THE VARIABILITY OF CLOUD CONDENSATION NUCLEI AND CLOUD DROPLET POPULATIONS IN CONVECTIVE CLOUDS OVER THE HIGH PLAINS: HOW OFTEN ARE CONTINENTAL CLOUDS CONTINENTAL? NEW CCN OBSERVATIONS

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Abstract: Daily observations of cloud condensation nuclei (CCN) concentrations active at 0.4% supersaturation were conducted at the surface near mid-day for 3 weeks in August, 2004, at Bowman, ND. Average concentrations of these CCN over ~30 minute-long sampling periods varied day-to-day from ~30 to ~200 cm^{-3}. Preliminary analysis of weather conditions and 3-day back-trajectories associated with these measurements suggests that air reaching Bowman after recent association (within prior two days) with precipitating clouds has generally lower CCN concentrations, while air that has not been recently associated with precipitation systems is more likely to have relatively higher CCN concentrations. Air from the west typically had the highest concentrations, while air from the north typically was the cleanest, other factors being equal.

1. INTRODUCTION

The cloud droplet concentration and size spectrum resulting from the activation of CCN in summertime convective cloud updrafts influences the mode by which, and speed with which, precipitation forms. When fewer CCN are present, the droplet concentration nucleating and growing in the updrafts of convective clouds is smaller, the droplet size spectrum broader, and the droplet mean size larger. This leads to more rapid collision and coalescence of cloud droplets to form drizzle and rain drops. When these drops are carried above the freezing level they freeze at relatively small supercoolings and this leads to the development of graupel at relatively lower levels in the cloud, resulting in earlier ice-phase precipitation. When higher concentrations of CCN are present, the resulting cloud droplet concentration is higher, the size spectrum is narrower, and the mean size smaller. Collision and coalescence operates much more slowly, rain drop formation is inhibited, and cloud droplets are lofted well above the freezing level before they begin to freeze, followed by growth to graupel by aggregation and riming. Precipitation formation occurs later and higher in the cloud. When low concentrations of active CCN are present, the resulting clouds are characterized as microphysically “maritime”, while if high concentrations of active CCN are present, the resulting clouds are commonly characterized as “continental”, based on characteristic CCN concentrations observed over the ocean and over continents.

Mather *et al.* (1997) proposed that if appropriate concentrations of smaller (~1 μm diameter) hygroscopic nuclei capable of nucleating cloud droplets at small supersaturations are introduced into convective cloud updrafts, they will readily nucleate droplets. These droplets will start to grow and deplete the excess vapor over saturation before sufficient supersaturation is reached in the updraft for nucleating the much higher concentrations of natural CCN active at higher supersaturations. If done correctly, this process could render a cloud that would naturally be continental into a more maritime microphysical state. Maritime clouds are more efficient precipitators due to their broader droplet size distribution with larger mean size, accelerating precipitation growth by droplet coalescence. Thus Mather and colleagues hypothesized that this hygroscopic seeding would increase precipitation on the ground from treated convective clouds. Silverman (2003) summarizes some of the initial work, both theoretical and experimental, that followed to investigate this possibility. He concludes that experimental results are suggestive, but not conclusive.

More recently, modeling studies by Seifert and Beheng (2005) and van den Heever *et al.* (2005) in which environmental conditions vary, suggest that in some convective environments more vigorous storms and larger storm precipitation totals may occur with continental CCN populations, and in other environments greater precipitation occurs with maritime CCN populations. Reasons for these differences are complex and still under investigation.

It often is taken for granted that near-surface air in interior continental regions is characterized by high cloud condensation nuclei (CCN) concentrations, i.e. many hundreds to thousands per cubic centimeter active near 1% supersaturation. Compilations of CCN observations from many locations (e.g. Fletcher, 1969, p. 101; Gotz *et al.,* 1991, pp. 98-99; Pruppacher and Klett, 1997, pp. 288-290) often show these characteristic differences between continental and maritime CCN populations. Such tables should be taken to represent typical conditions, much as a table of climatological average temperatures and rainfall might be used to characterize average conditions in various locations. When a series of CCN observations is made at one site, or in one region, for extended periods of time, a common result is that CCN concentrations vary between maritime (low) and continental (high) levels from day-to-day with changes in the air masses overlying the site or region. Hobbs *et al.* (1985) conducted an extensive series of airborne observations in the summertime boundary layer over the U.S. High Plains in the summers of 1975 and 1976. They observed that even in this continental interior region roughly 25% of the time the CCN population had distinctly maritime characteristics. Vali *et al.* (1982) present a two-month sequence of CCN observations made at the surface at two sites, one in northeast Colorado and the other in southwest Nebraska, separated by 60 km, in the summer of 1976. In this case, CCN concentrations were more maritime than continental on ~10% of all days. Detwiler (2004) examined cloud droplet spectra in clouds sampled during three convective storm field seasons on the High Plains, and found more maritime microphysical characteristics on ~25% of those days.

It is clear that practitioners of hygroscopic seeding, seeking to render a CCN population more maritime, need to be aware of the background CCN population, and other environmental characteristics, in order to target their efforts for situations where the seeding will have its intended effect. In pursuit of this goal, sampling of CCN characteristics was conducted at Bowman, North Dakota, during the summer of 2004 to learn more about natural CCN population characteristics in this region where there is potential interest in hygroscopic seeding.

2. INSTRUMENTATION

Concentrations of CCN were determined using an instrument based on the classic parallel-plate thermal diffusion chamber design. (See, e.g., Fletcher, 1969, pp. 81-82; Gotz *et al*., 1991, pp. 93-95.) The instruments used were commercially manufactured by Mee Industries in the early 1970's. In the design used by Mee, the top plate is at ambient temperature and the bottom plate is cooled thermoelectrically. Sampling is done by drawing a sample into the chamber, closing off the chamber and allowing equilibrium to be established, and then observing the droplet concentrations that result. The primary instrument, from the South Dakota School of Mines and Technology (SDSMT), was extensively refurbished by Mr. Brian Jesse, an associate of the Cloud and Aerosol Simulation Laboratory, Dept. of Atmospheric Sciences, Colorado State University (CSU). A video camera was installed to view nucleation of water vapor to droplets in the cloud chamber. A computer system was developed by Mr. Mike Pridgeon of SDSMT to record the video frames of cloud formation for later analysis. A view of the instrument is shown in Figure 1.

Figure 1: A view, looking down on the open case of the SDSMT Mee CCN counter. The cloud chamber is open, with the upper plate lying to the right, and the base of the chamber in the center left portion of the image. The blotter-paper-covered inner plate surfaces are 10 cm in diameter, and the spacing between the plates, when the chamber is assembled, is 1 cm. The video camera in the upper left of the picture looks into a mirror set at a 45^o angle to allow a view of the inner chamber. The white plastic bottle in the upper right of the image is the water reservoir for the system used to keep the inner plate surfaces moist.

The refurbished instrument was tested in the laboratory at CSU using artificially-generated ammonium sulfate aerosol with very narrow size distributions and a selectable mean size. Theory can be used to calculate the supersaturation at which ammonium sulfate aerosols of a given size will nucleate (e.g. Pruppacher and Klett, 1997, Ch. 6). The theoretical supersaturation at which particles of a given size nucleated versus measured temperature difference (ΔT) determined during these tests, using various ammonium sulfate particle mean sizes, is shown in Figure

2. Also shown is a family of supersaturation vs. ΔT curves for various chamber top plate temperatures computed by the manufacturer based on pure vapor/thermal diffusion theory. The supersaturations estimated by Mee based on classic diffusion theory, for a given ΔT, are seen to be almost a factor of two higher than supersaturations calculated based on the particle sizes activated under those conditions in our tests.

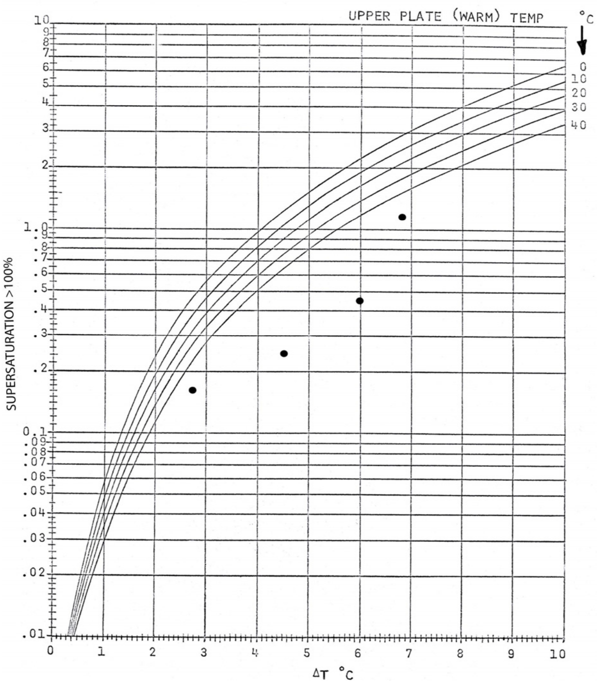

Figure 2: Peak supersaturation as a function of temperature difference between top and bottom plates of the Mee CCN counter, adapted from the Mee Instruments Model 130 CCN Counter manual. Curves sweeping upward and rightward are based on theory, each curve calculated for the indicated top plate temperature. The bottom plate is colder than the top plate by the ΔT indicated on the horizontal axis. Results from laboratory tests are indicated by large black dots. These were obtained by observing the ΔT required to produce nucleation on ammonium sulfate particles of known size. Theoretical supersaturation required to nucleate ammonium sulfate particles of these known sizes was calculated using theory based on the original work of Kohler, and described in Pruppacher and Klett, 1997, Ch. 6. Uncertainty in the supersaturations is a nonlinear function of uncertainty in the aerosol sizes and is roughly 0.05% around 0.4% supersaturation. Uncertainty in the subjective determination of the temperature difference at which they activated is roughly 0.25°C.

Nenes *et al.* (2001) suggest that the classic parallel plate chamber should function fairly closely to the way theory predicts, once a steady-state temperature difference is reached. However, it is possible for the actual temperature difference between the top and bottom inner surfaces of the chamber to be less than is measured by sensors embedded in the top and bottom plates if the tips of these sensors are some distance away from the inner surface. Paul DeMott (Colorado State University, personal communication) has tested two other identical Mee CCN counters in the same manner we did the counter used in this work, and obtained results similar to those shown in Figure 2. We tentatively attribute the difference between theoretical and inferred supersaturations to the indicated temperature difference being greater than the actual temperature difference between the inner surfaces of the upper and lower plates of the chamber. Whether this is the correct reason or not, the consistency between results for three different counters of the same design gives confidence in the supersaturation we assign to our measurements.

After calibration, additional work was done on the counter to improve the system for maintaining the moistness of the blotter paper on the inner top and bottom surfaces of the cloud chamber. The SDSMT counter was then shipped to Bowman and installed in an air conditioned shed housing the project radar electronics. A short ½" ID polyethylene tube conducted air from outdoors into the instrument. At the flow rates used (liters per minute) losses of tenths μm to few μm diameter particles in the sampling tube theoretically should been insignificant. Sampling tests involving alterations to the inlet position, tubing length, and sample flow rates, showed negligible variations in counts due to these alterations.

Problems developed after the SDSMT counter was set up in the field. The feedback-control circuit controlling the temperature difference between the top and bottom plate apparently lost the temperature difference signal, leading to a signal for maximum power to be fed to the thermoelectric cooling unit for an extended time, resulting in overheating the power transistor controlling this current. The counter became inoperable. An identical Mee CCN counter was obtained on loan from the University of North Dakota (UND) Department of Atmospheric Sciences. Although the UND Mee counter was not refurbished in the same manner as the SDSMT counter, nor tested in the CSU aerosol facility, based on the experience of DeMott showing similar supersaturation versus ΔT characteristics for different Mee counters tested in his laboratory, we use the calibration curve in Figure 2 with data obtained with the UND counter.

Total viewable illuminated volume in the UND counter was determined to be 0.06 cm^3 by removing the top plate and measuring the length, width, and height of the illuminated region with a transparent ruler graduated in millimeters, moving the ruler through and along the beam while observing it via the video output on the computer screen. Based on an ~10% uncertainty in the individual dimensions, we estimate the volume uncertainty to be ~30%. In addition, droplets could only be resolved in focus in the near half (lower left in the video view) of the illuminated volume, so the effective sample volume was estimated as 0.03 cm^3.

The internal pump in the UND counter did not work, so an external pump was rigged to draw air samples episodically through the cloud chamber in the counter. Flow rates were significantly higher with the external pump, further reducing aerosol losses in the inlet tubing. The external pump had to be operated manually, in contrast to automated control possible with the internal pump. This limited the number of samples that could be obtained as project personnel could not spare more than an hour per day to work manually with the CCN instrument. Sampling was conducted around the noon hour when other operational demands on personnel were normally light.

3. OBSERVATIONS AND RESULTS

On average, during the daily sampling sessions, 10 discrete samples were obtained over a time span of 30-45 min. The indicated chamber vertical temperature difference was maintained at 5.5°C, corresponding to a peak supersaturation of 0.4% midway between the plates in the chamber based on the empirical results presented in Figure 2. The video camera output was displayed on a computer display screen, and a digital camera was used to record an image of the screen when the cloud formation process peaked during a sampling cycle. A sample of such an image is shown in Figure 3. The number of droplets visible in the images from 10 sample cycles was manually counted and averaged.

Results are shown in Figure 4. Sampling was conducted on 14 days over a 3-week period. Average concentrations of CCN active at 0.4% supersaturation varied from near 30 cm^{-3} to near 200 cm^{-3}, or by a factor of 6, from day to day. The majority of the days had concentrations in the 100-150 cm^{-3} range, with fewer days at the lower and upper extremes. The vertical bars extend +/- one standard deviation from the mean, based on the standard deviation among all observations on a given day. Given the small sample volume, the standard deviations are relatively large. Most overlap each other significantly, although there is no overlap between the spreads around the few highest and lowest averages.

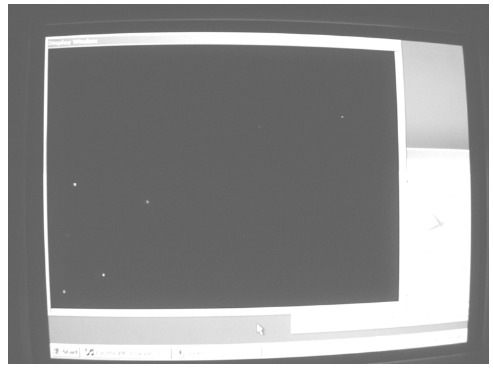

Figure 3: Image of a cloud forming in the CCN counter chamber displayed in a window on a computer monitor. Four droplets are visible in the lower left portion of the image. Lighting conditions and the ability of the optics to focus were such that droplets were only visible in the lower portion of the illuminated sample volume, which extends from the lower left towards the upper right of the display. The sample volume, corresponding to the lower left portion of the in-focus illuminated region, is 0.03 cm^{-3}.

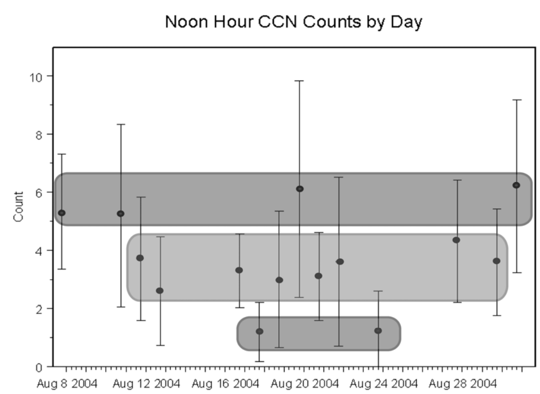

Figure 4: Results from CCN sampling conducted in this study. The observed number of droplets is indicated on the vertical axis, and the date on the horizontal axis. The black dots indicate mean counts per day, with vertical lines extending one standard deviation upward and downward. The results from the 4 days in the top box include the highest concentrations, while those enclosed in the lowest box represent the lowest. The majority of the observations fall within the middle region. Raw counts should be multiplied by 33 to compute concentration in number per cm^{-3}. Thus the range from 1 count to 6 counts corresponds to ambient concentrations ranging from 33 cm^{-3} to 200 cm^{-3}.

Concentrations near 100 cm^{-3} active at 0.4% supersaturation (shaded in green) are borderline between maritime and continental, compared to results collected in Fletcher (1969), Gotz *et al.* (1991), and Pruppacher and Klett (1997), while concentrations of 200 cm^{-3} at this supersaturation (shaded in pink) are approaching the low to middle range of continental observations. The frequency of very low CCN concentrations, characteristic of maritime situations, is 2 out of 14 days, or ~14%, falling between the frequencies for low concentrations in the sampling results of Vali *et al.* (1982) of ~10% of all days, and those of Hobbs *et al.* (1985) of ~25% of all days. It is perhaps not surprising that in a continental interior region far from major anthropogenic sources of aerosols CCN concentrations are typically in the lower range of those found in "continental" regions world-wide.

Preliminary efforts to relate CCN concentrations to the synoptic-scale trajectory of air arriving at the sampling site at the time of the sampling were made using trajectory calculations performed using the NOAA Air Resources HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) trajectory model available at http://www.arl.noaa.gov. Model documentation (Draxler and Hess, 2004) is available at http://www.arl.noaa.gov/data/web/models/hysplit4/win95/arl-224.pdf. Three-day back-trajectory calculations were performed for each day with observations, on-line, by specifying the geographic coordinates of Bowman and the time and date of the observation. The meteorological database used for the trajectory computations was the National Center for Atmospheric Research/National Centers for Environmental Prediction global reanalysis data set.

For much of the observational period, synoptic scale flows were weak, with some three-day back trajectories extending less than 500 km. In a few strong northwest flow cases, the three-day back trajectories extended over 1000 km. Stohl (1988) suggests that trajectory errors are on the order of 20% of distance traveled, making it possible to only indicate upstream source regions three days earlier in broad terms. In addition, sub-synoptic-scale mesoscale and convective motions, which in this season often dominate the synoptic-scale motions, can render trajectories determined using synoptic-scale gridded data less certain (Kahl and Samson, 1988).

With these limitations in mind, the results for the back trajectories on the 14 sampling days showed that when the air arriving at Bowman had been involved in precipitation processes within the previous day, as diagnosed from synoptic-scale precipitation included in the reanalysis data, CCN counts were lowest. An example is shown in Figure 5. It also was generally true that when air was coming from the northern Alberta or Northwest Territories, counts were lower, other factors being equal. The highest counts occurred in air that arrived at Bowman after moving slowly eastward across Montana without being involved in precipitation. An example of this is shown in Figure 6.

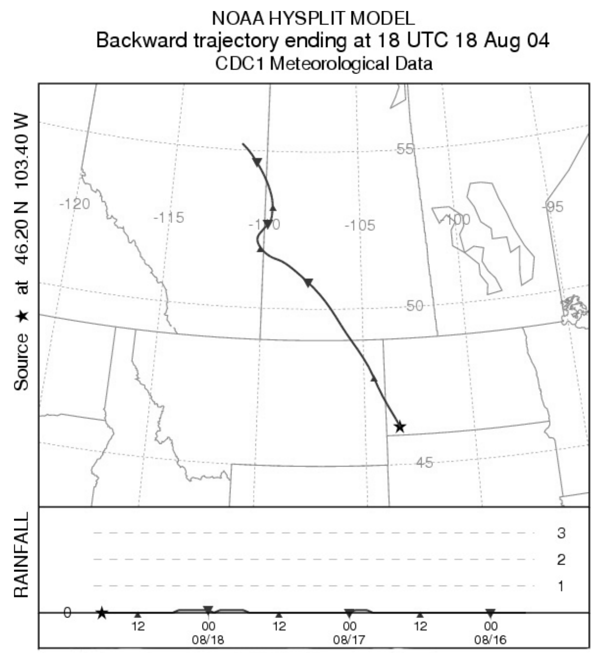

Figure 5: Output from HYSPLIT 3-day back trajectory calculation, for air arriving at 2 m AGL at Bowman at 18Z on 18 August, 2004. Upper panel shows trajectory. Lower panel shows rainfall rate in mm/hr experienced by air travelling along this trajectory.

These results support the conclusions of Hobbs *et al.* (1985) that air involved in precipitation-generating weather situations within a day or two of its arrival at the sampling site is likely to have fewer CCN active at low supersaturations. However, more study of these trajectories, and collection of data on a much larger number of days, is required before forming quantitative conclusions.

Days with significant storminess in the Bowman area were days with cloud seeding operations. Observations of CCN often were not taken on those days because project personnel were busy with seeding operations. For our future work, we hope to have a fully functional CCN counter that can run in an automated mode so that all weather regimes, particularly those when convective storms are in the local area at mid-day, are adequately sampled.

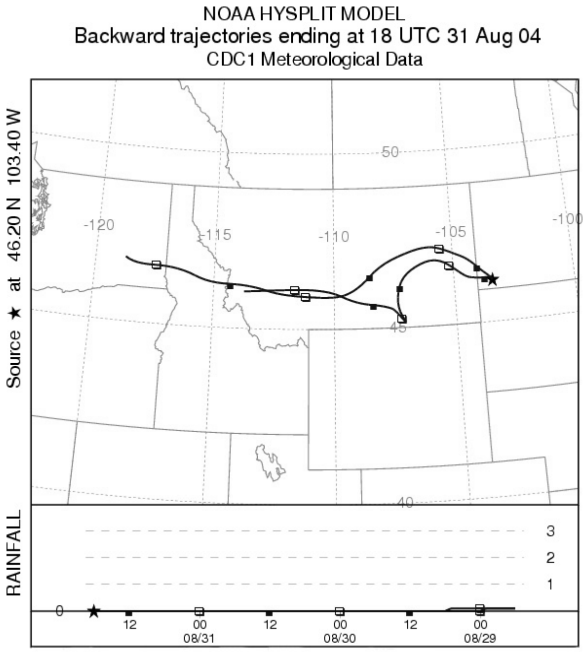

Figure 6: Output from HYSPLIT 3-day back trajectory calculation, for air arriving at 2 m and 500 m AGL at Bowman at 18Z on 18 August, 2004. Upper panel shows trajectory. Lower panel shows precipitation in mm/hr. There was no precipitation along this trajectory within two days of its arrival.

4. CONCLUSIONS

Concentrations of CCN active at supersaturation of ~0.4% were sampled near the local noon hour on 14 days spanning a 3-week period in August, 2004, at Bowman, North Dakota. These concentrations varied by a factor of six from day to day, from 30 to 200 cm^{-3}. Although this is a relatively small sample, results suggest that there is significant daily variation in the CCN population at this site. On the majority of days, the CCN concentrations active at 0.4% supersaturation could be characterized as borderline to mid-range continental. Following the suggestion of Hobbs *et al.* (1985), back trajectory analyses can be used to explain some of the variation in CCN population characteristics from day to day. Variation in geographic source region and in aerosol transformations by cloud and precipitation processes both are important.

Additional CCN sampling is needed to provide better guidance for hygroscopic cloud seeding experiments and observations. More frequent sampling conducted over a longer period of time, covering the diurnal cycle on each day, is needed. Samples should be obtained over a range of supersaturations to more completely characterize the CCN populations.

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