FINAL REPORT

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PROJECT:

Analysis of Ice Nucleating Aerosol Measurements during SUCCESS - April, May 1996

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PRINCIPAL INVESTIGATORS:

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Research Objectives and Major Findings:

Global climate studies have shown that clouds strongly influence the Earth's radiation balance, but many of the factors that are thought to control these influences are not well understood. It is known, for example, that water clouds and ice clouds have substantially different microphysical and radiative properties. Ice nuclei (IN) populations, and their response to anthropogenic emissions, are especially important in this context because the abundance and characteristics of IN can influence the concentration, shape and phase of cloud particles, the size and persistence of clouds, precipitation efficiency and radiative exchange processes. An evaluation of the effects that aircraft exhaust may have on climate, through effects upon ice nuclei abundance and natural cloud processes, is limited by the lack of appropriate measurements that characterize ice nuclei in both relatively unperturbed ("background") air and in air that has been influenced by aircraft exhaust. The central objective of the SUCCESS project is to assess the effect that aircraft exhaust can have on climate through direct effects (aerosols and contrails) and through indirect effects (clouds). Ice nuclei measurements in background and aircraft-affected air are crucial for this endeavor.

During the April-May 1996 SUCCESS project, we operated instruments on NASA's DC-8 Airborne Laboratory and obtained measurements of the IN and CN (total particle number). We also collected IN and non-IN aerosols for single particle electron microscope (EM) analysis of chemical composition, morphology and size. These data complemented the wide variety of observations from other participants and are among the first comprehensive observations of this type ever obtained in the upper troposphere.

Our participation in SUCCESS spanned three years. The first year was spent in preparation for the field project and was supported under grant NAG2-924. A supplement to that grant supported our second year efforts, in particular the field project. This report is for our work during the third year, which was supported under grant NAG2-1109. The results for all three years were summarized in a final report submitted in September 1997. The major accomplishments and findings during the third year were as follows:

1) Completion of data reduction and quality assurance analyses of SUCCESS measurements;

2) Submission of final data sets to the SUCCESS archive;

3) Participation in workshops to select and analyze SUCCESS case studies;

4) Organizing and conducting a CN instrument intercomparison workshop;

5) Analysis of selected cases and deriving functional descriptions of IN concentration and activity suitable for use in a numerical model;

6) Numerical model studies to investigate the effect of IN properties on cloud microphysical processes and properties using an adiabatic parcel model;

7) A finding that concentrations of heterogeneous IN were not significantly altered in aircraft exhaust, for the temperatures and supersaturation conditions of our measurements;

8) The first determination of the chemical composition of upper tropospheric IN, indicating enhanced contributions of crustal and carbonaceous components, relative to the numbers of those particle types in the total aerosol. Metallic species were also enhanced in those IN samples from aircraft exhaust-influenced air.

Summary of Research

This section describes our research activities during year three of this effort. In the second year, preliminary archive data sets were submitted to the SUCCESS archive. After additional analyses, final versions were prepared and submitted. These are included on the SUCCESS CD-ROM data editions that were recently released by NASA Ames.

Over the range of temperature and supersaturation conditions of our measurements (-15 to -40°C, and from ice saturation to ~15% water supersaturation), IN concentrations ranged from < 0.1 to ~500 per liter, being generally greater at colder temperatures and higher supersaturations. To estimate the potential of aircraft exhaust as a source of IN, we examined data from six days of the field project when the DC-8 was following closely behind other aircraft. CN concentrations were used to discriminate exhaust regions from background air, so that the IN concentrations from these two regions could be compared. During penetrations of aircraft exhaust plumes, CN exhibited a very strong response, but the IN did not. there was no strong evidence that exhaust is a significant source of ice nuclei for the temperature and humidity conditions of our measurements.

In April 1997, a microphysical workshop was convened at NCAR to select cases for in depth analyses and to address questions about the consistency of cloud ice crystal measurements (size distributions and mass concentrations) and aerosol size distributions. We attended this meeting and contributed to the discussions. A particular concern was identified in the CN measurements. On the DC-8, CN measurements were obtained by four different investigator groups, using commercially available instrumentation. The DC-8 SUCCESS CN data showed long periods where the measurements were in substantial agreement, but there were also periods with large discrepancies. Several possible factors were identified that could help explain these discrepancies, including minimum detectable particle size, response at reduced pressures, and location of sample inlet on the aircraft.

In August 1997, a CN instrument intercomparison workshop was organized at CSU to investigate the nature and origins of this problem. A separate summary of the workshop is being prepared, but a short description is included here. Participants included CSU, NCAR (C. Twohy) and NCAR (W. Cooper and C. Brock). Instruments included TSI models 3076, 3010 and 3025a. Three kinds of experiments were performed: (1) response versus aerosol particle size, (2) response at low pressures, corresponding to the upper troposphere/lower stratosphere (~150mb), and (3) response during rapid pressure changes. Monodisperse ammonium sulfate particles, from ~20nm to ~300nm, were generated and stored in a large pressure vessel. The various instruments sampled from a common manifold, with and without the sample lines that were used on the aircraft. Analyses of workshop data are continuing, but we can offer some preliminary conclusions. Losses within piping leading to the counters can be substantial, up to 40%; airborne measurements should be corrected for such losses. With steady pressure or slowly decreasing pressure, the CN concentrations were generally in agreement. Rapid pressure changes produced fluctuations in the apparent CN concentration. We attributed this to limitations of the instruments' critical orifices to regulate the flow. Because of this sensitivity to pressure changes, the placement of sample inlets on the aircraft can be very important.

Functional descriptions of IN activity were derived from measurements and used as input for simulating the formation of ice crystals in the two SUCCESS wave cloud cases, April 30 and May 2, with ambient temperatures ~-42 and ~-65°C. The simulations included both heterogeneous and homogeneous freezing nucleation. The results showed that in both cases, the heterogeneous contribution produces crystals earlier and in much lower concentrations than the subsequent homogeneous process. This modifies the sizes of cloud particles and yields a bimodal distribution.

Results from electron microscopy and x-ray analyses showed that crustal components and metals dominated the IN fraction in continental air, with a wider range of chemical composition in samples from the full aerosol spectrum. The IN fraction from marine air samples had fewer crustal components, while IN from exhaust samples were enriched in metals.

Results from our SUCCESS analyses were reported at a number of meetings, including the 1997 AEAP Meeting in Virginia Beach, the Spring Meeting of the American Geophysical Union in Baltimore, a WMO measurements workshop in Mexico, and the annual meeting of the American Association for Aerosol Research in Denver. Three manuscripts were prepared and submitted to the SUCCESS Special Issue in *Geophysical Research Letters*; they have since been accepted for publication. These presentations and papers are listed in the Publications and Conferences section below.

Publications and Conferences

Peer-Reviewed Publications

DeMott, P.J., D.C. Rogers and S.M. Kreidenweis. 1997: The susceptibility of ice formation in upper tropospheric clouds to insoluble aerosol components. *J. Geophys. Research*, 102 D16, 19575-19584.

Kreidenweis, S.M., Y. Chen, D.C. Rogers and P.J. DeMott, 1997: Isolating and identifying atmospheric ice-nucleating aerosols: A new technique. *Atmospheric Research*, (accepted)

Rogers, D.C., P.J. DeMott, S.M. Kreidenweis, Y.Chen, 1998: Measurements of Ice Nucleating Aerosols during SUCCESS. *Geophys. Res. Lett.* (accepted)

DeMott, P.J., D.C. Rogers, S.M. Kreidenweis and Y. Chen, 1998: The role of heterogeneous freezing nucleation in upper tropospheric clouds: Inferences from SUCCESS. *Geophys. Res. Lett.* (accepted)

Chen, Y., S.M. Kreidenweis, L.M. McInnes, D.C. Rogers and P.J. DeMott, 1998: Single particle analyses of ice nucleating aerosols in the upper troposphere and lower stratosphere. *Geophys. Res. Lett.* (accepted)

Gerber, H., C.H. Twohy, B. Gandrud, A.J. Heymsfield, P.J. DeMott and D.C. Rogers, 1998: Measurement of wave-cloud microphysics with two new aircraft probes. *Geophys. Res. Lett.* (accepted)

Conferences

Rogers, D.C., P.J. DeMott, S.M. Kreidenweis and Y. Chen, 1997: Airborne measurements of ice nucleating aerosols during NASA-SUCCESS. Conf. Atmospheric Effects of Aviation, Virginia Beach.

Chen, Y., S.M. Kreidenweis, P.J. DeMott and D.C. Rogers, 1997:The chemical characteristics of heterogeneous ice nuclei in the upper troposphere and lower stratosphere -- May 4 and May 8 SUCCESS case studies. Conf. Atmospheric Effects of Aviation, Virginia Beach.

DeMott, P.J., D.C. Rogers, S.M. Kreidenweis and Y. Chen, 1997: Use of the SUCCESS ice nuclei measurements to infer ice formation mechanisms in upper tropospheric clouds. Conf. Atmospheric Effects of Aviation, Virginia Beach.

Rogers, D.C., P.J. DeMott, C. Twohy, D. Hagen and W.A. Cooper, 1997: Intercomparison of condensation nuclei measurements on the DC-8 during SUCCESS. Conf. Atmospheric Effects of Aviation, Virginia Beach.

Rogers, D.C., P.J. DeMott, S.M. Kreidenweis, Y. Chen, 1997: Airborne measurements of ice nucleating aerosols. Annual meeting, American Association for Aerosol Research, Denver.

Chen, Y., S.M. Kreidenweis, P.J. DeMott and D.C. Rogers, 1997: The chemical characteristics of heterogeneous ice nucleating aerosols in the upper troposphere and lower stratosphere - May 4 and May 8 SUCCESS case studies. Annual meeting, American Association for Aerosol Research, Denver.

Rogers, D.C., F. Brechtel and S.M. Kreidenweis, 1997: Aerosol losses in metal bellows pumps used for aerosol sampling. Annual meeting, American Association for Aerosol Research, Denver.

DeMott, P.J., D.C. Rogers, S.M. Kreidenweis and Y. Chen, 1997: Upper tropospheric aerosols and ice nuclei: Case studies from the NASA SUCCESS program. Annual meeting, American Association for Aerosol Research, Denver.

DeMott, P.J., D.C. Rogers, S.M. Kreidenweis and Y. Chen, 1997: Physical and Chemical Studies of Ice Nuclei During SUCCESS. Annual meeting, American Geophysical Union, Baltimore.

Rogers, D.C., P.J. DeMott, S.M. Kreidenweis and Y. Chen, 1997: Ice nucleating aerosols: Measurement techniques, interpretation and potential for modeling applications. WMO Workshop on Measurements of Cloud Properties for Forecasts of Weather and Climate, World Meteorological Organization, WMO/TD No. 852, 203-211.