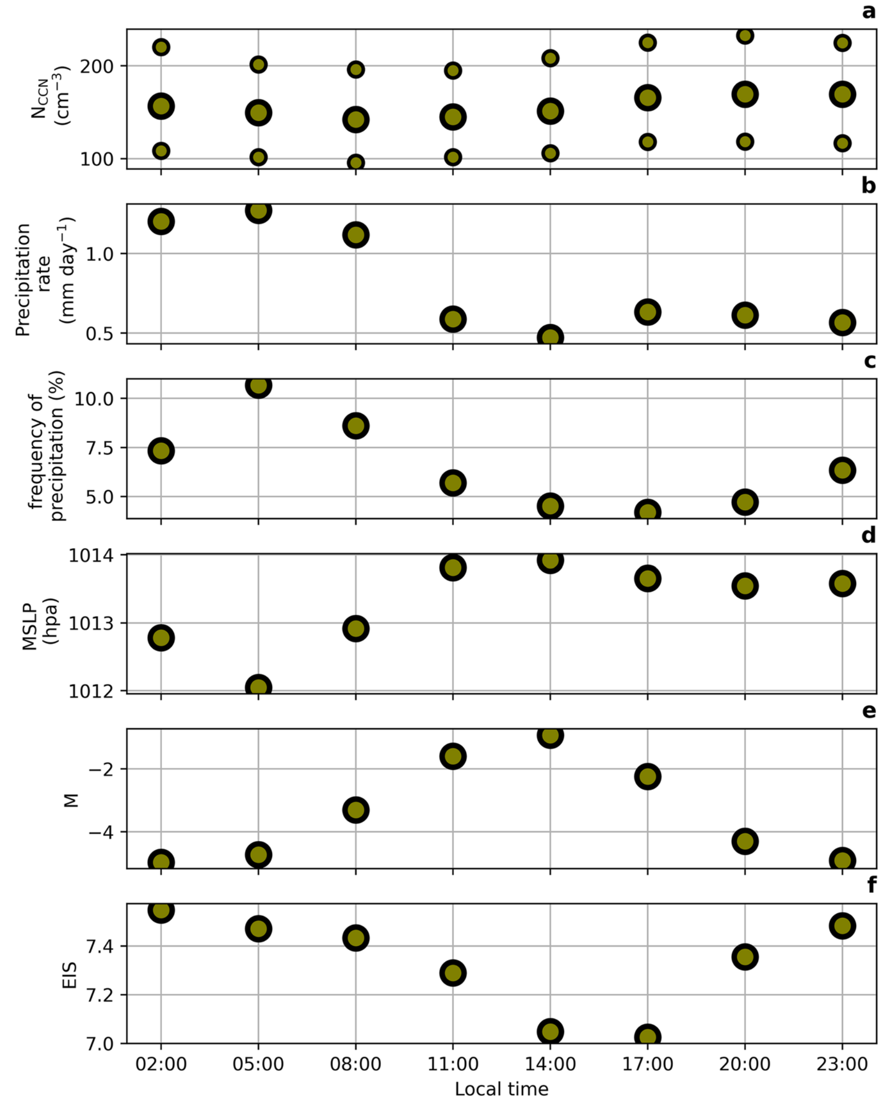


**Fig. 5 | Diurnal variations.** Diurnal cycle of $N_{CCN}$ ($cm^{-3}$) (median followed by 25th and 75th percentile) (**a**), average precipitation rate (mm day$^{-1}$) (**b**) and frequency of precipitation (%) (**c**), MSLP (**d**), M (**e**) and EIS (**f**) during the summertime (2011–2021) in baseline of CGO.

Overall, this analysis once again suggests that a greater understanding of the sinks of aerosols is required to more accurately close the CCN budget across the SO. Specifically, this analysis highlights the potential importance of precipitation in this regard, and the role of the large-scale circulation in driving precipitation across this region.

## Methods

### Data

The Baseline Air Pollution Monitoring (BAPMon) Station at Kennaook/Cape Grim (CGO) began operations in 1976 as part of the World Meteorological Organization BAPMon program of global atmospheric composition measurements relevant to climate. The observatory is situated at 40° 40′ 56″ S, 144° 41′ 18″ E, at the north-west tip of Tasmania, to ensure observations of SO air with minimal anthropogenic influences. The observatory building is located 94 m above sea level, roughly 100 m inland from the coastline break[20,37,70]. Figure 6 depicts the location of Cape Grim.

The $N_{CCN}$ for particles active at several supersaturations, but predominantly at 0.5% supersaturation (other supersaturations are not available hourly), was determined using a continuous-flow, streamwise thermal gradient CCN counter (CCNC, model CCN-100, Droplet Measurement Technologies, Longmont, CO, USA)[70]. The CCN counter supersaturation was calibrated annually using monodisperse ammonium sulfate particles[37]. The hourly $N_{CCN}$ over CGO for 11 years (2011–2021) was used to examine the role of precipitation and cloud morphology on SO $N_{CCN}$. The data are available in the World Data Centre for Aerosols (http://www.gaw-wdca.org/). However, it should be noted that 5 months of CCN records were lost during a COVID lockdown (from the end of September 2020 to March 2021) when the instrument was non-operational. The hourly precipitation data, expressed as mm day$^{-1}$, were from the Australian Bureau of Meteorology rain gauge at CGO (Station ID: 091331) for the same period of time.

The measurement of radon is also carried out hourly with the dual-flow-loop two-filter atmospheric radon detectors over the CGO station[79,80]. With its predominantly terrestrial source, unreactive nature, and 3.82-day radioactive half-life, radon is an unambiguous tracer of terrestrial influences on sampled air masses[81–83]. The fifth generation of European ReAnalysis (ERA5) wind data, produced by the European Centre for Medium-Range Weather Forecasts (ECMWF)[84], which is available through the Copernicus Climate Change Service Climate Data Store (https://cds.climate.copernicus.eu), were used along with the radon to determine baseline conditions for the same time period.

Finally, the hourly climatology of open and closed MCC is calculated from Himawari-8 imagery (13 µm channel) using a hybrid convolutional neural network (for more details, see Lang et al.[68]) for the six years (2016–2021). Himawari-8, a geostationary meteorological satellite, was launched by the Japanese Meteorological Agency in July 2015 and covers a large part of the SO[85]. The classification of MCC into open and closed categories was not based on cloud fraction or liquid water content, but rather relied on a convolutional neural network's pattern recognition capabilities, as demonstrated by Lang et al.[68]. They constructed and trained it to identify

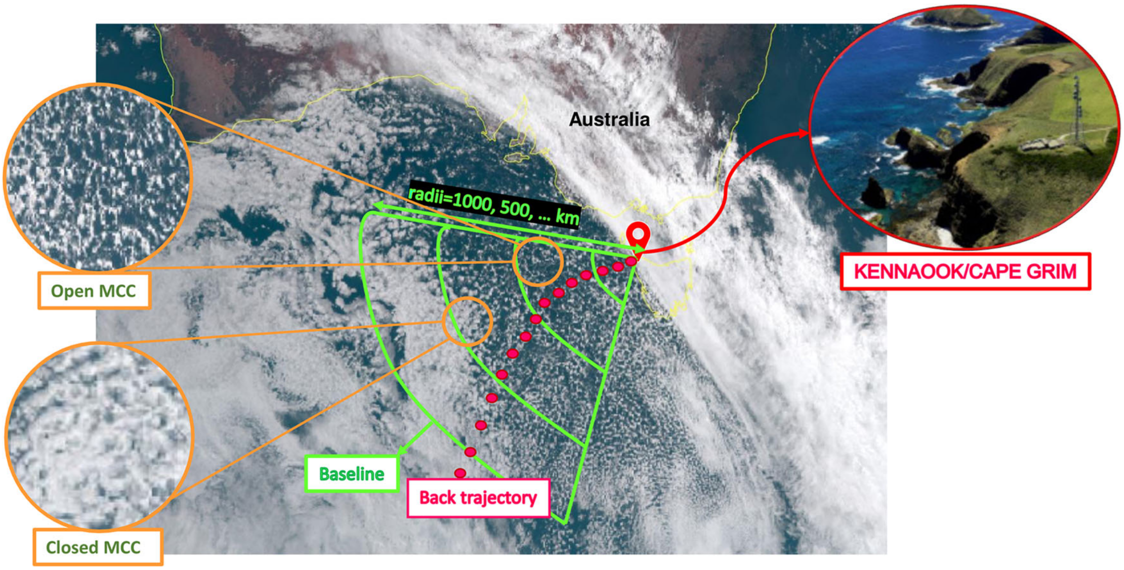


**Fig. 6 | Overview of the study area and methodology.** A true colour image of Himawari-8 (https://www.eorc.jaxa.jp/ptree/) on 14 January 2016, 00:00 UTC, supplied by the P-Tree System, Japan Aerospace Exploration Agency (JAXA), which illustrates the study area (CGO image sourced from https://capegrim.csiro.au/), the baseline sector defined based on different radii in green colour, symbolizing the varying analysis areas, a sample back trajectory in red colour started from CGO and also a snapshot of different upwind cloud morphologies.

highly ideal open and closed MCC imagery. All other scenes (e.g., clear sky, frontal clouds, cirrus clouds, stratus, disorganized MCC) are simply grouped together as "other" which restricts the ability to examine these circumstances. It should be noted that our analysis is limited by the availability of Himawari-8 records, which became operational in July 2015[85], for classifying the cloud morphology at the time of writing.

## Methodology

The initial hypothesis was evaluated using these datasets with two different methodologies. The first method defines baseline conditions, as local wind directions between 190 between 280 degrees (e.g.[37,86]) and ambient radon concentration less than 150 mBq $m^{-3}$, which includes approximately 80% of baseline sector observations[37]. To assess the sensitivity of the analysis to radon concentration thresholds, we also tested a lower threshold of 100 mBq $m^{-3}$. Qualitatively, the results are not sensitive to different radon concentration thresholds. Air sampled in the 'baseline' sector (190°–280°) frequently traveled thousands of kilometres across the SO since last land contact. Baseline sector and other detailed parameters for this method are depicted in Fig. 6 (green colours) and will be discussed later. The second method was based on back-trajectory calculations made with the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model[87] employing ERA5 meteorological data, providing the transit history of each air parcel (red colour in Fig. 6). It provided information on the impact of MCC type on each air parcel on its journey to CGO. Back trajectories were started at 1000 m elevation; nevertheless, the results are qualitatively similar to an initial elevation of 500 m. Since the results for both methods are consistent, we employed only the first method. This analysis is limited to 2016–2021 due to the availability of open and closed MCC climatology data from Himawari-8 observations. However, for the diurnal cycle analysis of precipitation and $N_{CCN}$ under baseline conditions, we used the full 11-year dataset to increase the sample size. Cloud morphology is not considered in the diurnal cycle analysis due to the small sample size for the period of summer 2016 to 2021.

Application of the baseline constraints, using ERA5 wind data and the radon constraint, leads to the elimination of 63% of all hourly records. For the remaining 17470 h, the fraction of coverage for both open and closed MCC ($FC_{MCC}$) was calculated using the Lang et al. classification[68] on Himawari-8 imagery over various upwind averaging times (1, 3 and 6 h) in the baseline sector with various upwind domain radii (50, 100, 200, 300, 500 and 1000 km) as shown in Fig. 6 (green colours). The upwind averaging time accounted for the travel time of air parcels in the baseline sector to reach CGO, while various domain radii were considered to assess the potential influence of upstream frontal systems on the results. For each upwind radius and averaging time, the mean domain $FC_{MCC}$ for both open and closed MCC was computed. The $FC_{MCC}$ is defined as the percentage of the baseline sector covered by either open or closed MCC within the specified radius. To distinguish whether a sample was primarily open or closed MCC, two $FC_{MCC}$ thresholds were used: 50% was considered as a basic requirement while 80% was considered as a more certain requirement. These trials assessed the sensitivity of the results to the upwind radius and upwind averaging time. For each configuration, we considered cases where either open or closed MCC covered more than 50% or 80% of the baseline sector ($FC_{MCC}$ >50% or 80% for open and closed). The median and the 5th and 95th percentiles of $N_{CCN}$ were determined for the times when each cloud class (open or closed MCC) was dominant ($FC_{MCC}$ >50% or 80%). Mean precipitation intensity and frequency were also determined for each case.

To briefly investigate potential meteorological factors influencing the observed cycles in precipitation from open and closed MCCs, we examined the stability parameters including the estimated inversion strength (EIS) and the marine cold air outbreak parameter (M). Each is calculated using ERA5 reanalysis. EIS estimates the strength of the planetary boundary layer inversion and is defined as[88]

$$EIS = LTS - \Gamma_m^{850}(z_{700} - LCL) \tag{3}$$

where the LTS is the lower tropospheric stability defined as the difference in potential temperature between 700 hPa and the surface (LTS = $\theta_{700}-\theta_{surf}$)[89], $\Gamma_m^{850}$ is the moist-adiabatic potential temperature gradient at 850 hPa, $Z_{700}$ is the altitude of the 700 hPa level and LCL is the lifting condensation level. Greater EIS indicates stronger temperature inversions, which can suppress vertical mixing and reduce the potential for cloud development and precipitation[72,88,90,91]. M was originally defined by Kolstad and Bracegirdle[92] and modified by Fletcher et al.[93] as the difference between the surface skin potential temperature and the 800 hPa potential temperature[93]. However, given that our study area is in the Southern Hemisphere, we used the 850 hPa potential temperature for the M calculation. This is consistent with Papritz et al.[94] over the South Pacific. Moreover, considering the surface skin temperature instead of sea surface temperature will exclude the areas of high sea-ice cover[93]. Positive M indicates a cold air mass over a relatively warmer surface, leading to an absolutely unstable boundary layer. This condition favours cellular convection, a typical signature of cold air outbreaks, which can result in precipitation[61,93].

HYSPLIT back-trajectories were checked to examine whether any differences between open and closed MCC were linked to airmass origins. Each back-trajectory was run from ERA5 fields for 72 h initiated at a height of 1000 m at CGO. To run the back-trajectories, instances were classified as either open or closed depending on whether the $FC_{MCC}$ for a 100 km upwind length and 3 h averaging time was greater than 80%.

## Data availability

The CCN concentration measurement, analyzed during the current study are available in the World Data Centre for Aerosols [http://www.gaw-wdca.org/]. The ECMWF-ERA5 reanalysis datasets are available through the Copernicus Climate Change Service Climate Data Store [https://cds.climate.copernicus.eu]. The precipitation data can be obtained by contacting [climatedata@bom.gov.au]. The radon data is available from the World Data Centre for Greenhouse Gases (WDCGG) [https://gaw.kishou.go.jp/] and from Alastair Williams from Australian Nuclear Science and Technology Organisation (ANSTO). The climatology of open and closed MCC available from the F. Lang on reasonable request.

## Code availability

All relevant codes used in this work are not publicly available but can be made available to qualified researchers upon reasonable request from the corresponding author.

Received: 24 April 2023; Accepted: 14 March 2024;
Published online: 22 March 2024

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## Acknowledgements

This research has been supported by the Australian Research Council Discovery Projects (DP190101362). Continued support for the Kennaook Cape Grim Program from the Australian Bureau of Meteorology and Commonwealth Scientific and Industrial Research Organisation (CSIRO) is also gratefully acknowledged.

## Author contributions

T. Alinejadtabrizi prepared the original draft of the paper and performed most of the data analysis. F. Lang & L. Ackermann prepared the climatology of open and closed MCC for 2016 to 2021. F. Lang also calculated stability parameters over the study area. All co-authors provided editorial feedback on the paper. All co-authors read and approved the final manuscript.

## Competing interests

The authors declare no competing interests.

## Additional information

**Supplementary information** The online version contains supplementary material available at https://doi.org/10.1038/s41612-024-00625-1.

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