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AEROSOL CHARACTERISTICS OBSERVED IN SOUTHEAST QUEENSLAND AND IMPLICATIONS FOR CLOUD MICROPHYSICS

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1. INTRODUCTION

Understanding the influence of aerosol on cloud microphysics and precipitation formation is of great importance, especially since such effects are potentially large yet not well understood (IPCC 2007). While it has been fairly well established that aerosols are necessary for cloud droplet nucleation and that as such aerosols could affect precipitation formation, it is still not well understood to what extent subsequent precipitation growth is affected by aerosol-influenced droplet spectra, especially in deep convection (Squires 1958, Rosenfeld 1999, 2000, Khain et al. 2005, Tao et al. 2007, Rosenfeld et al. 2008). Aerosol microphysical effects also apply to the concept of hygroscopic cloud seeding, which is based on the principle that adding large hygroscopic aerosols to a cloud could enhance the warm rain precipitation formation process, thereby enhancing the precipitation efficiency of the cloud and producing more overall rainfall at the surface (Bruintjes 1999, Silverman 2003). The concept of hygroscopic cloud seeding has often been studied as a counter response to warm rain suppression that might occur in a polluted environment (Rosenfeld 2000, Rosenfeld et al. 2008).

A recent study by Bigg (2008) concentrated on this issue for the region of southeast Queensland, Australia, by speculating that urban pollution (that has increased in recent years due to the growth of the Brisbane metro and Gold Coast population centers) could be related to the decrease in rainfall that this region has faced in the last 35 years. Bigg (2008) stated, however, that "the present concentrations of potentially precipitation-influencing particles is not known at any site or for any season [in Australia]". As such, physical relationships between precipitation-influencing aerosol and rainfall in the region cannot be established. This paper presents such in situ aerosol measurements, for the southeast Queensland region, that were collected during the Queensland Cloud Seeding Research Program (QCSRP; Tessendorf et al. 2010).

Having a good understanding of the natural aerosol that are present in the atmosphere and affecting cloud droplet formation is paramount toward understanding such implications on precipitation formation or whether or not using hygroscopic cloud seeding techniques could enhance rainfall in a given region. The goal of this study is to define the typical aerosol regimes observed in the southeast Queensland region, and present representative characteristics of each of those regimes. Such representations can be used for further detailed study of the precipitation processes in the region, including initializations for cloud resolving microphysical models.

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2. DATA SET AND METHODS

The measurements discussed herein were collected during the QCSRP between November 2008 and February 2009 in southeast Queensland, near Brisbane, Australia. The South African Weather Service (SAWS) Aerocommander was flown into and around clouds in the region equipped with a suite of microphysical instrumentation to obtain measurements of aerosol and droplet size spectra (see Table 1). Fine-mode aerosol measurements were obtained with the Differential Mobility Analyzer (DMA; Stolzenburg et al. 1998), accumulation mode aerosol were measured by a Passive Cavity Aerosol Spectrometer Probe (PCASP), and when flying out of cloud the Forward Scattering Spectrometer Probe (FSSP; Dye and Baumgardner 1984) was used to measure coarse mode aerosols. However, it should be noted that the small sampling volume of the FSSP limits its ability to measure coarse particles that are often present in very low concentrations and the FSSP was always being run in cloud droplet size range mode (see Table 1). The FSSP was primarily used to measure cloud droplet spectra in cloud. A Cloud Condensation Nuclei (CCN) counter (Roberts and Nenes 2005) was used to sample CCN to compare the fraction of aerosol measured at cloud base that served as cloud droplet nuclei. A Cloud Imaging Probe (CIP; on a CAPS instrument, see Baumgardner et al. 2001) and a Precipitation Imaging Probe (PIP) were used to measure the size, shape, and concentrations of cloud and precipitation particles.

For sampling sub-cloud aerosol at cloud base (hereafter, "cloud base aerosol"), the aircraft orbited below solid cloud bases, maintaining a constant altitude and remaining in the updraft as much of the time as possible. In order to remain below the cloud and to prevent the measurements from being contaminated by aircraft exhaust, the aircraft orbits drifted with the cloud. Cloud base aerosol sampling orbits typically were on the order of five minutes long, except when CCN supersaturation (SS) cycles were being run, in which case the aircraft maintained the cloud base orbit for 10 minutes to allow for the CCN counter to sample at three supersaturations (0.3%, 0.5%. and 0.8%). The CCN counter was set to sample at 0.3% supersaturation at all other times. Statistics on cloud base aerosol measurements used in this analysis (median PCASP concentrations, median 0.3% SS CCN concentrations, and mean aerosol size spectra) were then calculated for a 3-minute segment with relatively constant PCASP concentrations during each cloud base orbit. This allowed for a full up and down sizing cycle by the DMA to be represented in each measurement.

Table 1. List of instrumentation used to take measurements discussed herein. All of the instruments except the DMA were models from Droplet Measurement Technologies (DMT).

Instrument	Purpose	Range
CCN Counter	Cloud condensation nuclei concentration and spectra	Depends on supersaturation
DMA	Fine mode aerosol spectra and concentration	0.01 to .38 μm
SPP-200 PCASP	Aerosol concentration and spectra	0.1 to 3 μm
SPP-100 FSSP	Coarse mode aerosol spectra	3–47 μm
CIP	Cloud particle size, shape, concentration	25-1500 μm
PIP	Precipitation particle size, shape, concentration	100-6200 μm

After obtaining a cloud base aerosol sample, the aircraft always ascended to collect measurements of the initial cloud droplet spectra, at roughly 300 m above cloud base. Terrain occasionally impacted the aircraft's ability to fly at this altitude in cloud, and thus the aircraft would ascend to its minimum safe altitude to conduct these cloud base passes. For the purposes of our analysis, we only consider those cloud base passes that were within 300-600 m of cloud base. Furthermore, for our purposes we wanted to avoid analyzing data from the portions of cloud passes through cloud edge or areas with precipitation falling from above, therefore we only focused on FSSP measurements with a minimum concentration of 20 cm^{-3}, and excluded measurements with any particles measured larger than 100 µm (using the CIP and PIP).

a. Back trajectory modeling

The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Rolph 2010) was used to calculate 120-hour back trajectories in order to get an air mass history for each measurement. The model was run using the Global Data Assimilation System (GDAS) archived data, with a temporal resolution of 3 hours and spatial resolution of 1 deg x 1 deg in latitude and longitude. The GDAS data set is the only one available that covers the project domain at the time of the measurements; however, comparisons of trajectories calculated using GDAS and other data sets (i.e. NCEP/NCAR Reanalysis and FNL) for the same region in past years yielded highly similar results (not shown). The starting latitude, longitude, and altitude for each trajectory was based on the actual location of aircraft while taking measurements. The back trajectories, shown in Figure 1, illustrate the wide variety of source regions.

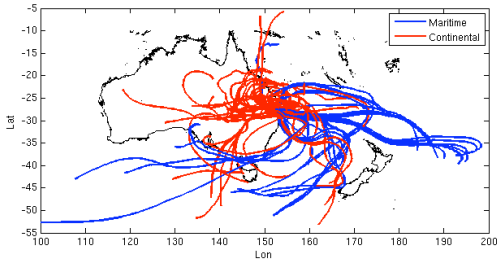


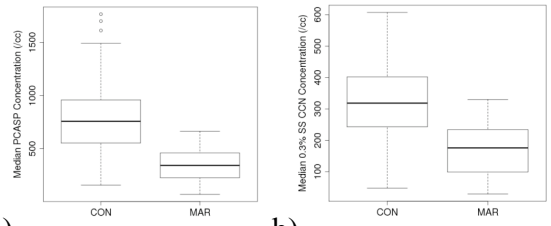
Figure 1. 120-hour HYSPLIT back trajectories for each cloud base aerosol measurement color-coded into two regimes based on how much time each trajectory spent over land: maritime regime <= 18 hrs over land (blue) and continental regime > 18 hrs over land (red).

3. REGIME CLASSIFICATION

Using the modeled back trajectories for each of the 69 cloud base aerosol measurements collected, we wanted to empirically determine different aerosol regimes that were observed in the region. Using the hours a trajectory spent over land as a primary influential factor on the measured aerosol conditions, we determined that splitting the data set into two regimes produced significantly distinct aerosol regimes (in aerosol concentration, as well as aerosol size spectra): maritime, identified as <=18 hours over land, and continental, with >18 hours over land (see Figure 1). With this classification, the two regimes exhibited generally distinct and more uniform PCASP concentrations than the season-wide dataset; however, there is still a fair amount of variability within each regime (Figure 2a).

These two regimes also exhibit distinctly different CCN concentrations, both at 0.3% supersaturation (Figure 2b) and across all supersaturations, on average, for those measurements with supersaturation cycles (Figure 3). Nonetheless, there is still considerable variability in the individual CCN SS cycle concentrations, that will be

explored in future analysis. However, those measurements with low (high) PCASP aerosol concentrations also tend to exhibit low (high) 0.3% SS CCN concentrations (Figure 4).



a) b)

Figure 2. Box and whisker plots of (a) median PCASP aerosol concentration and (b) median 0.3% SS CCN concentration split by regime (MAR: <=18 hours over land; CON: >18 hours over land).

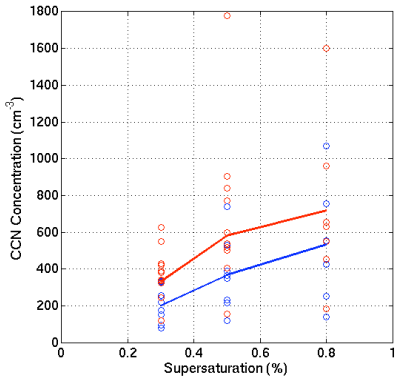


Figure 3. CCN concentrations at varying supersaturations measured during cloud base supersaturation cycles, colored by maritime (blue) and continental (red) regimes. The solid line connects the mean CCN concentration at each supersaturation for each regime.

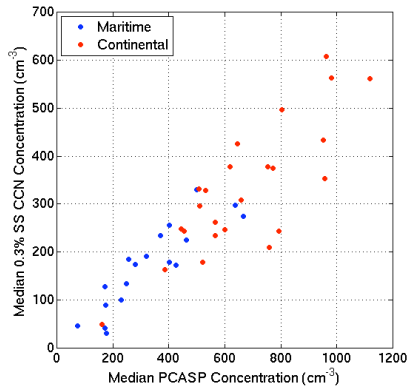


Figure 4. Scatter plot of the median PCASP concentration and 0.3% SS CCN concentration for each cloud base aerosol measurement, colored by maritime (blue) and continental (red) regimes.

a. Other influential factors

The variability in the measurements that still exists after classifying the regimes by the hours each trajectory spent over land can be explained by other influential factors, such as the number of fires along a trajectory, the proximity of the trajectory to the city of Brisbane (defined as the city center point at 27.5 S, 153.02 E), and the amount of rainfall in the prior 12-hour period before the measurement was taken. Fire locations and times were determined by MODIS satellite measurements and rainfall was calculated from the GDAS model along the last 12 hours of the trajectory.

For the maritime air masses, aerosol concentrations increase rapidly with hours over land (Figure 5a) and show a significant decrease as the trajectories get further away from Brisbane (Figure 5b). Very few of the maritime cases were influenced by any fires; which is not surprising due to the relatively short time these trajectories spent over land (and therefore fewer opportunities to be near a fire) as well as because fires were more common in months when maritime flow trajectories were less common (Figure 6). November and December experience far more fires, less rainfall along the trajectory prior to measurement, and generally more continental flow while the second half of the rainy season is more maritime in nature, with fewer fires and more rainfall along the trajectories. The fires in November and December are primarily from the sugar cane harvesting season, the effects of which have been previously discussed in Warner and Twomey (1967) and Warner (1968).

Conversely, the continental concentrations are highly dependent on the number of fires experienced along the trajectory (Figure 5c),

while showing less dependence on either hours over land or distance to Brisbane (Figure 5a-b). This was also part of the reasoning behind defining maritime as less than 18 hours over land given the dependence of aerosol concentration on this variable decreases significantly above 18 hours, as other factors become more dominant. The lack of dependence on distance to Brisbane in the continental flow is also unsurprising, given that the measurements made in continental flow were inland from Brisbane and could more often have been made upwind of the city.

Figure 5d illustrates how both regimes' aerosol concentrations are affected by recent rainfall. While noisy, the trend is for the concentrations to be lower with greater rainfall, as would be expected due to precipitation scavenging (e.g., Kerker and Hampl 1974, Barlow and Latham 1983).

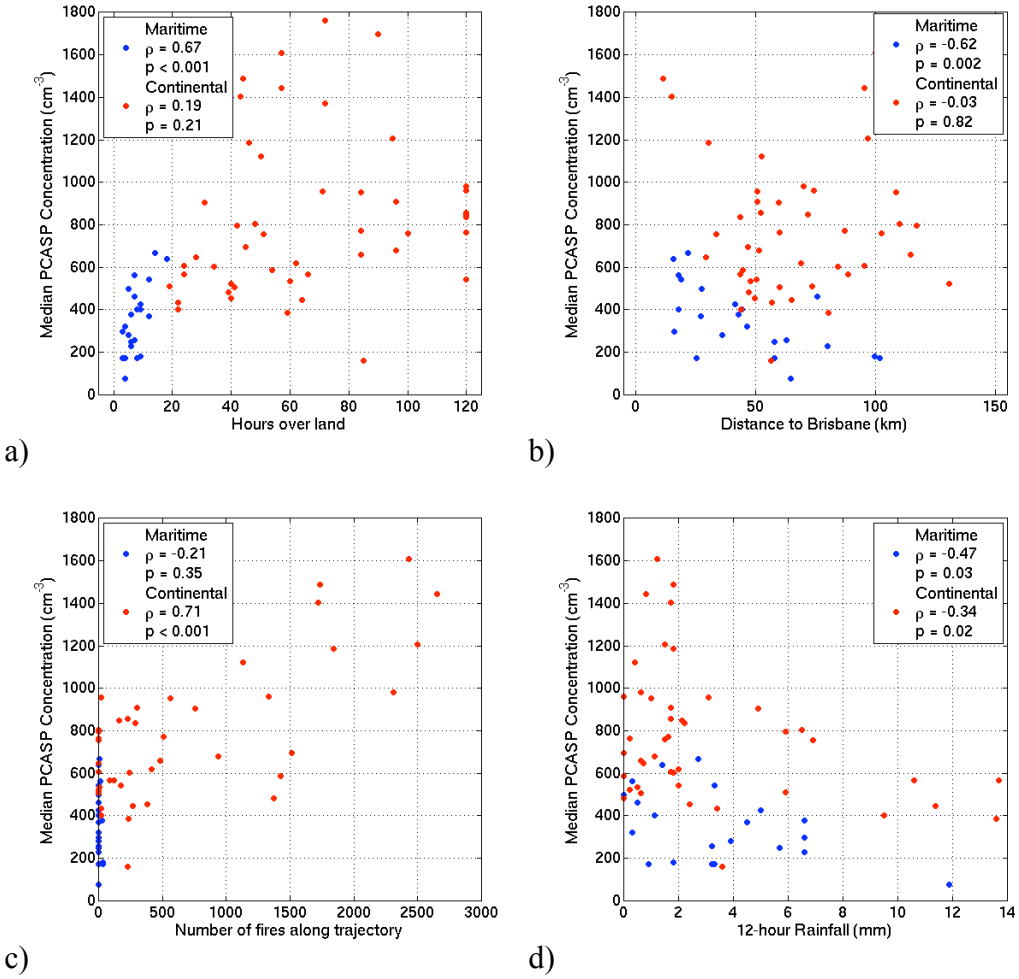


Figure 5. PCASP aerosol concentration variation based on the a) hours over land, b) distance to Brisbane, c) number of fires within 200 km of (and 12 hrs prior to) the trajectory, and d) 12-hour rainfall of each back trajectory colored by maritime (blue) and continental (red) regimes.

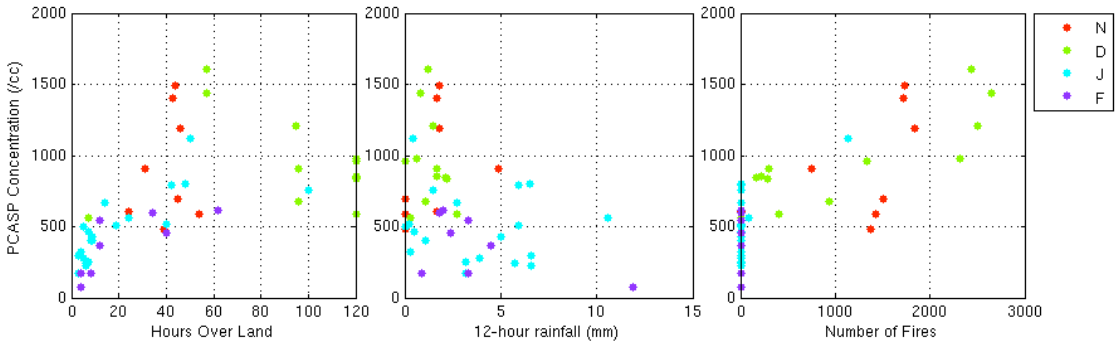


Figure 6. Monthly trends in concentration by hours over land, rainfall, and number of fires (N= November, D=December, J=January, F=February).

Based on these additional influences on aerosol concentration, each regime was broken into two sub-regimes (Figure 7): a city-influenced maritime regime was defined for trajectories with <= 18 hrs over land and < 50 km distance to Brisbane and a fire-influenced continental regime was define for trajectories with >18 hrs over land and >1000 proximity fires. The remaining measurements in each of the maritime and continental regimes were then included in respective "background" sub-regimes.

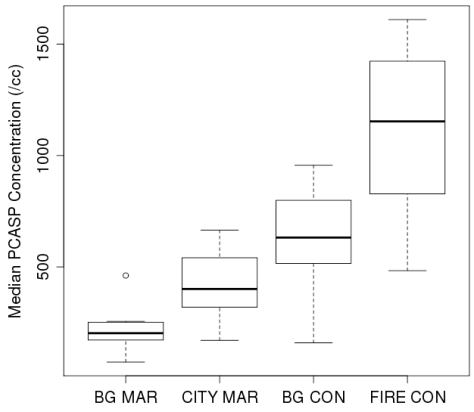


Figure 7. Box and whisker plots of median PCASP concentration for each of the sub-regimes: background maritime (BG MAR), city-influenced maritime (CITY MAR), background continental (BG CON), and fire-influenced continental (FIRE CON).

The means for the continental and maritime regimes, as well as their respective sub-regimes, are summarized in Table 2. A t-test was performed to determine if the means in each regime and sub-regime were significantly different. The t-test assumes a null hypothesis that two datasets are independent random samples from normal distributions with equal means and equal, but unknown, variances. The assumption of normality of PCASP concentrations within each regime was confirmed with the Lilliefors test (Lilliefors 1967). The t-test between the overall maritime and continental regimes yielded a p-value of $3.57*10^{-7}$ (the p-values for the tests between the sub-regime means are summarized in Table 3). Using a family-wise error rate of 0.05, all means are significantly different from one another.

Nonetheless, despite the regimes being classified in this manner, there are still some outlier measurements (for example, there is a very clean continental point with more than 80 hours over land, less than 500 fires and less than 4 mm rainfall). Additional analysis is currently underway to explain such outliers, and possibly revise the classification criteria if needed, in order to build a model that can predict the regional aerosol conditions based on the back trajectory in absence of in situ aerosol measurements.

Table 2. Regime and sub-regime mean cloud base PCASP concentrations and number of measurements.

Regime	Subregime	Mean PCASP Concentration (cm^{-3})	# of measurements
Maritime	All	351.3	22
	Background	223.3	10
	City-influenced	424.4	12
Continental	All	824.0	47
	Background	648.0	32
	Fire-influenced	1199.7	15

Table 3. P-values from the t-test for each pair of subregime means. For reference, a family-wise error rate of 0.05 would yield a critical p-value of $8.33*10^{-3}$ using the Bonferroni (Kutner et al. 2005) adjustment.

	Background Maritime	City-influenced Maritime	Background Continental	Fire-influenced Continental
Background Maritime	1	$2.61*10^{-3}$	$5.69*10^{-7}$	$1.13*10^{-6}$
City-influenced Maritime		1	$3.00*10^{-4}$	$2.03*10^{-7}$
Background Continental			1	$6.12*10^{-8}$
Fire-influenced Continental				1

4. REGIME CHARACTERISTICS

a. Aerosol characteristics

Aside from the significantly different PCASP (accumulation mode) aerosol concentrations discussed in Section 3 that were as such used to help define the two regimes, the maritime and continental regimes also have unique features in the full aerosol size spectra created by combining measurements from the DMA, PCASP, and FSSP[1] (Figure 8). Despite substantial variability in the measurements, the mean spectra do highlight the tendency for the maritime regime to have a stronger fine mode of aerosol particles compared to the continental regime. Interestingly the coarse modes in both regimes are quite similar, although the maritime regime tends to have a more robust coarse mode on average. When dividing into the four sub-regimes (Figure 9), some of the variability exhibited in Figure 8 is reduced. In particular, it is clear that the fire-influenced continental regime has a very weak coarse mode and an enhanced accumulation mode. Furthermore, the city-influenced maritime spectra exhibits a hybrid shape to the continental and maritime regimes, in that like the maritime regime it has a strong fine mode and slightly stronger coarse mode, but also has an enhanced accumulation mode more like its continental counterpart and the fine mode is even enhanced compared to the background maritime spectra.

[1] Due to the small sample volume of the FSSP and extremely low concentrations of large particles, there is considerable noise in the coarse mode end of these spectra.

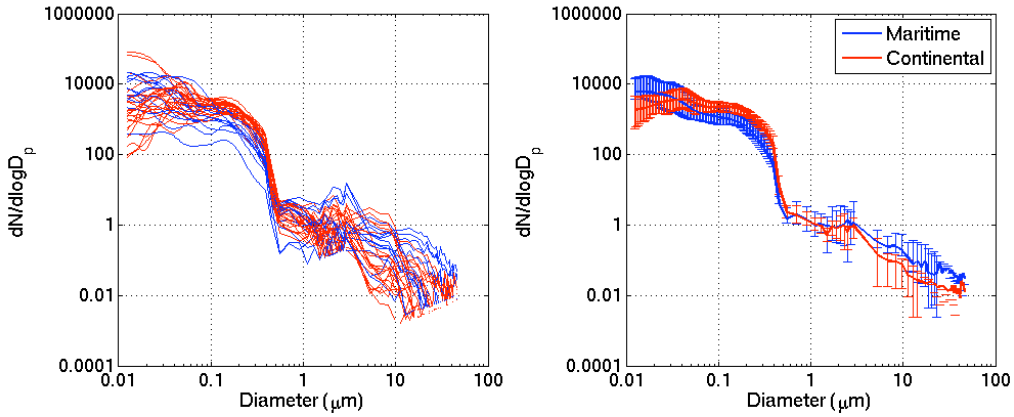


Figure 8. Aerosol size spectra for the maritime (blue) and continental (red) regimes (left) and the mean spectra for each regime (right). Whiskers denote the range of 25th and 75th percentile for each regime (right).

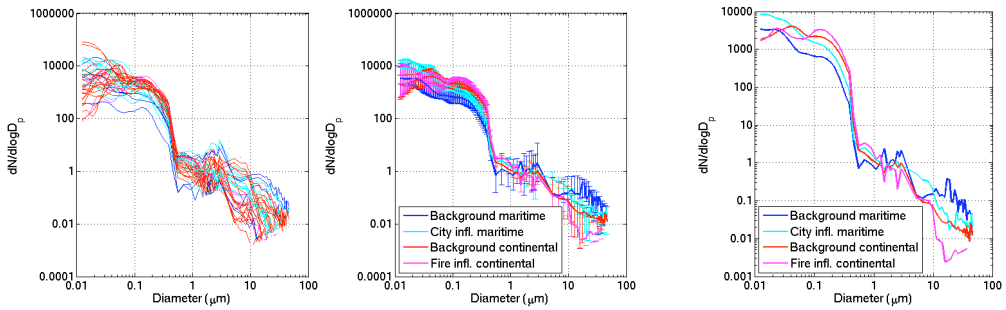


Figure 9. Same as Figure 8, except for the four sub-regimes (see color legend), and the mean spectra without whiskers have been included (far right) for clarity.

b. Cloud microphysics

Initial ("cloud base") maximum droplet concentrations measured 300-600m above cloud base were very similar on average in both the maritime and continental regimes (Table 4). This result was peculiarly surprising given the marked differences in aerosol and CCN concentrations observed between the two regimes. The cloud base droplet spectra, however, did exhibit some slight differences in that on average the maritime spectra were generally broader due to a tail of larger droplets (Figure 10). This could be the result of droplet nucleation on larger aerosol particles (exhibited in the coarse mode of Figure 8).

Table 4. Average cloud droplet concentrations and mean diameter statistics by regime.

Regime	Drop Conc (/cc)	Std Dev	Mean Diam (μm)	Std Dev
Maritime	431	138	12.1	1.6
Continental	423	100	11.1	0.7

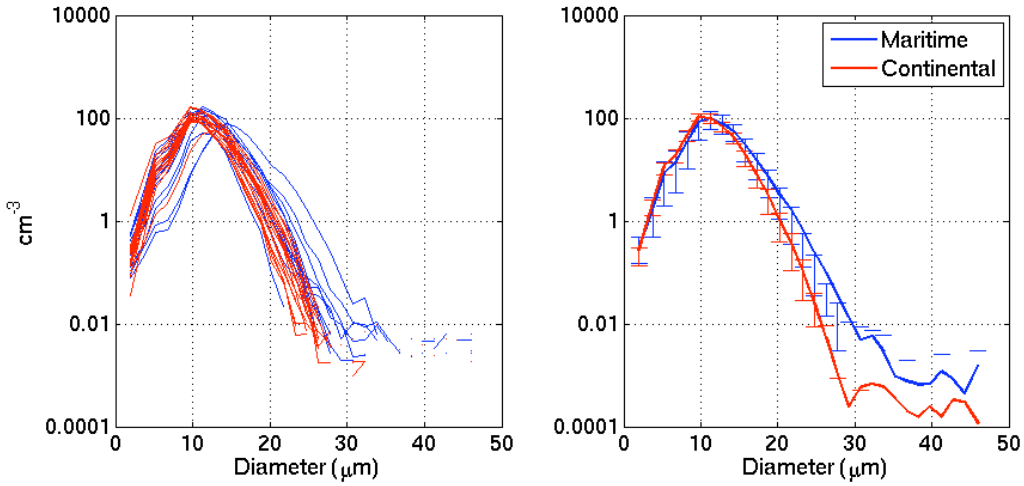


Figure 10. Same as Figure 8, except of cloud droplet spectra measured at 300-600 m above cloud base.

c. Thermodynamic environments

In addition to differences in aerosol, there may be inherent environmental differences between these regimes that could affect cloud microphysical processes. Here we present a few factors, such as moisture and instability, to investigate this possibility (Figure 11). Cloud base heights were consistently lower in the maritime regime, suggesting more humid surface conditions, although the cloud base temperatures were quite similar between the two regimes on average. Precipitable water was also fairly similar between the two regimes, on average, although the continental regime exhibited variability that extended to colder cloud base temperatures and slightly higher precipitable water. Instability is often measured by Convective Available Potential Energy (CAPE), however CAPE is highly dependent on surface conditions that can temporally and spatially vary. Given the 00Z sounding is taken at 10:00am local time, it was often the case that surface conditions measured by the sounding were not those reflected at the time of convection. Thus, here we used the Total Totals stability index (Miller 1972), which is calculated as

$$\text{Total Totals} = t_{d850} + t_{850} - 2 \times t_{500}$$

where t_{d850} is the dew point at 850 hPa, and t_{850} (t_{500}) is the temperature at 850 (500) hPa, and higher values indicate increased instability. Figure 11d indicates that both regimes have similar mean instability using this index. The continental regime, however, does exhibit a larger range of variability extending to slightly higher values of instability.

5. SUMMARY

The goal of this study was to document representative cloud base aerosol conditions in the southeast Queensland region, and investigate the relationship between the cloud base aerosol and initial cloud droplet spectra to establish implications of the aerosol on the cloud microphysics. Such representations could then be used to initialize models to further study potential aerosol impacts on precipitation formation in the region.

Two general regimes were identified with maritime versus continental influences based on the hours each measurement's back trajectory spent over land. However, within each of these two regimes there was still substantial variability in aerosol concentration that was determined to be

related to other factors, such as proximity to Brisbane or proximity to fires. Regardless, some outlier measurements are still not being explained by these factors, and thus additional analysis is currently underway to diagnose additional factors that may be necessary in order for our regime classifications to accurately predict aerosol conditions. Preliminary findings suggest revising the time a trajectory spent over land to be the time the trajectory spent over land *within the boundary layer* may be a better predictor than simply time over land. Future analysis will pursue this possibility, as well as develop a regression model to predict aerosol concentrations based on the factors that are determined to be most influential. Such a model could be used for future radar analysis and cloud seeding operations in the region, in the absence of in situ aerosol measurements. Representative aerosol and cloud droplet spectra will also be quantified for each regime in the model for use in initializing and validating cloud resolving models. Finally, additional analysis is underway to better understand the surprising result that both the maritime and continental regimes had similar initial droplet concentrations despite having significantly different aerosol concentrations. Preliminary analysis suggests that perhaps the maritime regime may have inherently achieved a higher supersaturation, given the consistently lower cloud base heights, which would allow for more of the aerosol particles to activate and form droplets raising the droplet concentrations to that of the continental regime.

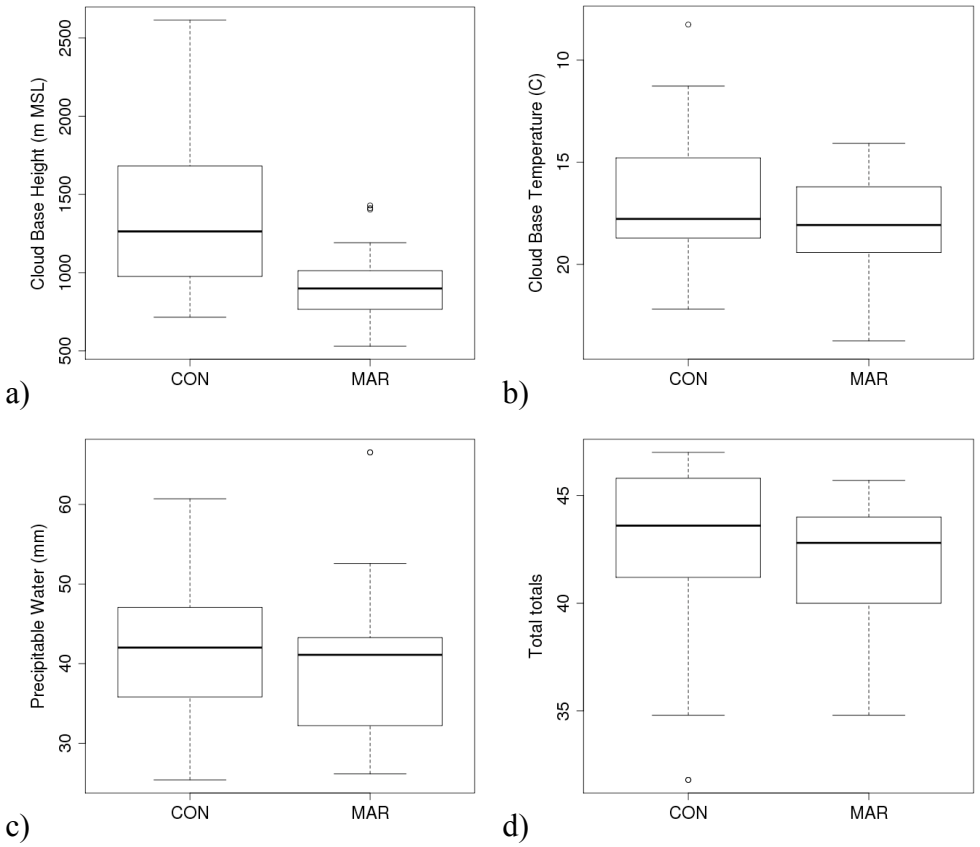


Figure 11. Box and whiskers plots of (a) cloud base height, (b) cloud base temperature, (c) precipitable water, and (d) the Total Totals stability index from 00Z Brisbane airport soundings on the day of each measurement.

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