Deep convection as a source of new particles in the midlatitude upper troposphere

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[1] A case study of new particle formation in the region downwind of a mesoscale convective system stretching across much of the central United States is presented. Airborne measurements were made of condensation nuclei (CN), cloud particle surface area, water vapor, and other gases. CN concentrations were greatly enhanced above and downwind of the cirrus anvil, with maximum concentrations of 45,000 per standard cm^3. Volatility and electron microscope measurements indicated that most of the particles were likely to be small sulfate particles. The enhancement extended over at least a 600-km region. Multivariate statistical analysis revealed that high CN concentrations were associated with surface tracers, as well as convective elements. Convection apparently brings gas-phase particle precursors from the surface to the storm outflow region, where particle nucleation is favored by the extremely low temperatures. Simple calculations showed that deep convective systems may contribute to a substantial portion of the background aerosol in the upper troposphere at midlatitudes. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0320 Atmospheric Composition and Structure: Cloud physics and chemistry; 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; *KEYWORDS:* particles, aerosols, sulfuric acid, nucleation, cirrus clouds, convection

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1. Introduction

[2] New aerosol particles may form from gas-phase sulfuric acid and water through homogeneous, bimolecular nucleation. Particle formation is favored by high concentrations of sulfur dioxide and hydroxyl radical (which form sulfuric acid vapor), high water vapor concentrations and cold temperatures. New particle formation is also thought to occur more often in clean air, since sulfuric acid will readily condense on existing particles if sufficient surface area is available. While conditions necessary for homogeneous nucleation do occur in the atmosphere, observations suggest that in some cases, new particles form more readily than models can predict for the homogeneous nucleation of sulfuric acid and water alone [*Weber et al.*, 1995, 1997]. Thus other species, including ammonia and organic vapors, may participate in nucleation of new particles in some parts of the atmosphere [e.g., *Ball et al.*, 1999; *Kulmala et al.*, 2000].

[3] This paper presents measurements of young particles formed in the outflow region of a mesoscale convective system (MCS) over the midwestern United States. A companion paper [*Clement et al.*, 2002] includes model calculations for this case, assuming homogeneous nucleation of sulfuric acid and water alone.

2. Previous Work

[4] Many of the studies where recently nucleated particles have been measured are given in Table 1. The observations are characterized by large number concentrations of fine particles. Detection of these small particles is possible using condensation nucleus counters (CNCs), which create an environment where small particles grow to sizes that can be detected by optical techniques [*Agarwal and Sem*, 1980]. Traditional CNCs measure the number concentration of particles larger than $\sim$0.01 μm at 1 atmosphere, while modifications used in ''ultrafine'' CNCs allow detection of even smaller particles [*Stolzenburg and McMurry*, 1991]. The counting efficiency of these instruments varies not only

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Table 1. Previous Observations Related to New Particle Production

Reference	Location[a]	Concentration Maximum,[b] cm^{-3}
Hegg et al. [1990]	midlat MBL (Pac), above, in clean stratus	6,000
Hoppel et al. [1990]	midlat MBL (Atl), cloud free	20,000
Hegg [1991]	midlat MBL (Pac), in marine stratus	600
Radke and Hobbs [1991]	midlat cont, near cumulus	12,000
Covert et al. [1992]	midlat clean MBL, broken cloud layer	3,200
Clarke [1992, 1993]	UT (Pac), especially above cloud	80,000
Gras [1993]	Antarctic coast, surface	600
Hofmann [1993]	midlat cont UT	25,000
Hoppel et al. [1994]	MBL and FT (Pac), especially above cloud	6,000
Perry and Hobbs [1994]	midlat, downwind of marine cumulus	4,200
Brock et al. [1995]	tropical UT	30,000
Ferek et al. [1995]	Arctic over ocean	5,400
Weber et al. [1995]	Pac MBL/FT boundary (Hawaii)	140
Covert et al. [1996]	Pac MBL (produced in FT?)	2,000
Clarke et al. [1997]	midlat free trop (Atl)	6,000
O'Dowd et al. [1997]	Weddell Sea MBL (Antarctic)	4,300
Raes et al. [1997]	subtropical free trop, cloud free	4,000
Schröder and Ström [1997]	midlat cont UT	10,000
Thornton et al. [1997]	tropical UT, near convection	18,000
Weber et al. [1997] and *Eisele and McMurry* [1997]	midlat clean cont, free trop air	600
Wiedensohler et al. [1997]	downwind of orographic cloud, night	1,200
Grenfell et al. [1999]	midlat MBL	100,000
Ström et al. [1999]	midlat UT, near cirrus anvil	25,000
Wang et al. [2000]	midlat UT (Atl), with convection	30,000
Mäkelä et al. [1997] and *Kulmala and Hämeri* [2000]	Finnish forest nucleation, multiple events	40,000
O'Dowd et al. [1999] and *O'Dowd and Hämeri* [2000]	coastal nucleation, 3 types, multiple events	1,000,000
Twohy et al. (this paper)	midlat cont UT, above cirrus anvil	45,000

[a] Definitions are as follows: MBL, marine boundary layer; FT, free troposphere; UT, upper troposphere; cont, continental; trop, troposphere; midlat, midlatitudes; Pac, Pacific Ocean; and Atl, Atlantic Ocean.

[b] Concentrations are given as reported in each paper and are not necessarily corrected to standard temperature and pressure, as the distinction was not always made. Also, some studies used fine-particle counters (minimum size 0.01–0.02 μm), while others used ultrafine particle counters (minimum size ~0.003 μm).

with particle size but also with ambient pressure [*Zhang and Liu*, 1991]. Table 1 includes observations using both types of counters and at a number of pressures; thus the particle size range for which these concentrations apply varies. In all cases, however, number concentrations were high enough to be identified by researchers as substantially above background conditions. In many cases, ancillary measurements, such as particle volatility or gas-phase measurements, were used to distinguish nucleation events from pollution incursions.

[5] New particles have been observed from the boundary layer to the tropopause, from the Antarctic to the Arctic, in or near clouds, and in cloud-free air. Most of these episodes occur over relatively small timescales and spatial scales. While measurements in the upper troposphere are relatively rare, these regions often contain the highest numbers of small particles observed [e.g., *Clarke*, 1992, 1993; *Brock et al.*, 1995; *Ström et al.*, 1999; *Nyeki et al.*, 1999]. Of particular interest in the upper troposphere is the outflow region of convective storms, where cold temperatures, high relative humidities, low aerosol surface area, and high concentrations of gas-phase precursors all coincide. Particles formed in the upper troposphere, if given sufficient time for growth by condensation and coagulation, may act as nuclei for cirrus or water clouds.

[6] Modeling studies have shown that small changes in temperature and relative humidity may produce large changes in the formation rate of new particles [*Yue and Hamill*, 1979]. Mixing of air from different layers may also produce the supersaturations needed to trigger particle formation [*Easter and Peters*, 1994]. Regions near clouds are expected to be favored sites because of high relative humidities and low aerosol surface area resulting from the scavenging effects of clouds. Enhanced actinic flux [*Madronich*, 1987] near clouds also can lead to high hydroxyl radical concentrations, which participate in particle formation. Within clouds, new particle production may be suppressed because of the large particle surface area, which provides preexisting sites for sulfuric acid condensation. However, *Hegg* [1991] presents observations of new particles apparently generated within clean stratus clouds.

[7] Two types of nucleation events have been observed in which the main condensing species are not sulfuric acid and ammonia: in Finnish forests [*Mäkelä et al.*, 1997] and off the Irish coast [*O'Dowd et al.*, 1999]. Their characteristics, covering many events, have been explored in large cooperative European campaigns, Biogenic Aerosol Formation in the Boreal Forest (BIOFOR) and New Particle Formation and Fate in the Coastal Environment (PARFORCE), whose results have recently been reported [*Kulmala and Hämeri*, 2000; *O'Dowd and Hämeri*, 2000].

3. Observations

[8] The observations presented here were made from the NASA DC-8 aircraft as part of the Subsonic Assessment:

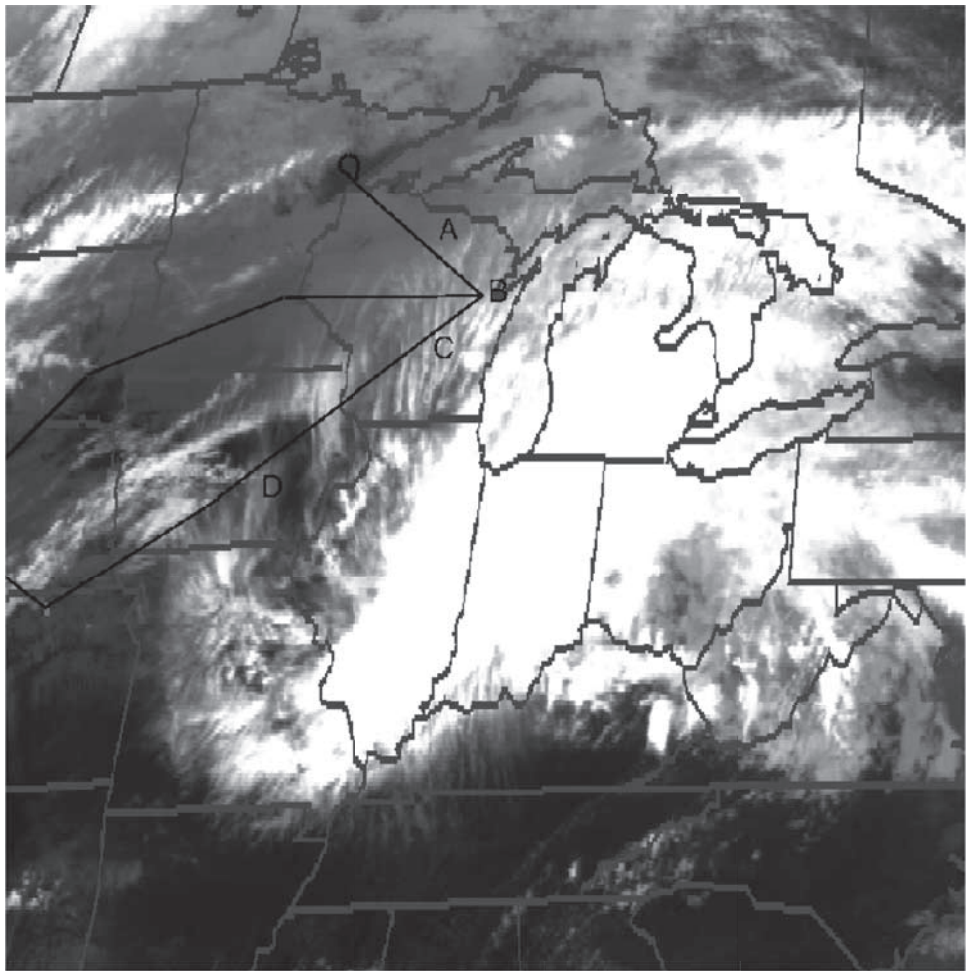

Figure 1. GOES 8 infrared satellite image of mesoscale convective system sampled on 8 May 1996, with approximate DC-8 flight track superimposed. Points A, B, C, and D are discussed in text.

Contrails and Cloud Effects Special Study (SUCCESS). Two flights were made near cirrus anvil outflow from convection in the upper troposphere. While some enhancement in fine-particle concentration was observed downwind of an isolated convective system on 21 April 1996, much higher concentrations were observed downwind of a mesoscale convective system on 8 May 1996. The second system was also sampled more extensively in both space and time. For these reasons the 8 May case is the focus of this paper.

[9] Figure 1 shows the cloud system that was sampled over the midwestern United States, with the aircraft flight track superimposed. The aircraft made a sounding up through the cirrus anvil outflow region of this large system starting at point A, ascending from 10.7 km to 12.5 km (pressure altitude). At point B it turned and flew roughly parallel to the major storm axis above the anvil outflow to point C. Here it descended to 11.9 km and flew at this altitude, in patchy anvil cirrus, before ascending again at point D to 12.5 km and exiting the area. Convective origins of this storm could be identified in satellite images at least 13 hours prior to sampling, and the storm, while extensive, was already beginning to decay at the time of sampling, which was near 1500 hours local time.

[10] A rear-facing fuselage-mounted inlet was used to collect small particles while excluding large cloud particles, if present. The number concentration of small particles was measured with a TSI 3760 CNC inside the aircraft cabin. *Zhang and Liu* [1991] measured the lower size limit (50% efficiency) of the Model 3760 to be $\sim$0.025 μm diameter at 200 mbar, utilizing a constant volume flow rate. Our unit has been verified to have similar detection characteristics [*Kofer et al.*, 2001]. A portion of the sample flow was heated to 250°C before being measured by a second matched CNC, and the percentage of heat-labile, or volatile, particles was calculated from the ratio of the two CNC measurements. The two units agreed within 1% when sampling ambient air without heating. They were also determined to be leak-tight by verifying that they detected no particles while sampling filtered air at typical in-flight pressures. Concentrations were corrected for coincidence using the manufacturer's recommended algorithm.

[11] For a portion of the flight through, above, and within the cirrus anvil outflow from the storm (A to past D), total condensation nuclei (CN) number concentration, ambient pressure and temperature, and cloud particle surface area [*Baumgardner and Gandrud*, 1998] are displayed in Figure 2. The gap in CN data between 72,400 and 73,200 s is due to a temporary switch in sampling inlets (to a counterflow virtual impactor [*Twohy and Gandrud*,

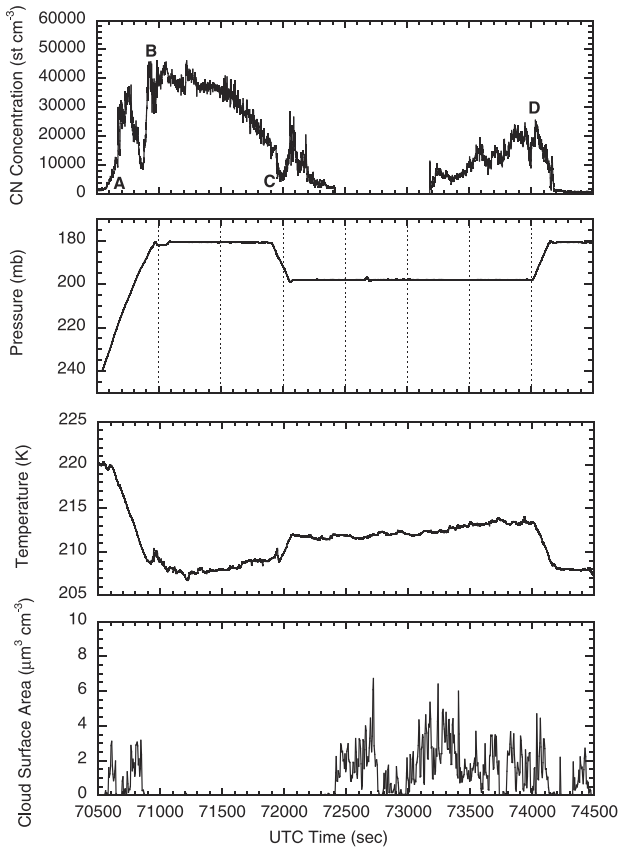

Figure 2. Time series of condensation nuclei (CN) concentration (>0.025 μm diameter), ambient pressure, temperature, and cloud surface area (particles between ~0.4 and 40 μm from multiangle spectrometer probe) in outflow region of 8 May storm. Cloud surface area should be viewed as qualitative, rather than quantitative, since the spectrometer's response to ice is not well characterized and larger ice crystals are not measured. Points A, B, C, and D are as discussed in text.

1998]). Figure 2 shows that CN concentrations were enhanced for a horizontal distance of ~640–820 km, depending on whether or not the sampling gap is included. This encompasses much of the states of Wisconsin and Iowa.

[12] Number concentrations are given relative to standard cubic centimeters (st cm^{-3}) which are independent of altitude changes (concentrations relative to actual cubic centimeters are about a factor of 4 lower). Average particle number concentrations in the background upper troposphere under conditions of clear skies and light surface winds are ~1100 st cm^{-3} [*Hofmann*, 1993] but vary with season. Average particle concentrations that we measured during SUCCESS away from convection or contrails were also ~1100 st cm^{-3} [*Twohy et al.*, 1997], although our CNC has slightly different characteristics and lower size limit (0.025 μm diameter) than Hoffman's (0.02 μm diameter or less).

[13] One period during the climb through the anvil region is shown in detail with a variety of companion measurements in Figure 3. The dip in CN concentration at ~70,870 s is just above the cirrus cloud top, but the concentration peaks again ~275 m above cloud top and remains high throughout the above-cloud leg. Particle volatility is ~99% for most of the time period, except for below cloud where the number concentration is low. On the basis of laboratory calibrations of this particular heating system, volatile percentages this high are indicative of sulfuric acid particles smaller than ~0.07 μm in diameter at this pressure and temperature.

[14] Various gas-phase species, including NO [*Campos et al.*, 1998], OH [*Brune et al.*, 1998], and CO [*Vay et al.*, 1998], are shown in Figure 3. All these gas-phase species show a strong correlation with small particle concentration. NO (and NO_y, which is not shown) is elevated in remarkable sync with CN concentration; these high levels of up to 1500 parts per trillion by volume are indicative of lightning production associated with the convective region of the storm. Carbon monoxide, methane (not shown), and the hydroxyl radical (OH) also are elevated in the region of enhanced CN and indicate that some polluted air from the surface has been brought up through the storm. In addition, increased actinic flux in the cirrus cloud region [*Madronich*, 1987] may have stimulated OH production. Correlations between the various measurements are discussed further in section 4.2 and by *Clement et al.* [2000].

4. Analysis

4.1. Electron Microscopy

[15] During the flight through and above the cirrus outflow (70,600–70,960 s), ambient particles were impacted onto coated grids for subsequent analysis by transmission electron microscope. The 50% cut point of the round-jet impactor was ~0.04 μm diameter at 200 mbar for particles with density 1.7×10^3 kg m^{-3} [*Marple and Willeke*, 1976]. A JEOL 200-CX electron microscope at the National Institute of Standards and Technology in Boulder, Colorado, was used for the analysis. Particles with diameters larger than ~0.10 μm diameter as they appeared on the SiO-coated nickel grid were visually sized, and primary elements with atomic numbers >4 were identified by X-ray analysis (excluding nickel, silicon, and oxygen, which were components of the grid themselves). Particles were grouped into the following categories with respect to their elemental signatures: pure sulfate (S only), mixed sulfate (S with other elements, most commonly C, Na, and/or K), carbonaceous (C only), other (usually Al, Fe, and/or Na), and undetermined (no elements detected). Since carbon is a light element for which this analysis is less sensitive, particles assigned to the undetermined category were most likely carbonaceous. There is also a possibility that small amounts of C were present in the "pure sulfate" category but could not be detected.

[16] Figure 4 shows the percentage of particles analyzed (n = 104) with different elemental signatures. Since the number of particles analyzed is relatively small, a large uncertainty (relative to the actual atmospheric percentages) is associated with the percentages shown in Figure 4. Still, the data allow us to make general inferences about the particle types prevalent in the atmosphere during this period. The majority of particles were pure or mixed sulfates, while a substantial fraction were carbonaceous. Sulfate particles often exhibited the characteristic satellite ring of sulfuric acid [*Gras and Ayers*, 1979]. Since sulfuric

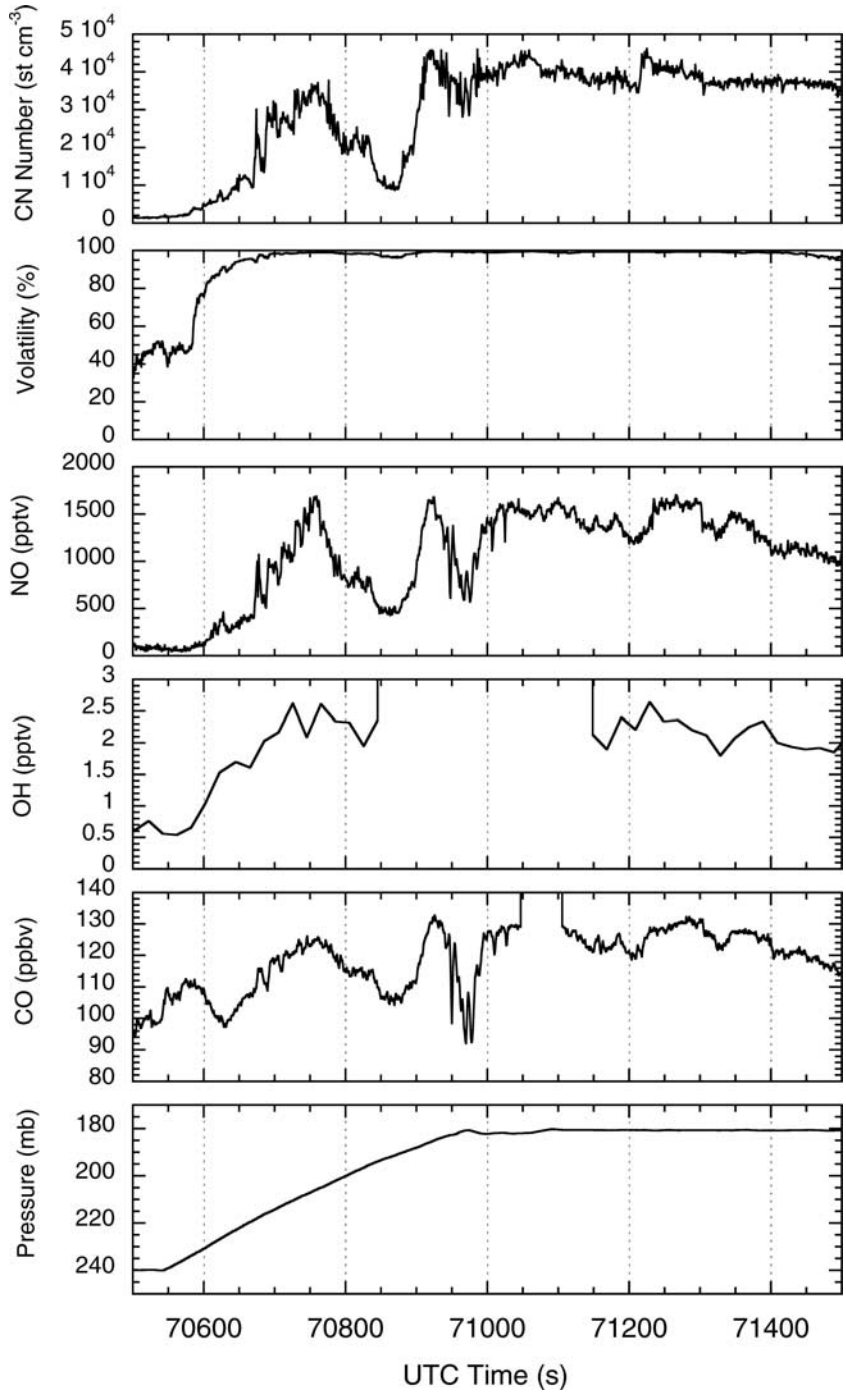

Figure 3. Time series of CN particle concentration, particle volatility, gas-phase variables, and ambient pressure on ascent through cirrus anvil outflow. The break in OH near the middle of the period is missing data.

acid and ammonium sulfate particles remain liquid at low humidity and spread out on the grid, their measured diameter is much larger than their actual diameter for spherical particles. A histogram of the actual sizes of sulfate particles during sampling, corrected for spreading on the grid and for drying after *Gras and Ayers* [1979], is presented in Figure 5a. The distribution peaks just above 0.04 μm, which is the cut size of the impactor. Since collection efficiency curves are relatively sharp for impactors of this type [e.g., *Marple and Willeke*, 1976], we can assume that this distribution represents the majority of particles >0.04 μm and very few of those <0.04 μm.

[17] In Figure 5b the distribution has been corrected back to ambient conditions assuming sulfuric acid and using measured temperature and water vapor content (yielding particles that were an estimated 36% by weight of sulfuric acid). The impaction cut size of 0.04 μm is equivalent to ~0.06 μm for the ambient aerosol. On the basis of the particle loading on the grid and the CN data it is estimated that <1% of the particles counted by the CN counter were

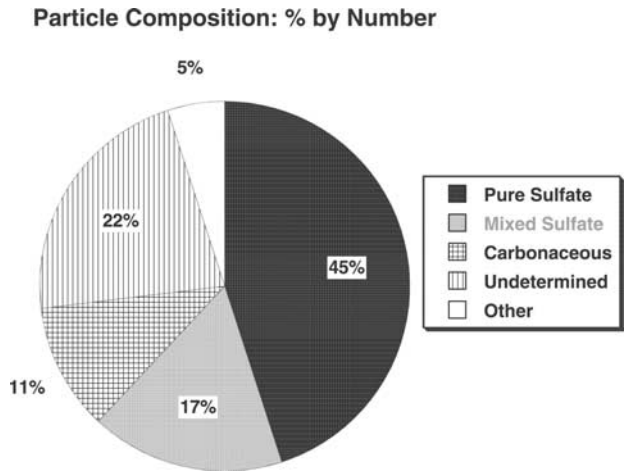

Figure 4. Particle composition (percent by number of 104 total particles analyzed) determined by transmission electron microscopy for a particle sample taken during ascent through cirrus anvil outflow during the time period 70,600–70,960 s (ice crystals excluded).

actually collected by the impactor. (Losses of liquid particles within the impactor are expected to be small [e.g., *Turner et al.*, 2000]). Thus we conclude that the majority of particles measured by the CNC were <0.04 μm at impaction or <0.06 μm in the atmosphere. Volatility measurements also indicated that most of the particles were less than ~0.07 diameter. The minimum size of the detected particles can be estimated from the CNC cut size (50% efficiency) of 0.025 μm, which we estimate to correspond to ~0.038 μm at ambient conditions. We can thus establish that the approximate ambient diameter range for the majority of particles measured was 0.038–0.06 μm. (Many more particles <0.038 μm may have been present but not detected by the CNC.) These relatively small sizes and high concentrations suggest that observed particles were formed relatively recently, probably earlier in the lifetime of this storm.

[18] Since some carbonaceous particles were observed, it is possible that both sulfuric acid and organic particles, or combinations thereof, were formed in the storm outflow region. Both SO_2 and organic vapors would be expected to be carried up from the boundary layer through the convective region of the storm, with at least some fraction surviving [e.g., *Walcek and Taylor*, 1986; *Thornton et al.*, 1997]. However, *Clement et al.* [2002] present nucleation and growth calculations for this particular case, assuming homogeneous nucleation of sulfuric acid and water only. Their model is able to approximate both the mass and number concentrations of the observed CN aerosol within a timescale consistent with that of the storm. Therefore organics or other species are not required to explain the high particle concentrations we observed; the rather extreme conditions were favorable for nucleation of sulfuric acid and water alone.

4.2. Principal Components Analysis

[19] Principal components analysis (PCA) is a multivariate statistical technique, related to factor analysis [*Jackson*, 1991]. PCA extracts the important information in a data matrix by transforming it into a smaller number of factors (or components) that are linear combinations of the original data set. These factors are orthogonal and can often be attributed to physical phenomena or sources. A weight, or loading, is assigned to each variable on a factor; the loading describes how important a variable is to that particular factor. Data variables with large positive loadings on a factor all vary together and are negatively correlated with variables that have large negative loadings.

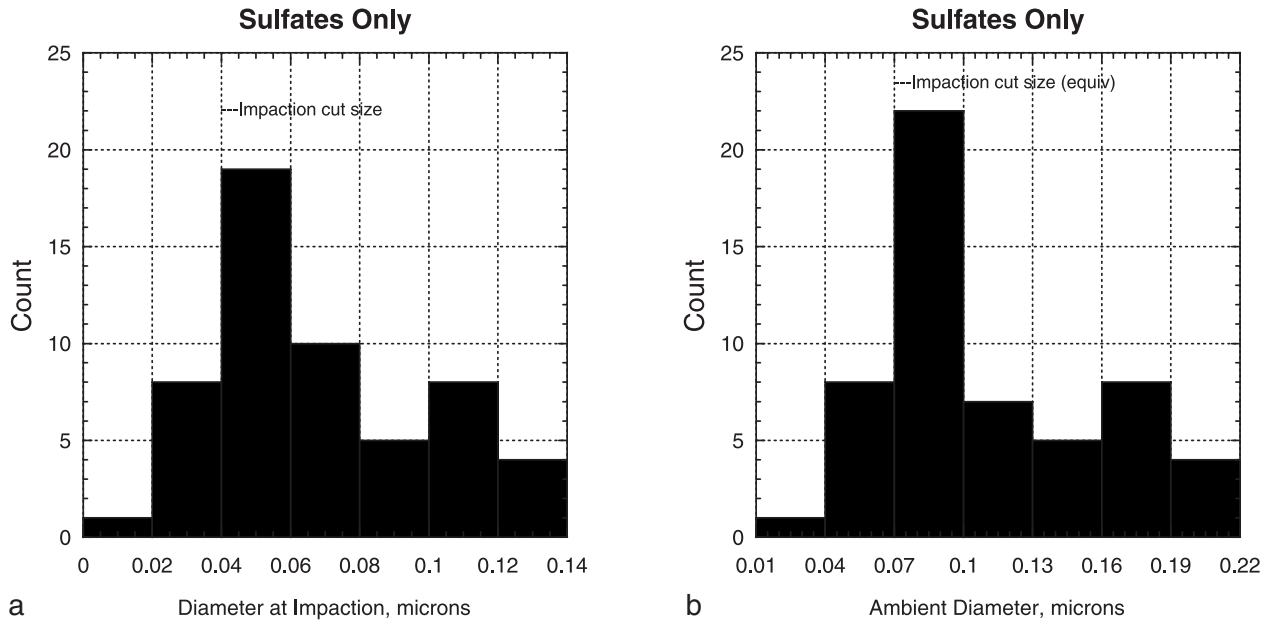

Figure 5. Histogram of particle size for pure sulfate particles identified in sample of Figure 4 (a) after correction for spreading on the grid and evaporation following *Gras and Ayers* [1979] and (b) corrected back to ambient temperature of 208K.

Table 2. Results of Principal Components Analysis Ascent Through Cirrus Anvil 8 May 1996[a]

Variable	Factor 1 (62.5%)	Factor 2 (19.3%)	Factor 3 (5.2%)
CH_4	0.958	...	...
NO	0.938	...	...
V	0.931	...	...
OH	0.923	...	...
NO_y	0.922	...	...
HO_2	0.890	...	...
CO	0.888	...	...
CN	0.883	...	...
Volume	0.855	...	...
Pressure	−0.798	...	0.526
Temperature	−0.750	...	0.533
U	−0.707	...	0.618
Surface	...	0.836	...
H_2O	...	0.776	...
O_3	...	−0.743	...
W	...	...	0.704
Turbulent energy dissipation rate	...	...	...

[a] Varimax rotation was used to transform the original matrix.

[20] A commercial statistical package, SPSS 6.1, was used to generate factors from the 1-s data for the time period shown in Figure 3. (For OH and HO_2, only one data value every 5 s was available, so this value was used for each second in that interval.) All of the variables shown in Figures 2 and 3, as well as NO_y, HO_2, O_3, and CH_4, were included. Ambient pressure, temperature, the three-dimensional wind components *U*, *V*, and *W*, and the turbulent energy dissipation rate [*Chan et al.*, 1998] were also used. Variable loadings on the three primary factors, responsible for 87% of the variance in the data, are given in Table 2. Values are given only for loadings >0.50 or <−50; otherwise the loading is of little importance for that particular factor.

[21] Factor 1 accounts for 62.5% of the variance in the data and relates to the origin of the sampled air mass. The high positive loadings for CH_4, CO, NO, NO_y, OH, and HO_2 indicate that all these gas-phase species are highly correlated with each other and with CN and particle volatility. These variables are also correlated with the *V* wind component (positive values from the south) and are anticorrelated with the *U* wind component (positive values from the west). This suggests that the CN and gas-phase species have a common source and originate from the southeast, where the main part of the storm resides. The correlation with both convective (NO) and surface tracers (CH_4 and CO) supports the hypothesis that convection brings gas-phase species (SO_2 and OH precursors) to the upper troposphere, where they can participate in new particle formation. CN and most of the gas-phase species are anticorrelated with pressure and temperature, which both decrease with altitude. No apparent correlation with the turbulent energy dissipation rate was found; however, this does not preclude that turbulence had contributed to particle formation earlier in the storm's lifetime.

[22] The second factor (19.3% of variance) has a high loading for particle surface area and for water vapor and relates to the anvil ice cloud. The strong anticorrelation with ozone is probably due to the difference in their source regions (water vapor primarily at the surface and ozone primarily in the stratosphere). The variables associated with the ice cloud show no correlation with CN, which was formed separately in one of two air masses which were subsequently mixed [*Clement et al.*, 2002]; the ice cloud was probably formed during the mixing process.

[23] The third factor accounts for only 5.2% of the variance and shows correlations between pressure, temperature, and rising air with a more westerly component. This probably represents the influence of another air mass not associated with the convection. While only 1-s data were used in this analysis, *Clement et al.* [2000] also showed that correlation strength may vary with the time interval of the measurements or the correlation analysis, as well as with location relative to the storm.

4.3. Assessment of Large-Scale Consequences

[24] To our knowledge, these observations represent the largest atmospheric nucleation event published to date. The vertical transport of gas-phase precursors by large convective systems to the upper troposphere and the extremely low temperatures found there are conducive to particle nucleation on a vast scale. Smaller convection will not lift precursors to as cold temperatures nor be as pervasive in spatial extent or lifetime. There is no reason to believe this particular MCS was unique in terms of particle formation, as several other observations near convection have been made with similar CN number concentrations reported (Table 1). Measurements near deep convective systems are relatively rare, however, since safety considerations often preclude sampling too near the convective core, which may contain severe turbulence and hail. Because of the large size of this system, however, the outflow region extended far from the storm center. This allowed relatively new particles to be detected in nonturbulent regions well removed from the intense convection. These particles may have been generated much earlier and closer to the active convection.

[25] Simple box-type calculations are used here to assess the potential importance of deep convection in maintaining the background level of small particles in the midlatitude upper troposphere, where background is defined to be areas apart from convection or other short-term sources. Calculations assume that this storm is typical of other deep convective systems and that particles generated by these systems eventually become mixed throughout the midlatitude upper troposphere (UT), which is assumed here to be the upper 5 km of the troposphere. Only the upper troposphere at midlatitudes and over land areas is considered here to be conservative. Most of the observations of new particles have been made at midlatitudes, and gas-phase precursors are expected to be more prevalent over land, which is mostly polluted at midlatitudes. However, the same general mechanisms may operate over the oceans and in the tropics. Convective transport of dimethyl sulfide and subsequent conversion to SO_2 have been reported over the North Pacific Ocean [*Thornton et al.*, 1997], and high CN concentrations were regularly observed by *Brock et al.* [1995] in the upper troposphere near the Intertropical Convergence Zone.

[26] The contribution of deep convection in maintaining midlatitude particle concentrations, C_{ML}, can be estimated as follows:

$$C_{ML} = C_{CS} F_Z F_A F_D F_L, \tag{1a}$$

where C_{CS} is the typical small particle concentration associated with the outflow region of the convective system. F_Z is the fraction of UT height over which particles are initially enhanced, and F_A is the fraction of area covered by cirrus anvils associated with deep convection. F_D is the fraction of time during daylight (particle production is assumed to be inhibited at night when OH is not actively produced), and F_L is the fraction of land relative to total midlatitude area (particle production is assumed to occur primarily over land, where gas-phase precursors are highest).

[27] Some of these fractions are simple to estimate, while others are more difficult with larger uncertainties. Since enhancements were observed over about a 2-km depth, 0.4 is used for F_Z. For F_D, 0.50 was used, and F_L, the fraction of midlatitude land area, was estimated to be 0.47. For the fraction of area F_A covered by cirrus anvil associated with deep convection, data from the International Satellite Cloud Climatology Project [*Rossow and Schiffer*, 1991] were utilized. The 11-year mean D2 database for daytime high cloud amount (tops 50–440 mbar) over midlatitude land areas shows that deep convective clouds cover ~3%, cirrostratus clouds cover ~8%, and cirrus are highly variable between ~6 and 30%. It is unknown exactly what percentage of the cirrostratus and cirrus is anvil cirrus associated with deep convection; however, *Machado and Rossow* [1993] showed that these systems produce about 4 times the amount of anvil cirrus cover relative to the amount of deep convective cloud cover. From this and the deep convective cloud amount of 3% we can estimate F_A to be 0.12, and the relationship becomes

$$C_{ML} = 0.011 C_{CS}. \quad (1b)$$

On the basis of our measurements a value of 20,000 st cm^{-3} was used for C_{CS}, yielding C_{ML} = 220 st cm^{-3}.

[28] *Hofmann* [1993] compiled extensive particle measurements in the upper troposphere over Laramie, Wyoming, for a 20-year period. On the basis of Figure 1 of *Hofmann et al.* [1998] it was assumed that the characteristics of the fall and spring aerosol were more similar to the winter season than summer season characteristics given by *Hofmann* [1993]. (Hofmann hypothesized that the substantially higher particle number concentrations and mass in the summer season were due to photochemical production of new particles.) While large seasonal and day-to-day variation exists, annual mean background number and mass concentrations of ~1100 st cm^{-3} and 0.4 μg st m^{-3} for particles >0.02 μm in the 5- to 10-km-altitude range were extracted from the Hofmann data sets. Our measurements during SUCCESS [*Twohy et al.*, 1997], while more limited, yielded similar values for number concentration of small particles. Our calculated C_{ML} of 220 st cm^{-3} is ~20% of the approximate background number concentration of 1100 st cm^{-3}.

[29] Small particle mass was estimated to be ~60–90% of the total particle mass in the UT background aerosol [*Hofmann*, 1993]. In theory, either number or mass concentrations can be related by equations (1a) and (1b). Mass concentrations are more difficult to extract from our data, however, since the exact size of the particles we measured is unknown. *Hofmann* [1993] fitted two lognormal particle modes to their upper tropospheric data, with the small particles having a mode diameter of 0.02–0.04 μm, depending on season. Assuming the particles we measured were of similar size, then the contribution of deep convection to small particle mass would be similar to its contribution to small particle number. Our previous estimates that the particles we measured were between ~0.038 and 0.06 μm diameter (ambient conditions) mean that they actually may have been on the large end of the small particle mode presented by *Hofmann* [1993].

[30] On the basis of the large uncertainties associated with the values used in equation (1a), C_{ML} may have an uncertainty as large as 100% or more. However, the number chosen for C_{CS} is conservative, since many particles may have been smaller than the detection limit of the CNC used. While crude, the analysis does suggest that deep convective systems are an important source of new particles to the background upper troposphere at midlatitudes. More detailed model calculations, which include storm dynamics and life cycle, as well as particle nucleation, coagulation, and growth, are needed for a more quantitative assessment.

5. Conclusions

[31] Deep convection is a potential source of new particles in the upper troposphere. High concentrations of gas-phase precursors from lower in the atmosphere and cold temperatures may contribute to extensive formation of new particles. Low preexisting surface area and high actinic flux may also be important. Mesoscale convective systems have the potential for great impact, because of their large spatial scales and lifetimes and the high (cold) altitudes to which they extend. Very high particle concentrations, up to 45,000 st cm^{-3}, were observed during flights in the outflow region of a mesoscale convective system over the midwestern United States. Enhanced concentrations were observed over 600–800 km horizontally and 2 km vertically, making this potentially the largest nucleation event ever reported.

[32] The enhanced CN were highly correlated with methane, carbon monoxide, nitrogen oxides, and hydroxyl radicals, indicating that air in which the particles formed had traveled through the convective region of the storm before being ejected near the tropopause. No correlation or anticorrelation with the anvil cirrus cloud was found, suggesting that the particles and cirrus had formed independently, although they were in the same region at the time of sampling. Both volatility measurements and electron microscopy indicate that these particles were primarily small (≤0.06 μm), young sulfate particles.

[33] Simple calculations were used to assess the potential importance of deep convective systems to the background aerosol concentrations in the upper troposphere at midlatitudes. While large uncertainties are inherent in these calculations, they suggest that deep convection could contribute to 20% or more of the background small particle number concentration and mass.

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