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FINAL REPORT

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PROJECT:

Measurements of Ice Nucleating Aerosols in the Upper Troposphere and Lower Stratosphere

NASA Grant #: NAG 2 - 924

PRINCIPAL INVESTIGATORS:

S.M. Kreidenweis, D.C. Rogers and P.J. DeMott
Department of Atmospheric Science
Colorado State University

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May 1, 1994 to April 30, 1997

Effective Project Start Date:
September 1, 1994

September 2, 1997

FINAL REPORT:

Measurements of Ice Nucleating Aerosols in the Upper Troposphere and Lower Stratosphere

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. Ice nuclei (IN) populations, and their response to anthropogenic emissions, are especially important for high-altitude cold clouds. An evaluation of the effects that aircraft exhaust may have on climate, through effects upon ice nuclei abundance and thus upon natural cloud processes, is limited by the lack of appropriate measurements that characterize ice nuclei in both relatively unperturbed ("background") air masses, and in air masses strongly influenced by aircraft exhaust.

The focus of this project was the measurement and characterization of IN in the upper troposphere and lower stratosphere. The design of the airborne version of the CSU continuous flow diffusion (CFD) chamber, used to detect IN at controlled temperatures and ice supersaturations, was finalized under this project. The instrument was deployed in the Spring 1996 SUCCESS project, where our contributions were (1) the measurement of the number concentrations of IN in background air, and in aircraft exhaust regions; (2) the measurement of total number concentrations of particles less than 3 μm in diameter, to determine the fraction of those particles active as IN; and (3) the separation of the IN and non-IN fractions, and subsequent determination of the elemental composition and morphologies of both fractions.

To accomplish item (3), we exploited the rapid growth of activated IN to supermicron-sized ice crystals in the CFD, which permitted inertial separation of the activated from the non-activated particles. Each fraction was then impacted onto appropriate substrates and analyzed using scanning and transmission electron microscopy (SEM and TEM) and energy-dispersive x-ray (EDS) spectrometry. The development of the inertial separator and the testing of the separation and single-particle analysis techniques were carried out in our laboratory in proof-of-concept experiments.

Our major accomplishments and findings under this grant were as follows:

(1) measurements of total particle number concentrations in the mid- and upper-troposphere, adding significantly to the current limited data base of vertical profiles of particle concentrations and their variations with location and meteorological conditions (archived data);

(2) the first real-time, in-situ measurements of upper tropospheric heterogeneous IN concentrations, and their variations with location and meteorology (archived data);

(3) a finding that concentrations of heterogeneous IN were not significantly altered in aircraft exhaust, for the temperatures and supersaturations at which our data were taken;

(4) 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:

Our major accomplishments during Year 1 were:

- Development of aerosol and crystal collection techniques
- Development of single-particle chemical characterization techniques
- Modification of system design for DC-8 and initial testing

Our major accomplishments during Year 2 were:

- Completion of system design and proof-of-concept experiments
- Participation in SUCCESS mission
- Reporting of theoretical and laboratory findings in the scientific literature
- Reporting of initial SUCCESS results at scientific meetings

Our major accomplishments during Year 3 were:

- Data reduction and submission of all SUCCESS data to the archive
- Participation in SUCCESS data comparison workshops
- Reporting of SUCCESS results at the AEAP Meeting
- Organization of CN counter intercomparison workshop
- Preparation of three manuscripts submitted to the *GRL* SUCCESS Special Issue

These activities are described in more detail in the following sections.

1 Completion of the airborne CFD instrument and flight testing

Although primarily supported under separate funding from NSF, the development of the airborne version of the CFD was critical to this project, and our progress in support of the SASS research is briefly reviewed here. A new compact refrigeration system for the airborne CFD was assembled. Laboratory tests showed that the chamber wall temperatures are uniform, and the chamber can be cooled for sampling as cold as -40 °C. Development was completed of software for the aircraft instrument system, which provides for instrument control, real-time display, and data recording.

Through NCAR and NSF, we applied for and received flight time on the NSF-facility University of Wyoming King Air. The CFD system was mounted the aircraft, and four test flights were conducted in Wyoming early in February 1996.

2 Development of aerosol and crystal collection and analysis techniques

Our field experiment methodology involved continuous sampling of ambient air, outside of cloud regions, and exposing the sample stream to controlled ice-supersaturation conditions and cold temperatures inside the CFD. Ice nuclei active at these conditions will grow, as they flow through the CFD chamber, to ice crystals with diameter (D_p) larger than about 3 μm. Inertial separation of these large ice crystals from unactivated aerosol particles (D_p < 2 μm) was then used to obtain the two aerosol fractions - IN and non-IN - for subsequent chemical analyses. Both fractions were analyzed dry; that is, the ice was evaporated from the IN after separation, and the residual particles were analyzed.

We designed a five-jet impactor for crystal collection, with a 50% cutoff size near 2 μm and a specially-designed stage that accepts a TEM grid as collection substrate. Calibrations were performed for various flow rates and for one, two or three jets blocked, since in operation the total flow rate through the instrument may be varied, changing the cutoff diameter unless the flow rate through each jet is also adjusted. Theoretical estimates of the change in cutoff diameter with pressure changes expected during in-flight operation showed that this would have a relatively small influence on the cutoff size. For comparison with the IN fraction, we also collected ambient aerosol to obtain information on the overall aerosol composition. Since the number concentrations of the total ambient aerosol was orders of magnitude higher than the concentrations of IN, there was a much larger sample of non-IN particulate matter to deal with. Separation by size was necessary to avoid masking of the chemical characteristics of the more numerous smaller particles by those of the less numerous, but more massive, large particles. A seven-stage, commercial PIXE cascade impactor was used in the laboratory and field experiments to size-segregate the total aerosol sample stream. For this purpose, the impactor stages were loaded with TEM grids, which were subsequently subjected to TEM / EDS single-particle analyses.

Both the five-jet and PIXE impactor grid samples from SUCCESS, together with blanks, were analyzed at the CSU Electron Microscopy Center by Yalei Chen, a Ph.D.-level graduate student. In preparation for this project, he completed training and certification on the scanning and transmission electron microscopes at CSU, and on the use of the energy-dispersive x-ray spectrometer. Lab studies were used to develop grid handling techniques, sample recording methodologies, single-particle location, recording and classification techniques, and methods for the interpretation of EDS spectra. Computer / video image capture hardware and image analysis software were used as means for storing information on collected particle sizes and concentrations. Dr. Lynn McInnes, presently at NOAA in Boulder, CO, is an expert in single-particle EM analysis techniques, and served as a consultant to our work in chemical characterization. In addition to working at the CSU facilities, she also performed analyses for us at the National Center for Electron Microscopy in Berkeley, CA. These specialized facilities permitted identification of lower-atomic weight elements and some quantification of relative amounts of elements in mixed particles.

3 Proof-of-concept experiments

Laboratory proof-of-concept experiments were completed and reported in a poster and an extended abstract presented at the 14th International Conference on Nucleation and Atmospheric Aerosols, 26-30 August 1996, Helsinki, Finland; a manuscript on this work has been accepted for publication in *Atmospheric Research*. The experiments demonstrated the separation of IN from non-IN in the CFD, and also showed qualitative agreement between optically-counted IN and total particles deposited on the sample grid, confirming good crystal collection efficiency for our impactor design. Two types of experiments were performed. In the first, a mixed stream of AgI and NaCl particles of known, submicron size was processed in the CFD; AgI serves as an ice-nucleating agent, whereas NaCl does not. The processed stream was then fed through the PIXE impactor. It was expected that ice crystals, formed on AgI particle and grown to sizes larger than 2 μm, would be collected on the upper stages of the PIXE, with smaller unactivated particles deposited on lower stages. This was confirmed by chemical analyses of the stages: only AgI particles were found on the two largest (D_p > 2 μm). In the second type of experiment, a natural air sample was processed in the CFD, and particles in the processed stream again collected in the PIXE. The IN fraction (the particles collected as crystals on the largest stages) was dominated by mineral particles containing Ca and Si. The unactivated aerosol contained additional elements, primarily S, associated with more hygroscopic material.

4 Participation in SUCCESS Mission

Our activities from February through May 1996 were focused on preparing for and participating in the Spring 1996 SUCCESS Mission. Preparations included installing the system in a DC-8 rack and testing, and shipping to Ames for inspection and installation on the DC-8. We met all schedule target dates and our instrumentation operated during every DC-8 mission flight. IN were measured over a wide range in temperature (-15 to -40 °C) and humidity (ice saturation to 20% water supersaturation). To our knowledge, our experiments were the first attempt to obtain relatively fast response continuous measurements of IN concentrations, in real time, from an airborne platform. We also made continuous measurements of condensation nuclei (CN) which were used to determine when the DC-8 was in an exhaust plume, and to define atmospheric vertical layers. IN and ambient aerosols were collected using the impaction techniques described above, and were processed at the CSU Electron Microscopy Center.

Our full CN data set was submitted to the archive shortly after the conclusion of the mission. Data requiring post-experiment processing (IN concentrations and single-particle chemical composition) were added as they became available; this process has been completed.

5 Post-Mission Data Analyses

Immediately following the conclusion of the mission, our efforts turned to post-calibration of instruments and to analysis of mission data. The CN data clearly identified aircraft exhaust penetrations, and were used to segregate IN concentrations and chemical composition data into background and contrail-influenced subpopulations. Typical background IN concentrations were on the order of about 1 per liter, with a range from 0 to several; large increases or decreases in freezing nuclei (at ~ -30 °C) were not apparent in jet exhaust regions. The responses of IN concentrations to temperature at high supersaturations, and particle mixing states deduced from chemical analyses, were used in a modeling study to estimate the effects of heterogeneous IN on cloud microphysics. We found that the heterogeneous IN activated first and formed larger crystals, sometimes leading to a bimodal cirrus crystal size 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 in samples from marine air had fewer crustal components, while IN in exhaust samples were enriched in metals.

The results from these analyses were summarized in three manuscripts which we prepared and submitted to *Geophysics Research Letters* for publication in the SUCCESS Special Issue.

6 Extensions

Our laboratory work using the prototype CFD and theoretical work in support of experiment design suggested a number of additional studies of interest to the science issues addressed in our SASS project. We explored some of these in conjunction with the tasks performed as part of this project.

First, a theoretical study of the effects of heterogeneous nucleation upon ice formation in clouds was completed and accepted for publication in the *Journal of Geophysical Research*. Second, through his involvement with this research, graduate student Yalei Chen developed a proposal for laboratory studies aimed at investigation of homogeneous and heterogeneous ice nucleating properties of known aerosols, and was awarded a NASA Global Change Fellowship to pursue this work. His Ph.D. dissertation, under the direction of PI Kreidenweis, will be focused upon IN characterization, and upon potential climate impacts of aircraft-induced changes in ambient IN characteristics. The SUCCESS mission data, in addition to data he obtains in controlled laboratory experiments, will form the foundation for this work.

Third, we initiated a CN measurement intercomparison, with SUCCESS PIs Twohy, Cooper, and Hagen. We hosted a 2-day instrument intercomparison workshop at CSU in August 1997, and are distributing results to the archive and to the community.

7 Conference Participation, Publications and Presentations

The three Principal Investigators - Kreidenweis, Rogers, and DeMott - attended the SASS Workshop at NASA Ames Research Center in June 1994. Our research objectives and scientific approach were outlined in a presentation, and we participated in working groups examining the effects of aircraft on cirrus.

PI Rogers attended the 5th Annual AEAP Conference in Virginia Beach in April 1995, and presented two papers from our group.

PI DeMott attended the 14th International Conference on Nucleation and Atmospheric Aerosols, 26-30 August 1996, Helsinki, Finland, and presented two papers.

The three PIs attended the October 1996 SUCCESS mission meeting in Colorado, where we presented papers discussing initial analyses of our observations of IN number concentrations and chemical composition in different regimes sampled during SUCCESS.

PIs Rogers and Kreidenweis attended the AEAP Meeting in VA Beach in March 1997 and presented four papers on our SUCCESS findings.

The three PIs attended the SUCCESS microphysics data workshop at NCAR in April 1997 to discuss selection of case studies for further analyses.

PI DeMott attended the Spring Meeting of the AGU in May 1997 and presented an invited paper on our SUCCESS data and analyses.

Presentations and Proceedings

"Ice nucleating aerosols: Plans for high altitude measurements and chemical characterization," D.C. Rogers, P.J. DeMott, Y. Chen, and S.M. Kreidenweis, presented at the 5th Annual AEAP Conference in Virginia Beach, VA, April 23 - 28, 1995.

"Susceptibility of ice formation in upper tropospheric clouds to the quantity and size of insoluble components in mixed-aerosols," P.J. DeMott, D.C. Rogers, and S.M. Kreidenweis, presented at the 5th Annual AEAP Conference in Virginia Beach, VA, April 23 - 28, 1995.

"Isolating and identifying atmospheric ice-nucleating aerosols: A new technique," Y. Chen, S.M. Kreidenweis, D.C. Rogers, and P.J. DeMott, presented at the 14th International Conference on Nucleation and Atmospheric Aerosols, 26-30 August 1996, Helsinki, Finland.

"Studies of homogeneous and heterogeneous ice formation by aerosols in upper tropospheric cloud conditions," P.J. DeMott, Y. Chen, D.C. Rogers and S.M. Kreidenweis, presented at the 14th International Conference on Nucleation and Atmospheric Aerosols, 26-30 August 1996, Helsinki, Finland.

"Analysis of IN and total aerosol composition," S.M. Kreidenweis, Y. Chen, L. McInnes, D.C. Rogers, and P.J. DeMott, presented at the SUCCESS Meeting, October 1996.

"Is aircraft exhaust a source of ice nuclei?," D.C. Rogers, P.J. DeMott, S.M. Kreidenweis, and Y. Chen, presented at the SUCCESS Meeting, October 1996.

"Single particle analyses of IN and total aerosol composition," Y. Chen, S.M. Kreidenweis, L. McInnes, D.C. Rogers, and P.J. DeMott, presented at the AEAP Conference in Virginia Beach, March 10-14, 1997.

"Intercomparison of condensation nuclei measurements on the DC-8 during SUCCESS," D.C. Rogers, C. Twohy, D. Hagen, P.J. DeMott, and A. Cooper, presented at the AEAP Conference in Virginia Beach, March 10-14, 1997.

"Airborne measurements of ice nucleating aerosols during NASA-SUCCESS," D.C. Rogers, P.J. DeMott, S.M. Kreidenweis, and Y. Chen, presented at the AEAP Conference in Virginia Beach, March 10-14, 1997.

"Use of the SUCCESS ice nuclei measurements to infer ice formation mechanisms in upper tropospheric clouds," P.J. DeMott, D.C. Rogers, S.M. Kreidenweis, and Y. Chen, presented at the AEAP Conference in Virginia Beach, March 10-14, 1997.

"Physical and chemical studies of ice nuclei during SUCCESS," P.J. DeMott, D.C. Rogers, S.M. Kreidenweis, and Y. Chen, invited paper, presented at the AGU Spring Meeting, June 1997.

Peer-Reviewed Publications

P.J. DeMott, D.C. Rogers, and S.M. Kreidenweis, "On the susceptibility of ice formation in upper tropospheric clouds to insoluble aerosol components," accepted for publication in *J. Geophys. Res.*

S.M. Kreidenweis, Y. Chen, D.C. Rogers, and P.J. DeMott, "Isolating and identifying atmospheric ice-nucleating aerosols: A new technique," accepted for publication in *Atmos. Research.*

D.C. Rogers, P.J. DeMott, S.M. Kreidenweis, and Y. Chen, "Measurements of ice nucleating aerosols during SUCCESS," submitted to *Geophys. Res. Lett.*

P.J. DeMott, D.C. Rogers, S.M. Kreidenweis, Y. Chen, C. Twohy, D. Baumgardner, A. Heymsfield and K. Chan, "The role of heterogeneous freezing nucleation in upper tropospheric clouds: Inferences from SUCCESS," submitted to *Geophys. Res. Lett.*

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

8 Negative Inventions Statement

I, as Principal Investigator on this project, certify that to the best of my knowledge, no invention or new process which might be patentable was conceived or reduced to practice by personnel working under this award.

Sonia M Kreidenweis 9/2/97

Sonia M. Kreidenweis