#### 747 CLOUD BASE AEROSOL CHARACTERISTICS AND IMPLICATIONS FOR MICROPHYSICS IN SOUTHEAST QUEENSLAND

Sarah A. Tessendorf\*, Courtney E. Weeks, Roelof T. Bruintjes, Duncan Axisa

National Center for Atmospheric Research, Boulder, CO

### 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 aerosolinfluenced 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 potentially precipitationof 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 This paper presents such in situ established. aerosol measurements, for the southeast Queensland region, that were collected during the Queensland Cloud Seeding Research Program (QCSRP; Tessendorf et al. 2010, 2011).

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.

### 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 bequipped 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 a Differential Mobility Analyzer (DMA; Stolzenburg et al. 1998), accumulation mode aerosol were measured by a Passive Cavity Aerosol Spectrometer Probe (PCASP-100X), and when flying out of cloud a Forward Scattering Spectrometer Probe (FSSP; Dve 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) since it was primarily used to cloud droplet spectra in cloud. measure Furthermore, FSSP instruments measure particles at ambient relative humidity, and depending upon the hygroscopicity of the observed particles may

<sup>&</sup>lt;sup>\*</sup> *Corresponding Author:* Sarah A. Tessendorf, NCAR/RAL, P.O. Box 3000, Boulder, CO 80307. Email: saraht@ucar.edu

measure larger sizes than the PCASP and DMA instruments that measure dry particle sizes. The PCASP and FSSP were calibrated using latex spheres and glass beads, respectively, of known size and refractive index, and each was operated with 30 separate size bins utilizing the Signal Processing Package (SPP) electronics (SPP-100 for the FSSP and SPP-200 for the PCASP) from Droplet Measurement Technologies (DMT).

A DMT single column Cloud Condensation Nuclei (CCN-100) counter (Roberts and Nenes 2005) was used to sample the CCN concentration. A constant pressure controller unit was utilized to keep the CCN sample at a constant pressure of approximately 600 hPa throughout the flight. Measured CCN concentrations were then corrected to ambient pressure (Roberts and Nenes 2005). 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. The CIP and PIP were calibrated using their respective spinning-disk calibration tools.

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.

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<sup>-3</sup>, and excluded measurements with any particles measured larger than 100 µm (as measured by 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 was the highest resolution set available for HYSPLIT that covered the project domain at the time of the measurements<sup>1</sup>. The starting latitude, longitude, and altitude for each trajectory was based on the location of aircraft while actual taking measurements. The back trajectories, shown in Fig. 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 below 2 km: maritime regime <= 12 hrs over land below 2 km (blue) and continental regime > 12 hrs (red).

<sup>&</sup>lt;sup>1</sup> Sensitivity tests of trajectories calculated using GDAS and other data sets (i.e. NCEP/NCAR Reanalysis and FNL) for the same region in past years when those other data sets were available yielded highly similar results. Thus, we feel the GDAS was the most appropriate and available data set for the present study.



Figure 2. Sensitivity tests showing median PCASP concentration vs. trajectory hours over land beneath 2 km as calculated using the a) GDAS data set using modeled wind fields (as presented in rest of paper), b) NCEP/NCAR Global Reanalysis Data using modeled wind fields, and c) GDAS data using isentropically modeled vertical motion.

Since the aerosol characterization herein relies on the results of the HYSPLIT model run with GDAS data, a sensitivity study was performed to test the influence of the model data resolution and vertical motion calculation on the results. We recreated the results using the NCEP/NCAR Global Reanalysis Data (spatial resolution of 2.5 x 2.5 degrees and 6-hour output) with all other conditions held the same, and again with the GDAS data but computing vertical motion differently (isentropically rather than from modeled wind fields). Relationships of median PCASP concentration and hours over land, as calculated from these test runs, are shown in Fig. 2. The 12hour threshold determined in the original analysis lies within the 95% confidence intervals for the most significant regime threshold determined by the reanalysis data and the isentropic vertical motion runs. However, most notably from the lower resolution data run (Fig. 2b), there are a few outlier points that would be misclassified.

### 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 within the boundary layer (estimated at 2 km; hereafter, "hours 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 <=12 hours over land, and continental, with >12 hours over land (see Fig. 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 (Fig. 3a).



Figure 3. Box and whisker plots of (a) median PCASP aerosol concentration and (b) median 0.3% SS CCN concentration split by regime (MAR: <=12 hours over land below 2 km; CON: >12 hours over land below 2 km).

These two regimes also exhibit distinctly different CCN concentrations, both at 0.3% supersaturation (Fig. 3b) and across all supersaturations, on those measurements average, for with supersaturation cycles (Fig. 4). 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 (Fig. 5).

### 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 the MODIS Fire and Thermal Anomalies Product (Giglio et al. 2003) and processed as in Wiedinmyer et al. (2006). The number of fires along a trajectory (hereafter, proximity fires) was then counted by accumulating the number of MODIS fire locations within a 200 km radius of the trajectory at each hour along the 120-hr back trajectory. This method may count the same fire multiple times, which we feel is acceptable as it implies that it had more time to influence the air mass. Rainfall was calculated from the GDAS model and accumulated along the last 12 hours of the trajectory.



Figure 4. 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 5. 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.

For the maritime air masses, there was little relationship between aerosol concentrations and hours over land (Fig. 6a), however the aerosol concentrations exhibited a considerable increase as the trajectories get closer to Brisbane (Fig. 6b). 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 (Fig. 7). November and December experienced far more fires, less rainfall along the trajectory prior to measurement, and generally more continental flow while the second half of the rainy season was more maritime in nature, with fewer fires and more rainfall along the trajectories. We expect that the fires in November and December were 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 were highly dependent on the number of fires observed along the trajectory (Fig. 6c), while showing less dependence on hours over land and no relationship with distance to Brisbane (Figs. 6a-b). The lack of dependence on distance to Brisbane in the continental flow is unsurprising, given that the measurements made in continental flow were inland from Brisbane and were more often made upwind of the city.

Figure 6d illustrates how both regimes' aerosol concentrations were affected by recent rainfall. While noisy, the trend suggests that the concentrations will be lower with greater rainfall, as would be expected due to precipitation scavenging (e.g., Kerker and Hampl 1974, Barlow and Latham 1983).

The trajectory and locations of fires throughout the 120-hour duration of the trajectory for examples of two extremes in aerosol concentration are illustrated in Figure 8. Figure 8a highlights the measurement with the highest PCASP aerosol concentration (1610 cm⁻³), measured on 6 December 2008, and in Fig. 8b for the cleanest case (PCASP aerosol concentration 73 cm<sup>-3</sup>) measured on 14 February 2009. The 6 December case spent 51 hours over land, was in proximity to nearly 2436 fires throughout its 120-hr path, and had very little (1.2 mm) rainfall to scavenge aerosol prior to the measurement. The 14 February trajectory, on the other hand, spent only 4 hours over land, did not come closer than 50 km to Brisbane, and had nearly 12 mm of rainfall in the previous 12 hours before the measurement.



Figure 6. PCASP aerosol concentration variation based on the a) hours over land below 2 km, 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 7. Monthly trends in concentration by hours over land below 2 km, rainfall, and number of fires (N= November, D=December, J=January, F=February).



Figure 8. Trajectories corresponding to (a) the highest (6 December 2008) and (b) lowest (14 Febraury 2009) PCASP aerosol concentrations. Red asterisks represent the locations of fires within the 120-hr trajectory period. The light gray shaded region represents a 50 km radius around the Brisbane city center, for reference.

Based on these additional influences on aerosol concentration, each regime was broken into two sub-regimes (Fig. 9): a city-influenced maritime regime was defined for trajectories with <= 12 hrs over land below 2 km and at least one time step along the trajectory that was < 50 km to Brisbane; and a fire-influenced continental regime was define for trajectories with >12 hrs over land below 2 km and >1000 proximity fires. The remaining measurements in each of the maritime and continental regimes were then included in respective "background<sup>2</sup>" sub-regimes.

The mean aerosol concentrations 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<sup>3</sup> with equal means and equal, but unknown, variances.

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.



Figure 9. 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).

Furthermore, splitting the PCASP data into the maritime and continental regimes reduced the overall variability in measured aerosol concentration (measured as a sum-squared deviation) by 58.5%. Splitting into sub-regimes reduced the variability in the maritime and continental regimes by another 39.8%. The sum-

<sup>&</sup>lt;sup>2</sup> The use of the term background here refers to the remainder of the measurements that did not meet the sub-regime criteria, and does not imply these remaining measurements were "remote" background measurements in each of the maritime or continental regimes.

<sup>&</sup>lt;sup>3</sup> The assumption of normality of PCASP concentrations within each regime was confirmed with the Lilliefors test (Lilliefors 1967).

squared deviations for each of the sub-regimes (Table 3) indicate that the two maritime sub-regimes have very minimal variability.

### b. Regression model

One goal of this characterization analysis is to be able to predict general aerosol concentrations, in the absence of in situ aerosol measurements, with a tool that is readily available (e.g., HYSPLIT is open source). Based upon this analysis, various features related to the back trajectory for a given measurement could be used to predict the resulting aerosol concentration. Therefore, PCASP concentrations were regressed on the most relevant trajectory "predictor" variables (hours over land below 2 km, distance to Brisbane, number of fires, and prior 12-hr accumulated rainfall). Two models were created, and will be discussed below.

As a first pass we built a regression model that treated the data as if it were more homogeneous, even though we've seen that the maritime- and continental-influenced flows have different predictor relationships to the variables. Nonetheless, with this method, the following model produced to estimate the PCASP was concentration:

### PCASP\_est = 361.10 + 4.01\*HOLBL + 0.28\*Fires

where "Fires" is the number of proximity fires, and "HOLBL" is the hours the trajectory spent over land below 2 km. In this case, neither distance to Brisbane nor recent rainfall was a significant predictor of the final aerosol concentration. The adjusted  $R^2$  for this model was 0.66.

Based on the results of the aerosol characterization analysis, however, we do expect that the maritime and continental regimes may have different significant predictors. Thus, we created a model that took the maritime versus continental regime classification into account. This model produced the following relationship to estimate PCASP concentration:

PCASP\_est = 544.49 + C\*(0.24\*Fires + 2.41\*HOLBL) – M\*(3.02\*DTB – 22.60\*Rain)

where C = 1 (and M = 0) if HOLBL > 12 hrs (continental) and M = 1 (and C = 0) if HOLBL <= 12 hrs. In this relationship, "DTB" is the distance to Brisbane city center, and "Rain" is the amount of modeled rainfall in the 12-hr period prior to measurement time. The adjusted  $R^2$  for this model was higher, as we expected, at 0.71.

# 4 REGIME CHARACTERISTICS

### a. Thermodynamic environment

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 (Fig. 10). Cloud base heights were consistently lower in the maritime regime, suggesting more humid surface conditions, although the cloud base temperatures were guite 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

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. Fig. 10d 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.

# b. Aerosol characteristics

Aside from the significantly different PCASP (accumulation mode) aerosol concentrations discussed in Section 3 that were 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<sup>4</sup> (Fig. 11). Despite substantial variability in the measurements, the mean spectra highlight the

<sup>&</sup>lt;sup>4</sup> 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 10. 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.



Figure 11. Aerosol size spectra for the maritime (blue) and continental (red) regimes (a) and the mean spectra for each regime (b-c). Whiskers denote the range of  $25^{th}$  and  $75^{th}$  percentile for each regime in (b), but have been removed for clarity in (c).



Figure 12. Same as Fig. 11, except for the four sub-regimes (see color legend).

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 (Fig. 12), some of the variability exhibited in Figure 11 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 cityinfluenced 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.

# c. Cloud microphysics

Initial ("cloud base") maximum droplet concentrations measured 300-600m above cloud base should in theory be correlated with the aerosol that serve as CCN found at cloud base if they enter the cloud and become activated. However, taking in situ droplet measurements in cloud just above cloud base has several caveats. making it difficult to isolate the region of the cloud would expect where vou the droplet measurements to be most related to the sub-cloud aerosol. Such issues are related to entrainment of cloud-free air, which dilutes the cloud water in the parcel below adiabatic conditions and typically affects cloud edges; drizzle and precipitation formation that may be falling from above, which may artificially broaden the drop spectra; and varying supersaturations in the cloud, in part due to varying updraft velocities, will also affect the number and sizes of particles that are activated. As described in Section 2, we have done some processing of the data to omit measurements that may have been affected by precipitation (defined as measurements having particles > 100 µm on the CIP and PIP probes). Nonetheless, there are still several methods for characterizing the droplet concentrations in clouds, and each can vield different results (Yum et al. 1998, Hudson and Yum 2001, 2002). For example, one simple method is to calculate flight-averaged droplet concentrations, as has been done in some aerosol characterization studies, however the cloud edge measurements will act to dilute the mean and would not provide the best measurements with which to compare the aerosol observations for the purposes of the present study. Using the

maximum droplet concentration should perform better, however, absolute maximum measurements can be the result of artificial spikes in the data.

Despite these challenges, we have attempted three different methodologies for calculating the typical droplet concentration, mean diameter, and size spectra just above cloud base that could result from nucleation on the aerosol particles we measured below cloud base. Each method is described below, followed by a discussion of the results. As one may expect, each yielded different results, and we cannot speculate which is more correct, rather they are all possible scenarios that need to be considered.

# i. Steady-state method

This method is the most subjective of the three we utilized. Here we based the method on the principle that when the aircraft is well inside the cloud and the near adiabatic core of the cloud, the liquid water content and drop concentrations should be fairly steady (for a *minimum* of 3 seconds, which translates to roughly 250 m in distance covered by the aircraft). Thus, these periods of level drop measurements were noted and means of the concentrations, mean diameter, and size spectra over the course of these steady periods were calculated for each cloud penetration.

# ii. Maximum drop concentration method

As stated earlier, in theory the maximum droplet concentration should be related to the sub-cloud aerosol. However, spikes that may occur in the 1-Hz data prevent this correlation from always being the case. Thus, we implemented an objective strategy that calculated the maximum droplet concentration in a given penetration, and then took a mean of the concentration, mean diameter, and size spectra over the course of a 3-second period; plus or minus one second from the 1-Hz maximum measurement. This method ideally helped to smooth out spikes, but some the 3-second averages may still have some influence from data spikes.

# iii. "Binmax" method

This method was presented in Yum et al. (1998) as an objective way to identify the near adiabatic droplet concentrations in cloud passes, while reducing the chances of contamination from spikes in the data. In their study, this method had high correlations of droplet and CCN concentrations. All 1-Hz droplet concentration measurements during passes through a particular cloud<sup>5</sup> were binned into 10 cm<sup>-3</sup> frequency distribution bins of droplet concentration. The data points in the bin with the highest concentration that also contained at least three 1-Hz data points were then used to calculate the droplet characteristics for that particular cloud. The droplet concentrations, mean diameters, and drop size spectra were then averaged from those data points in that maximum bin and used to represent the drop characteristics for the given cloud.

### iv. Cloud base droplet characteristics

Droplet concentration and mean diameter results using the steady-state and maximum methods were very similar on average in both the maritime and continental regimes (Table 4), with the maximum method resulting in the highest drop concentration measurements, as may be expected. The similarities between the two regimes were peculiarly surprising given the marked differences in aerosol and CCN concentrations that had been observed. The "binmax" method yielded the most notable differences between the two regimes, in a manner that would be expected given the lower (higher) aerosol concentrations and thus lower (higher) droplet concentrations in the maritime (continental) regime (Table 4). Mean droplet diameters were also larger in the maritime regime than the continental regime, which is also expected given the lower droplet concentrations and slightly broader aerosol size distributions in the maritime regime.

Despite the similarities in droplet concentration between the regimes using the steady-state and maximum methods, the cloud base droplet spectra did exhibit some slight differences in that, on average, the maritime spectra were generally broader due to a tail of larger droplets (Fig. 13). This was also the case using the binmax method. This result was most notable in the steady-state and binmax methods, in which the difference in the concentration of drops larger than 20 µm was statistically significant between the two regimes for both of these methods. The mean diameter was significantly different between the regimes (i.e. larger in the maritime regime) under the binmax method only. This tendency for a tail of larger drops in the maritime regime could be the result of

droplet nucleation on larger aerosol particles (exhibited in the coarse mode of Fig. 11).



Figure 13. Mean cloud droplet spectra measured at 300-600 m above cloud base for each regime (continental = red, maritime = blue) and each calculation method (solid lines for binmax, dashed for steady-state, and solid with dots for the max method).

#### 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. Furthermore from these measurements, regression model а was developed that could be used for future radar analysis and cloud seeding operations in the region, in the absence of in situ aerosol measurements, to give an estimate of what the (accumulation mode) aerosol concentration would be on a given day. Representative mean aerosol and cloud droplet spectra were also presented for each regime, which could be used as guidance in initializing and validating cloud resolving models, especially those with detailed (bin) microphysics.

<sup>&</sup>lt;sup>5</sup> In Yum et al. (1998) they included all cloud droplet measurements over a particular flight.

The thermodynamic environments of the continental and maritime regimes were guite similar in instability and total precipitable water, however the most notable difference was in the cloud base heights, which were consistently lower in the maritime regime. This environmental influence on the clouds that formed under the maritime regime could have contributed to the higher concentrations of coarse mode aerosol particles by nature of the measurements being taken closer to the ground surface. Our initial cloud base droplet spectra did often tend to have a tail of larger drops in the maritime regime compared to the continental regime, which may be the result of the differing aerosol conditions. By having defined these aerosol regimes in a quantitative way, we hope that future observational studies in the region can further diagnose the potential effects of the aerosol and that numerical modeling studies can help fill in the gaps where our observations are incomplete and unable to control for aerosol differences alone.

Acknowledgements: This research was sponsored by the Queensland Government Department of Environment and Resource Management through the Queensland Climate Change Centre of Excellence. The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model used in this publication. MODIS fire data was processed and provided by Christine Wiedinmyer (NCAR).

### REFERENCES

- Barlow, A.K. and J. Latham, 1983: A laboratory study of the scavenging of sub-micron aerosol by charged raindrops. *Quart. J. Royal. Met. Soc.*, **109**, 763–770
- Baumgardner, D., H. Jonsson, W. Dawson, D. O'Connor and R. Newton, 2001: The cloud, aerosol and precipitation spectrometer (CAPS): A new instrument for cloud investigations, *Atmos. Res.*, **59-60**, 251-264.
- Bigg, E.K., 2008: Trends in rainfall associated with sources of air pollution. *Environ. Chem.*, **5**, 184-193. doi:10.1071/EN07086.
- Bruintjes, R. T., 1999: A review of cloud seeding experiments to enhance precipitation and some new prospects, *Bull. Amer. Meteorol. Soc.*, **80**, 805–820.
- Chen, J.-P., 1994. Predictions of saturation ratio for cloud microphysical models. J. Atmos. Sci. 51, 1332–1338.

- Clark, T.L., 1974. On the modeling nucleation and condensation theory in Eulerian spatial domain. *J. Atmos. Sci.* **31**, 2099–2117.
- Draxler, R.R. and Rolph, G.D., 2010: HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model access via NOAA ARL READY Website (http://ready.arl.noaa.gov/HYSPLIT.php). NOAA Air Resources Laboratory, Silver Spring, MD.
- Dye, J.E. and D. Baumgardner, 1984: Evaluation of the forward scattering spectrometer probe:
  I. Electronic and optical studies. *J. Atmos. Ocean. Tech.*, 1, 329-344.
- Giglio, L., J. Descloitres, C.O. Justice, Y.J. Kaufman, 2003: An enhanced contextual fire detection algorithm for MODIS. *Remote Sens. Environ.*, **87**, 273-282.
- IPCC, 2007: Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change [Solomon, S., D. Qin, M. Manning, Z. Chen, M. Marquis, K.B. Averyt, M.Tignor and H.L. Miller (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.
- Kerker, M., and V. Hampl, 1974: Scavenging of aerosol particles by a falling water drop and calculation of washout coefficients. *J. Atmos. Sci.*, **31**, 1368-1376.
- Kutner, M., C. Nachtsheim, J. Neter, W. Li, 2005: Applied Linear Statistical Models, McGraw-Hill, New York, NY, 1396 pp.
- Lilliefors, H., 1967: On the Kolmogorov–Smirnov test for normality with mean and variance unknown, Journal of the American Statistical Association, Vol. 62. pp. 399–402.
- Miller, R.C., 1972: Notes on analysis and severe storm forecasting procedures of the Air Force Global Weather Central. *Tech. Rept. 200(R)*, Headquarters, Air Weather Service, USAF, 190 pp.
- Roberts, G.C., and A. Nenes, 2005: A continuousflow streamwise thermal-gradient CCN chamber for atmospheric measurements. *Aerosol Sci. and Tech.*, **3**, 206-221.
- Rosenfeld, D., 1999: TRMM observed first direct evidence of smoke from forest fires inhibiting rainfall. *Geophys. Res. Lett.*, **26**, 3105-3108.
- Rosenfeld, D., 2000: Suppression of rain and snow by urban and industrial air pollution. *Science*, **287**, 1793-1796.
- Rosenfeld, D., U. Lohmann, G.B. Raga, C.D. O'Dowd, M. Kulmala, S. Fuzzi, A. Reissell, and M.O. Andreae, 2008: Flood or drought:

How do aerosols affect precipitation? *Science*, **321**, 1309-1313.

- Silverman,B. A., 2003: A critical assessment of hygroscopic seeding of convective clouds for rainfall enhancement. *Bull. Amer. Meteor. Soc.*, **84**, 1219-1230.
- Squires, P., 1958: The microstructure and colloidal stability of warm clouds. I. The relation between structure and stability, *Tellus*, **10**, 256-271.
- Stolzenburg, M., N. Kreisberg, and S. Hering, 1998: Atmospheric size distributions measured by differential mobility optical particle size spectrometry. *Aerosol Sci. and Tech.*, **51**, 402-418.
- Tao, W.-K., X. Li, A. Khain, T. Matsui, S. Lang, and J. Simpson, 2007: Role of atmospheric aerosol concentration on deep convective precipitation: Cloud-resolving model simulations. J. Geophys. Res., 112, doi 10.1029/2007JD008728.
- Tessendorf, S.A., and Co-authors, 2010: Overview of the Queensland Cloud Seeding Research Program. *J. Wea. Modification*, **42**, 33–48.

- Tessendorf, S.A., and Co-authors, 2011: The Queensland Cloud Seeding Research Program. *Bull. Amer. Meteor. Soc.*, in review.
- Twomey, S., 1959: The nuclei of natural cloud formation-Part II: The supersaturation in natural clouds and the variation of cloud droplet concentration. *Geofis. Pura Appl.*, **43**, 243-249.
- Twomey, S., 1977: *Atmospheric Aerosols*. Elsevier, 302 pp.
- Warner, J., 1967: A reduction in rainfall associated with smoke from sugar-cane fires—An inadvertent weather modification? *J. Appl. Meteor.*, **7**, 247-251.
- Warner, J., and S. Twomey, 1967: The production of cloud nuclei by cane fires and the effect on cloud droplet concentration. *J. Atmos. Sci.*, 24, 704-706.
- Wiedinmyer, C., B. Quayle, C. Geron, A. Belote, D. McKenzie, X. Zhang, S. O'Neill, and K.K. Wynne, 2006: Estimating emissions from fires in North America for Air Quality Modeling. *Atmospheric Environment*, **40**, 3419-3432.

### TABLES

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

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).

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

Regime	Sub-regime	Mean PCASP Concentration (cm-3)	
Maritime	All		319
	Background		237.9
	City-influenced		386.7
Continental	All		786.6
	Background		670.2
	Fire-influenced		1097

	BG Mar	City Mar	BG Con	Fire Con
BG Mar	1.0	0.008	2.04E-08	1.55E-06
City Mar		1.0	3.99E-07	2.59E-06
BG Con			1.0	1.67E-04
Fire Con				1.0

Table 3. P-values from the t-test for each pair of subregime means.

Table 4. Average cloud droplet concentrations ("Conc"), mean diameter ("Diam"), and concentrations of droplets greater than 20  $\mu$ m ("Conc>20") by regime for each of the three calculation methods. Standard deviations ("StdDev") and p-values ("P") are also provided.

Method	Regime	Conc	StdDev	Р	Diam	StdDev	Р	Conc>20	StdDev	Р
Steady- state	MAR	411	135	.94	12.1	1.7	.13	5.9	7.8	.03
	CON	431	111		11.1	0.8		1.7	1.5	
Maximum	MAR	596	62	20	11.1	1.2	E۸	4.0	5.2	65
	CON	534	113	.30	11.1	0.8	.04	2.6	3.6	.05
BinMax	MAR	353	207	41	12.8	2.3	01	7.9	9.0	00
	CON	414	154	.41	10.9	1.6	.01	3.4	7.7	.00