Final Report

Work Carried out on NASA Grant NAG 1 1804

"Theoretical Studies of Processes Affecting the Stratospheric and Free Tropospheric Aerosols"

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Introduction

This report describes the work done with funding from NASA Grant NAG 1 1804 during the past three years. Funding commenced in June, 1996 and had a planned duration of three years. This report covers the time period June 1996 to June 1999. Here we present a short description of the projects carried out and documentation of the work done in terms of publications, papers presented, and conferences attended.

1. Microphysical Modeling

This category consist of two related tasks, namely the development of a simple microphysical model for modeling the Pinatubo plume and carrying out a study of sulfate particle formation in a volcanic plume.

The work on modeling the Pinatubo plume was carried out in collaboration with Dr. Howard Houben at NASA Ames Research Center. Some of the work was presented at conferences and in published papers as described below. The main thrust of the more recent research was work related to the microphysical and dynamical model named CARMA that was developed at Ames Research Center, primarily by Drs. Eric Jensen and Andrew Ackerman. This work is not entirely finished, as we are still actively involved in incorporating sulfuric acid microphysics into the model. However, the model itself does work and has been used extensively by several researchers to study the formation of uppper tropospheric clouds and aerosols.

Concerning the study of sulfate particle formation in a volcanic plume, we have developed a new approach to the problem of particle formation methods in an environment containing gas phase sulfuric acid. The technique which we developed was

described in a journal article and submitted to Science, but it was not accepted for publication due to a negative review. The paper is being re-written and will be submitted to some other journal. The basic idea is the fact that for extreme conditions such as encountered in a volcanic plume, the nucleation process can be described as the coagulation of small molecular clusters containing only one or two sulfuric acid molecules.

2. Analysis of Sun Photometer Measurements

We have been involved in studies of the data obtained by the NASA Ames Sun photometer. These studies are led by Dr. Phillip Russell. I have been meeting on a regular basis with Dr. Russell and his group. I have been also collaborating with a member of his group (J. Bauman) on analysis of SAGE satellite data. Of particular interest to us have been the data obtained during the TARFOX and ACE2 measurement campaigns which contain a significant amount of new information on tropospheric aerosols. Results from the TARFOX project were presented by me at a meeting of the Russian Aerosol Society in St. Petersburg, Russia in July, 1998.

A part of the analysis has been to consider the problem of incorporating new values for vapor pressures and optical parameters in the studies of the optical properties of sulfate aerosols as a function of environmental conditons such as temperature, relative humidity, etc.

Another part of this analysis involved analyzing the ELSIE program that is used in the aerosol community to determine aerosol extinction from impactor data or from a given aerosol size distribution. We were able to show that the technique used by ELSIE to evaluate indices of refraction lead to values that are not consistent with laboratory studies of the index of refraction of sulfuric acid solutions.

3. Wire Impactor Analysis

I have been working with Dr. Rudolph Pueschel and Dr. Anthony Strawa on analyzing and understanding results obtained with the Ames Wire Impactor. Some of this work is described in a paper whose principal author is Dr. J. Goodman.

One of our projects is to compare extinctions calculated from the number of particles on the wire with those measured by SAGE. Some of this work is being done by one of my graduate students at San Jose State University. The results obtained

so far indicate a persistent discrepancy between the two instruments. The reason for this discrepancy has not yet been determined.

We are also using a simple box model to evaluate observations made by Drs. Pueschel and Strawa during the SONEX mission. This model indicates that some of the particle concentration measurements can be explained by the nucleation of sulfuric acid droplets. The results will be incorporated into a paper describing the SONEX mission.

4. Other Theoretical Studies

In this area we carried out a number of interesting studies. The most interesting ongoing study, as mentioned above, is the study of the formation of sulfate particles. We believe this process is due to the coagulation of small stable sulfuric acid hydrates rather than via homogeneous nucleation.

We have also been working with Dr. Richard Turco of UCLA on a new theoretical explanation for the formation of NAT in polar stratospheric clouds. Some interesting and promising preliminary results have been obtained.

It has been observed that a layer of enhanced nitric acid appears at high altitudes over the poles during winter. For example, it has been observed that between June 15 to July 15 over the South Pole there is a region of enhanced HNO_3 between about 35 and 45 km altitude. We have carried out modeling studies to determine if the layer can be explained by heterogeneous reactions on sulfuric acid droplets. We have developed a box model to carry out the analysis. We are now planning to incorporate our box model into a model for the diabatic descent of air in this region. Results were presented at the Spring AGU meeting in Boston.

We considered the possibility that tropical cyclones are a source for the background stratospheric aerosol layer. Studies carried out during the STEP program showed that ascending air associated with tropical cyclones is injected into the stratosphere. This was proved conclusively by the presence of radon in the stratosphere during studies of the outflow from cyclones. The temperature of this air is also cold enough to explain the extreme aridity of stratospheric air. We carried out studies of the possibility that tropical cyclones inject sulfuric acid particles into the stratosphere. This work involved using both the STEP data and SAGE II data. Future work will include modeling studies with sulfate microphysics coupled to dynamical processes.

Publications

"Global to microscale evolution of the Pinatubo volcanic aerosol derived from diverse measurements and analyses", P. B. Russell, J. M. Livingston, R. F. Pueschel, J. J. Bauman, J. B. Pollack, S. L. Brooks, Patrick Hamill, L. W. Thomason, L. L. Stowe, T. Deshler, E. G. Dutton and R. W. Bergstrom *J. Geophys. Res., 101*, 18745-18763, 1996.

"Microphysical Processes Affecting the Pinatubo Volcanic Plume", Patrick Hamill, Howard Houben, Richard Young, Richard Turco and Jingxia Zhao, Pages 49-59, The Mount Pinatubo Eruption: Effects on the Atmosphere and Climate, NATO ASI Series, vol 142, G. Fiocco, D. Fua and G. Visconti, editors., 1966.

"New evidence of size and composition of polar stratospheric cloud particles", J. Goodman, S. Verman, R. F. Pueschel, Patrick Hamill, G. V. Ferry and D. Webster, *Geophys. Res. Lett, 24*, 615-618, 1997.

"A time-dependent solution for the cluster concentrations in homogeneous nucleation", T. Olson and Patrick Hamill, in "Nucleation and Atmospheric Aerosols, 1996", M. Kulmala and P. Wagner, Eds, Pergamon, 1996, pp 184-187.

"The Life Cycle of Stratospheric Aerosol Particles," Patrick Hamill, Eric Jensen, P.B. Russell and J.J. Bauman, *Bull. Am. Met. Soc, 78*, 1395-1410, 1997.

"A new parameterization of H2SO4/H20 aerosol composition: Atmospheric implications" A. Tabazadeh, O. B. Toon, S. L. Clegg and Patrick Hamill, *Geophys. Res. Lett, 24*, 1931-1934, 1997.

"Soot aerosol in the lower stratosphere: Pole-to-pole variability," R. F. Pueschel, K. A Boering, S. Verma, S. D. Howard, G. V. Ferry, J. Goodman, D. A. Allen and P. Hamill, *J. Geophys. Res., 102*, 13113-13118, 1997.

"Satellite and aircraft observations of tropical cyclones," P. Hamill and H. Selkirk, extended abstract published in Proceedings of International Conference on Tropical Cyclones, Havana, May, 1998.

Papers Presented

"Particle Formation in the Atmosphere," Heterogeneous Chemistry Workshop, Strasbourg, Oct. 1996.

"Sulfate Aerosol Formation Mechanisms," A Seminar Presented at the Department of Civil Engineering, Stanford University, Jan 1966.

"Sulfate Particle Formation - A New Approach," A Seminar presented at NASA AMES Research Center, SGG Weekly Seminar Series, January 1996.

"A Time-Dependent Solution for the Cluster Concentrations in Homogeneous Nucleation." D. Olson and P. Hamill, Fourteenth International Conference on Nucleation and Atmospheric Aerosols, Helsinki, Finland, August 26-30, 1996.

"Particle Formation in the Atmosphere," P. Hamill, Heterogeneous Chemistry Workshop, Strasbourg, France, October 21-23, 1996.

"Hydrate Coagulation as a Mechanism for Particle Formation in the Troposphere," P. Hamill, C. S. Kiang, Azadeh Tabazadeh, IAMAS-IAPSO Joint Assembly, Melbourne, Australia, July, 1997.

"Life Cycle of the Background Stratospheric Aerosol," Patrick Hamill, P. B. Russell, J. Bauman, AGU Spring Meeting, Baltimore, MD, June, 1997.

"Satellite and aircraft observations of tropical cyclones," Patrick Hamill and H. Selkirk, International Conference on Tropical Cyclones, Havana, May, 1998.

"Microphysical Processes Affecting the Formation of the Background Stratospheric Aerosol," Patrick Hamill, 4th International Aerosol Symposium, St. Petersburg, Russia, July, 1998.

"Anthropogenic Aerosol Properties and Radiative Effects: An Overview and Selected Results from TARFOX and ACE 2," P. B. Russell, P. V. Hobbs, L. L. Stowe, J. Livingston, B. Schmid, A. Chien, P. A. Durkee, T. S. Bates, P. K. Quinn and Patrick Hamill (Presented by Patrick Hamill), 4th International Aerosol Symposium, St. Petersburg, Russia, July, 1998.

"Stratosphere-troposphere exchange in the tropics," Patrick Hamill and Eric Jensen, Tropico '99, Havana, Cuba, March, 1999.

" Study of the Formation of Nitric Acid at High Altitudes over the Antarctic Winter Pole," Patrick Hamill, Azadeh Tabazadeh, J. Mergenthaler, J. Kumer, Spring Meeting, American Geophysical Union, Boston, Mass, June 1999.

"Arctic Ozone Hole in a Denitrified Future Colder Stratosphere," Azadeh Tabazadeh, M. L. Santee,H. C. Pumphrey, P. A. Newman, M. Y. Danilin, J. L. Mergenthaler, P. Hamill, and N. Livesey, Spring Meeting, American Geophysical Union, Boston, Mass, June 1999.

Meetings attended

Fourteenth International Conference on Nucleation and Atmospheric Aerosols, Helsinki, Finland, August 26-30, 1996.

Heterogeneous Chemistry Workshop, Strasbourg, France, October 21-23, 1996.

AGU Annual Meeeting, San Francisco, December, 1996.

AGU Annual Meeeting, San Francisco, December, 1997.

AGU Spring Meeeting, Baltimore, June, 1997.

SAGE III Science Team Meeting, August, 1997.

Russian Aerosol Society, St. Petersburg, Russia, July, 1998.

AGU Annual Meeeting, San Francisco, December, 1998.

Tropico '99, Cuban Meteorological Society, Havana, Cuba, April 1999.

AGU Spring Meeeting, Boston, June, 1999.

Gordon Conference, Newport R.I., June, 1999.

SELECTED REPRINTS