

African dust aerosols as atmospheric ice nuclei

Paul J. DeMott,¹ Kenneth Sassen,² Michael R. Poellot,³ Darrel Baumgardner,⁴ David C. Rogers,⁵ Sarah D. Brooks,¹ Anthony J. Prenni,¹ and Sonia M. Kreidenweis¹

Received 26 March 2003; revised 9 May 2003; accepted 20 May 2003; published 17 July 2003.

[1] Measurements of the ice nucleating ability of aerosol particles in air masses over Florida having sources from North Africa support the potential importance of dust aerosols for indirectly affecting cloud properties and climate. The concentrations of ice nuclei within dust layers at particle sizes below $1\ \mu\text{m}$ exceeded $1\ \text{cm}^{-3}$; the highest ever reported with our device at temperatures warmer than homogeneous freezing conditions. These measurements add to previous direct and indirect evidence of the ice nucleation efficiency of desert dust aerosols, but also confirm their contribution to ice nuclei populations at great distances from source regions. **INDEX TERMS:** 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0320 Atmospheric Composition and Structure: Cloud physics and chemistry; 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry. **Citation:** DeMott, P. J., K. Sassen, M. R. Poellot, D. Baumgardner, D. C. Rogers, S. D. Brooks, A. J. Prenni, and S. M. Kreidenweis, African dust aerosols as atmospheric ice nuclei, *Geophys. Res. Lett.*, 30(14), 1732, doi:10.1029/2003GL017410, 2003.

1. Introduction

[2] The effects of dust transport on aerosol composition and deposition are known to extend globally from major desert regions [Husar *et al.*, 2001; Prospero, 1996, 1999]. Dusts are important atmospheric aerosols for direct climate forcing due to their effect on scattering and absorption of solar radiation. Evidence is mounting for the indirect effects of dust aerosols on clouds. Rosenfeld *et al.* [2001] noted the effect of Saharan dust was to reduce precipitation in shallow convective clouds near the source. This was hypothesized to be due to lowering of the coalescence efficiency of clouds resulting from increases in cloud condensation nuclei. The dramatic correlation of the presence of Asian dust and ice formation in modestly supercooled altocumulus clouds was previously demonstrated by Sassen [2002]. Levi and Rosenfeld [1996] have confirmed relative increases in ice nuclei concentrations during dust storm periods in Israel, and Rosenfeld and Nirel [1996] alluded to the action of Saharan dust particles as ice nuclei in attempting to explain impacts on cloud seeding experiments in Israel.

[3] Laboratory studies have indicated the strong ice nucleating behavior associated with dust in air [Isono *et al.*, 1959], clay minerals [Roberts and Hallett, 1968; Zuberi *et al.*, 2002] and many of the metal oxide components of desert dust [Hung *et al.*, 2003]. The study of dust aerosols as ice-forming nuclei has been motivated by the recognition of their potential importance to ice phase transitions in cold clouds, observations documenting the wide distribution and frequent occurrence of dust in the atmosphere, and the realization that changes in land use worldwide could lead to greater dust loadings in the atmosphere.

[4] Relatively few measurements have documented the ice nucleating function of dust aloft in the atmosphere. Evidence is provided in this study for the strong ice nucleating function of Saharan dust aerosols as measured at long distances from the source. These measurements were collected in July 2002 as part of the National Aeronautics and Space Administration (NASA) CRYSTAL-FACE (Cirrus Regional Study of Tropical Anvils and Cirrus Layers - Florida Area Cirrus Experiment) program, a study focused on understanding the formation, properties and atmospheric effects of tropical cirrus clouds.

2. Methods

[5] Ice nuclei (IN) measurements were obtained using the Colorado State University continuous flow diffusion chamber (CFDC). The basic instrument design and performance characteristics have been described in detail by Rogers *et al.* [2001]. Recent modifications of the instrument include the construction of a refrigeration system permitting operation to -65°C . The device exposes air containing aerosol particles to steady state conditions of temperature and relative humidity for periods of 10 to 30 s. The temperatures of two concentric ice-coated plates through which sample air is focused determine the “processing” conditions. Optical detection of ice particles growing to larger sizes indicates ice nucleation.

[6] The CFDC instrument was installed on the University of North Dakota Citation II aircraft, which profiled atmospheric conditions and aerosol particle and cloud characteristics up to 13 km above mean sea level (MSL). Aerosol particles were sampled through an exterior inlet that was located within less than 1 m air transit length into the CFDC. The inlet was a forward-facing diffuser with a 1.5 mm diameter hole expanding in a 6° cone to a 9.5 mm bore that exhausted through the back. The axis of the inlet tip was 122 mm above the aircraft skin, estimated to be double the boundary layer thickness at the sampling location. The decelerated air sample was drawn at right angles into a 6 mm tube that led to the CFDC. Aerosol particles larger than about $1\ \mu\text{m}$ (aerodynamic diameter) were removed by two identical impactors of the same design as described by

¹Department of Atmospheric Science, Colorado State University, Fort Collins, Colorado, USA.

²Geophysical Institute, University of Alaska Fairbanks, Fairbanks, Alaska, USA.

³Atmospheric Sciences Department, University of North Dakota, Grand Forks, North Dakota, USA.

⁴Universidad Nacional Autonoma de Mexico, Mexico City, Mexico.

⁵National Center for Atmospheric Research, Boulder, Colorado, USA.

Rogers *et al.* [2001] in order to remove the possibility of falsely identifying large aerosol particles as nucleated ice crystals in the CFDC.

[7] Lidar data are also used in this study. The mobile, dual-wavelength, University of Utah Polarization Diversity Lidar (PDL) was located at Ochopee in the western Everglades about 135 km to the north of flight operations in Key West [Sassen *et al.*, 2003]. Linear depolarization ratios (δ , the ratio of the returned laser powers in the planes of polarization orthogonal and parallel to that transmitted) are presented here from the 0.532 μm channel. Relatively large, non-spherical dust particles can be detected by their strong depolarization [Sassen, 2002].

[8] Supporting data are presented from aerosol particle sensors on the Citation aircraft and on the NASA WB-57 aircraft.

3. Measurements

[9] Measurements suggest the presence of Saharan dust aerosols at varied concentration levels in the Florida project area during CRYSTAL-FACE. This paper focuses on 28–29 July 2002 when surface dust concentration in Miami exceeded $30 \mu\text{g m}^{-3}$ and the dust layer reached at times to above 5 km height over sea level [Sassen *et al.*, 2003]. A representative isentropic backward air trajectory terminating in the project area, determined using the NOAA HYSPLIT model (HYbrid Single-Particle Lagrangian Integrated Trajectory Model, 1997, <http://www.arl.noaa.gov/ready/hysplit4.html>) is shown in Figure 1. This indicates a likely source of air from Africa about 1 week prior to aircraft measurements. High aerosol optical depths derived from the MODIS (Moderate-Resolution Imaging Spectroradiometer) instrument on the Terra satellite, also shown in Figure 1, document the dust transport during 20–27 July, consistent with trajectories.

[10] The vertical distribution of dust aerosols is indicated in Figure 2, based on lidar and aircraft data on 28 July 2002. Lidar relative retrieved power indicates an aerosol layer with a top at just below 4 km. This layer persisted throughout the day. Linear depolarization values as high as 0.12 indicate that aerosols in the noted layer were more characteristic of desert dust rather than pollution, smoke aerosols or the salt particles found below 1 km in maritime air. The δ values are somewhat less than those indicated for Asian dust over Utah [Sassen, 2002], but the amount of aerosol depolarization may be similar considering the added molecular scattering at sea level. No depolarization is expected for sulfate/organic aerosols.

[11] Aircraft measurements of aerosols, larger than 3.63 μm from the Citation FSSP (Forward Scattering Spectrometer Probe) and in two size ranges above 0.5 μm from the CAS (Cloud Aerosol Spectrometer) probe [Baumgardner *et al.*, 2002] on the WB-57 support the presence of a dust layer at 1 to 4 km. Total particle (condensation nuclei, not shown) numbers were not higher in this layer compared to other levels. The altitude of the dust layer is consistent with the location of Saharan aerosol layers [Carlson and Prospero, 1972]. The vertical profiles of aerosol data and nearly constant water vapor mixing ratio and potential temperature through the dust layer are similar to those obtained by Reid *et al.* [2002].

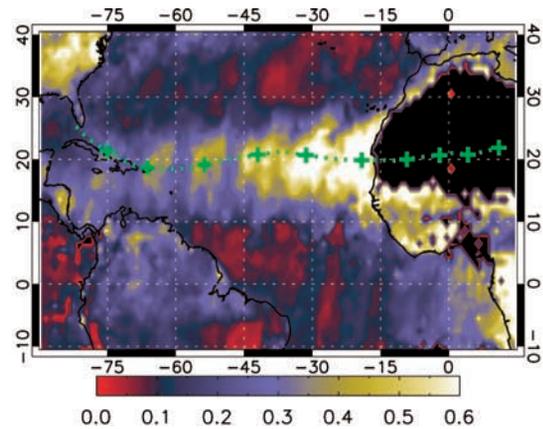


Figure 1. Ten-day HYSPLIT back trajectory (green) for the 3 km flight level over Key West at 19Z on 29 July overlain on color-coded MODIS aerosol mean optical depth data for the 8 day period starting on 20 July 2002. Plus marks are days in the trajectory. Latitude and longitude are shown.

[12] Ice nuclei concentrations increased coincidentally with and correlated to the aerosol particle signals in the location of the dust layer. The CFDC processing conditions were approximately $-36.5 \pm 0.5^\circ\text{C}$ and $86.5 \pm 0.5\%$ relative humidity with respect to water (123% relative humidity with respect to ice). These conditions were selected to obtain some data on the maximum contribution of heterogeneous nuclei to ice formation in cumulus clouds at temperatures warmer than where homogeneous freezing is expected to occur (i.e., $>-38^\circ\text{C}$). Relative humidity below water saturation assured that homogeneous freezing was not possible. Heterogeneous IN concentrations exceeded 1 cm^{-3} through a depth of nearly 2 km. Previous CFDC measurements of ice nuclei concentrations in the heterogeneous nucleation regime warmer than -38°C range from 10^{-4} to 10^{-1} cm^{-3} throughout the troposphere [e.g., Rogers *et al.*, 1998]. Median ice nuclei concentrations are typically below 0.01 cm^{-3} . Elevated ice nuclei concentrations for the same processing conditions were also noted in coincidence with a dust layer on 29 July 2002 [Sassen *et al.*, 2003]. Sassen *et al.* [2003] also show evidence for the unusual glaciation of modestly supercooled altocumuli at the top of the dust layer on that day.

[13] CFDC data on ice nuclei at lower altitudes were not obtained on every flight during CRYSTAL-FACE due to emphasis on sampling in and around anvil cirrus clouds. Nevertheless, vertical profile data of ice nuclei concentrations in the lower troposphere were obtained on at least one day (18 July 2002) for which only modest dust influences are inferred. The PDL instrument and WB-57 were not operating on this day. Although Miami surface dust measurements indicate some enhancement over average background values on this day [Sassen *et al.*, 2003], Angström exponent values for aerosols inferred from AERONET (Aerosol Robotic NETWORK) sun photometer measurements near Miami exceeded 1.3 at the time of the Citation flight. This indicates a predominance of aerosols aloft that were smaller than on 28–29 July when Angström exponents ranged from 0.3 to 0.5. Additionally, only weak transport of Saharan dust was evident from MODIS retrieved aerosol optical depth during the period around 18

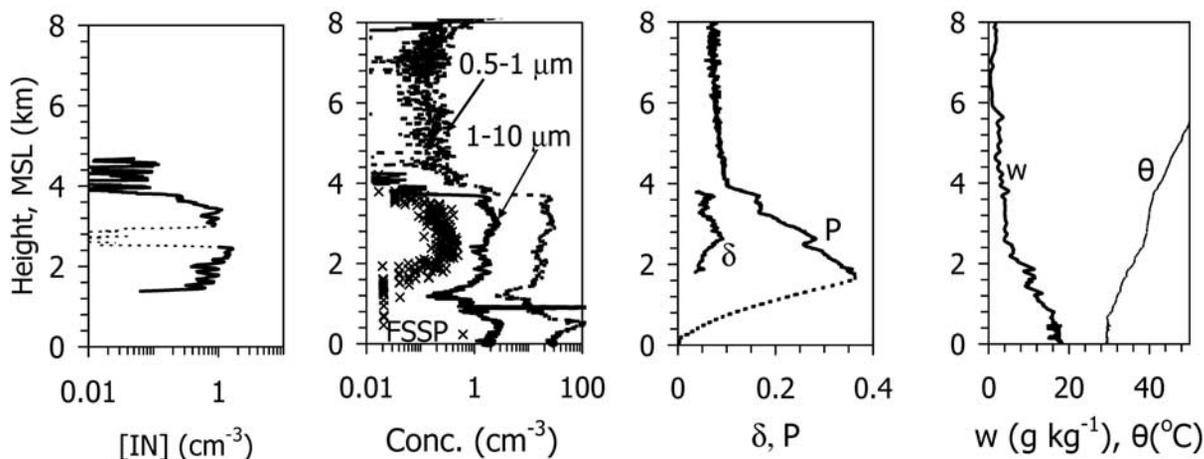


Figure 2. Ice nuclei (IN) concentration (5 s running mean), aerosol concentration, lidar linear depolarization ratio (δ) and relative returned power (P), and water vapor mixing ratio (w) and potential temperature (θ) as a function of altitude on 28 July. IN, FSSP ($>3.6 \mu\text{m}$) and thermodynamic data were collected from the Citation aircraft at ~ 2200 UTC. CFDC processing conditions for ice nuclei were approximately -37°C and 86% (123%) relative humidity with respect to water (ice). The dashed curve segment in the IN panel indicates the action of filtering sample air. Aerosol measurements in 0.5 to 1 and 1 to 10 μm size ranges are from the CAS instrument on the WB-57 aircraft at ~ 2330 UTC. Lidar data are averages from 1953 to 1958 UTC. Low-altitude lidar signals cannot be used to calculate δ -values because of strong, off-scale signals.

July. We therefore consider this day to represent “no-dust” conditions for July in Florida.

[14] Mixing ratio and potential temperature profiles (Figure 3) are also consistent with the absence of a dust layer above 2 km on 18 July. The CFDC instrument was processing particles at warmer temperatures (-24°C) during the aircraft descent sounding on 18 July. Nevertheless, processing ice relative humidity, which often correlates with ice nucleation activity, was 134%, higher than on 28 July. Figure 3 shows that IN concentrations were slightly elevated from typical values in and around the base of a cirrus anvil at 6.4 km and in a lower clear-air region where modest amounts of large aerosols were detected by the FSSP. Measurements

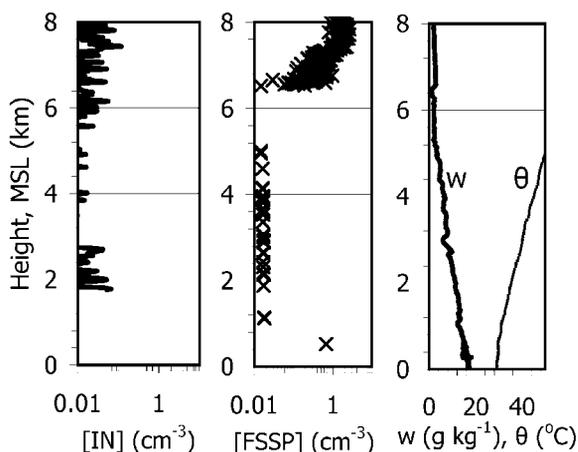


Figure 3. Mixing ratio, potential temperature, and IN and FSSP concentrations for the 18 July 2002 “no dust” case. IN processing conditions were approximately constant at -24°C and 104% (134%) relative humidity with respect to water (ice). IN measurements were within a cirrus anvil above 6.4 km. There were no IN measurements below 1.8 km.

of IN in cirrus anvils will be addressed in a future publication. Ice nuclei concentrations in clear air regions down to 1.5 km MSL never exceeded about 0.05 cm^{-3} on this day. These IN concentrations are consistent with peak values measured previously in this temperature regime in the free troposphere [Rogers *et al.*, 1998], but are at least 20 times lower than peak concentrations noted in the dust layer on 28–29 July. These lower IN concentrations on 18 July, consistent with the absence of strong desert dust loading above the marine boundary layer, emphasize the unusual nature of the IN measurements within dust layers.

4. Discussion and Conclusions

[15] The vertical descent profiles of ice nuclei concentrations on the three mentioned days are compared directly in Figure 4. When Saharan dust layers are present (28 and

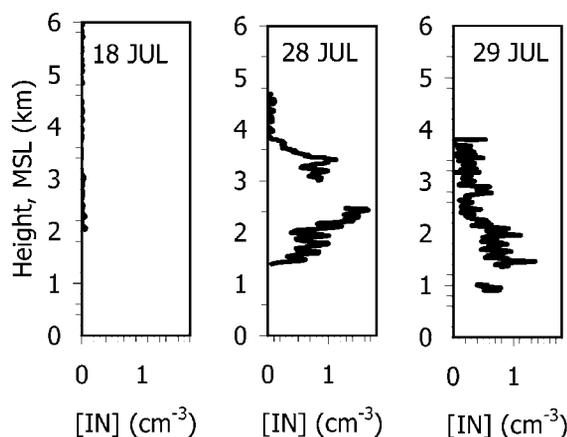


Figure 4. Comparison of ice nuclei concentration profiles on 18, 28 and 29 July. Nuclei processing conditions, which varied, are described in the text.

29 July), ice nuclei concentrations are strongly perturbed, even at great distances from the source. The IN concentrations measured on 28 and 29 July 2002 ($>1 \text{ cm}^{-3}$) exceed typical IN concentrations by at least 20 to 100 times in the heterogeneous ice nucleation regime warmer than -38°C .

[16] High concentrations of dust particles acting as ice nuclei in clouds could lead to changes in cloud microphysical and radiative properties, latent heating and precipitation compared to more pristine conditions. While detailed numerical modeling studies to elucidate such effects are outside of the scope of this paper, simple considerations based on previous model sensitivity studies are instructive. For example, if IN concentrations of 1 cm^{-3} reach the upper troposphere, heterogeneous ice nucleation could dominate the formation of ambient cirrus in preference to homogeneous freezing of more abundant haze (e.g., sulfate) particles [Gierens, 2003] and thereby alter cloud forcing and upper tropospheric relative humidity. The simple exercise (not detailed here) of increasing IN concentrations by 100 times (to noted levels) in a microphysical model of adiabatic cumulus cloud parcels [Rogers *et al.*, 1994] suggests that ice formation could deplete cloud water and prevent homogeneous freezing in maritime clouds with updrafts as large as 12 m s^{-1} . Investigations are underway to compare cumuli and anvil cirrus properties on dusty versus less dusty days during CRYSTAL-FACE.

[17] Although large dust storms are episodic, dust particles are often present throughout the atmosphere and so must also be their general effects on ice formation in clouds. In the United States, for example, dust from African [Perry *et al.*, 1997] and Asian [VanCuren and Cahill, 2002] sources can dominate fine aerosol mass at times in spring through summer. The annual dust minimum occurs during winter, with potential implications for the natural efficiency of winter precipitation and the formation of supercooled drizzle, an aircraft icing hazard. It is therefore important to further quantify the annual dust cycle, how it may be altered and the indirect effects of dust on ice phase clouds and precipitation.

[18] **Acknowledgments.** This research was supported by NASA grants NAG5-11476 and NAG5-11503. The authors gratefully acknowledge the MODIS Atmosphere Discipline Group (<http://modis-atmos.gsfc.nasa.gov/index.html>) at NASA for the aerosol optical depth data, NASA's AERONET group (<http://aeronet.gsfc.nasa.gov>) for sun photometer data and NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model. Thanks to Liz Zarovy, Kristi Gebhart, Katie Walters and Ben Ruston for assistance with back trajectory calculations and MODIS data presentation. Brian Jesse, Cindy Twohy, Paul LeHardy, Mark Askelson, Jeff Schild and David Fahey are specially thanked for assistance at various times during this research program.

References

Baumgardner, D., H. Jonsson, W. Dawson, D. O. Connor, and R. Newton, The cloud, aerosol and precipitation spectrometer (CAPS): A new instrument for cloud investigations, *Atmos. Res.*, 59–60, 251–264, 2002.

Carlson, T. N., and J. M. Prospero, The large-scale movement of Saharan air outbreaks over the north equatorial Atlantic, *J. Appl. Meteorol.*, 11, 283–297, 1972.

Gierens, K., On the transition between heterogeneous and homogeneous freezing, *Atmos. Chem. Phys.*, 3, 437–446, 2003.

Hung, H.-M., A. Malinowski, and S. T. Martin, Kinetics of heterogeneous ice nucleation on the surfaces of mineral dust cores inserted into aqueous ammonium sulfate particles, *J. Phys. Chem. A*, 107, 1296–1306, 2003.

Husar, R. B., et al., Asian dust events of April 1998, *J. Geophys. Res.*, 106, 18,317–18,330, 2001.

Isono, K., M. Komabayasi, and A. Ono, The nature and origin of ice nuclei in the atmosphere, *J. Meteorol. Soc. Japan*, 37, 211–233, 1959.

Levi, Y., and D. Rosenfeld, Ice nuclei, rainwater chemical composition, and static cloud seeding effects in Israel, *J. Appl. Meteorol.*, 35, 1494–1501, 1996.

Perry, K. D., T. A. Cahill, R. A. Eldred, D. D. Dutcher, and T. E. Gill, Long-range transport of North African dust to the eastern United States, *J. Geophys. Res.*, 102, 11,225–11,238, 1997.

Prospero, J. M., Saharan dust transport over the North Atlantic Ocean and Mediterranean: An overview, in *The Impact of Desert Dust Across the Mediterranean*, edited by S. Guerzoni and R. Chester, Kluwer, Dordrecht, 1996.

Prospero, J. M., Long-term measurements of the transport of African mineral dust to the southeastern United States: Implications for regional air quality, *J. Geophys. Res.*, 104(D13), 15,917–15,927, 1999.

Reid, J. S., D. L. Westphal, J. M. Livingston, D. L. Savoie, H. B. Maring, H. H. Jonsson, D. P. Eleuterio, J. E. Kinney, and E. A. Reid, Dust vertical distribution in the Caribbean during the Puerto Rico Dust Experiment, *Geophys. Res. Lett.*, 29, 1151, doi:10.1029/2001GL014092, 2002.

Roberts, P., and J. Hallett, A laboratory study of the ice nucleating properties of some mineral particulates, *Q. Jl. R. Meteorol. Soc.*, 94, 25–34, 1968.

Rogers, D. C., P. J. DeMott, and L. O. Grant, Concerning primary ice nuclei concentrations and water supersaturations in the atmosphere, *Atmos. Research*, 33, 151–168, 1994.

Rogers, D. C., P. J. DeMott, S. M. Kreidenweis, and Y. Chen, Measurements of ice nucleating aerosols during SUCCESS, *Geophys. Res. Lett.*, 25, 1383–1386, 1998.

Rogers, D. C., P. J. DeMott, S. M. Kreidenweis, and Y. Chen, A continuous flow diffusion chamber for airborne measurements of ice nuclei, *J. Atmos. Oceanic Technol.*, 18, 725–741, 2001.

Rosenfeld, D., and R. Nirel, Seeding effectiveness - The interaction of desert dust and the southern margins of rain cloud systems in Israel, *J. Appl. Meteorol.*, 35, 1502–1510, 1996.

Rosenfeld, D., Y. Rudich, and R. Lahav, Desert dust suppressing precipitation: a possible desertification feedback loop, *Proceedings of the National Academy of Sciences*, 98(11), 5975–5980, 2001.

Sassen, K., Indirect climate forcing over the western US from Asian dust storms, *Geophys. Res. Lett.*, 29, doi:10.1029/2001GL014051, 2002.

Sassen, K., P. J. DeMott, J. M. Prospero, and M. R. Poellot, Saharan dust storms and indirect aerosol effects on clouds: CRYSTAL-FACE Results, *Geophys. Res. Lett.*, 30(12), doi:10.1029/2003GL017371, 2003.

VanCuren, R. A., and T. A. Cahill, Asian aerosols in North America: Frequency and concentration of fine dust, *J. Geophys. Res.*, 107, doi:10.1029/2002JD002204, 2002.

Zuberi, B., A. K. Bertram, C. A. Cassa, L. T. Molina, and M. J. Molina, Heterogeneous nucleation of ice in $(\text{NH}_4)_2\text{SO}_4\text{-H}_2\text{O}$ particles with mineral dust immersions, *Geophys. Res. Lett.*, 29, doi:10.1029/2001GL014289, 2002.

P. J. DeMott, S. D. Brooks, A. J. Prenni, and S. M. Kreidenweis, Department of Atmospheric Science, Colorado State University, Fort Collins, CO 80526-1371, USA. (pdemott@lamar.colostate.edu)

K. Sassen, Geophysical Institute, University of Alaska Fairbanks, Fairbanks, AK, USA.

M. R. Poellot, Atmospheric Sciences Department, University of North Dakota, Grand Forks, ND, USA.

D. Baumgardner, Universidad Nacional Autonoma de Mexico, Mexico City, Mexico.

D. C. Rogers, National Center for Atmospheric Research, Boulder, CO, USA.