Charakterization of ambient aerosol from measurements of cloud condensation nuclei during the 2003 Atmospheric Radiation Measurement Aerosol Intensive Observational Period
Characterization of ambient aerosol from measurements of cloud condensation nuclei during the 2003 Atmospheric Radiation Measurement Aerosol Intensive Observational Period at the Southern Great Plains site in Oklahoma


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Measurements were made by a new cloud condensation nuclei (CCN) instrument (CCNC3) during the Atmospheric Radiation Measurement (ARM) Program’s Aerosol Intensive Observational Period (IOP) in May 2003 in Lamont, Oklahoma. An inverse aerosol/CCN closure study is undertaken, in which the predicted number concentration of particles available for activation \( N_P \) at the CCNC3 operating supersaturations is compared to that observed \( N_O \). \( N_P \) is based on Köhler Theory, with assumed and inferred aerosol composition and mixing state, and the airborne aerosol size distribution measured by the Caltech Dual Automatic Classified Aerosol Detector (DACAD). An initial comparison of \( N_O \) and \( N_P \) assuming the ambient aerosol is pure ammonium sulfate \((\text{NH}_4)_2\text{SO}_4\), results in closure ratios \( (N_P/N_O) \) ranging from 1.18 to 3.68 over the duration of the IOP, indicating that the aerosol is less hygroscopic than \((\text{NH}_4)_2\text{SO}_4\). \( N_P \) and \( N_O \) are found to agree when the modeled aerosol population has characteristics of an external mixture of particles, in which insoluble material is preferentially distributed among particles with small diameters (<50 nm) and purely insoluble particles are present over a range of diameters. The classification of sampled air masses by closure ratio and aerosol size distribution is discussed in depth. Inverse aerosol/CCN closure analysis can be a valuable means of inferring aerosol composition and mixing state when direct measurements are not available, especially when surface measurements of aerosol composition and mixing state are not sufficient to predict CCN concentrations at altitude, as was the case under the stratified aerosol layer conditions encountered during the IOP.


1. Introduction

One of the largest uncertainties in aerosol radiative forcing is associated with the indirect effect, which results from the relationship between atmospheric aerosols and cloud formation, properties, and lifetime [Intergovernmental Panel on Climate Change, 2001]. Twomey [1977] postulated that an increase in the number concentration of atmospheric aerosol particles would increase the number of cloud droplets formed for a given air mass. For fixed liquid water content, the cloud droplets would also be smaller than those formed under conditions with lower particle concentrations. This increase in number and decrease in mean diameter of cloud droplets would have two indirect effects on climate. Cloud albedo is greater for clouds with more numerous, smaller droplets; this has been termed the first indirect climatic effect of aerosols. Also, the lifetime of a cloud with smaller cloud droplets is greater than that of a cloud with larger droplets because the rain forming mechanisms are less efficient [Albrecht, 1989]. This is referred to as the second indirect climatic effect of aerosols. Both effects create clouds that are more reflective and more persistent, leading to the cooling of the Earth’s surface [Twomey, 1977].

[1] One of the largest uncertainties in aerosol radiative forcing is associated with the indirect effect, which results from the relationship between atmospheric aerosols and cloud formation, properties, and lifetime [Intergovernmental Panel on Climate Change, 2001]. Twomey [1977] postulated that an increase in the number concentration of atmospheric aerosol particles would increase the number of cloud droplets formed for a given air mass. For fixed liquid water content, the cloud droplets would also be smaller than those formed under conditions with lower particle concentrations. This increase in number and decrease in mean diameter of cloud droplets would have two indirect effects on climate. Cloud albedo is greater for clouds with more numerous, smaller droplets; this has been termed the first indirect climatic effect of aerosols. Also, the lifetime of a cloud with smaller cloud droplets is greater than that of a cloud with larger droplets because the rain forming mechanisms are less efficient [Albrecht, 1989]. This is referred to as the second indirect climatic effect of aerosols. Both effects create clouds that are more reflective and more persistent, leading to the cooling of the Earth’s surface [Twomey, 1977].
cloud condensation nuclei (CCN), and aerosol size distribution and composition, in addition to meteorological conditions, are central to the indirect climatic effect of aerosols. For a given particle composition and size, the supersaturation above which the particle undergoes spontaneous condensational growth (activation) into a cloud droplet, the so-called critical supersaturation, is described by Köhler Theory. The activation diameter, the dry diameter at which a particle of known or assumed composition will activate, can also be calculated for a given supersaturation. Prediction of aerosol activation from Köhler Theory is very successful for aerosols composed of soluble, inorganic salts, such as ammonium sulfate ((NH₄)₂SO₄), sodium chloride (NaCl), and ammonium bisulfate (NH₄HSO₄). However, Köhler Theory needs to be augmented when considering chemical components, such as organic compounds, that are partially soluble, insoluble, or affect the surface tension of the aqueous solution. The chemical composition of atmospheric aerosol can be complex and include many different chemical species, which may affect aerosol activation in competing ways.

[5] The ability of Köhler Theory to predict ambient CCN concentrations can be studied by comparing atmospheric CCN measurements at a given supersaturation with CCN concentrations calculated using aerosol size distribution and composition measurements. This type of study, called an aerosol/CCN closure, compares the observed CCN concentration (Nₐ) at the operating supersaturation of the CCN instrument to that predicted from the aerosol size distribution and composition (Nₚ) in a closure ratio, defined here as Nₚ/Nₐ. Nₚ is determined from the aerosol size distribution by summing the concentration of particles with diameters greater than the activation diameter calculated from Köhler Theory [VanReken et al., 2003]. When direct measurements of aerosol composition are unavailable, Nₚ can be calculated using an assumed aerosol composition or one that is inferred from other available measurements. An “inverse” aerosol/CCN closure study (explained further in section 5.2) can be undertaken to determine aerosol composition and mixing states that are most consistent with CCN observations when direct measurements of these aerosol characteristics are unavailable.

[5] In this paper, data measured by a new CCN instrument (CCNC3) during the Atmospheric Radiation Measurement (ARM) Aerosol Intensive Observational Period (IOP) in May 2003 are used in an inverse aerosol/CCN closure study of the midcontinental aerosol sampled near the Southern Great Plains (SGP) Central Facility (CF) to determine aerosol composition and mixing states consistent with the CCN measurements at the operating supersaturations of the instrument. The Texas A&M Differential Mobility Analyzer/Tandem Differential Mobility Analyzer (DMA/TDMA) data from the surface are used to infer the mixing state and insoluble fraction of the aerosol population as a function of dry diameter. This information is used with the airborne CCN and aerosol size distribution measurements to determine conditions under which aerosol composition and mixing states inferred from surface measurements are able to reproduce CCNC3 measurements at altitude. Aerosol properties, categorized by closure ratio and aerosol size distribution shape, during pollution and smoke events are also discussed.

2. ARM Aerosol IOP

[5] The ARM Aerosol IOP occurred from 5 to 31 May 2003 at the Department of Energy’s (DOE’s) ARM SGP CF in Lamont, Oklahoma. There were a total of 16 science flights, with a total of 60.6 flight hours, conducted by the Center for Interdisciplinary Remotely-Piloted Aircraft Studies’ (CIRPAS) Twin Otter aircraft on 15 days during this period. The ARM Aerosol IOP flight tracks for flights 6–10 and 12–17 (the flights for which there are CCN data) are shown in Figure 1, which illustrates that most flights took place over or near the SGP CF. The last flight, flight 17, was coordinated with the Moderate Resolution Imaging Spectroradiometer (MODIS) overpasses of four ARM sites (SGP CF, Extended Facility (EF)-12, EF-20, and EF-19 in Figure 1).

[7] The ARM SGP site is located in a midlatitude, continental area, surrounded by agricultural land and dirt roads. The site is influenced by local emissions from nearby industrial and power plants and local aerosol sources, such as vehicle and agricultural aerosols. Sulfur dioxide (SO₂) emissions from nearby oil refineries and power plants, such as the Conoco and Ponca City Power Plants, are major local sources of sulfate aerosols over the SGP site. Anthropogenic, agricultural related aerosol sources include local fertilizer application and production, field burning, and animal byproducts. Local biomass burning is greatest from May through July [Ziogou and Lohmann, 2003]. The particles that are commonly found at the ARM SGP site are a complex mix of these aerosol types, with smoke- and dust-dominated events, which are commonly characterized by decreased aerosol hygroscopicity, occurring occasionally [Sheridan et al., 2001]. Routine condensation particle counter (CPC) measurements from the SGP site generally show a strong, diurnal cycle of aerosol number concentration, with peak concentrations in the afternoon and early evening. Over the 4-year period from July 1996 to June 2000, the daily average condensation nuclei (CN) concentration ranged from less than 1000 cm⁻³ to about 20,000 cm⁻³, with a mean around 5000 cm⁻³. The hourly average CN concentration ranged from about 4000 cm⁻³ from 1100 to 1500 UTC (Universal Time Coordinated) to about 18,000 cm⁻³ from 1800 to 2000 UTC [Sheridan et al., 2001]. (The difference between local time (LT) at the ARM site and UTC is 5–6 hours, depending on daylight savings. During the ARM Aerosol IOP, LT was 5 hours behind UTC, so that 1200 LT corresponds to 1700 UTC in this paper.) These high CN concentrations could result from build-up and advection of pollutant aerosols from local sources, photochemical particle production [Sheridan et al., 2001], coagulation, or the evolution of the boundary layer [Ziogou and Lohmann, 2003].

[8] Most of the Twin Otter flights during the ARM Aerosol IOP were conducted under clear or partly cloudy skies to assess aerosol impacts on solar radiation. Additional flights targeted mostly cloudy conditions to assess aerosol/ cloud interactions, test theoretical understanding of aerosol activation, and to test surface remote sensing of the indirect effect. Ground and airborne measurements, which included
aerosol absorption, scattering, extinction, size distribution, and CCN concentration, are compared in a variety of closure studies to help resolve differences in measurements and models. Routine ARM SGP aerosol measurements (absorption, total scattering and hemispheric backscattering, light scattering as a function of relative humidity, total condensation particle concentration, number concentration of particles with diameters from 0.1 to 10 \( \mu m \), vertical aerosol optical thickness, etc.) continued throughout the IOP.

3. Instrument Descriptions

3.1. Twin Otter Inlet System

In order to minimize sampling losses, the aerosol inlet on the CIRPAS Twin Otter is designed to admit the air sample prior to any bending of flow lines and slow the sample down before transport to the instruments. The intake extends forward from the roof of the cabin to a position 1.2 m directly above the aircraft’s nose. The sampled air is initially slowed down by a factor of 5 by means of an aerodynamically lipped diffuser. A second diffuser, positioned at the centerline of the first diffuser, reduces the flow speed by another factor of two, while excess flow from the first diffuser exits along the sides of the second diffuser. The sample then flows down a 7.62 cm diameter duct and enters the cabin after a 45° bend. Inside the cabin the duct is straightened out again with another 45° bend, and samples are drawn to the various instruments from ports mounted on the side of the duct. The ports are flush inside the duct, but extend outward at 45° angle to the flow. At an aircraft speed of 50 m/s, approximately 1000 liters per minute (lpm) flow down the duct. Air not ciphered off to the instruments is dumped out of the cabin.

3.2. CCN Instrument (CCNC3)

The CCN instrument (CCNC3) deployed on the Twin Otter consists of three columns, each of which is physically modeled after a previous CCN instrument column design [Chuang et al., 2000] with some changes to the physical design. Three temperature-controlled sections are used to create the desired temperature gradient, instead of fourteen sections, as in the work by Chuang et al. [2000]. The instrument was designed to be fully automated and software controllable in normal operation and to operate with a different supersaturation in each column. CCNC3 operating conditions, some aspects of which differ from those given by Chuang et al. [2000], are discussed in the following paragraphs. Only column 1 operated properly during the ARM Aerosol IOP, the first field mission in which the CCNC3 was deployed; thus CCN data were obtained at one supersaturation per flight.

Each CCNC3 column (Figure 2) consists of a stainless steel growth tube 0.4 m in length with a 1.9 cm outer diameter and a 1.6 mm wall thickness. The inner wall of the growth tube is lined with filter paper, which is rewetted by a small peristaltic pump every 90 min. A linear temperature profile was used to develop a constant super-
saturation at the centerline of the growth tube [Rogers and Squires, 1977; Roberts and Nenes, 2005]. (A linear temperature gradient was also used in the airborne CCN instruments during the Cirrus Regional Study of Tropical Anvils and Cirrus Layers – Florida Area Cirrus Experiment (CRYSTAL-FACE) [VanReken et al., 2003; Roberts and Nenes, 2005].) The sampled aerosol is focused onto the centerline of the growth tube and introduced to the column with filtered sheath air; the droplets that form within the growth tube are then counted by a detector.

[12] The CCNC3 detector consists of a laser, an optics tube, and an avalanche photo-diode (APD) module, and was designed on the basis of that of Laser Particle Counter (LPC) Model 3755 (TSI, Incorporated), scaled down for CCN application. A 670 nm, 10 mW Lasiris MFL Micro-Focus Laser is positioned at one end of an optics tube, so that the droplet inlet to the optics tube is at the 30 mm working distance of the laser. When a droplet falls through the laser beam, the laser light is scattered in the forward direction, and a pair of aspherical condenser lenses collects the scattered light and focuses it into a fiber optic at the other end of the optics tube. The signal from the scattered laser light is sent to a Hamamatsu Photonics C5460-01 APD module, which sends the resulting digital pulse to the data acquisition system.

[13] During the IOP, the column operated at three different linear temperature gradients, with one temperature gradient per flight. The total flow rate of the column was about 0.56 lpm with a sheath to sample ratio around 10. (NH₄)₂SO₄ calibrations for column 1 at its different linear temperature gradients are shown in Figure 3. The activation diameters and supersaturations associated with each linear temperature gradient are given in Figure 3. For each linear temperature gradient calibration, a solution of (NH₄)₂SO₄ was atomized to create droplets that were then dried and introduced into a differential mobility analyzer (DMA). Certain dry diameters were selected using the DMA and then split to the CCNC3 inlet and the inlet to a CPC (TSI, Inc., Model 3010). The activated ratio (AR) was calculated as the ratio of the number concentration of CCN measured by the CCNC3 (N_CCNC) to the number concentration of total particles measured by the CPC (N_CPC). The activation diameter (d_act) is the dry diameter at which 50% of the particles are activated (AR = 0.5). The uncertainty limits given on the calibration curves in Figure 3 result from the uncertainty in the diameter produced by the DMA (generally taken to be ±5%) and the combined uncertainties associated with the concentrations measured by the CPC and the CCNC3. These uncertainties associated with the column calibrations are folded into the overall measurement uncertainty, which is estimated for each flight on the basis of criteria explained in section 4.3.

[14] The supersaturation corresponding to dry (NH₄)₂SO₄ particles with diameter d_act and thus the operating supersaturation of the column, was calculated theoretically by Köhler Theory. Droplet density is calculated from data of Tang and Munkelwitz [1994]; the full Pitzer model [Pitzer, 1973; Pitzer and Mayorga, 1973] is used to calculate the osmotic coefficient; and values from Pruppacher and Klett [1997] are used for surface tension. The model calculates the critical supersaturation for particles that contain certain soluble salts, certain organics, and generalized insoluble material [Brechtel and Kreidenweis, 2000a, 2000b].

Figure 2. A schematic of a CCNC3 column. A second layer of insulation covers the growth tube and heating/cooling sections (to prevent temperature transfer to the outside air) and the detector (to prevent condensation within the optics tube).

Figure 3. (NH₄)₂SO₄ calibration curves for column 1 of the CCNC3 at the temperature gradients used during the ARM Aerosol IOP. The column was not functioning properly for flights 1–5 and 11. The activation ratio is defined as the ratio of the number concentration of CCN measured by the CCNC3 to the number concentration of particles measured by the CPC.
3.3. Dual Automatic Classified Aerosol Detectors (DACAD)

[16] The Caltech Dual Automatic Classified Aerosol Detectors (DACAD) have been deployed in several previous airborne experiments, and their characteristics are well documented [Wang et al., 2002, 2003; VanReken et al., 2003]. The DACAD consists of two DMA systems operated in parallel. One of the DMA systems measures the dry aerosol size distribution, while the other measures the aerosol size distribution at ambient relative humidity (RH) by using an active RH controller [Wang et al., 2003]. The aerosol wet and dry size distributions are obtained separately and independent of each other; therefore no size-resolved information is obtained. The main components of the measurement system are a cylindrical DMA (TSI Inc., Model 3081) and a CPC (TSI Inc., Model 3010), which has a 50% counting efficiency at 10 nm. Using the scanning mobility technique, each DMA system generates a size distribution for particle diameters from ~17 to ~720 nm every 72.5 s [Wang et al., 2002]. (This scanning time was reduced from 103 s [VanReken et al., 2003] prior to the ARM Aerosol IOP.)

3.4. Tandem Differential Mobility Analyzer (DMA/TDMA)

[17] The Texas A&M DMA/TDMA measured aerosol size distributions and size-resolved hygroscopic growth at the SGP Cloud and Radiation Testbed (CART) trailer at the CF during the IOP [Gasparini et al., 2006]. The main measurement section of the DMA/TDMA consists of two Aerosol Dynamics, Inc., high-flow DMAs (HF-DMAs [Stolzenburg et al., 1998]), a charger, two Nafion tubes, and a CPC. One DMA/TDMA measurement sequence (~1 hour) consists of two different operational modes to obtain both the aerosol size distribution and the size-resolved hygroscopic growth. For both modes, the sample air is first dried in a Nafion tube and then introduced to a charger before entering the first DMA. During the single DMA mode (~5 min), the aerosol size distribution is measured by scanning the voltage applied to the first DMA [Wang and Flagan, 1990] and counting the size-selected aerosol particle concentration with the CPC. In TDMA mode, the voltage supplied to the first DMA is fixed to produce a monodisperse aerosol of known particle size. The monodisperse aerosol is exposed to a controlled, elevated relative humidity (RH; 85% in this IOP) before entering the second DMA. The humidified aerosol is classified by scanning the voltage applied to the second DMA and the size- and hygroscopicity-resolved aerosol particle concentration is observed with the CPC. The second mode sequence is repeated for other particle dry diameters by changing the voltage applied to the first DMA [Gasparini et al., 2004]. A third mode was implemented during the IOP, in which the dry monodisperse aerosol was exposed to a wide range of RH to determine the deliquescence properties of the aerosol [Gasparini et al., 2006].

[18] The aerosol particle soluble fraction and mixing state is inferred by comparing the dry diameter separated by the first DMA with the hydrated size distribution measured with the second DMA. The comparison results in a normalized distribution expressed in terms of the hygroscopic growth factor (GRH), which is defined as the ratio of the hydrated particle diameter to that of the dry particle. The aerosol is then divided into four hygroscopicity-based categories: pure insoluble, pure soluble, mixed insoluble, and mixed soluble. The full technique used to determine the relative contributions of soluble and insoluble components to the dry particle composition is described by Gasparini et al. [2004].
was measured using the Particle-Into-Liquid Sampler–Ion Chromatography (PILS-IC) technique [Weber et al., 2001], and the aerosol total mass concentration was measured using the Tapered Element Oscillating Microbalance (TEOM) technique [Patashnik and Rupprecht, 1986]. The PILS-IC technique has a \( <0.05 \) \( \mu g \) \( m^{-3} \) limit of detection for all of the ions. The TEOM exhibited a temperature-related oscillation behavior during the IOP that resulted in an uncertainty of \( \pm 40\% \). The time resolution of the PILS-IC and TEOM were 8 min and 30 min, respectively, during the IOP. A sharp cut cyclone and two glass honeycomb denuders in series remove particles greater than 1 \( \mu m \) diameter and water-soluble vapor phase species, respectively, from the sample flow to both the PILS-IC and the TEOM. To prevent condensation of water within the tapered element, the TEOM inlet was maintained at 35\(^\circ\)C [Patashnik and Rupprecht, 2006].

[20] Routine aerosol light scattering coefficient \( (\sigma_{sp}) \) and light scattering hygroscopic growth factor \( (f(RH)) \) measurements for total and fine (submicron) mode aerosol continued at the ground site during the IOP. Measurements of \( \sigma_{sp} \) and \( f(RH) \) complement those from the PILS-IC and TEOM. Total scattering and backscattering were measured with a three-wavelength integrating nephelometer (TSI Inc., Model 3563) [Sheridan et al., 2001]. The light scattering hygroscopic growth factor, \( f(RH) \), is different from the hygroscopic growth factor \( (G(RH)) \) determined from DMA/TDMA data. \( G(RH) \) is based on the increase in particle diameter with increasing RH (see section 3.4), whereas \( f(RH) \) is defined as the change in aerosol light scattering with changing RH [Covert et al., 1972; Rood et al., 1987] and is determined as the ratio of \( \sigma_{sp} \) at 80\% to that at 40\% RH. \( f(RH) \) was measured using a humidograph [Carriço et al., 1998], which consists of two nephelometers separated by a humidity control system that ramps up the RH in the second nephelometer. Data from the humidograph are used to relate \( \sigma_{sp} \) at any RH to that at 40\% RH with a three-parameter curve-fitting model [Sheridan et al., 2001].

4. Data Trends During the ARM Aerosol IOP

4.1. Particle Composition (PILS-IC, TEOM, and DMA/TDMA) and Aerosol Mixing State (DMA/TDMA)

[21] Of the major inorganic species (\( NH_4^+ , SO_4^{2-} , NO_3^- , K^+ , Ca^{2+} , Mg^{2+} , Na^+ , Cl^- \)) measured at the SGP site during the IOP, \( NH_4^+ \) and \( SO_4^{2-} \) were dominant. \( NO_3^- \) was observed in \( \sim 10\% \) of the samples and only when \( SO_4^{2-} \) was completely neutralized by \( NH_4^+ \). \( K^+ \), a tracer for biomass burning, and \( Ca^{2+} \), a tracer for dust, were occasionally observed in appreciable levels. On average, the ratio of \( NH_4^+ \) to \( SO_4^{2-} \) was 2, with ratios significantly greater than 2 observed on 20 May (no flight) and 22 May (flight 13) and less than 2 observed on 21 May (flight 12) [Pahlow et al., 2006].

[22] Aerosol organic fraction increased continuously during the daytime hours, while the inorganic concentration remained fairly constant. Overall, the aerosol organic content, which is estimated as the difference between total mass (TEOM) and total inorganic mass (PILS-IC), accounted for \( >40\% \) of the aerosol mass. Increases in organic fraction, values of which were as high as 80\%, were accompanied by a lowering of \( f(RH) \), suggesting that the particulate organic fraction had a lower hygroscopicity than the inorganic fraction. Also, the amount of aerosol organic mass correlated strongly with the amount of black carbon inferred from the aerosol absorption coefficient [Pahlow et al., 2006], which was measured using a filter-based light absorption photometer [Sheridan et al., 2001].

[23] Gasparini et al. [2006] inferred size-resolved aerosol particle composition and hygroscopic growth properties from data collected by the Texas A&M DMA/TDMA at the SGP site using the technique from Gasparini et al. [2004]. The \( G(RH) \) of the observed aerosol during the IOP was found to increase with increasing dry diameter, and the average median \( G(RH) \) at 85\% RH was 1.20 at 12 nm and 1.37 at 300 nm, which indicates that the smallest analyzed particles were largely composed of carbonaceous compounds. The largest particles were either very hygroscopic or slightly hygroscopic, but rarely exhibited hygroscopicity in between these extremes. At times during the IOP, a nonhygroscopic mode with median \( G(RH) \) less than 1.10 was observed at 450 and 600 nm and, more rarely, at 200 and 300 nm [Gasparini et al., 2006].

4.2. Aerosol Size Distribution (DACAD-Airborne)

[24] The aerosol size distributions measured by the DACAD were analyzed for flights 6–10 and 12–17, for which CCN data were collected. Aerosol size distributions differed greatly between flights, as well as at different altitudes within a single flight. For example, Figure 4 shows DACAD aerosol size distributions from six level legs and five different altitudes from flight 16. At the lower altitudes, 351 m and 720 m AGL (Above Ground Level; all reported altitudes are AGL), the aerosol size distributions are multimodal with median diameters less than the cutoff diameter of the DACAD (\( \sim 17 \) nm), which may indicate that the sampled aerosols are freshly emitted or formed locally in the atmosphere. The aerosol populations at 1028 m and 1337 m have similar size distributions that are multimodal with median diameters that may be smaller than the cutoff diameter of the DACAD, which may indicate mixed aerosol layers that are a combination between fresh and aged aerosol populations. The aerosol population at 2281 m during flight 16 is multimodal but with a less pronounced second mode at larger diameters, which is indicative of background aerosol. Also note that the \( dN/d\log D_p \) values are greater for lower-altitude level legs than higher legs, which is a typical feature of ambient aerosol. Aerosol size distributions such as those shown in Figure 4 were seen throughout the IOP flights, along with some simple unimodal distributions. The differences in aerosol size distributions are consistent with sampled aerosol layers that had different sources and/or were of different ages.

[25] Rapidly changing aerosol size distributions were observed frequently during the IOP. Since the DACAD size distribution scan time is 72.5 s, some characteristics of a rapidly changing aerosol size distribution may not have been resolved. The CCNC3 has a sampling rate of 1 s and, therefore, is capable of observing rapid changes in the aerosol size distribution. For this reason, predicted and observed CCN concentrations may not agree well during
times of rapidly changing aerosol characteristics. This will be discussed further in section 6.

4.3. CCN Concentration (CCNC3-Airborne)

The trends in the CCN concentration data follow those seen in both the airborne CPC and airborne DACAD data. When present, homogeneous boundary layers are easily discerned from plots of CCNC3, CPC, and DACAD data, e.g., Figure 5 for flight 14. A more specific CCN trend analysis is discussed in section 6.

The airborne CPC had a cutoff diameter of 13 nm, and the DACAD scanned particle sizes down to ~17 nm.

Figure 4. Aerosol size distributions from different level legs during flight 16. The legend gives the average altitudes (AGL) and times analyzed for each level leg. The times given are UTC; 1700 UTC corresponds to 1200 LT.

Figure 5. Flight traces of CPC, CCNC3, and DACAD data for flight 14. The DACAD traces give the total concentrations of particles with diameters greater than those noted in the legend. A well-mixed boundary layer is clearly shown from about 2010 to 2045. The times given are UTC; 1700 UTC corresponds to 1200 LT.
The CCN number concentrations are always less than the total particle number concentrations measured by the CPC or the DACAD in any of the eleven flights, even though the activation diameters for pure (NH₄)₂SO₄ particles at the operating supersaturations of the CCNC3 are similar to the cutoff diameters for the CPC and DACAD. To determine the uncertainty in the CCN measurements, the CPC concentration was used as a high-end limit for CCN concentration on all flights. The concentration measured by the CCNC3 column 2, when operating, was used as the lower limit for the column 1 CCN concentration. The operating supersaturation of column 2 was unable to be determined with any certainty, but it was always operating at some supersaturation lower than that of column 1. Therefore measurements by column 2 are used as estimates of the lower uncertainty limits for the measurements of column 1, when both columns were operating, but are not used in any other part of the analysis. Table 1 gives the uncertainty limits for column 1.

5. Inverse Aerosol/CCN Closure Study: Results and Discussion

Comparison of the CCN data to the aerosol size distribution data allows certain conclusions to be drawn about the characteristics of the measured CCN and the aerosol population. Certain aerosol population properties, such as particle insoluble fraction or mixing state, can be estimated as those that lead to closer agreement between predicted and observed CCN concentrations at the operating supersaturations of the CCN instrument. These estimated properties can be compared with those inferred from or measured by other instruments to determine whether the estimates are reasonable.

Figure 6. Predicted CCN concentration (Nₚ; assuming pure (NH₄)₂SO₄) versus observed CCN concentration (Nₒ) for ARM Aerosol IOP. Only flights for which CCN data are available are shown. Nₚ is determined from the DACAD size distribution for all flights except flight 9, for which Nₚ is determined from the CPC total particle (>13 nm) concentration. The linear fit in log-log space for all flight closure data is shown, and corresponding fit parameters are given. The mean and standard deviation of the ratio of Nₚ to Nₒ averaged over all flights are also given. The fit parameters and mean and standard deviation of the ratio of Nₚ to Nₒ are also shown for the “Insoluble Limit” case, for which Nₚ is calculated using the insoluble fractions calculated in section 5.2.1.
aerosol/CCN closure analysis generally does not include data above this altitude.

5.1. Inverse Aerosol/CCN Closure: Initial Comparison, Assuming 100% (NH₄)₂SO₄

[30] Figure 6 shows an initial comparison of the predicted CCN number concentrations (Nₚ) to those observed by the CCNC3 instrument (Nₒ) for all eleven flights and includes all the data points from each flight that fit the criteria outlined in section 5. For this initial comparison, Nₚ is determined as the sum of the number concentration of particles in the DACAD size distributions with diameters greater than the activation diameter of an aerosol consisting of pure (NH₄)₂SO₄ for the operating supersaturation of the CCN instrument. Pure (NH₄)₂SO₄ is assumed as a starting point because Pils-IC data from the ground indicated that NH₄⁺ and SO₄²⁻ were dominant species and that the average ratio of NH₄⁺ to SO₄²⁻ was near 2 during the ARM Aerosol IOP. For flight 9, Nₚ is determined from the CPC total particle concentration, since the activation diameter of the CCNC3 column is about the same as the cutoff diameter for the CPC. The average value of Nₚ/Nₒ (μ(Nₚ/Nₒ)) for all flights is 1.92, with a standard deviation (σ(Nₚ/Nₒ)) of 1.29, and Table 2 shows this ratio for each flight. (μ(Nₚ/Nₒ) is used when values of the ratio, Nₚ/Nₒ, have been averaged over more than one DACAD scan.) The average value of Nₚ/Nₒ for each flight, and thus for all flights averaged together, is greater than unity. This indicates that fewer particles activated than would be expected if the particles were composed of pure (NH₄)₂SO₄. The average Nₚ/Nₒ also differs from flight to flight, which indicates that the aerosol CCN activity properties changed from flight to flight.

[31] The Nₚ versus Nₒ data for each flight were fit to a straight line in log-log space, and the results for the slope and intercept of those fits are given in Table 2. The fit for all data is shown in Figure 6, and Figure 7 shows the fit for flights 8, 12, 14, and 17. Nonunity slopes and nonzero intercepts of these linear fits in log-log space offer some insight as to the characteristics of the ambient aerosol population through its deviation from the activation properties of pure (NH₄)₂SO₄ particles.

[32] The slopes of the log-log fit of Nₚ versus Nₒ for the IOP flights are generally not unity, which indicates that the chemistry and mixing state of the aerosol population does not affect the CCN ability at all concentrations equally. This imbalance results from sampling different air masses with different aerosol properties, which lead to different Nₚ to Nₒ ratios, during the same flight. Aerosol populations with low CCN concentration, which often result from those with low total particle concentration (Nₜₒₜᵃˡ), tend to have “clean” aerosol sources or be well-aged aerosols, in which particle coagulation has decreased Nₜₒₜᵃˡ and cloud processing has likely increased soluble fractions. These types of air masses with low CCN concentrations would have Nₚ to Nₒ ratios nearer to unity. High Nₜₒₜᵃˡ, on the other hand, is often indicative of fresh, polluted aerosol sources, possibly with high levels of insoluble material and a greater degree of external mixing, which could lead to higher values (greater than unity) of Nₚ/Nₒ. In Figure 7, the clustering of flight 12’s closure data points at low, medium, and high CCN concentrations could indicate that three different air masses were sampled. The air mass with midlevel CCN concentrations could be a mixture of the air masses with high- and low-level CCN concentrations, instead of a distinct air mass. Figure 8 illustrates trends of Nₚ/Nₒ with altitude as a function of longitude for flights 12 (Figure 8a) and 14 (Figure 8b). The high CCN concentration data points (Figure 7) for flight 12 correspond to Nₚ/Nₒ points (Figure 8) at altitudes of about 600 m (Nₜₒₜᵃˡ = 2.77), 850 m (Nₜₒₜᵃˡ = 1.99–2.42), and 1380 m (Nₜₒₜᵃˡ = 2.00); the mid CCN concentration data points correspond to the Nₚ to Nₒ ratios at about 550 m (Nₜₒₜᵃˡ = 1.51–2.02), 600 m (Nₜₒₜᵃˡ = 1.77, 1.80), and 1380 m (Nₜₒₜᵃˡ = 1.67); and the low CCN concentration points correspond to the Nₚ to Nₒ ratios at about 2100 m (Nₜₒₜᵃˡ = 1.23–1.53).

[33] The situation is quite different for flight 14 (see Figures 7 and 8), for which the generalized high and low Nₜₒₜᵃˡ classifications do not hold. All of the closure data points from flight 14 are clustered at CCN concentrations less than 2000 cm⁻³, and the slope is less than unity. Contrary to what was observed in flight 12, the sampled aerosol with lower Nₜₒₜᵃˡ has a greater deviation from idealized, pure (NH₄)₂SO₄ particles than those with higher Nₜₒₜᵃˡ. As will be discussed later in section 6, flight 14 was apparently influenced by a Siberian smoke event [Schmid et al., 2004]. Smoke aerosols that have been transported a long distance are likely to have low particle concentrations through particle coagulation and cloud processing. However, the aged smoke particles at low Nₜₒₜᵃˡ in flight 14 did not

### Table 2. Average (μ(Nₚ/Nₒ)) and Standard Deviation (σ(Nₚ/Nₒ)) of the Ratios of Predicted CCN Concentration to Observed CCN Concentration for All Flights for Which There Were CCN Data

<table>
<thead>
<tr>
<th>Flight</th>
<th>Day in May</th>
<th>μ(Nₚ/Nₒ)</th>
<th>σ(Nₚ/Nₒ)</th>
<th>m</th>
<th>b</th>
<th>R²</th>
<th>Number of Data Points</th>
</tr>
</thead>
<tbody>
<tr>
<td>6</td>
<td>14</td>
<td>1.30</td>
<td>0.18</td>
<td>0.63</td>
<td>1.32</td>
<td>0.63</td>
<td>80</td>
</tr>
<tr>
<td>7</td>
<td>14</td>
<td>1.29</td>
<td>0.13</td>
<td>0.81</td>
<td>0.77</td>
<td>0.96</td>
<td>11</td>
</tr>
<tr>
<td>8</td>
<td>15</td>
<td>1.18</td>
<td>0.07</td>
<td>1.05</td>
<td>-0.10</td>
<td>0.96</td>
<td>55</td>
</tr>
<tr>
<td>9</td>
<td>17</td>
<td>1.29</td>
<td>0.35</td>
<td>0.85</td>
<td>0.54</td>
<td>0.96</td>
<td>78</td>
</tr>
<tr>
<td>10</td>
<td>18</td>
<td>1.34</td>
<td>0.08</td>
<td>0.97</td>
<td>0.21</td>
<td>0.98</td>
<td>27</td>
</tr>
<tr>
<td>12</td>
<td>21</td>
<td>1.79</td>
<td>0.36</td>
<td>1.14</td>
<td>-0.22</td>
<td>0.99</td>
<td>23</td>
</tr>
<tr>
<td>13</td>
<td>22</td>
<td>1.78</td>
<td>0.51</td>
<td>0.91</td>
<td>0.51</td>
<td>0.90</td>
<td>38</td>
</tr>
<tr>
<td>14</td>
<td>25</td>
<td>1.84</td>
<td>0.48</td>
<td>0.77</td>
<td>0.88</td>
<td>0.96</td>
<td>54</td>
</tr>
<tr>
<td>15</td>
<td>27</td>
<td>1.87</td>
<td>0.45</td>
<td>1.14</td>
<td>-0.18</td>
<td>0.97</td>
<td>38</td>
</tr>
<tr>
<td>16</td>
<td>28</td>
<td>3.68</td>
<td>2.96</td>
<td>1.40</td>
<td>-0.68</td>
<td>0.65</td>
<td>42</td>
</tr>
<tr>
<td>17</td>
<td>29</td>
<td>3.45</td>
<td>0.86</td>
<td>0.93</td>
<td>0.74</td>
<td>0.77</td>
<td>71</td>
</tr>
<tr>
<td>All</td>
<td></td>
<td>1.92</td>
<td>1.29</td>
<td>0.90</td>
<td>0.54</td>
<td>0.74</td>
<td>517</td>
</tr>
</tbody>
</table>

*Slope (m) and intercept (b) values for linear fits of Nₚ to Nₒ in log-log space are also given. Assumes pure (NH₄)₂SO₄ in the calculation of Nₚ.
become more CCN active than the background aerosol, which is probably the type of aerosol sampled under about 1400 m (see Figure 8).

[34] An intercept that deviates from zero indicates that some fraction of the aerosol population is unavailable for activation at the measurement supersaturation, although this fraction would activate under the assumption of pure \((\text{NH}_4)_2\text{SO}_4\) particles. This nonactive fraction could consist of particles from an internal or external mixture that have some fraction of insoluble, hydrophobic material that prevents activation. Even if the slope were near unity, the intercept can, as explained above, deviate from zero if particles exist that do not exhibit predicted CCN activity characteristics. Flights 8 and 17 (Figure 7) have slopes near unity but have very different intercepts, even though their observed CCN concentrations are within a similar range. DACAD size distributions from the analyzed times during flight 8 are bimodal, with no indication of sampled pollution events, and \(dN/d\log D_p\) values were less than 5000 cm\(^{-3}\). Flight 17, however, was dominated by pollution events, as is indicated by its primarily multimodal size distributions that are dominated by particles with diameters smaller than 50 nm. Measured \(dN/d\log D_p\) values for flight 17 were as high as 60,000 cm\(^{-3}\), but most were under 10,000 cm\(^{-3}\). It makes sense, then, that the deviation of flight 17’s intercept from zero is greater than that of flight 8 because fewer particles are available for activation under polluted than clean conditions. When the slope is not near unity, the intercept can be affected by differences in CCN activities among the sampled air masses and may not have a simple interpretation.

On the basis of this initial comparison of \(N_O\) and \(N_P\) determined from DACAD size distributions with the assumption of 100% \((\text{NH}_4)_2\text{SO}_4\), it is apparent that more detailed aerosol properties, such as chemical composition and the mixing state, are needed to predict the CCN measurements at the operating supersaturations of the CCNC. Because such composition and mixing data were, however, unavailable on the Twin Otter during the IOP, an inverse closure study is performed, in which the comparison of \(N_O\) and \(N_P\) based on the assumption of pure \((\text{NH}_4)_2\text{SO}_4\) is used to infer the aerosol composition and mixing state that are consistent with CCN observations.

5.2. Inverse Aerosol/CCN Closure: Inferring Deviations From 100% \((\text{NH}_4)_2\text{SO}_4\)

An inverse aerosol/CCN closure evaluates the extent to which particle composition alone can explain the discrepancy between \(N_P\) and \(N_O\). Particle composition can be modeled as an internal or an external mixture and can consist of completely soluble and insoluble material or include partially soluble material, as well. For the purposes of the study presented here, particle components are considered to be either completely soluble or completely...
insoluble, and both internally and externally mixed aerosol populations are considered. Although the customary definition of an internal mixture is that all particles of the same size have the same chemical composition, a more simplified, size-independent internal mixture is used here in the absence of airborne measurements of size resolved chemistry. Thus, under the assumption of a size-independent internally mixed aerosol population, all particles of all diameters have the same fractional insoluble/soluble composition in this analysis. An external mixture can include purely soluble particles, purely insoluble particles, and particles composed of both insoluble and soluble material.

5.2.1. Estimating Aerosol Particle Composition From CCNC3 and DACAD Measurements

[37] The initial comparison of \( N_P / N_O \) (from section 5.1) can be used to determine the particle volumetric fraction of insoluble material (\( \varepsilon_{\text{ins}} \)) that would be necessary for \( \mu(N_P/N_O) \) to approach unity, assuming that the ambient aerosol is a size-independent internal mixture, with particles of all sizes having insoluble fraction \( \varepsilon_{\text{ins}} \). In order to calculate \( \varepsilon_{\text{ins}} \) for each flight, the cumulative aerosol size distribution is determined, with each size bin containing the concentration of all particles with diameters greater than the bin diameter. For each analysis period, the DACAD midbin diameter for which the cumulative aerosol concentration approaches the measured CCN concentration is chosen as the cutoff diameter, defined as the dry diameter above which sampled ambient particles would be able to activate under the assumption of size-independent internal mixing. A value for \( \varepsilon_{\text{ins}} \) is determined by inputting the cutoff diameter and different values of \( \varepsilon_{\text{ins}} \) (with the balance \((\text{NH}_4)_2\text{SO}_4\)) into the Köhler Theory model, until the particle’s calculated critical supersaturation approaches the operating supersaturation of the CCNC3.

[38] Since the sampled air mass properties typically varied widely during a flight, this analysis is carried out for each level leg during each flight. A level leg is defined as one for which consecutive static pressure measurements are constant within ±3 mbar. Some level legs are further divided if it appears that drastically different air masses were sampled during the same level leg. This usually occurs for level legs that cover long distances but was observed even during some shorter-distance legs. Table 3 gives the range and mean of \( \varepsilon_{\text{ins}} \) values for each flight. Most of the values of \( \varepsilon_{\text{ins}} \) that are required for \( \mu(N_P/N_O) \) to approach unity exceed 0.90, with only 14 out of the 113 analyzed level legs having \( \varepsilon_{\text{ins}} \) values less than 0.90. While insoluble mass fractions in aerosol particles in continental areas have been found as high as 0.98, values less than 0.6 are more

Figure 8. Altitude versus longitude trace for flights (a) 12 and (b) 14. Closure data points are indicated by the solid circles. The color of the closure data points indicates the magnitude of \( N_P/N_O \), assuming pure \((\text{NH}_4)_2\text{SO}_4\), for the corresponding DACAD scan.

Figure 9. Three-day HYSPLIT back trajectories for 15 May 2003 (flight 8) at the SGP site at altitudes of 450, 650, and 1000 m. The times given are UTC; 1700 UTC corresponds to 1200 LT.
common [Pruppacher and Klett, 1997]. Lower insoluble fractions would be expected in the ambient aerosol, especially if the aerosol were truly internally mixed independently of particle size. Under the assumption of a size-independent internal mixture of particles, the values of $e_{\text{ins}}$ that would be required to achieve agreement between $N_P$ and $N_O$ exceed the average fraction of insoluble material commonly found in continental aerosols. Other aerosol properties likely contributed to the discrepancy between $N_P$ and $N_O$.

### 5.2.2. CCN and DACAD Derived Insoluble Fraction (Flight 10)

[39] During the IOP, there was one flight (flight 10) during which CCN concentrations and DACAD wet and dry aerosol size distributions were all measured. From the wet and dry aerosol size distributions and assuming size-independent internally mixed aerosol population, an estimate of $e_{\text{ins}}$ can be obtained by comparing the cumulative particle concentrations of the wet and dry size distributions at each diameter. The average difference in wet and dry

#### Table 3. Insoluble Volumetric Fractions ($e_{\text{ins}}$) and Fraction of Particles Unavailable for Activation ($f_{\text{NA}}$) for Which $\mu(N_P/N_O) \approx 1$

<table>
<thead>
<tr>
<th>$s_i$, %</th>
<th>$d_{\text{aer}}$, nm</th>
<th>Flight</th>
<th>$e_{\text{ins}}$ (Range$^c$)</th>
<th>$e_{\text{ins}}$ (Mean$^c$)</th>
<th>$f_{\text{NA}}^d$ (Range$^c$)</th>
<th>$f_{\text{NA}}^d$ (Mean$^c$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.8</td>
<td>15</td>
<td>6</td>
<td>0.66 – 0.96</td>
<td>0.90</td>
<td>0.39 – 0.90</td>
<td>0.22</td>
</tr>
<tr>
<td>2.8</td>
<td>15</td>
<td>7</td>
<td>0.93 – 0.96</td>
<td>0.94</td>
<td>0.27 – 0.94</td>
<td>0.21</td>
</tr>
<tr>
<td>2.8</td>
<td>15</td>
<td>8</td>
<td>0.97 – &gt;0.99</td>
<td>0.98</td>
<td>0.37 – &gt;0.99</td>
<td>0.15</td>
</tr>
<tr>
<td>2.8</td>
<td>15</td>
<td>10</td>
<td>&gt;0.99 (all)</td>
<td>&gt;0.99</td>
<td>0.26 – 0.99</td>
<td>0.25</td>
</tr>
<tr>
<td>3.6</td>
<td>13</td>
<td>9</td>
<td>0.91 – &gt;0.99</td>
<td>0.91</td>
<td>0.61 – &gt;0.99</td>
<td>0.22</td>
</tr>
<tr>
<td>2.1</td>
<td>12</td>
<td>12</td>
<td>0.78 – 0.96</td>
<td>0.96</td>
<td>0.64 – 0.96</td>
<td>0.42</td>
</tr>
<tr>
<td>2.1</td>
<td>18</td>
<td>13</td>
<td>0.95 – 0.98</td>
<td>0.96</td>
<td>0.61 – 0.96</td>
<td>0.40</td>
</tr>
<tr>
<td>2.1</td>
<td>18</td>
<td>14</td>
<td>0.98 – &gt;0.99</td>
<td>&gt;0.99</td>
<td>0.63 – &gt;0.99</td>
<td>0.42</td>
</tr>
<tr>
<td>2.1</td>
<td>18</td>
<td>15</td>
<td>&gt;0.99 (all)</td>
<td>&gt;0.99</td>
<td>0.70 – &gt;0.99</td>
<td>0.44</td>
</tr>
<tr>
<td>2.1</td>
<td>18</td>
<td>17</td>
<td>&gt;0.99 (all)</td>
<td>&gt;0.99</td>
<td>0.91 – &gt;0.99</td>
<td>0.63</td>
</tr>
</tbody>
</table>

$^a$Size-independent internal mixing is assumed in the determination of $e_{\text{ins}}$.

$^b$The ranges of values for $e_{\text{ins}}$ and $f_{\text{NA}}$ are determined from those averaged over each of the level legs during a flight.

$^c$The mean values of $e_{\text{ins}}$ and $f_{\text{NA}}$ are averaged over all analysis points.

$^d$NP is determined under the assumption of pure (NH$_4$)$_2$SO$_4$ in the determination of $f_{\text{NA}}$. 

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**Figure 10.** Three-day HYSPLIT back trajectories for 29 May 2003 (flight 17) (a) at the SGP site at altitudes of 400, 800, and 3000 m and (b) at site EF-19 at altitudes of 650, 800, and 1000 m. The times given are UTC; 1700 UTC corresponds to 1200 LT.
The soluble particle fraction is assumed to be composed

\[ e = \text{sol} \]

where \( e_{\text{sol}} \) is the growth factor of soluble material. If it is assumed that no soluble organic material is present in the aerosol particles, then

\[ G(RH) = e_{\text{ins}} G(RH)_{\text{sol}} \]

is the growth factor of insoluble material for the determined value of \( G(RH)_{\text{Total}} \). Then, equation (1) can be solved for \( e_{\text{ins}} \) as the insoluble fraction estimated from the DACAD wet and dry aerosol size distributions,

\[ e_{\text{ins}} = G(RH)_{\text{sol}} = \frac{G(RH)_{\text{Total}} - G(RH)_{\text{sol}}}{1 - G(RH)_{\text{sol}}} \]

The soluble particle fraction is assumed to be composed of deliquesced (NH₄)₂SO₄, for which the value of \( G(RH)_{\text{sol}} \) is 1.35 at the approximate DACAD operating RH of 70%.

The average value of the growth factor for the aerosol populations during flight 10, \( G(RH)_{\text{ave}} \), is 1.05, and the average \( e_{\text{ins}} \) is 0.78. The \( G(RH)_{\text{Total}} \) and \( e_{\text{ins}} \) values for each DACAD scan, and the resulting \( \mu(N_p/N_O) \), are given in Table 4. \( N_p \) is the summation of particles from the DACAD size distribution that have diameters greater than a cutoff diameter, \( D_c \), which is the activation diameter, because \( G(RH)_{\text{Total}} \) is based on the difference between the wet and dry diameters of the particle, which is directly related to the water uptake by the particle and is more directly applicable to CCN activity. The total growth factor can be used to estimate \( e_{\text{ins}} \) from,

\[ G(RH)_{\text{Total}} = e_{\text{ins}} G(RH)_{\text{org}} + e_{\text{sol}} G(RH)_{\text{sol}} \]

where \( e_{\text{org}} \) is the volumetric fraction of organic material, \( G(RH)_{\text{org}} \) is the growth factor of organic material, \( e_{\text{ins}} \) is the insoluble fraction of soluble material, and \( G(RH)_{\text{sol}} \) is the growth factor of soluble material. If it is assumed that no soluble organic material is present in the aerosol particles, then

\[ G(RH)_{\text{Total}} = e_{\text{ins}} G(RH)_{\text{sol}} \]

is the volumetric fraction of organic material, \( G(RH)_{\text{org}} \) is the growth factor of organic material, \( e_{\text{ins}} \) is the insoluble fraction of soluble material, and \( G(RH)_{\text{sol}} \) is the growth factor of soluble material. If it is assumed that no soluble organic material is present in the aerosol particles, then

\[ G(RH)_{\text{Total}} = e_{\text{ins}} G(RH)_{\text{sol}} \]

is the growth factor of insoluble material for the determined value of \( G(RH)_{\text{Total}} \). Then, equation (1) can be solved for \( e_{\text{ins}} \) as the insoluble fraction estimated from the DACAD wet and dry aerosol size distributions,

\[ e_{\text{ins}} = G(RH)_{\text{sol}} = \frac{G(RH)_{\text{Total}} - G(RH)_{\text{sol}}}{1 - G(RH)_{\text{sol}}} \]

The soluble particle fraction is assumed to be composed of deliquesced (NH₄)₂SO₄, for which the value of \( G(RH)_{\text{sol}} \) is 1.35 at the approximate DACAD operating RH of 70%.

The average value of the growth factor for the aerosol populations during flight 10, \( G(RH)_{\text{ave}} \), is 1.05, and the average \( e_{\text{ins}} \) is 0.78. The \( G(RH)_{\text{Total}} \) and \( e_{\text{ins}} \) values for each DACAD scan, and the resulting \( \mu(N_p/N_O) \), are given in Table 4. \( N_p \) is the summation of particles from the DACAD size distribution that have diameters greater than a cutoff diameter, \( D_c \), which is the activation diameter, because \( G(RH)_{\text{Total}} \) is based on the difference between the wet and dry diameters of the particle, which is directly related to the water uptake by the particle and is more directly applicable to CCN activity. The total growth factor can be used to estimate \( e_{\text{ins}} \) from,

\[ G(RH)_{\text{Total}} = e_{\text{ins}} G(RH)_{\text{org}} + e_{\text{sol}} G(RH)_{\text{sol}} \]

where \( e_{\text{org}} \) is the volumetric fraction of organic material, \( G(RH)_{\text{org}} \) is the growth factor of organic material, \( e_{\text{ins}} \) is the insoluble fraction of soluble material, and \( G(RH)_{\text{sol}} \) is the growth factor of soluble material. If it is assumed that no soluble organic material is present in the aerosol particles, then

\[ G(RH)_{\text{Total}} = e_{\text{ins}} G(RH)_{\text{sol}} \]

is the volumetric fraction of organic material, \( G(RH)_{\text{org}} \) is the growth factor of organic material, \( e_{\text{ins}} \) is the insoluble fraction of soluble material, and \( G(RH)_{\text{sol}} \) is the growth factor of soluble material. If it is assumed that no soluble organic material is present in the aerosol particles, then
such that $\mu(N_p/N_\Omega) \approx 1$ is calculated from the following equation,

$$f_{NA} = \frac{N_{PEA}}{N_p} = \frac{N_p - N_\Omega}{N_p} = 1 - \frac{N_\Omega}{N_p}$$

(3)

where $N_{PEA}$ is the number concentration of the externally mixed particles that are unavailable for activation. The statement that $N_{PEA} = N_p - N_\Omega$ assumes that the CCNC3 was able to count all particles that were capable of activating at the operating supersaturations, which is likely at such high supersaturations where kinetic effects would not be dominant. Table 3 gives the average $f_{NA}$ for each flight and the results are discussed further in section 5.3.3.

5.3.2. Aerosol Mixing State Inferred With DACAD Derived Insoluble Fraction (Flight 10)

[43] Along with the $e_{ins, DACAD}$ estimates, which were not sufficient to obtain agreement between $N_p$ and $N_\Omega$, an additional estimate of the extent of external mixing of the aerosol population can be calculated for flight 10. For this analysis, the $f_{NA}$ that leads to $\mu(N_p/N_\Omega) \approx 1$ is calculated by considering $e_{ins, DACAD}$ (constant for particles of all diameters) with the balance (NH$_4$)$_2$SO$_4$ in the calculation of $N_\Omega$ Table 4 shows the values of $f_{NA}$ that satisfy $\mu(N_p/N_\Omega) \approx 1$ for each DACAD scan during flight 10. The average value of $f_{NA}$ in Table 4 is 0.22, which is close to the average value of $f_{NA}$ (0.25) for flight 10 with $e_{ins} = 0$. The similar values of $f_{NA}$ calculated with and without the maximum value of $e_{ins}$ for a size-independent internal mixture of particles ($e_{ins, DACAD}$) indicates that the aerosol was an external mixture that included some purely insoluble particles. Particles with an insoluble fraction of $e_{ins, DACAD}$ that were unable to activate at the operating supersaturations of the CCNC3 made up only 3% of the total aerosol population; 22% were purely insoluble particles (that did not activate), and 78% of the particles were able to activate.

5.3.3. Aerosol Mixing State Discussion

[46] The values of $f_{NA}$ follow a similar trend to that seen for $\mu(N_p/N_\Omega)$ in section 5.1, with more particles unavailable for activation during flights that encountered polluted conditions. The aerosol size distribution feature that distinguishes between polluted and clean conditions is the predominance of small particles with diameters less than 30 nm under polluted conditions. The increase in $f_{NA}$ values with increasing polluted conditions, therefore, indicates that the particles that are unavailable for activation at the operating supersaturations of the CCNC3 are predominantly those with diameters less than 30 nm. This is consistent with the findings of Gasparini et al. [2006], that the smallest analyzed particles were largely composed of nonhygroscopic compounds (see section 4.1). At the relatively high operating supersaturations of the CCNC3, these small (<30 nm), largely nonhygroscopic particles would make up the majority of the particles that remained unactivated. Considering that the insoluble fractions in a size-independent internal mixture required for $N_p$ and $N_\Omega$ agreement in section 5.2 exceed 0.90 for 99 out of the 113 level legs analyzed, $f_{NA}$ probably consists of some pure insoluble particles, with a range of diameters. An internal mixture of particles with an even distribution of insoluble material across particle diameters is not expected, especially at the location of the SGP site, where sampled aerosol varies in both source and age.

5.4. CCN and DMA/TDMA Derived Mixing State and Particle Composition

[45] Size-resolved particle composition and mixing state of ambient aerosol were derived from the Texas A&M DMA/TDMA ground data from the IOP [Gasparini et al., 2006]. For each size bin, the DMA/TDMA measurements can be used to calculate the fraction of the particles in that size bin that are purely soluble material, purely insoluble material, and a mixture of soluble and insoluble material. The fraction of insoluble material in the mixed particles is also calculated. It was assumed that the purely insoluble particles did not activate at the operating supersaturations of the instrument. The purely soluble particles were assumed to activate if the size bin diameter was greater than the critical diameter of (NH$_4$)$_2$SO$_4$ at the operating supersaturation of the CCNC3 instrument. The mixed particle soluble and insoluble fractions were input into the Köhler model to determine whether particles with this chemical composition and diameters equal to the middiameter of the size bin were able to activate at the operating supersaturation of the CCNC3 instrument.

[46] The flights for which there were simultaneous CCN measurements, DACAD size distributions, and DMA/ TDMA-derived aerosol population properties were flights 8, 9, and 16. For this analysis, included level legs were limited to being within 8047 m (5 miles; horizontal distance) of the SGP CART site and within the boundary layer, where the ground DMA/TDMA-derived aerosol properties are more likely to be valid for flight data. The boundary layer height was calculated using the procedure described by Delle Monache et al. [2004], which uses Balloon-Borne Sounding System data. The boundary layer height is defined as the lowest altitude within a potential temperature inversion at which a critical lapse rate and threshold potential temperature are exceeded. The following thresholds were determined by Delle Monache et al. [2004] to apply at the SGP CF: $\theta_{Top} - \theta_{Bottom} = 2$ K and $\Delta \theta/\Delta z = 0.001$ K m$^{-1}$, where $\theta_{Top}$ is the potential temperature at the top of the boundary layer in Kelvin (K), $\theta_{Bottom}$ is the potential temperature at the bottom of the boundary layer in K, $\theta$ is the potential temperature in K, and $z$ is the altitude in m. DMA/TDMA data were used to classify aerosol particle components as externally mixed insoluble, externally mixed soluble, internally mixed insoluble, and internally mixed soluble, and these aerosol properties were averaged over the diameter ranges of the DACAD. DACAD scan times within the DMA/TDMA size distribution scan times (~5 min) were used.

[47] The $\mu(N_p/N_\Omega)$ values resulting from the DMA/ TDMA-derived properties are given in Table 5. For flights 8 and 9, the $\mu(N_p/N_\Omega)$ values are within instrument uncertainty, but they are still significantly greater than unity for flight 16, even though using the DMA/TDMA-derived aerosol properties decreased the $\mu(N_p/N_\Omega)$ value significantly, by about 73%. The flight traces for flights 8 and 9 (Figures 11 and 12) show well-mixed boundary layers for the time periods during which the TWIN OTTER was sampling within the boundary layer and within 8 km of the ground site. The boundary layer during the time period studied for
flight 16 is less well mixed, as is illustrated by its flight trace in Figure 13. The time period within which the Twin Otter was sampling within the boundary layer and within 8 km of the ground site was near the beginning of flight 16, from about 1849 to 1851 UTC. Considering that the DMA/TDMA data were measured at the ground, it is not unexpected that the agreement between $N_P$ and $N_O$ is more satisfactory for well-mixed boundary layers than non-well-mixed boundary layers.

A comparison of the DMA/TDMA and DACAD aerosol size distributions illustrates this point further. Figure 14 compares the DMA/TDMA and DACAD aerosol and predicted CCN size distributions for flights 8 and 9; Figure 15 illustrates the same for flight 16. Both of the CCN size distributions (DMA/TDMA and DACAD) are calculated using the size-resolved chemistry derived from the DMA/TDMA measurements. For the DACAD CCN size distributions, the DMA/TDMA size-resolved chemistry is applied to the DACAD aerosol size distributions, which assumes that the DMA/TDMA-derived chemistry applies at the sampling altitude of the DACAD. As can be seen, this assumption seems to hold for flights 8 and 9 but not flight 16.

During the sampling period 1849 to 1851 UTC during flight 16, the CPC and DACAD exhibit sharp peaks in number concentration. The DACAD aerosol size distribution shows that these peaks are caused by increases in small particles that do not show up in the DMA/TDMA size distribution for the same time period. This indicates that the layers that were sampled by the Twin Otter did not necessarily reach the ground and that the aerosol layers within the boundary layer were stratified. The DELTA (Detection and Evaluation of the Long-Range Transport of Aerosols) rotating drum impactor aerosol composition results from the SGP site measurements also indicate that the layers sampled aloft on 25–29 May did not reach the ground [Cahill et al., 2004]. Another possibility is that the DMA/TDMA, which has a longer total sequence scan time ($\tau$) than the DACAD (72.5 s), was unable to resolve rapid changes in the aerosol size distribution, which may have occurred during the time period of interest. The DMA/TDMA size-

### Table 5. $\mu(N_P/N_O)$ and $\sigma(N_P/N_O)$ Values for Flights 8, 9, and 16 Using $\varepsilon_{int}$ and External Mixing Properties Derived From DMA/TDMA Data

<table>
<thead>
<tr>
<th>$\delta$, %</th>
<th>$d_{a,b}$, nm</th>
<th>Flight</th>
<th>Number of DACAD Scans Included</th>
<th>Without DMA/TDMA Derived Properties ($\varepsilon_{int} = 0$, $f_{mix} = 0$)</th>
<th>With DMA/TDMA Derived Properties</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.8</td>
<td>15</td>
<td>8</td>
<td>4</td>
<td>1.18</td>
<td>1.00</td>
</tr>
<tr>
<td>3.6</td>
<td>13</td>
<td>9</td>
<td>1</td>
<td>1.14</td>
<td>1.10</td>
</tr>
<tr>
<td>2.1</td>
<td>18</td>
<td>16</td>
<td>3</td>
<td>5.38</td>
<td>1.47</td>
</tr>
</tbody>
</table>

*The results without DMA/TDMA derived properties are also given for the same DACAD scans. This table only includes times during which the Twin Otter was within 5 miles of the SGP CART site and within the boundary layer.

Figure 11. Flight trace for flight 8, showing a well-mixed boundary layer between approximately 1730 and 1840 UTC when the Twin Otter was sampling beneath ~1000 m. The error bars on the CCN trace indicate the 10% uncertainty level. The times given are UTC; 1700 UTC corresponds to 1200 LT.
resolved composition does not apply to the air masses sampled by the Twin Otter for flight 16 because the shape of the DMA/TDMA and DACAD sampled aerosol size distributions are not similar. If the DMA/TDMA and DACAD size distribution shapes are similar, as is true for flights 8 and 9, then the DMA/TDMA composition results can be coupled with airborne aerosol size distribution measurements to predict CCN concentrations with-

![Figure 12.](image1.png)

**Figure 12.** Flight trace for flight 9, showing a well-mixed boundary layer between approximately 2200 and 2300 UTC when the Twin Otter was sampling beneath ~1400 m. The error bars on the CCN trace indicate the 10% uncertainty level. The times given are UTC; 1700 UTC corresponds to 1200 LT.

![Figure 13.](image2.png)

**Figure 13.** Flight trace for flight 16, showing the lack of a well-mixed boundary layer from approximately 1820 to 1940 UTC when the Twin Otter was sampling beneath ~2250 m. The error bars on the CCN trace indicate 20% uncertainty level. The times given are UTC; 1700 UTC corresponds to 1200 LT.
in the boundary layer and within a reasonable horizontal distance from the ground measurement site.

### 6. Case Studies: “Clean” Conditions, Local Pollution, and Siberian Smoke Events

The inverse aerosol/CCN closure study (assuming 100% (NH₄)₂SO₄) indicates a division of the flights into three groups, based on the value of the closure ratio of \( N_p \) to \( N_D \). Flights 6 through 10 (14–18 May) have \( \mu(N_p/N_D) \) values between 1.18 and 1.34, while values for flights 12 through 15 (21–27 May) are between 1.78 and 1.87, and values for flights 16 and 17 (28 and 29 May) are 3.68 and 3.45, respectively. This division seems independent of the instrument operating supersaturation. Operating supersaturations of 2.8% and 3.6% are included in flights 6 through 10, which have similar \( \mu(N_p/N_D) \) values. The operating supersaturation (2.1%) is the same for flights 12 through 17, although each flight has different \( \mu(N_p/N_D) \) values. ARM Aerosol IOP flights fall into categories consistent with relatively clean conditions with intermittent pollution events (flights 6–10), conditions influenced by local pollution and/or smoke events (flights 12–15), and conditions dominated by local pollution events (flights 16–17) [Schmid et al., 2004]. Aerosol size distributions from the DACAD are especially illustrative of the different categories of sampled air masses. In the following discussions, only DACAD scan times that were used in the inverse closure analysis are considered and presented.

During the first two weeks of the ARM Aerosol IOP (5–18 May, flights 6–10), relatively clean atmospheric conditions were noted. Also during this time, severe weather moved through north-central Oklahoma, including a “thunderstorm and high wind” event on 16 May (NOAA National Climatic Data Center (NCDC) access via Department of Commerce (DOC) NOAA National Environmental Satellite, Data, and Information Service (NESDIS) NCDC website, http://lwf.ncdc.noaa.gov/oa/ncdc.html). This may be one reason that the atmosphere was relatively clean. DACAD size distributions early in flights 6–10 (14–
(18 May) have unimodal or bimodal distributions with median diameters between about 40 and 160 nm and peak \( \frac{dN}{d\log D_p} \) values less than 5000 cm\(^3\)/C0. Aerosol size distributions for flight 6 begin to deviate from these characteristics about 45 min after takeoff and continue to deviate until landing. Aerosol size distributions exhibit these “clean” characteristics throughout the analyzed times during flights 7, 8 and 10. For flight 9, aerosol size distributions exhibit these “clean” characteristics near the end, as well as early in, the flight, but deviate for midflight times. The \( \mu(\frac{N_P}{N_O}) \) values for these clean background periods are 1.19, 1.33, 1.18, 1.21, and 1.34 for flights 6–10, respectively.

[52] Background closure ratios for flights 9 and 10 (1.21 and 1.34, respectively) on 17 and 18 May may include data points related to a sulfur event in the SGP area. Around 17 May, Cahill et al. [2004] noted, on the basis of their ground site data, an aerosol sulfur episode caused by an air mass that they predict was over Ohio 2 days earlier. Most of the aerosol size distributions from flights 9 and 10 are unimodal with median diameters greater than 100 nm, which could be indicative of an aged aerosol population and which is consistent with the findings of Cahill et al. [2004]. Some aerosol size distributions from flight 9 indicate the influence of local aerosol sources, as will be discussed.

[53] After 18 May, the aerosol size distributions are unimodal or bimodal with median diameters between 40 and 80 nm and peak \( \frac{dN}{d\log D_p} \) values less than 10,000 cm\(^3\)/C0. Aerosol size distributions with these “dirtier” background aerosol characteristics were found during flights 12–17 (21–29 May). The \( \mu(\frac{N_P}{N_O}) \) values for background conditions during flights 12–17 are 1.36, 1.28, 1.70, 1.54, 2.30, and 2.30, respectively.

[54] At later times during flight 6, midtimes during flight 9, and times throughout flights 12–17, measured aerosol size distributions indicated the presence of local pollution sources. Elevated aerosol layers were visually observed at altitudes of \( \sim 3700 \) m during flight 6, \( \sim 2500 \) m during flight 9, \( \sim 3000 \) and 3700 m during flight 14, and \( \sim 2000, 3000, 3700, \) and 5000 m during flight 15. Some DACAD size distributions from flights 6, 7, 9, and 12–17 have characteristically low median diameters (sometimes less than the cutoff diameter of the DACAD, \( \sim 17 \) nm) and particularly large concentrations of particles with diameters less than 30 nm. These aerosol size distributions are consistent with those from locations that are downwind of a fresh pollution source, such as a coal-fired power plant [Brock et al., 2002]. Ground-based PILS-IC results show that for flights 12, 14, and 15 (21, 25, and 27 May) NH\(_4\)+ to SO\(_4^{2-}\) molar ratios much less than 2 were observed [Pahlow et al., 2006], which could indicate addition of sulfate to particles through vapor and aqueous phase reactions. The SGP site is near two power plants: the Ponca City and Conoco power plants, and IOP flight patterns passed near at least four other industrial and/or power plants, as well.

Figure 15. Comparison of DMA/TDMA and DACAD size distributions for flight 16. The error bars give the standard deviation of the DACAD aerosol size distributions averaged over the DMA/TDMA scan times. The times given are the DACAD midscan times in UTC; 1700 UTC corresponds to 1200 LT. The area under the aerosol size distribution curve gives the total aerosol number concentration, and the area under the CCN size distribution curve gives the predicted CCN number concentration. Both DMA/TDMA and DACAD predicted CCN distributions were calculated using the insoluble fraction and external mixing properties determined from the DMA/TDMA data.
Table 6. Summary of the Values of $\mu(N_p/N_O)$ for Each Flight, Divided Into Categories Determined From DACAD Size Distributions*

<table>
<thead>
<tr>
<th>$s$, %</th>
<th>$d_{np}$, nm</th>
<th>Flight</th>
<th>Background</th>
<th>Polluted</th>
<th>Smoke</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.8</td>
<td>15</td>
<td>6</td>
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<td>2.8</td>
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<td>1.33</td>
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<tr>
<td>3.6</td>
<td>13</td>
<td>9</td>
<td>1.21</td>
<td>1.26</td>
<td>NE</td>
</tr>
<tr>
<td>2.1</td>
<td>18</td>
<td>12</td>
<td>1.36</td>
<td>1.89</td>
<td>NE</td>
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<td>1.54</td>
<td>2.73</td>
<td>1.61</td>
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<td>16</td>
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</tr>
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<td>18</td>
<td>17</td>
<td>2.30</td>
<td>3.75</td>
<td>NE</td>
</tr>
</tbody>
</table>

*NE indicates that the condition was “not encountered” during the DACAD scan times used in the analysis.

Table 6 summarizes the $\mu(N_p/N_O)$ values for all flights when divided into categories based on the shape and magnitude of the DACAD aerosol size distribution. In general, the $\mu(N_p/N_O)$ values are lower for the clean conditions than the more obviously polluted conditions.

Siberian smoke layers were observed in the SGP CF area during the period 25–28 May, 2003 (flights 14–17) [Schmid et al., 2004]. DACAD aerosol size distributions indicate that smoke layers were sampled at altitudes of $\sim$3700 m on flight 14 and $\sim$3100 m on flight 15. The Siberian fires were detected in April but escalated greatly in May. In transport simulations beginning on 10 May 2003, the smoke plume from Siberia reached Canada in about 11 days, with a smoke maximum on 23 May in Manitoba, Canada [Damoah et al., 2004]. HYSPLIT [Draxler and Rolph, 2003; Rolph, 2003] back trajectories for flight 14 on 25 May 2003 show that air masses 3000 and 4000 m over the ARM SGP site were over Canada 2 days prior to sampling, which corresponds with the smoke maximum in Manitoba on 23 May.

On 25 May 2003 (flight 14), the observed elevated layers were characterized by large values of dry extinction and backscatter, which are consistent with aged smoke particles [Ghan et al., 2006]. The DACAD size distributions from these smoke events tend to have a large accumulation mode, with median diameters greater than 100 nm. This is consistent with aged aerosol populations originating from biomass burning, in which processes such as particle coagulation and cloud processing have occurred and have decreased the peak concentration and increased the median diameter of the size distribution [Radke et al., 1995].

The ability of smoke particles to act as CCN has been shown to increase with smoke plume age [Andreae and Merlet, 2001]. The critical supersaturation of smoke particles is reduced, and thus their CCN ability increased, through cloud processing, in which condensational uptake of water-soluble material is possible. Past studies have found that the ratio of CN to CCN at 1% supersaturation is about 1.7 in a slightly aged (1–2 hours) smoke plume [Andreae and Merlet, 2001]. The Siberian smoke aerosols were transported over a large distance, which included time spent over industrial and desert regions. Therefore, along with aged smoke particles, it is likely that industrial pollutants and some mineral dust were present in these smoke layers [Jaffe et al., 2004]. This may explain why the $\mu(N_p/N_O)$ values, given in Table 6, differ from those of other studies of aged smoke plumes.

The layers that were analyzed in the closure analysis for flights 16 and 17 have aerosol size distributions that are more consistent with local pollution sources than with aged smoke particles. Flight 17 was a MODIS overpass flight, which sampled above three ARM ground sites, in addition to the SGP CF. During flight 17, the Twin Otter flew near five power plants, so the aerosol size distribution characteristics are not unexpected.

7. Conclusions

The inverse aerosol/CCN closure study has revealed possible features of the aerosol sampled during the ARM Aerosol IOP in the absence of direct airborne aerosol composition measurements. The ARM SGP site is greatly influenced by local aerosol sources, as well as long-range sources, and the aerosol CCN activity at high supersaturations was greatly influenced by the chemical characteristics and mixing state of the aerosols in the sampled air masses. The characteristics of air masses sampled during the ARM Aerosol IOP ranged from those of relatively clean air masses to those of air masses influenced by local and long-range pollution events to those of aged smoke plumes. These air masses can be categorized by aerosol size distribution, as well as by CCN behavior as indicated in the ratio of $N_p$ to $N_O$. It has been shown that the general low aerosol CCN activity that was observed in this inverse aerosol/CCN study is consistent with other measurements from the IOP.

The DACAD aerosol size distributions measured during the ARM Aerosol IOP were highly variable, even on level legs of approximately constant altitude. This could account for some of the disagreement between $N_p$ and $N_O$ because the DACAD is not able to resolve rapid changes in aerosol size distributions because of its longer measurement time compared to that of the CCNC3.

If the CCNC3 instrument had operated at lower supersaturations that are more representative of those found in the atmosphere, the inverse aerosol/CCN closure study may have resulted in significantly different $N_p$ to $N_O$ ratios. The only indication that we have of the size resolved aerosol composition in this area is the DMA/TDMA derived results. These results indicate that, in general, material of low hygroscopicity was preferentially distributed among the smallest measured particles. This is consistent with in-cloud aerosol processing and/or fresh pollution sources, and it is likely that both of these scenarios were encountered during the ARM Aerosol IOP because of the close proximity of power and industrial plants and the verified Siberian smoke events near the end of the month.

Although some of the chemistry effects may not have been captured at smaller supersaturations, future measurements should be made at lower operating supersaturations to better determine the ability to predict CCN concentration using surface data. However, for a continental location such as the ARM SGP site that is influenced by many different aerosol sources, ranging from local to international, predicting CCN concentrations from aerosol size distributions and surface aerosol composition measurements alone is proba-
bly not adequate. Future studies should also include airborne aerosol composition measurements to allow more complete aerosol/CCN closure studies.

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