NCAR Annual Scientific Report 1995

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A Message from NCAR's Director
Robert Serafin

Dear Friends and Colleagues,

This has been a year of many significant accomplishments at NCAR. Progress has been made in fundamental and applied science, in modeling, in new instrument and facility development, in education, in technology transfer and in societal impacts assessments. We have continued our collaborative efforts with universities and laboratories around the world and our programs remain national and international in scope and influence. I urge you to peruse the descriptions of NCAR's recent accomplishments that are available on the WWW through the NCAR and division home pages.

NCAR has been a leader in implementing change. Our science has become more cross disciplinary and more interactive than ever. Support for NCAR's programs has broadened, with many U.S. government agencies contributing and with substantial augmentation by international organizations and private sector groups. We continue to change as the framework within which science is conducted evolves.

This is a time when considerable stress is being placed on the support base for science in our country. It seems that financial stress will not be substantially relieved, as the nation attempts to balance its budget. It is encouraging however, that funding for the National Science Foundation in particular is being supported by the Administration and by many groups in Congress as a high national priority. Yet, in no one's crystal ball is there an impression that substantial budget increases are likely to occur. This budget climate has led to some growth in selected areas at NCAR, but we have also had to reduce some programs and to reduce staff.

For more than a year, we have been working to establish and implement strategies for ensuring the continued success of NCAR in this budget climate. We believe that there are three characteristics that will be common to successful organizations in the coming years. (1) The work done must be of the highest quality. (2) Organizations must be responsive to the needs of their constituencies. (3) Organizations must be able to adapt to change. We have embraced these principles in planning for the future.

In the coming years I see many opportunities. The public in our country and societies around the world place very high value on the environment. This was evident as the nation responded negatively and vocally to proposed congressional reductions in environmental programs. Our planet is becoming relatively smaller as population grows and technology advances. Because our planet is figuratively shrinking, the environment--weather, climate, air and water quality will all become increasingly important to society. NCAR's research will play an integral and necessary role in achieving a sound scientific understanding of how the earth system works and will contribute substantially to the decisions that society must make in order to protect the environment for future generations.

I believe that NCAR will chart the correct course into the future based on the creative ideas and dedication of its staff. For me it has been and remains a distinct pleasure to be a part of NCAR and to have the opportunity to

work with the many people who make it such an outstanding organization.

Sincerely yours,

Bob Serafin

NCAR's Mission:

The **National Center for Atmospheric Research** focuses on important atmospheric research problems that require major commitments of resources and a wide range of scientific talent over extended periods of time. By housing scientific programs and providing a wide variety of research facilities, facilities support, and related services, NCAR serves the specialized needs of the university research community.

Science

Science programs are carried out by four scientific divisions:

- **Atmospheric Chemistry**,
- **Climate and Global Dynamics**,
- **Mesoscale and Microscale Meteorology**, and the
- **High Altitude Observatory**.

Facilities

Facilities are developed, operated, and maintained by two other divisions:

- **Atmospheric Technology** and
- **Scientific Computing**.

Programs

There are three additional programs.

- The **Research Applications Program** is working to improve predictions of a variety of weather hazards that affect aviation, and to develop forecasting and visualization products for the aviation industry.
- The **Environmental and Societal Impacts Group** is made up of scientists who research the impacts of climate change, both naturally occurring and man-made.
- The **Advanced Study Program** selects and appoints graduate and postdoctoral fellows from numerous disciplines who wish to broaden their research experience during a two year visit at NCAR.

The center is dedicated, in partnership with the universities, to excellence in the atmospheric and related sciences to the benefit of humankind.

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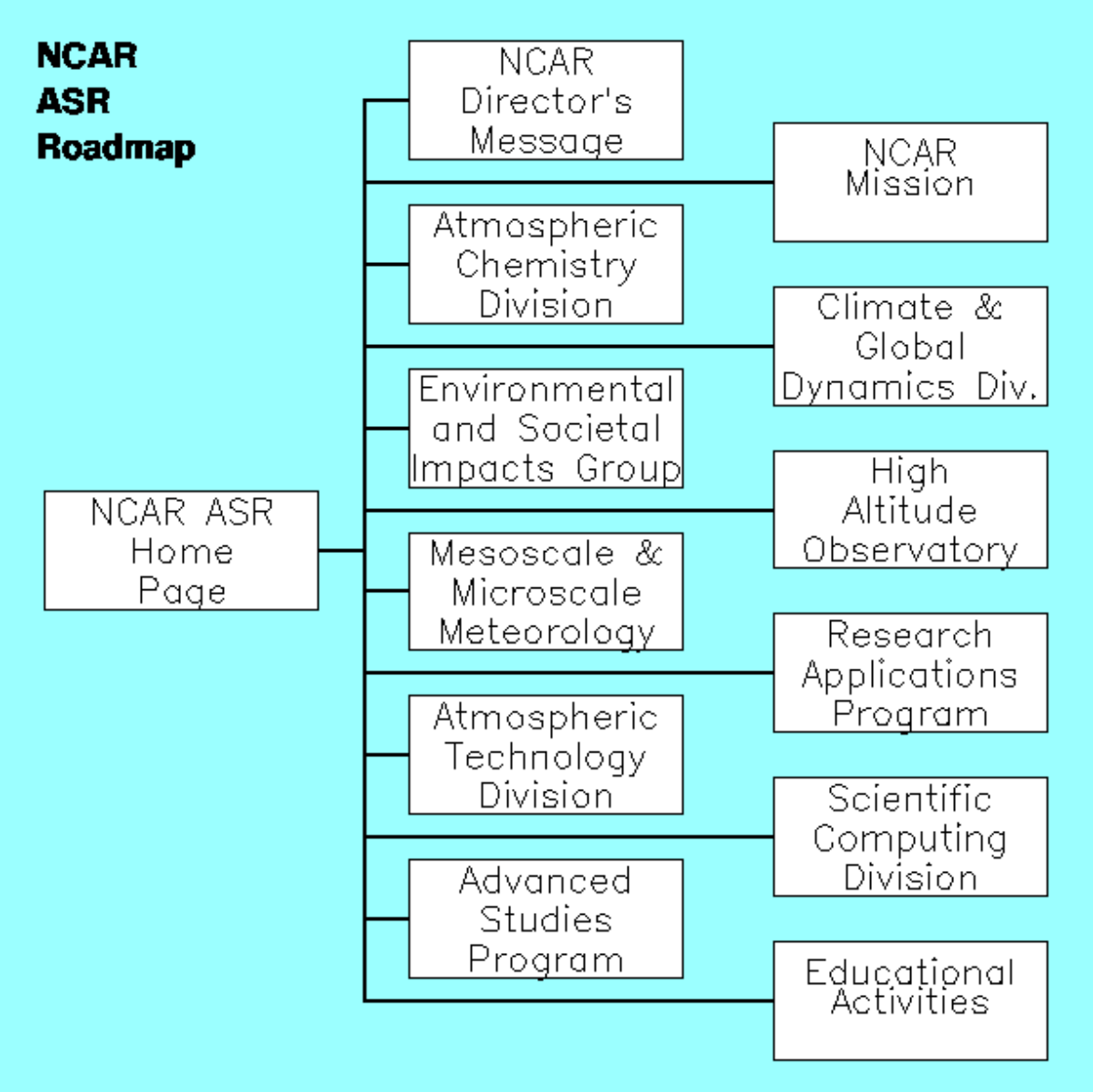

Atmospheric Chemistry Division

ACD Mission

ACD ASR Roadmap

- Significant Accomplishments
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- Publications
- Community Service

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Atmospheric Chemistry Division

I. Introduction

The overall ACD objective is to foster improved understanding of chemical processes that affect the Earth's atmosphere. This is accomplished through interdisciplinary studies that probe the complex interactions between oceans, land, ecosystems, and the atmospheric interface they share. ACD serves the broader scientific community through model development, collaborative studies, implementation of aircraft instrumentation, and development of satellite data and instrumentation. ACD maintains a balance between theoretical and experimental research techniques while developing a hierarchy of numerical models to both interpret measurements and extend our understanding to future predictions.

ACD's research program focuses on chemical and photochemical processes in the global troposphere, chemical interactions between the biosphere and atmosphere, and on dynamical and chemical processes in the middle atmosphere. In collaboration with university and NOAA investigators, ACD has helped to plan a series of missions making use of the NCAR WB-57 to probe questions relating to aerosols, radiation, chemistry and ozone in the stratosphere and troposphere (STERAO). ACD scientists also began fabrication of key instruments for use onboard the WB-57, including measurements of ozone, NO_y, CO, and trace gases. A flexible system to measure the key trace gases OH and H_2SO_4 onboard aircraft was also designed, tested, and deployed; it provides a key component of the Aerosol Characterization Experiment (ACE). With this broad complement of instrumentation, a payload to dramatically improve our understanding of the troposphere and lower stratosphere is being developed.

ACD is playing a leading role in EXPRESSO (Experiment for Regional Sources and Sinks of Oxidants) in Africa. During FY95, preliminary studies were carried out to characterize the field site, and screen over 150 species of vegetation for isoprene and terpene emissions, in preparation for intensive campaigns during the burning and non-burning seasons next year.

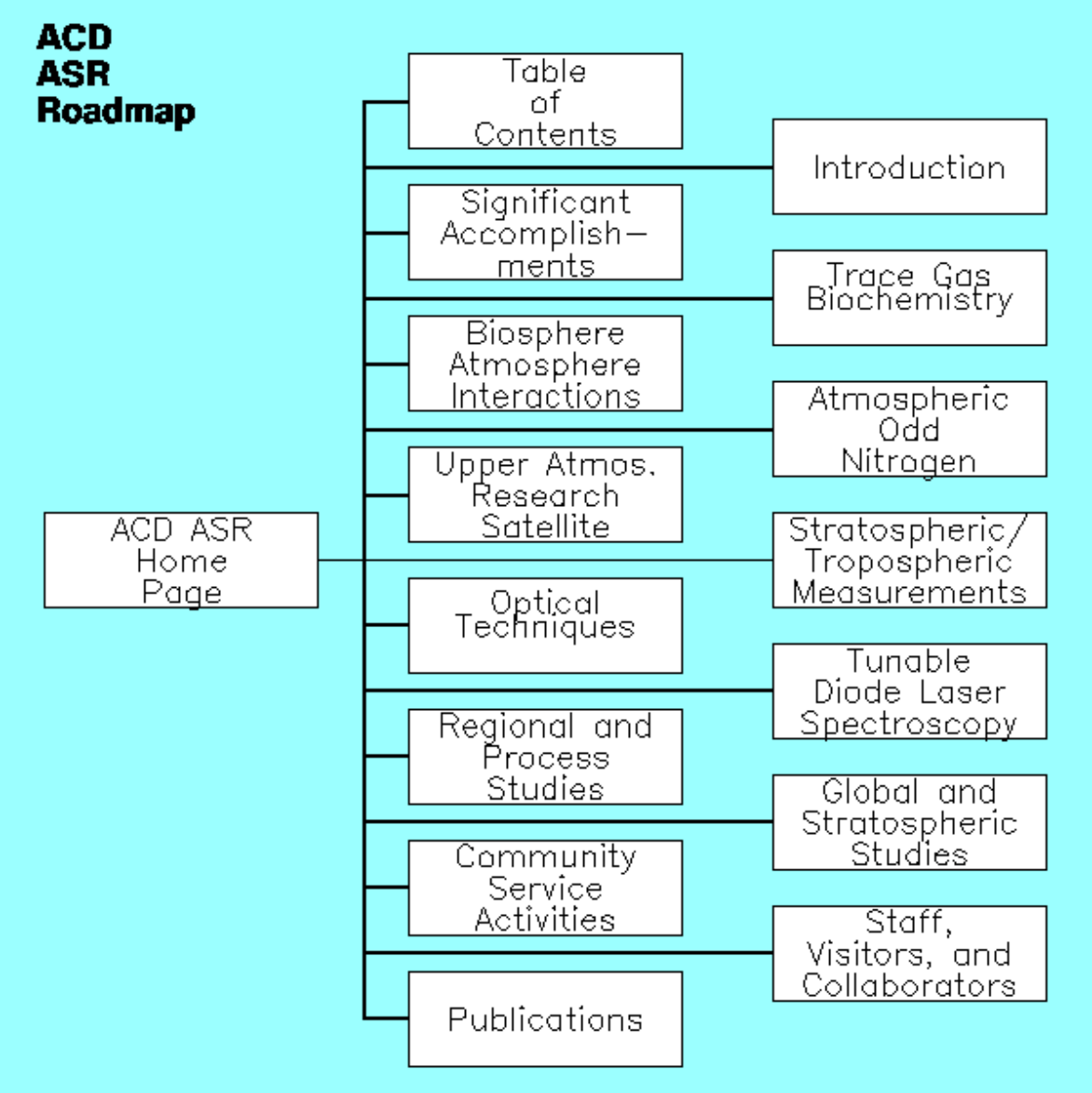
Domestic field studies have also been a major ACD focus, including measurements of biogenic compounds such as terpenes during the Southern Oxidant Study (SOS) Nashville campaign. ACD has also played a leading role in a series of intercomparisons aimed at improving the community's ability to measure formaldehyde, volatile oxygenated organic compounds, and other non-methane hydrocarbons. These trace gases are critical to tropospheric ozone and oxidant formation. Laboratory kinetic studies focussed on a range of questions including the chemistry of tropospheric non-methane hydrocarbons, in particular, the rates of reaction of chlorine atoms with hydrocarbons such as ethane, butane, propane, methanol, and acetaldehyde.

Modeling studies to develop and use a range of models, from the mesoscale to the global scale, have advanced considerably in the past year, including the implementation of detailed chemistry in the Climate System Model (CSM). In addition, research studies have focused on topics such as the impact of biomass burning on tropospheric ozone, effects of subsonic aircraft, and the increases in surface radiation and changes in tropospheric chemistry due to ozone depletion.

Development of satellite instrumentation and algorithm has also advanced together with concurrent scientific analyses. A range of questions relating to the database from the Upper Atmosphere Research Satellite (UARS) have been addressed, including development of algorithms to infer information about aerosol distributions that are critical for stratospheric chemistry.

Through this coordinated research and implementation approach, ACD continues to serve the scientific objectives of a variety of national and international programs of interest to the broad community, particularly the Global Tropospheric Chemistry Program (GTCP). ACDI

This is the roadmap to the ACD Annual Scientific Report. You may click on any box in this image to move to the part of the report that is named in the box.



II. Significant Accomplishments

- Intercomparisons: ACD scientists played leading roles in several intercomparison campaigns in FY95 for key trace gases that are critical to tropospheric ozone formation, including volatile oxygenated organic compounds and formaldehyde. The studies involved a number of universities and agencies and the results will serve the entire scientific community.

- EXPRESSO: Botanists from the Congo and Central African Republic helped classify over 150 regional vegetation species and samples were analyzed for volatile organic carbon. Enclosure measurements in Africa provided initial estimates of isoprene and monoterpene fluxes from major vegetation types along a transect from dry woodland savanna to primary tropical forest, thus providing the beginnings of a comprehensive study of biogenic emissions under this international experiment. Aircraft survey photography and reflectance measurements were also taken to compare with satellite data, and logistics information gathered confirmed the experiment could be conducted.

- Lightning Source of NO_y: Using measurements of reactive nitrogen during a thunderstorm experiment in New Mexico, a direct estimate of the global strength of the lightning source of NO_y has been provided and its importance for the global budget has also been evaluated using models.

- Mission Planning for the Community: Together with university and NOAA investigators, ACD has planned a new series of aircraft experiments to probe key aspects of Stratosphere-Troposphere Experiments: Radiation, Aerosols and Ozone (STERAO). In addition, ACD investigators began the design and fabrication of instruments for the WB-57 to provide high-quality measurements of a variety of important variables including ozone, NO_y, PAN, CO, and long-lived tracers.

- Measurements of OH and H_2SO_4: An aircraft instrument to measure these two gases, which are critical both for the oxidizing capacity of the troposphere and for aerosol formation, was designed, built, and deployed as an essential cornerstone of the Aerosol Characterization Experiment (ACE) on the NCAR C-130. The OH instrument was also modified to enable simultaneous measurements of multiple species and an advanced design inlet with internal calibration port was developed.

- Laboratory Measurements: Among the kinetic studies carried out in FY95, the measurement of the rates of reaction of chlorine atoms with several organic molecules are of particular importance. These studies were done using the complementary techniques of flash photolysis and resonance fluorescence, and are essential for studies of the role of chlorine in determining the distributions and lifetimes of organic compounds such as ethane, butane, propane, methanol, and acetaldehyde.

- Satellite Studies: Algorithm development related to UARS, HIRDLS, and MOPITT has progressed, and complementary scientific studies are illustrated by a detailed analysis of TOMS, SBUV, and SBUV/2 data using a coordinate system based on the structure of the polar vortex. This study has allowed better quantification of trends in ozone by showing that ozone losses occur outside of the polar vortices in both hemispheres and has improved the understanding of their seasonal dependence. Following a successful System Concept Review (SCR), NASA approved HIRDLS as the first instrument to proceed into phase C/D implementation on the CHEM-1 platform scheduled for a December, 2002 launch.

- Trace Gases: Using aircraft observations of chlorofluorocarbons, methyl bromide, and halons, the chlorine and bromine budgets of the lower stratosphere were inferred as functions of latitude and altitude, allowing for quantification of the active chlorine and bromine that is critical for stratospheric ozone depletion.

- Model Development and Application: A hierarchy of models is being developed by the ACD staff for use in interpreting atmospheric measurements and predicting the future state of the atmosphere. These range from a regional

scale chemical model using the MM5 dynamical model fields which has been developed for examining local chemistry, to the global MOZART model that includes about 40 chemical species and makes use of the three-dimensional CSM dynamics model. Modeling applications to current critical scientific questions include the study of the impacts of subsonic aircraft and biomass burning on tropospheric ozone.

ACD Staff

Division Director's Office

Guy Brasseur (director)
Michael Coffey
Teresa Rivas
Donna Sanerib
Paul Sperry
Sharon Vieyra (to July 1995)

Trace Gas Biogeochemistry

Eric Apel
Timothy Gilpin
James Greenberg
Patrick Zimmerman (leader)

Biosphere Atmosphere Interactions

Alex Guenther (leader)
Peter Harley
Lee Klinger

Atmospheric Odd Nitrogen

Frank Grahek
Brian Ridley (leader)
James Walega
Andrew Weinheimer

HO_x Measurements

Christopher Cantrell
Fred Eisele (leader)
Richard Shetter
David Tanner

Laboratory Kinetics

John Orlando
Geoffrey Tyndall (leader)

UARS

Charles Cavanaugh
Cheryl Craig
John Gille (manager)
Lawrence Lyjak
Steven Massie
Daniel Packman

William Randel (leader)
Fei Wu

MOPITT

Paul Bailey (leader)
John Caron
David Edward
John Gille (manager)
Liwen Pan
Laura Rokke
Mark Smith
Charles Spaur
Jinzue Wang

HIRDLS

Philip Arter
Michael Dials
John Gille (leader)
Chris Halvorson
Linda Henderson
Michael Howard
Karl Kneisel (to January 1995)
Joanne Loh (manager)
Judy Oeltjenbruns (to January 1995)
David Wilson
Douglas Woodard

Stratospheric/Tropospheric Measurements

Elliot Atlas (leader)
Frank Flocke
Richard Lueb
Sue Schauffler

Regional and Process Studies

Mary Barth
John Caron
Chris Fischer
Siri Flocke
Claire Granier
Peter Hess
Sasha Madronich (leader)

Global and Stratospheric Studies

Guy Brasseur (leader)
David Erickson
Rolando Garcia
Elisabeth Holland
Anne Smith
XueXi Tie
Stacy Walters

Optical Techniques

Michael Coffey
James Hannigan
William Mankin (leader0

Tunable Diode Lasers

Alan Fried (leader)
Bruce Henry

Technical Support

William Bradley
Edward Ellert
Timothy Fredrick
Paul Sperry (leader)

Administrative Support

Trinh Guenther (to June 1995)
Janice Powell
Teresa Rivas (leader)
Donna Sanerib
Marilena Stone
Sharon Vieyra (to July 1995)
Jack Wainwright

Affiliate Scientists

Aaron Goldman, University of Denver
Susan Solomon, NOAA Aeronomy Laboratory

XII. Visitors and Collaborators

Dates refer to visitor's stay at NCAR during FY95. No dates are given for collaborators who did not visit NCAR.

David Andrews; Oxford University; HIRDLS

Bernard Aumont; University of Paris; June 1995 to July 1995; atmospheric chemical modeling

Linnea Avallone; University of California, Irvine; AASE II

Bradford Baker; University of Colorado; June 1995 to August 1995; Trace gas biogeochemistry

Dennis Baldocchi; NOAA, Oak Ridge; Southern Oxidant Study

John Barnett; Oxford University; HIRDLS

Timothy Bates; NOAA Pacific Marine Environment Laboratory; global distribution of trace gases

William Baugh; University of Colorado; July 1994 to December 1996; Ecosystem studies

Gufran Beig; M. L. Sukkadia University, India; 9-20 July 1995; Global and stratospheric studies

Richard Bevilacqua; Naval Research Laboratory; steering committee, NDSC

Adelrhani Boucham; Belgian Institute for Space Aeronomy; January 1995 to July 1995; Globaland stratospheric studies

William Brune; Pennsylvania State University; photochemistry and heterogeneous processes

Susan Canney; Green College, Oxford, Great Britain; May 1995 to June 1995; EXPRESSO

Simon Chabrillat; Free University of Brussels; March 1994 to May 1996; Global and stratospheric studies

Phillippe Cias; Direcion des Sciences de la Matiere, France; February 1995; Global and stratospheric studies

Candis Claiborne; Washington State University; Southern Oxidant Study

Julie Cole; June 1994 to December 1995; Ecosystem studies

Glen Cota; University of Tennessee; organic halogens in the Arctic

John Daniel; NOAA Aeronomy Laboratory, Boulder; AASE II

Purnendu Dasgupta; Texas Technical University; May 1995; hydrocarbon intercomparison

Gary Davis; University of Saskatchewan; MOPITT

Gregory Deem; University of Colorado; October 1994 to May 1995; Trace gas biogeochemistry

Anne DeRudder; CNRM, METEO-France; June 1995 to July 1995; Global and stratospheric studies

Terry Deshler; University of Wyoming; balloon-borne aerosol size distribution data

Sarah Dizick; University of Colorado; November 1994 to April 1995; Trace gas biogeochemistry

Mark Dombrowski; NASA Goddard Space Flight Center; July 1995 to December 2002; HIRDLS Mark Drier; University of Colorado; October 1994 to September 1995; Ecosystem studies

James Drummond; University of Toronto; MOPITT; field TDLAS system

Lee Elson; Jet Propulsion Laboratory; HALOE

Francis Eparvier; NOAA Space Environment Laboratory; October 1994 to September 1995; Atmospheric odd nitrogen

Paul Eriksen; Danish Meteorological Institute; infrared spectrometry; SESAME

Raymond Fall; University ofColorado; effect of light on the emission of terpenes and other biogenic trace gases

Hans Fast; Atmospheric Environment Service, Canada; Network for Detection of Stratospheric Change

François Figarol; University of Pierre and Marie Curie, France; January 1995 to May 1996; Global and stratospheric studies

Jean-Marc Fracheboud; Oxford University, England; ozone photochemistry

Gene Francis; University of Colorado; August 1993 to August 1996; Global and stratospheric studies

Lars Franzen; Göteborg University, Sweden; successional and Gaia theory

Paul Fraser; Commonwealth Sscientific Industrial Research Organization, Australia; nitrous oxide and halocarbon intercomparison experiment

Verity Fridd; York University, Canada; November 1994 to March 1995; Trace gas biogeochemistry

David Fritts; University of Colorado; gravity wave breaking in the middle atmosphere

Kochy Fung; Atmospheric Assessment Association, Calabasas, California; oxygenated hydrocarbon intercomparison

Zhang Genfafor; Texas Technical University; May 1995 to June 1995; Hydrocarbon intercomparisons

Rolf Gepraegs; Institut Fuer Atmosphaeren Physik , Germany; June 1995 to August 1995; Global and stratospheric studies

Christopher Geron; U. S. Environmental Protection Agency; non-methane hydrocarbon emission modeling

Paul Ginoux; University of Brussels; September 1991 to December 1996; Global and stratospheric studies

Arun Gopalan; State University of New York, Stony Brook; September 1994 to December 1996; CLAES

Peter Gray; Rutherford Appleton Laboratory, UK; HIRDLS

Patrice Gregoire; University of Clermont-Ferrand, France; October 1994; Global and stratospheric studies

David Griffith; University of Wollongong, Australia; Network for Detection of Stratospheric Change

Sylviane Haberkorn; September 1995 to June 1996; atmospheric kinetics

Brad Hall; Washington State University; Southern Oxidant Study

Chris Halvorson; October 1994 to March 1996; HIRDLS

Wei Min Hao; USDA Forest Service; January 1995; Global and stratospheric studies

Brian Heikes; University of Rhode Island; May 1995 to June 1995; Hydrocarbon intercomparison

Detlev Helmig; University of California, Riverside; April 1992 to December 1996; Trace gas biogeochemistry

Daniel Hereid; University of Colorado; January 1995 to September 1996; Ecosystem studies

C. Nicholas Hewitt; Lancaster University, Great Britain; October 1994 to February 1995; Trace gas biogeochemistry

James Holton; University of Washington; HIRDLS

Theresa Huang; University of Michigan; May 1995 to August 1996; Global and stratospheric studies

Andrew Hudak; University of Colorado; EXPRESSO field campaign

Roger Hunneman; Reading University; HIRDLS

Jud Isebrandes; U. S. Forest Service; isoprene emissions from a poplar plantation

Dan Jaffe; University of Alaska; sprintime tropospheric ozone maximum

Anne Jefferson; Georgia Institute of Technology; September 1993 to August 1995; HO_x measurements

Gregory Jenkins; Pennsylvania State University; July 1995 to August 1995; Trace gas biogeochemistry

Brian Johnson; University of Michigan; July 1995 to April 1996; HIRDLS

James Johnson; NOAA Pacific Marine Environment Laboratory; global distribution of trace gases

Nicholas Jones; National Institute for Water and Atmosphere; New Zealand; Network for Detection of Stratospheric Change

Tørben Jørgensen; Danish Meteorological Institute; infrared spectrometry; SESAME

Prasad Kasibhatla; Georgia Institute of Technology; January 1995 to January 1996; UARS

William Kaiser; Ford Motor Company; chlorine chemistry

Carla Kegley-Owen; University of Colorado; July 1993 to July 1996; Atmospheric kinetics

Ken Kim; University of Colorado; May 1995 to October 1995; Trace gas flux measurements

Andrew Kisselelv; A. I. Voeikov Main Geophysical Observatory, Russia; October 1994 to December 1994; Global and stratospheric studies

Thaddeus Kleindienst; Mantech Environmental Laboratory, North Carolina; Hydrocarbon intercomparisons

Kunihiko Kodera; Meteorologial Research Institute, Japan; June 1995 to July 1995; atmospheric chemical modeling

Heidi Krapfl; University of Colorado; May 1995 to May 1996; Stratospheric/tropospheric measurements

Robert Kremer; Colorado State University; EXPRESSO field campaign

Jack Kumer; Lockheed Palo Alto Research Laboratory; CLAES

Michael Kurylo; National Aeronautics and Space Administration; steering committee, NDSC

Brian Lamb; Washington State University; Southern Oxidant Study

Frederic Lacquer; University of Nebraska, Omaha; analytical studies of atmospheric alcohols

Jean-François Lamarque; Catholic University of Louvain; September 1994 to August 1996; Regional chemical transport models

Frederic Laquer; University of Nebraska, Omaha; June 1995 to July 1995; Stratospheric/tropospheric measurements

Yin-Nan Lee; Brookhaven National Laboratories; May 1995 to June 1995; Hydrocarbon intercomparisons

Conway Leovy; University of Washington; HIRDLS

Ann Lee Liu; University of Colorado; June 1995 to August 1995; Stratospheric/tropospheric measurements

Shaw Liu; NOAA Aeronomy Laboratory, Boulder; May 1988 to December 1996; various ongoing collaborations

William Lonneman; U. S. Environmental Protection Agency; oxygenated hydrocarbon intercomparison

Manuel Lopez-Puertas; Instituto de Astrofisica, Granada, Spain; CLAES

Rachel Lum; Georgia Institute of Technology; February 1995 to March 1995; HO_x measurements

Robert MacQueen; Rhodes College; atmospheric research spectrometer

Paul Manning; University of Denver; September 1994 to October 1995; Optical techniques

Gloria Manney; Jet Propulsion Laboratory; ISAMS

Edward Martell; October 1984 to July 1995; Aerosols

James Marti; University of Minnesota; April 1995 to May 1995; HO_x measurements

Andrew Matthews; National Institute of Water and Atmosphere, New Zealand; Network for Detection of Stratospheric Change

R. Leon Mauldin; NOAA; April 1994 to April 1996; HOx measurements

Michel M'Bangui; Central African Institute of Agronomical Research; EXPRESSO

Michele McCarthy; University of Colorado; December 1994 to May 1995; Trace gas biogeochemistry

John McConnel; York University, Ontario, Canada; MOPITT

Stuart McDermid; Jet Propulsion Laboratory; steering committee, NDSC

Michael McIntyre; Cambridge University; HIRDLS

Gerard Mégiè CNRS, France; steering committee, NDSC

Alvin Miller; NOAA; steering committee, NDSC Norman Miller; Lawrence Livermore National Laboratory; November 1994 to December 1994; Ecosystem studies

Kenneth Minschwaner; New Mexico Institute for Mining and Technology; CLAES

Denise Montzka; NOAA; July 1991 to September 1996; Atmospheric odd nitrogen

Jean-Marie Moutsambote; University of Brazzaville, Republic of the Congo; EXPRESSO

Michael Mozurkewich; York University, Toronto; heterogeneous studies on sulfuric acid aerosols

H. G. Muller; University of Sheffield; HIRDLS

Jean-François Müller; Belgian Institute for Space Aeronomy; atmospheric chemical modeling

David Murcray; University of Denver; Network for Detection of Stratospheric Change

Frank Murcray; University of Denver; Network for Detection of Stratospheric Change

Daniel Murphy; NOAA Aeronomy Laboratory, Boulder; aerosol formation and composition

C. T. Mutlow; Rutherford Appleton Laboratory; HIRDLS

Jason Neff; Stanford University; June 1995 to July 1995; Ecosystem studies

Paul Newman; NASA Goddard Space Flight Center; STRAT; steering committee, NDSC

Ole-John Nielsen; Risø National Laboratory, Denmark; acetonitrile oxidation

Justus Notholt; Alfred Wegener University, Germany; Network for Detection of Stratospheric Change

Alan O'Neil; Reading University; HIRDLS

Luanne Otter; University of Witwatersrand, South Africa; June 1995 to August 1995; Trace gas biogeochemistry

Christopher Palmer; Oxford University; HIRDLS

David Parish; NOAA Aeronomy Laboratory; Boulder; P-3 aircraft

Dieter Perner; Kernforschungzentrum, Juelich, Germany; polar stratospheric chemistry; SESAME

Guy Peskett; Oxford University; MOPITT

Thomas Pierce; U. S. Environmental Protection Agency; non-methane hydrocarbon emission modeling

Jean-Pierre Pommereau; Servide d'Aeronomie, Paris; steering committee, NDSC

Robert Portmann; University of Colorado; September 1994 to August 1996; Global and stratospheric studies

Michael Prather; University of California, Irvine; AASE II

Barbara Prezelin; University of California, Santa Barbara; ultraviolet and visible atmospheric radiation

Ronald Prinn; Massachusetts Institute of Technology; steering committee, NDSC

Joseph Prusa; Iowa State University; modeling small-scale gravity wave forcing, propagation and breaking

Manuel Puertas-Lopez; Institude de'Astrofisica de Andulucia, Spain; Remote sensing

John Pyle; Cambridge University; HIRDLS

Richard Ramaroson; Office National d'Etudes et Rechersches Aerosptiales, France; July 1995 to August 1995; Global and stratospheric studies

David Raymond; New Mexico Institute of Technology; New Mexico thunderstorm experiment

Adrienne Regamey; Swiss Federal Institute of Technology; November 1994 to March 1995; Trace gas biogeochemistry

Henry Reichle; North Carolina State University; MOPITT

William Rison; New Mexico Institute of Technology; New Mexico thunderstorm experiment

Clive Rodgers; Oxford University; August 1994 to July 1995; HIRDLS; steering committee; NDSC

Nigel Roulet; McGill University; MOPITT

James M. Russell III; NASA Langley Research Center; HALOE Steven Rutledge; Colorado State University; thunderstorm study

Joseph Sabutis; University of California, Los Angeles; mechanistic model of the stratosphere

Murry Salby; University of Colorado; Augusut 1984 to December 1995; Global and stratospheric studies

Fabrizio Sassi; Universita deglia Studie-L'Aquila, Italy; July 1990 to March 1996; Global and stratospheric studies

Mark Schoberl; NASA Langley Research Center; Vortex Ozone Transport and Tropical Ozone Transport Experiments

Mary Scholes; University of Wittwatersrand, South Africa; April 1994 to october 1994; Ecosystem studies

Jens Sehested; Risø National Laboratory, Denmark; acetonitrile oxidation

Dominique Serca; Paul Sabatier University, Toulouse, France; February 1995 to February 1996; Trace gas biogeochemistry

Scott Sewell; University of Colorado; January 1992 to May 1995; Tunable diode laser

Paul Shepson; Purdue University; oxygenated hydrocarbon intercomparison

Masato Shiotani; Hokkaido University, Japan; July 1995; Global atmospheric changes

Knut Stamnes; University of Alaska; February 1995; Global and stratospheric studies

Gary Starkey; October 1994 to December 1994; Trace gas biogeochemistry

John Streete; Rhodes College; June 1995 to December 1995; Optical techniques

William Sturges; University of East Anglia; organic halogens in the arctic

James Sulzman; June 1994 to July 1996; Ecosystem studies

Elizabeth Sulzman; Colorado State University; EXPRESSO field campaign

Carine Suter; September 1995 to June 1996; Stratospheric/tropospheric measurements

Fredrick Taylor; Oxford University; HIRDLS

John Taylor; Australian National University; successional and Gaia theory

Darin Toohey; University of California, Irvine; AASE II

Geoffrey Toon; Jet Propulsion Laboratory; Network for Detection of Stratospheric Change

O. B. Toon; NASA Ames Research Center; Vortex Ozone Transport and Tropical Ozone Transport Experiments

Ian Tosh; Rutherford Appleton Laboratory, UK; HIRDLS

Klereti Tourpali; Aristole University of Thessaloniki, Greece; March 1995 to May 1995; Global and stratospheric studies

Adrian Tuck; NOAA Aeronomy Laboratory, Boulder; thunderstorm study

Geraint Vaugham; University College of Wales; HIRDLS

James Vedder; NASA Ames Research Center; AASE-II

Peter Venters; Oxford University; HIRDLS

Jean-Bruno Vickos; Central African Instittue of Agronomical Research; EXPRESSO

Lee Vierling; University of Colorado; May 1993 to May 1996; Trace gas biogeochemistry

Andreas Voltz; Kernforschungzentrum, Juelich, Germany; NO_2 photodissociation comparison

Timothy Wallington; Ford Motor Company; alkoxy radical chemistry

Yonghua Wang; Georgia Institute of Technology; mechanistic model of the stratosphere

Zhong Wang; Tianjin University, China; February 1995; Global and stratospheric studies

Elisabeth Weatherhead; Cooperative Institute for Research in Environmental Sciences, University of Colorado; March 1995 to June 1995; Global and stratospheric studies

Alex Weaver; NOAA Aeronomy Laboratory; studies of organic hologen variations

Jeffrey Welker; Colorado State University; June 1994 to December 1994; Ecosystem studies

Robert Wells; Oxford University; HIRDLS

Bryan Wert; University of Colorado; June 1995 to June 1999; Tunable diode laser

Halvore Westberg; Washington State University; June 1995; Hydrocarbon intercomparison

John Whitney; Oxford University; HIRDLS

Eric Williams; NOAA Aeronomy Laboratory, Boulder; measurements, technology and standards group of the Southern Oxidants Study for NO_y

E. James Williamson; Oxford University; HIRDLS

Steven Wofsy; Harvard University; STRAT

Rudolf Zander; University of Liege; Network for Detection of Stratospheric Change

Beiying Wu; Institute of Atmospheric Physics, China; July 1995; Remote sensing

Wanli Wu; University of Michigan; July 1993 to December 1995; modeling the middle atmosphere

Yoko Yokouchi; National Institute for Environmental Studies, Japan; Stratospheric/tropospheric measurements

Jun Zeng; University of Alaska; May 1995 to April 1996; Regional and process studies

Jiangfen Zheng; NOAA Aeronomy Laboratory, Boulder; September 1993 to December 1995; AASE-II

Rodney Zika; University of Miami; oxygenated hydrocarbon intercomparison

XIII. Publications

APEL, E., J. CALVERT, and F. FEHSENFELD*, 1994: The nonmethane hydrocarbon intercomparison experiment (NOMHICE): Tasks 1 and 2. *Journal of Geophysical Research*, **99**, 16,651-16,664.

AVALLONE, L., D. TOOHEY, S. SCHAUFFLER, W. POLLOCK, L. HEIDT, E. ATLAS, and K. CHAN*, 1995: In situ measurements of BrO during AASE II, *Geophysical Research Letters*, **22**, 831-834.

CALVERT, J., G. YARWOOD*, and A. DUNKER*, 1994: An evaluation of the mechanism of nitrous acid formation in the urban atmosphere. *Res. Chem. Intermed.*, **20**, 463-502.

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Editorships of Peer Reviewed Journals

Guy Brasseur, Editor-in-Chief, *Journal of Geophysical Research* (Atmospheres), 1992.

Brian Ridley, Co-Editor with Paul Crutzen and Dieter Ehhalt, *Journal of Atmospheric Chemistry*, 1994.

Elisabeth Holland, Associate Editor, *Journal of Geophysical Research*, 1995.

Committees/Advisory Boards

David Erickson III, United Nations Environment Program (UNEP), Panel Member on the Environmental Effects of Ozone Depletion, 1994 - present.

David Erickson III, Advisory Board Member, Tropical Atmospheric Science Center, University of Puerto Rico, 1995 - present.

David Erickson III, Steering Committee Member, IGAC/GEIA project on Reactive Chlorine Inventory Assessment, 1995 - present.

Michael Coffey, Chairman, NSF Observing Facilities Advisory Panel, 1994-1996.

Michael Coffey, Professor Advisory Panel at University of Toronto for appointment of new chair in Atmospheric Sciences Department.

Elisabeth Holland, Steering Committee, NASA Distributed Active Archive Center, Oak Ridge National Laboratory, 1994 - present.

Elisabeth Holland, Steering Committee, Biosphere-Atmosphere Research Training Grant to EPOB, University of Colorado, 1994 - present.

Elisabeth Holland, Steering Committee, U.S. Trace Gas Network, IGAC/IGBP Project Trace Gas Exchange between Mid-latitude Terrestrial Ecosystems and Atmosphere, 1992 - present.

Elisabeth Holland, Steering Committee, NCAR Student Employment Program for Minority Students, 1995 - present.

Elisabeth Holland, Member Review Panel, National Institute for Global Environmental Change, 1995.

Christopher Cantrell, Nominations Committee of Atmospheric Sciences Section of American Geophysical Union, 1995-1997.

Alan Fried, Treasurer/Secretary Rocky Mountain Optical Society, 1995-1996.

Alan Fried, Member of the International Advisory Committee for the 1996 American Optical Society's Laser Applications for Chemical Analysis Symposium.

Alan Fried, Member of the International Advisory Committee for the 1995 Russian High Resolution Spectroscopy Conference held in Moscow Russia, 1995.

Honors

Miscellaneous

Guy Brasseur, Director, NATO Advanced Study Institute on "The Stratosphere and Its Role in the Climate System," 1995.

Elisabeth Holland, Director, NATO Advanced Study Institute on "Soils and Global Change: Carbon Cycle, Trace Gas Exchange and Hydrology," 1995.

Atmospheric Chemistry Division

I. Introduction

The overall ACD objective is to foster improved understanding of chemical processes that affect the Earth's atmosphere. This is accomplished through interdisciplinary studies that probe the complex interactions between oceans, land, ecosystems, and the atmospheric interface they share. ACD serves the broader scientific community through model development, collaborative studies, implementation of aircraft instrumentation, and development of satellite data and instrumentation. ACD maintains a balance between theoretical and experimental research techniques while developing a hierarchy of numerical models to both interpret measurements and extend our understanding to future predictions.

ACD's research program focuses on chemical and photochemical processes in the global troposphere, chemical interactions between the biosphere and atmosphere, and on dynamical and chemical processes in the middle atmosphere. In collaboration with university and NOAA investigators, ACD has helped to plan a series of missions making use of the NCAR WB-57 to probe questions relating to aerosols, radiation, chemistry and ozone in the stratosphere and troposphere (STERAO). ACD scientists also began fabrication of key instruments for use onboard the WB-57, including measurements of ozone, NO_y, CO, and trace gases. A flexible system to measure the key trace gases OH and H_2SO_4 onboard aircraft was also designed, tested, and deployed; it provides a key component of the Aerosol Characterization Experiment (ACE). With this broad complement of instrumentation, a payload to dramatically improve our understanding of the troposphere and lower stratosphere is being developed.

ACD is playing a leading role in EXPRESSO (Experiment for Regional Sources and Sinks of Oxidants) in Africa. During FY95, preliminary studies were carried out to characterize the field site, and screen over 150 species of vegetation for isoprene and terpene emissions, in preparation for intensive campaigns during the burning and non-burning seasons next year.

Domestic field studies have also been a major ACD focus, including measurements of biogenic compounds such as terpenes during the Southern Oxidant Study (SOS) Nashville campaign. ACD has also played a leading role in a series of intercomparisons aimed at improving the community's ability to measure formaldehyde, volatile oxygenated organic compounds, and other non-methane hydrocarbons. These trace gases are critical to tropospheric ozone and oxidant formation. Laboratory kinetic studies focussed on a range of questions including the chemistry of tropospheric non-methane hydrocarbons, in particular, the rates of reaction of chlorine atoms with hydrocarbons such as ethane, butane, propane, methanol, and acetaldehyde.

Modeling studies to develop and use a range of models, from the mesoscale to the global scale, have advanced considerably in the past year, including the implementation of detailed chemistry in the Climate System Model (CSM). In addition, research studies have focused on topics such as the impact of biomass burning on tropospheric ozone, effects of subsonic aircraft, and the increases in surface radiation and changes in tropospheric chemistry due to ozone depletion.

Development of satellite instrumentation and algorithm has also advanced together with concurrent scientific analyses. A range of questions relating to the database from the Upper Atmosphere Research Satellite (UARS) have been addressed, including development of algorithms to infer information about aerosol distributions that are critical for stratospheric chemistry.

Through this coordinated research and implementation approach, ACD continues to serve the scientific objectives of a variety of national and international programs of interest to the broad community, particularly the Global Tropospheric Chemistry Program (GTCP). ACDI

III. Trace Gas Biogeochemistry (TGB)

Research in the Trace Gas Biogeochemistry project (James Greenberg, Detlev Helmig, Patrick Zimmerman) focused on biogenic volatile organic compounds released into the atmosphere, their sources, and the products of their atmospheric reactions. These emissions have a central place in the carbon cycle (because of their magnitude) and atmospheric chemistry (because of their reactivity and that of their reaction products). In FY95, TGB, in collaboration with the Biosphere Atmosphere Interactions (BAI) project, developed and applied analytical technology to the measurement of trace gas concentrations and fluxes in several ecosystems and conducted laboratory studies to isolate and identify emission processes. In addition, TGB served the atmospheric chemistry community through the organization of intercalibration exercises and analytical technology transfers (Eric Apel, Timothy Gilpin).

Effect of Light on the Emission of Terpenes and Other Biogenic Trace Gases

A series of laboratory experiments were conducted by BAI with C. Nicholas Hewitt (Lancaster University, Great Britain) and Adrienne Regamey (Swiss Federal Institute of Technology) to characterize the light dependence of the emissions of monoterpenes and methyl butenol from Quercus ilex (Mediterranean oak) and other species of oak, Sitka spruce, Magnolia grandiflora, eucalyptus,and ponderosa pine. In conjunction with these experiments, Greenberg, Gregory Deem and Raymond Fall (University of Colorado) extracted essential oils from needles and leaves, and stored monoterpenes were measured by GC-FID. Several species showed terpene emissions which were light dependent. Ponderosa pine proved to be a significant emitter of methyl butenol; methyl butenol emissions were also light dependent.

CFC Calibration Exercises

The GC-AED was applied to the quantitation of CFC concentrations (F12, F22, F11, F113,methyl chloroform, carbon tetrachloride, methyl bromide, bromoform, methyl iodide, and several others) in several analytical standards (with Elliot Atlas, ACD). Laboratories measuring these CFCs often use different analytical standards, which sometimes do not agree. Consequently, an independent technique is useful to evaluate these standards. GC-AED quantitation was made with respect to a single analytical standard of n-butane (NIST special mixture), utilizing the elemental carbon response of the atomic emission detector. GC-AED proved to be an accurate and completely independent technique for the calibration of standards containing these CFCs.

Methyl Bromide Soil Deposition Experiment

Increased methyl bromide emissions have been suggested as a potential threat to stratospheric ozone. Natural and anthropogenic sources have been reported but few measurements of deposition losses have been made. Measurements were made with Dominique Serca (Paul Sabatier University, Toulouse, France) of the uptake of methyl bromideby a variety of soil types, including peat bogs, agricultural land, and local soils. An enclosure technique which included the injection of inert tracer (F113) to quantify enclosure leak rates was used. Samples were analyzed by GC-MS with standards calibrated by GC-AED analysis. Initial results indicate that the deposition velocities of methyl bromide for the soils tested are similar to those reported for other soil classifications (approximately 0.1 cm sec^{-1}).

NOMHICE

The non-methane hydrocarbon intercomparison experiment (NOMHICE, an activity of the International Global Atmospheric Chemistry program (IGAC)) was designed to evaluate current analytical methods used to determine the ambient levels of various atmospheric, non-methane hydrocarbons (NMHCs), to identify and correct existing problems in these analyses, and to help ensure quality control. During the past year, the data analysis for Task 3 (identification and quantitation of components of a gravimetrically prepared standard of 62 non-methane hydrocarbons, C2 to C11) was

completed. Major portions of the experiment and data analysis for Task 4 (identification and quantitation of components of a whole air sample collected near NCAR) were also completed. There are currently 30 laboratories participating throughout the world.

Measurements, Technology and Standards Group (MTS) of the Southern Oxidants Study (SOS)

The Measurements, Technology, and Standards group is one of the four major SOS scientific groups. MTS (Eric Apel, ACD, leader) has three major sub-programs: MTS-Hydrocarbons (Apel), MTS-Carbonyls (Yin-Nan Lee, Brookhaven National Laboratory), and MTS-NO_y (Eric Williams, NOAA). Some accomplishments during FY95 included: developed strategy for VOC measurements for the 1995 SOS Nashville intensive; coordinated and carried out quality assurance activities for VOC measurements including intercomparisons before and after intensive, data monitoring and analysis; continued development and improvement of techniques of SOS scientists; supplied evaluation standards and test mixtures to participants; modified commercially available Entech hydrocarbon preconcentator system for use by Nashville SOS participants; developed techniques to extended analytical capabilities through C14; coordinated SOS Nashville level 2 sites and aircraft inorganic analysis intercalibration activities for SO_2, NO_y, and O_3;

- Formaldehyde Intercomparison-Phase 1

In June 1995, Gilpin and Apel conducted an intercomparison of formaldehyde measurement techniques in Boulder (Yin-Nan Lee, Brookhaven National Laboratory; Thaddeus Kleindienst, Mantech Environmental Laboratory, North Carolina; Halvore Westberg, Washington State University; Brian Heikes, University of Rhode Island; Purnendu Dasgupta, Texas Technical University) using the Tunable Diode Laser Absorption Spectroscopy (TDLAS), Alan Fried (ACD), as the reference method. The experiment employed a well-defined series of reference standards and potential interferences added to a zero air matrix in a glass manifold. Accurate measurements of formaldehyde are needed in order to both constrain and validate tropospheric photochemical models. Data processing is in progress.

- Oxygenated Hydrocarbon Intercomparison-Phase 2

A field comparison was conducted during the July 1995 Nashville intensive of techniques for measurement of formaldehyde and other volatile oxygenated organic compounds (Kochy Fung, Atmospheric Assessment Association, Calabasas, California; Thaddeus Kleindienst, Mantech Environmental Laboratory, North Carolina; William Lonneman, US EPA; Rodney Zika, University of Miami; Paul Shepson, Purdue University). The study included comparisons of standards and ambient air samples.

IV. Biosphere Atmosphere Interactions (BAI)

The BAI project conducts research in collaboration with the Trace Gas Biogeochemistry project to characterize ecosystem structure and function with respect to trace gas emissions. Their focus is on the characterization and modeling of biological and environmental controls over trace gas fluxes.

Non-Methane Hydrocarbon Emission Modeling

Non-methane hydrocarbons (NMHCs) play a major role in the chemistry of the atmosphere. Significant improvements in NMHC emission modeling techniques have been made and are being incorporated into regional and global chemistry and transport models (CTMs). This effort is led by Alex Guenther and Peter Harley and includes collaboration with David Erickson (ACD), Nicholas Hewitt (University of Lancaster), Christopher Geron and Thomas Pierce (USEPA). A global model was developed and released as part of the International Global Atmospheric Chemistry (IGAC) program and is currently being used in several global CTMs. A regional model was developed for use by the USEPA in regulatory and research applications. This model predicts regional isoprene emission rates that are about a factor of five higher than was previously assumed.

Biogenic Hydrocarbon Emissions from Southeastern U.S. Forests (Southern Oxidants Study, Oak Ridge, Tennessee)

The 1995 Southern Oxidant Study (SOS) Nashville/Middle Tennessee ozone study investigated ozone production, transport and effects in order to provide the tools to develop efficient and cost-effective ozone management strategies. Biogenic hydrocarbon emissions from individual leaves, whole forest canopies, and entire landscapes were measured at the SOS field site near Oak Ridge by Pat Zimmerman, Guenther, James Greenberg, Harley, Detlev Helmig, Lee Klinger, Brad Baker, Bill Baugh, Dan Hereid, and Dominique Serca, in collaboration with Dennis Baldocchi (NOAA), and Hal Westberg, Brian Lamb, Brad Hall, and Candis Claiborne (Washington State University). Isoprene emission measurements were made on leaves of a number of trees at various canopy depths. An automated relaxed eddy accumulation system with in-situ GC-PID analysis provided continuous measurements of 30 min average isoprene fluxes above the forest canopy. A tethered balloon system flown up to 2 km carried up to six air sampling packages and tethered atmospheric observation systems (TAOS). These systems were used to characterize winds, temperature, water vapor, and hydrocarbon concentrations and estimate regional isoprene fluxes using mixed-layer gradient and mass balance techniques. Regional biogenic emission models are being evaluated and improved with the field measurement database. Methods were developed to use satellite measurements to identify oak trees which are the major isoprene emission source in this region. The land cover database and improved biogenic emission model will be used by SOS collaborators to model and interpret the Nashville study results.

Isoprene Emissions from Trees

A permanent field site has been established in a poplar plantation near Boardman, Oregon, in collaboration with Westberg and Lamb (Washington State University) and Jud Isebrandes (U.S. Forest Service). The site includes a large flat expanse of genetically identical trees providing an exceptional site for evaluating isoprene flux measurement systems. Above canopy isoprene fluxes were measured by eddy correlation using a fast response chemiluminescense isoprene analyzer by Guenther, Alan Hills, and Serca. These measurements are the first attempt to estimate isoprene fluxes with a direct eddy correlation technique. The site also includes genetic trial plots with a wide range of planted clones of known genetic history. Intra-species isoprene emission rate variations were investigated by Harley and Luanne Otter (University of Witwatersrand) by measuring emissions of a select group of clones under controlled environmental conditions.

Large uncertainties have been associated with isoprene emissions from landscapes in California. Isoprene emission rate measurements reported for Californian oak species were an order of magnitude lower than emission rates reported for

eastern U.S. oak species. Isoprene emission rates of the six major California oak species were measured by Guenther and Harley at three field sites in California. The results demonstrate that California oaks emit isoprene at rates comparable to eastern U.S. oak species. This should lead to a substantial increase in the isoprene emission rates used for regional photochemistry modeling in California.

Phytotron Controlled Environment Studies

Experiments were conducted in the NCAR Phytotron (greenhouse/ environmental control chamber) to survey biogenic emissions from a wide range of temperate and tropical plants and examine environmental controls over emissions. Studies included characterizing light and temperature dependent monoterpene emissions from *Quercus ilex* and methyl-butenol emissions from *Pinus ponderosa*. The results show that methyl-butenol and monoterpenes are emitted from some plant species at very high rates and have light and temperature responses that are similar to isoprene. This effort is led by Harley in collaboration with Greenberg, Guenther, Hereid, Hewitt, and Adrienne Regamey (Swiss Federal Institute of Technology). The results are being used to develop numerical algorithms that can be incorporated into regional and global models.

EXPRESSO (EXPeriment for REgional Sources and Sinks of Oxidants)

Two EXPRESSO field campaigns were conducted in FY95. The first campaign in Jan- Feb 1995 focused on the savanna and secondary forest ecosystems in the Central African Republic (CAR). This trip was conducted by Lee Klinger, Elizabeth Sulzman, Robert Kremer (Colorado State University), and Andrew Hudak (University of Colorado) Michel M'Bangui and Jean-Bruno Vickos (Central African Institute of Agronomical Research, Central African Republic), and Jean-Marie Moutsambote (University of Brazzaville, Republic of the Congo). Major vegetation types in CAR were determined and intensive study sites were selected. Ecological field measurements included species lists, aerial photography and videography, leaf area index, and radiation-reflectance measurements. Comparison of field versus satellite data were made for several regions in the CAR.

The second trip in May-June 1995 focused on more detailed landscape characterization and chemical screening of vegetation emissions for the four distinct vegetation regions defined by remote sensing in the EXPRESSO study domain. This trip was carried out by Klinger, Greenberg, Susan Canney (Green College Oxford, Great Britain), and Lee Vierling (University of Colorado). Over 150 species of vegetation were screened for isoprene and terpene emissions. Together these species probably account for over 50% of the leaf biomass of vegetation in the study region. Volatile Organic Carbon emissions were collected and analyzed for about forty of the vegetation species and for ambient air samples. Spectrometer, leaf area index, photosynthetically active radiation, and global positioning system measurements were taken to relate surface spectral parameters to remote sensing data.

Successional Theory

Important contributions were made to studies of successional and Gaia theory. A paper in press by Klinger, John Taylor (Australian National University), and Lars Franzen (Göteborg University) uses modeling and observed data to show that peatlands likely play an important role in ice age initiation and Gaian regulation. Two other papers in press by Klinger and Susan Short (University of Colorado) provide strong evidence from field studies in Canada that landscape successions converge onto climax bog communities. These findings support the notion, arising from complexity theory, that self-regulation at the landscape and global scales is an emergent property resulting from the tight coupling of biotic and environmental components of the landscape.

V. Atmospheric Odd Nitrogen (AON)

The group's interest is in the measurement of trace constituents which are influential players in determining the photochemical production and destruction of ozone in the troposphere and stratosphere and the oxidizing capacity of the troposphere. AON focuses on (a) the development of instrumentation for measurements in, but not limited to, the remote atmosphere, (b) the development of aircraft and ground-based measurement programs to test our understanding of chemistry and transport processes, and (c) through collaborations, the analysis and interpretation of observations by modeling exercises.

Instrument Development

A priciple effort of the group has been to modify $NO_{x,y}$, and O_3 instrumentation for the upcoming Vortex Ozone Tranport (VOTE) and Tropical Ozone Transport (TOTE) Experiments on the NASA DC-8 aircraft (Principle investigators O. B. Toon, NASA Ames and Mark Schoberl, NASA Langley). Both experiments are designed to investigate exchange processes in the upper troposphere and lower stratosphere (UT/LS) between mid-latitudes and the tropical or polar regions in the fall and winter period. They will also provide an opportunity for us to extend the "climatology" of odd nitrogen species in the upper troposphere and lower stratosphere. Studies in this region, where net ozone production can be highly non-linear, are also of current importance in relation to programs designed to assess the effects of aircraft emissions on atmospheric ozone. Specially designed inlets have been constructed for two NO_y channels to investigate the abundance of odd nitrogen in solid and liquid aerosols in the atmosphere or in cirrus clouds. Improved ozone generators have also been assembled for the instrument. This activity has been led by Andrew Weinheimer, Frank Grahek, James Walega, Frank Eparvier, and Denise Montzka.

Another effort by Grahek, Walega, Steve Shertz and Brian Ridley has been to start the design and fabrication of a five channel chemiluminescence instrument to operate autonomously on the recently acquired WB-57 aircraft. At least three channels of this instrument need to be operational for a study of convective transport of trace species and the production of odd nitrogen by thunderstorms in the Colorado region. One weakness of large scale models used to assess global ozone trends (due to anthropogenic precursors or aircraft emissions) is that the global lightning source of odd nitrogen and its vertical distribution are not well-known. The thunderstorm study is scheduled as part of the Stratosphere-Troposphere Experiments: Radiation, Aerosols, and Ozone (STERAO) program for late spring and early summer of 1996.

Gregory Dubois-Felsman and Frank Flocke have also progressed well on a gas chromatographic technique for eventual measurements of peroxyacetyl nitrate (PAN) on the WB-57 aircraft. Although the group has measured this species on previous aircraft missions, recent advances will allow a much more refined instrument. PAN is one of the major reservoirs of odd nitrogen species in the remote middle atmosphere but whether it plays a major role in the upper troposphere or perhaps lower stratosphere is not well known.

Analysis and Modeling

The group has also progressed on the analysis and manuscript preparation from previous field measurements, specifically the MLOPEX II ground and aircraft experiment, the New Mexico thunderstorm study, and AASE-II data. Analysis of the NO_2/NO steady state and implied radical concentration for the four seasons has been completed in collaboration with the modeling section (Sasha Madronich, Didier Hauglustaine, and Guy Brasseur) and compared with actual measurements of OH and peroxy radicals at the Hawaii site. In general, the agreement with the OH measrements is satisfactory, but steady-state derived peroxy radical concentrations are higher than directly measured. The aircraft component of MLOPEX II has shown very convincingly that the atmosphere in spring over the Hawaii region is stronly impacted by transport of constituents from higher latitudes. Vertical soundings with the aircraft demonstrate that a highly structured atmosphere is quite common. A second manuscript nearing completion on the New Mexico thunderstorm experiment summarizes estimates of the global strength of the lightning source of NO . This work has allowed a very nice comparison with the

sensitivity analysis to the lightning source strength completed using the IMAGES three-dimensional model (Brasseur, Jean-François Lamarque, and Jean-François Müller (Belgian Institute for Space Aeronomy)) and has been completed with James Dye (MMM) and William Rison and David Raymond (New Mexico Institute of Technology).

Anna Zheng (visitor, NOAA) in collaboration with Shaw Liu (NOAA Aeronomy Laboratory) has completed further analysis of small scale "spikes" in the AASE-II NO, NO_y, O_3, CO, CO_2, and CH_4 data base and has been able to identify very probable sources from aircraft, from ground emissions, and perhaps from lightning. However, the data does not identify the aircraft corridor effect over the North Atlantic that is so often discussed in reports and literature.

A long standing issue in the broad-band photolysis technique used for aircraft measurements of NO_2 has been the possibility of interferences from $ClONO_2$, N_2O_5, and especially HO_2NO_2 due to heterogeneous decomposition in the sampling lines or photolysis cell. All of these species are difficult to work with in the laboratory, however, Weinheimer, John Orlando and Geoffrey Tyndall have been able to run laboratory tests. Of the three, HO_2NO_2 is an important interferent since the tests suggest 20-30% decomposition in a typical sampling system. Since the HO_2NO_2 abundance predicted by models is comparable to NO_2 in the upper troposphere, changes in current instrumentation will be required for NO_2 measurements. This is a critical result, since the broad-band technique is utilized by a number of aircraft research groups.

Community Service

Several of the group were actively involved in the organization, development, and proposal preparation of the STERAO program. Ridley and Shaw Liu (NOAA Aeronomy Laboratory) as co-chairs of the program were responsible for organizing a workshop of approximately 80 scientists from universities, NASA, NOAA, NSF and NCAR to define a program of upper tropopheric and lower stratospheric research that require the WB-57 aircraft. That workshop has led to the development of three research programs to be conducted over the next 3-5 years. The first centers on a study of thunderstorms over Colorado and a formal proposal (Steven Rutledge, Colorado State University; Dye, MMM; and Adrian Tuck, NOAA Aeronomy Lab) was submitted to the Observing Facilities Deployment Pool this summer. The second defined project, co-chaired by Daniel Murphy (NOAA Aeronomy Laboratory) and Andrew Heymsfield (MMM), will research aerosol formation and composition and the microphysics and radiation in deep convective systems, such as those in the ITCZ. The third project, co-chaired by William Brune (Pennsylvania State University), and Fred Eisele (NCAR and Georgia Institute of Technology), will focus on the photochemistry and heterogeneous processes that might occur in the UT/LS region.

The group also supplied ozone instrument drawings to David Parish (NOAA) for equipping their P-3 aircraft. Ridley also continues to serve as the American editor for the *Journal of Atmospheric Chemistry*.

A. HO_x Project

C-130 Aircraft OH/H_2SO_4 Instrument

The hydroxyl radical plays a central role in atmospheric photochemistry and is also probably the single most important cleansing agent in the troposphere. It is responsible for the initial oxidation and subsequent removal of a wide variety of otherwise relatively stable atmospheric compounds. Despite its importance, OH has proved to be quite elusive because of its low concentration.

During the past year, an aircraft version of our ion-assisted OH measurement apparatus has been constructed and tested under simulated flight conditions. In addition to hardware modification required for aircraft installation, several unique components also had to be designed, constructed and tested before successful aircraft OH measurements could be made. The first of these was a virtual iris which could vary the amount of gas and ions entering the mass spectrometer vacuum system while allowing essentially no contact between ions and aperture surfaces. The second component was an inlet for OH and H_2SO_4 which straightened and slowed the airflow so that it could be introduced into the instrument's ion source region without becoming turbulent or contacting any surfaces. Finally, a temperature controlled, nitrogen flushed, automated OH calibration source had to be constructed inside of the inlet housing.

Several tests were conducted both in the NCAR and the U.S. Air Force Academy wind tunnels to verify that the proper flow characteristics were being achieved in the new inlet. Temperature, pressure and H_2O concentrations typical of anticipated flight regimes were then reproduced in a large and fast pumped flow tube coupled to the inner portion of the sample inlet. The new inlet appeared to function well throughout the laboratory and wind tunnel tests. This instrument is presently measuring OH, H_2SO_4 and MSA as part of the ACE-1 (Aerosol Characterization Experiment) on the NCAR C-130.

HO_2, HO_2/RO_2 Instrument

Peroxy radicals also play key roles in tropospheric chemistry involving photochemical ozone formation and production of peroxides. The NCAR chemical amplifier has been used in a number of recent field studies and participated in an intercomparison exercise last year. Since the instrument presently only retrieves the sum of HO_2 and the organic peroxy radicals, work has begun on a new instrument that utilizes the ion assisted mass spectrometric measurement of OH with chemical conversion using NO to measure and speciate HO_2 and the organic peroxy radicals. Based on preliminary laboratory and field experiments this year, it is anticipated that this system will be much smaller, yet possess a detection limit of a fraction of a pptv with superior stability compared to the present chemical amplifier.

Selected Ion Chemical Ionization Mass Spectrometric Measurement of Nitric Acid

Several new chemical ionization schemes were investigated for the measurement of atmospheric nitric acid vapor. The first few of these involved the use of compounds that would transfer F- ions to HNO_3. One of these schemes appeared practical but only at low H2O concentrations such as those found in the upper troposphere and stratosphere. Alternatively, this scheme could be used in a reduced pressure ion source, but this introduced the problem of transferring the HNO_3 from atmospheric pressure to a low pressure reaction vessel while minimizing wall contact. It was thus, decided to use HSO_4^- ions as the initial reactant species, and to use an ion source similar to that used to measure H_2SO_4. The typical H_2SO_4 ion reaction region, however, was replaced by a crude drift tube region in order to shorten and vary the ion reaction time. The latter reaction scheme appears to work quite well and is nearly unaffected by atmospheric H_2O concentrations. The instrument has a response time of a second or less and should be able to detect about 10 ppt of HNO_3 with about a 1 second integration time.

Atmospheric Photolysis Rate Coefficient Measurements using Actinic Flux Spectroradiometry

Photochemical reactions provide the driving force for much of the chemistry in the atmosphere. The in situ rates of these photolysis reactions are important in understanding production and loss terms for key atmospheric species like OH and ozone. We have been developing an spectroradiometric instrument to measure the atmospheric actinic flux spectra to use for calculations of atmospheric photolysis rate coefficients.

Analysis and Publication of Results

In the past year, data has been analyzed and prepared for publications from three field campaigns in which the AON group was involved: MLOPEX II, Tropospheric OH Photochemistry Experiment and the Peroxy Radical Intercomparison Exercise. Papers were authored or co-authored by members of the HO_x group as a result of these three campaigns.

B. Laboratory Kinetics

Bromine Chemistry

Bromine has been implicated in the mechanism of ozone destruction in the lower stratosphere. The Laboratory Kinetics group (Geoffrey Tyndall and John Orlando) has been involved with projects which address the chemistry of the bromine-containing source gases; and the reactions of the bromine radicals after their release. In collaboration with Timothy Wallington (Ford Motor Company), laboratory studies investigated the kinetics and mechanism of methyl bromide (CH_3Br) and methylene bromide (CH_2Br_2) degradation, two of the major sources of bromine in the atmosphere. Rate constants were measured for the reactions of chlorine atoms with both species over a range of temperatures, and for the reaction of OH radicals with methylene bromide at room temperature. The studies resolved considerable discrepancies in the literature regarding the rates of these reactions. In addition, the two groups carried out a joint study of the oxidation mechanisms of both molecules. The current work shows that the CH_2BrO radical produced from methyl bromide decomposes to HCHO + Br, even at 230 K, whereas three previous studies had all inferred that the radical reacts with oxygen under atmospheric conditions.

One of the major temporary reservoir species for bromine in the stratosphere is bromine nitrate (BrONO2). However, most of our information on this molecule is inferred from its chlorine analog ClONO2. A study was carried out to measure the rate of the thermal decay of bromine nitrate between 320 and 340K. These experiments provided a measurement of the BrO-NO2 bond strength, and thus the heat of formation of bromine nitrate. Surprisingly, bromine nitrate was found to be more stable than chlorine nitrate. The new data allow more accurate estimates of the reactivity of bromine nitrate to be made.

Chlorine Chemistry

Chlorine atoms are important not only in regard to ozone destruction but also potentially in the marine boundary layer where they can contribute to hydrocarbon oxidation. Studies of the reactions of chlorine atoms with several organic molecules were conducted using the complementary techniques of flash photolysis with resonance fluorescence, and the relative rate method with FTIR or gas chromatographic detection (with Wallington and William Kaiser, Ford Motor Company). The experiments provided rate constants for the reactions of Cl atoms with ethane, butane, propane, methanol and acetaldehyde, and highlighted discrepancies in the literature data on these reactions. An improved formulation was proposed which allows the prediction of rate constants for reactions of chlorine atoms with alkanes.

The rate constants and mechanisms for the reactions of Cl and F atoms with acetonitrile were measured in collaboration with Wallington and Jens Sehested and Ole-John Nielsen (Risø National Laboratory, Denmark). The rate constant for the chlorine atom reaction was measured as a function of temperature and found to be substantially larger than the accepted literature value. The photolysis of chlorine nitrate was found to produce chlorine atoms in their excited spin state ($^2P_{1/2}$). Rate constants have now been measured for the quenching of these excited chlorine atoms by a number of atmospheric gases (with Carla Kegley-Owen, University of Colorado). It has been suggested that such excited atoms may react more rapidly with methane than ground state atoms, and that the atoms produced in the photolysis of chlorine nitrate could thus impact the lifetime of methane in the stratosphere. However, it was shown that methane simply quenches the excited atoms, and that the residual concentration in the atmosphere is too low to affect any chemistry.

Technique Development

A new flash photolysis system was built (with Alan Fried) which uses infrared diode laser detection to monitor the kinetics of reaction products. The method offers the advantages of high sensitivity and selectivity of the species detected in a time-resolved experiment. Kegley-Owen constructed the apparatus and tested it by studying the production of HCl in the reaction of Cl atoms with C_2H_6 and CH_3CHO.

Experiments are in progress to characterize flowing sources of chlorine nitrate, nitrogen pentoxide and peroxynitric acid to test for interferences in an NO_2 field instrument (Orlando and Andy Weinheimer). The instrument converts NO_2 to NO by ultraviolet photolysis, and could also respond to the three reservoir species given above. However, the results suggest that such interferences should be minimal.

VI. Upper Atmosphere Research Satellite (UARS)

The Upper Atmosphere Research Satellite hosts ten instruments that measure the global distributions of a dozen chemical species in the stratosphere and mesosphere, in addition to temperature, winds and solar inputs. This NASA project is currently distributing the UARS data to the research community. Work in ACD focused upon validating the UARS data and developing scientific applications of the data.

UARS Data Validation

A special issue of the *Journal of Geophysical Research* (*JGR*) will contain validation papers which discuss the accuracy and precision of the UARS data. Meaningful comparisons of theory and observation are possible only when one understands the strengths and weaknesses of data sets. John Gille, Steven Massie, and William Mankin are co-editors for this special *JGR* issue.

Each validation paper compares UARS observations to correlative variable measurements (e.g., ozone mixing ratios, as observed by different instruments at locations and times which coincide with UARS observations). Correlative data came from other UARS experiments, other satellite experiments, balloon-borne instrumentation, conventional meteorological soundings (for temperature-pressure profiles), and ground based facilities (e.g. lidar stations, mesospheric radars). The correlative measurements were obtained by research groups at numerous university, NASA, and foreign facilities. Cheryl Craig and Charles Cavanaugh processed the correlative data from the numerous research groups.

Gille, Paul Bailey, Massie, Lawrence Lyjak, David Edwards, Craig, and Cavanaugh co-authored the validation papers on temperature, ozone, and aerosol data from the CLAES (Cryogenic Limb Array Etalon) experiment on UARS. Numerous papers were co-authored with the CLAES team at the Lockheed Palo Alto Research Laboratory and with the ISAMS team at Oxford, England. Gloria Manney (Jet Propulsion Laboratory) and Massie contributed a paper to the special issue which compares National Meteorological Center and the United Kingdom Meteorological Office temperature analyses in the polar regions, and compared these analyses with radiosonde data.

UARS Research

William Randel and Massie authored papers which appeared in the October 1994 issue of the *Journal of the Atmospheric Sciences* entitled "The Upper Atmosphere Research Satellite: Early Scientific Results." Randel, in collaboration with Byron Boville (CGD), studied the global variability of N_2O using the NCAR Community Climate Model and the CLAES N_2O observations from UARS. The study quantified the importance of planetary wave constituent transport in the stratosphere.

Massie used CLAES aerosol extinction data to develop and apply a multi-wavelength method which can be used to study polar stratospheric clouds (PSCs). PSCs alter the chemistry of the stratosphere, and lead to the development of the Antarctic ozone hole phenomena. [To see examples of UARS observations of PSCs, and the cold stratospheric temperatures which lead to their formation, click here and here. These figures show PSC extinction and cold temperatures over Scandinavia at 20 km altitude on January 10, 1992.] In related work, Massie used balloon-borne aerosol size distribution data (in collaboration with Terry Deshler of the University of Wyoming) and ER-2 data (in collaboration with James Dye and Darrel Baumgardner, CGD and ATD) to develop techniques by which the UARS extinction data can be transformed into sulfate aerosol and PSC area and volume densities. These densities are important quantities for modeling studies which simulate chemistry upon particle surfaces.

Gille and Lyjak studied the diurnal variation of $ClONO_2$ using CLAES data. Its rate of increase at sunset was compared with theoretical calculations which used CLAES NO_2 and MLS (Microwave Limb Sounder) ClO, and was shown to be in good agreement under most conditions. Comparisons for a complete diurnal cycle between CLAES $ClONO_2$ and three-dimensional chemical model calculations of Guy Brasseur were performed as a function of local time and showed generally

good agreement. Using a coordinate system based on the structure of the stratospheric polar vortex, Gille and Lyjak used isopleth motions of CLAES CH_4 to study the formation, evolution, and disintegration of the vortex in both the Northern and Southern Hemispheres. [To view an example of the distribution of CH4 for June 29, 1992, as observed by the CLAES instrument, click here.]

Anne Smith investigated the longitudinal variability of the horizontal winds in the winter mesosphere measured by the HRDI (High Resolution Doppler Imager) instrument. Wind variations in the upper mesosphere (75-85 km) show a consistent negative correlation with those in the stratosphere. The structure and variations of the winds provide strong evidence that the mesospheric longitudinal variations are caused by gravity waves that have been filtered by the planetary waves in the stratosphere.

David Edwards performed a study of non-local thermodynamic equilibrium (non-LTE) limb radiance near 10 microns as measured by the CLAES experiment. The analysis shows good agreement between measured and calculated radiances and confirms previous theoretical modeling of the non-LTE populations of molecular states. The diurnal variation of the measured emission points to solar dependent mechanisms selectively populating molecular vibrational levels in CO_2 and O_3. The model developed will be of use in correcting past and future broad-band radiometer (LIMS, HIRDLS) O_3 data above about 40 km. This work was carried out in collaboration with Jack Kumer (CLAES instrument team) and Manuel Lopez-Puertas (Instituto de Astrofisica, Granada, Spain). Edwards collaborated with Kenneth Minschwaner (New Mexico Institute for Mining and Technology) in using CLAES global measurements of N_2O, CH_4, and CFC12 to investigate the role of the stratosphere in the radiative forcing of climate.

UARS data have been a great benefit for testing and verifying numerical models. Brasseur and XueXi Tie compared chemical fields measured by the MLS and CLAES experiments with simulations from a complex dynamical chemical model of the stratosphere. The model simulations of ozone and other species in the Antarctic stratosphere show many similarities with the UARS observations. [To look at model and UARS observations of ozone, ClO, and ClONO2 click here and here.] The model was also used to simulate ozone chemistry under pre-industrial conditions to give a quantitative description of the differences between the unperturbed and present-day stratosphere.

Randel used MLS ozone and HALOE (Halogen Occultation Experiment) ozone, nitrogen dioxide, methane and water vapor data in a study of interannual variability in the stratosphere during the first four years of the UARS mission. Randel's results quantify the changes of ozone and nitrogen dioxide following the eruption of Mt. Pinatubo in June 1991, and present the first observations of quasi-biennial oscillation variations in stratospheric methane and water vapor. Randel collaborated with James Russell III (NASA Langley) and Lee Elson (Jet Propulsion Laboratory) in this work.

Analysis and Interpretation of Global Ozone Data

There is considerable interest in determining time trends of ozone, temperature, and other atmospheric variables, i.e. to determine if these variables are increasing or decreasing with time. Randel contributed a chapter to volume 28 of the book series "Methods of Experimental Physics." Randel's chapter discusses mathematical techniques that filter and preprocess data for time series analysis.

Randel and Fei Wu applied trend analysis to study ozone depletion as measured by the TOMS (Total Ozone Mapping Spectrometer) and SBUV and SBUV/2 (Solar Backscatter Ultraviolet) instruments. The ozone depletions were studied in a coordinate system based on the structure of the polar winter vortex (i.e. in terms of the dynamical potential vorticity field). TOMS trends clearly show that ozone losses occur outside of the polar vortices in both the Northern and Southern Hemispheres. [To see a TOMS map of column ozone (in Dobson units) on March 15, 1986, click here. Low column ozone amounts are observed within the northern polar vortex, represented by the black lines of potential vorticity.]

Long-term variability of stratospheric ozone was investigated by Randel and Wu using data from the SBUV instrument on the Nimbus-7 satellite and with SBUV/2 data from the NOAA-11 satellite. These data were combined into a continuous daily global data set covering sixteen years (1978-1994). Both ozone profile (above 25 km) and column ozone amounts are available to the research community by electronic transfer. This data set can be used for analysis of natural and anthropogenic ozone variability, and for various stratospheric modeling activities. A comprehensive NCAR Technical Note has been produced documenting this data set and climatological ozone statistics derived from it.

Global satellite observations were used by Randel to document the changes in stratospheric ozone and temperature resulting from the eruption of Mt. Pinatubo in June 1991. Ozone observations showed decreases in column ozone of 5-10% during 1992-93, mainly in winter middle-high latitudes (particularly large throughout 1993 in the NH). The observed variations have a characteristic space-time structure that provides a fingerprint for simulations of Pinatubo- associated losses.

Dynamic variability of the mesosphere was analyzed using Pressure Modulated Radiometer satellite data covering three years, in a collaborative study between Randel and Oxford University. These temperature observations were used to derive global wind variations over the altitude range 35-85 km; this is the first data set used to analyze both interannual and day-to-day variability in the mesosphere.

A collaborative study between Randel and the NOAA Geophysical Fluid Dynamics Laboratory demonstrated that the observed cooling of the lower stratosphere during the past two decades (observed by satellite and radiosonde data) is attributable, at least in part, to the contemporaneous depletion of ozone in the lower stratosphere. The researchers found that the space-time signature of modeled temperature changes matched those observed strongly suggesting that ozone depletion is the principal cause of the temperature change.

A. Measurement of Pollution in the Troposphere (MOPITT)

The MOPITT instrument scheduled for launch aboard the Earth Observing System AM-1 satellite in mid 1998 is among the first significant applications of space based remote sensing to global tropospheric chemistry research. The goal of the experiment is to support studies of the oxidizing capacity of the lower atmosphere on large scales by measuring the global distributions of carbon monoxide and methane. Work at NCAR on the Canadian-built MOPITT is proceeding in close collaboration with the principal investigator, James Drummond (University of Toronto). Additional collaborators on the Science Team include Brasseur, Gary Davis (University of Saskatchewan), John McConnel (York University), Guy Peskett (Oxford University), Henry Reichle (North Carolina State University), and Nigel Roulet (McGill University).

The algorithm and software group at NCAR which includes Bailey, Pan, Edwards, Rokke, Wang, Spaur, Craig, Cavanaugh and Packman is scheduled to make the first delivery of MOPITT science data product software to the NASA data center in early 1996. Edwards has developed a fast radiative transfer model describing MOPITT for use with the production retrieval algorithm developed by Pan and with the cloud detection and correction algorithm being developed by Rokke. Spaur, Craig, Cavanaugh and Packman have begun to assemble these algorithms into deliverable software systems. Wang and Smith have produced updated versions of the Algorithm Theoretical Basis Documents which describe the scientific basis for the instrument and algorithmic approaches.

In parallel with these activities, ACD team members Gille, Smith and Shertz are constructing the MOPITT Algorithm Test Radiometer (MATR), an aircraft instrument which utilizes the same correlation cell technology as the space based instrument. MATR will be used to verify prior to launch the radiative transfer models and data retrieval algorithms which are the basis of the operational data product software. The first aircraft flight of MATR is scheduled to take place in mid 1996.

B. High Resolution Dynamics Limb Sounder (HIRDLS)

HIRDLS is a 21 channel infrared limb-scanning radiometer being jointly developed by NCAR and Oxford University for flight on the Earth Observing System Chemistry Platform. The Principal Investigators are John Gille and John Barnett (Oxford University). The goals of HIRDLS are to measure the outgoing limb radiance in the infrared, from which the temperature and concentrations of 10 trace gases can be derived. The resulting data will have horizontal and vertical resolutions better than previous observations, extend from the mesopause down into the upper troposphere and provide pole to pole coverage.

The Lockheed Martin Palo Alto Research Laboratories were selected in a competetive procurement as the Instrument Integrator. The same group had previously been selected to build the Telescope Subsystem, and having them fill both roles will greatly simplify the technical and managerial interfaces. Joanne Loh (program manager) led the international HIRDLS team in an Implementation Review, where the scientific requirements and plans for the experiment development were presented. Subsequently extensive work was done on both sides of the Atlantic to develop conceptual designs for all 9 subsystems, as well as initial plans for instrument integration, testing, calibration, and scientific activities. Gille, Loh, Michael Dials (technical manager), Douglas Woodard, Philip Arter, Russell Howard with their British colleagues of the HIRDLS team presented this work to NASA at a System Concept Review, where it was well received. In the same time frame a proposal for the Execution and Post Launch phases was submitted to NASA.

HIRDLS co-investigators include Bailey, Byron Boville (CGD), Brasseur, Michael Coffey, and William Mankin (all of NCAR), James Holton and Conway Leovy (University of Washington), David Andrews, Clive Rodgers, Fredrick Taylor, Robert Wells, John Whitney, and E. James Williamson (Oxford University), Roger Hunneman and Alan O'Neil (Reading University), Michael McIntyre and John Pyle (Cambridge University), H.G. Muller (University of Sheffield), C.T. Mutlow (Rutherford Appleton Laboratory), and Geraint Vaugham (University College of Wales).

VII. Stratospheric/Tropospheric Measurements (S/TM)

The S/TM project investigates the sources, budgets, distribution and variations of trace gases in the stratosphere and troposphere, with an emphasis on those species related to ozone production and destruction. An integral part of the program is to evaluate and develop state-of-the-art sampling and analytical facilities for trace gas measurement from different environments.

Stratospheric Tracers of Atmospheric Transport (STRAT)

The STRAT program is a NASA-sponsored mission to measure the morphology of long-lived tracers and dynamical quantities as functions of altitude, latitude, and season to help determine rates of global-scale transport and distributions of trace gases. A secondary goal is to further characterize atmospheric photochemistry of the lower stratosphere and upper troposphere. Mission scientists for the STRAT project are Steven Wofsy (Harvard University) and Paul Newman (NASA Goddard Space Flight Center).

The Whole Air Sampler (WAS) is being used in STRAT to measure tracers with atmospheric lifetimes from weeks to months, to provide data on some of the less abundant hydrochlorofluorocarbons (CFC "replacement" compounds), and to characterize and quantify organic bromine and organic iodine speciation in the lower stratosphere. Some of the target trace gases for this study include methane, ethane, ethyne, propane, chloroform, tetrachloroethylene, HCFC 22, HCFC 141b, HCFC 142b, methyl bromide, dibromomethane, and methyl iodide.

Tests of the WAS during flights in March and May assessed in detail the performance of the WAS to accurately and reproducibly determine concentrations of these trace gas species. Results from these tests indicate that present techniques can provide measurements reproducible within 2-6% over the range of conditions encountered in the lower stratosphere. In addition, application of new analytical techniques (negative ion chemical ionization mass spectrometry) have lowered the detection of a number of halogenated trace gases to <.01 pptv. This level of detection is necessary to track the distributions of some trace gases from the tropopause further into the stratosphere. Detailed vertical profiles of most trace gases measured with the WAS indicated a layered structure in the 12-20 km altitude region with evidence of equatorial intrusions into mid-latitude air masses.

Airborne Arctic Stratospheric Experiment II (AASE-II)

AASE II was a major research expedition, organized and coordinated by NASA, to investigate the chemistry and dynamics of the Arctic polar vortex in relation to stratospheric ozone depletion in the northern hemisphere winter. Whole air samples collected during the mission were analyzed for over 20 individual organic chlorine and organic bromine compounds.

A. Organic bromine in the stratosphere

Measurements of the primary compounds that contribute to organic bromine in the stratosphere from WAS samples collected during AASE II represent the most comprehensive data set available for the northern hemisphere. The vertical and latitudinal distributions of individual compounds plus correlations with other trace gases are especially useful in evaluating the dynamics and bromine chemistry in the lower stratosphere in two- and three-dimensional models (in collaboration with Susan Solomon and John Daniel (NOAA Aeronomy Lab) and Michael Prather and Linnea Avallone (University of California, Irvine)).

B. Evaluation of the age of stratospheric air and inorganic chlorine and bromine release

In collaboration with Daniel and Solomon, we estimated the average transport time from the tropical tropopause to various regions of the northern hemisphere lower stratosphere (stratosphere age) using simultaneous mixing ratio measurements of

CFC-115 and CO_2 measured by the WAS during AASE II. Below 525 K, we estimated air outside the vortex to be less than 4.5 years old, with ages generally increasing with increasing latitude and latitude. Inside the polar vortex air is between about 3 and 6 years old. We calculated inorganic chlorine and bromine as functions of latitude and potential temperature using the WAS organic chlorine and bromine measurements in combination with modeled tropospheric halocarbon trends and with the ages estimated from CFC-115 and CO_2. Inferred inorganic chlorine and inorganic bromine increase with increasing latitude and altitude even though air at higher latitudes and altitudes is generally older and entered the stratosphere with lower organic chlorine and bromine abundances. For our analyzed locations inside the polar vortex, we estimated a maximum Cl_y abundance of 2670 pptv and a maximum Br_y abundance of 14.3 pptv. The locations of the maxima correspond to an average N_2O value of about 120 ppbv.

C. Organic bromine/total bromine relationships

In a collaborative effort with Linnea Avalone and Darin Toohey (University of California, Irvine) measurements of brominated sources gases collected by the WAS during AASE II were correlated with N_2O to determine the total organic bromine along a given ER-2 flight track. The corresponding inorganic bromine (Br_y) was calculated as the difference between the total organic measurements and the tropospheric abundance at the time of entry into the stratosphere, which was determined using CFC 115 to calculate the age of the air and a 2% yr^{-1} growth rate for total organic bromine. The derived Br_y was used to determine the ratios of measured BrO to available inorganic bromine. These ratios showed only slight increases in polar regions relative to midlatitudes. A comparison between observed latitudinal and diurnal variations of these ratios and those calculated by photochemical models shows reasonable agreement in behavior, but significant discrepancies in magnitude. It is unclear whether this difference is due to errors in measurements, models, or both.

Global Distribution of Trace Gases

Halocarbon analysis was completed on atmospheric samples collected in the remote Pacific from a NOAA/RITS cruise (October/November 1993, Seattle to Punta Artenas, Chile). A collaboration with Timothy Bates and James Johnson (NOAA Pacific Marine Environment Laboratory), this data will be combined with measurements in the same region collected six months earlier to evaluate changes in the abundances and latitudinal distributions of individual halocarbons. We are especially interested in the brominated organic species, the CFC replacement compounds such as HCFC 141b, and organic nitrates. These data indicate sources of brominated organics and organic nitrates from productive marine areas into the marine boundary layer.

In conjunction with Julia Lee (ASP), David Erickson, and J.-F. Müller (Belgian Institute of Space Aeronomy), we are examining the atmospheric budget of methyl bromide using the IMAGES model and calculations from the NCAR CCM2, constrained by the measured distributions. The measurements of relatively short lived halogens (e.g. C_2Cl_4, C_2HCl_3, etc.) allow us to establish the tropospheric abundance and distribution of those species that are measured in stratospheric samples as part of the STRAT campaign to evaluate dynamics in the lower stratosphere.

Calibrations

Calibration of the numerous compounds measured in whole air samples is an ongoing process that involves standard preparations using both static and flow dilution systems. We are reevaluating old standards and a series of commercially prepared, multi-component, 100 ppbv level standards using a new, combined instrumental approach. The method being developed allows absolute calibration of the high concentration standards using an atomic emission detector (AED) which can quantify compounds independent of molecular structure. Thus carbon in heteroatomic molecules (such as halocarbons) provides a response equivalent to an equal weight of carbon in a hydrocarbon. This equivalence allows calibrations of different molecular species to be linked to well characterized hydrocarbon standards certified by the National Institute of Standards and Technology (NIST). Further comparisons using flame ionization detection are used to monitor changes in standard mixtures over time. Ultimately, the standard mixtures are referenced to working standards of air at normal ambient concentrations. This work is being done in collaboration with James Greenberg.

Whole Air Sampler Design and Construction

A new whole air sampler (WAS) is being designed and built for the recently acquired WB-57F high altitude aircraft. The new version of the WAS will be able to accommodate up to 60 samples per flight, twice the current capacity being flown on the ER-2. The new sampler will have the ability to automatically pressurize canisters with 3 atmospheres of sample air according to a pre-programmed schedule. First use of the new sampler is in summer of 1996 as part of the STERAO (Stratosphere-Troposphere Experiments: Radiation, Aerosols, and Ozone) experiments.

Additional Studies and Collaborations

Additional studies were performed during the year in collaboration with outside university and government scientists. Briefly, these include: Alex Weaver and Susan Solomon (NOAA-Aeronomy Laboratory) on studies of organic halogen variations in the atmosphere over Greenland; Frederic Lacquer (University of Nebraska at Omaha) conducting analytical studies related to measurement of atmospheric alcohols; Glen Cota (University of Tennessee) and William Sturges (University of East Anglia) on sources of organic halogens in the arctic region.

VIII. Optical Techniques (OT)

Polar Stratospheric Chemistry

Continuing their long involvement in field measurements of stratospheric composition in the polar regions, William Mankin, Michael Coffey, and James Hannigan of the Optical Techniques (OT) project participated in the Second European Stratospheric Arctic and Midlatitude Experiment (SESAME) to study the evolution of the arctic stratospheric chemistry during the winter. Collaborating with Dieter Perner (KfA Jülich) and Tørben Jørgensen and Paul Eriksen (Danish Meteorological Institute), they located their infrared spectrometer at the DMI facility near Søndre Strømfjord, Greenland, last winter and recorded over 20,000 infrared spectra of the sun. From these high resolution spectra, column amounts of trace gases including HCl, HF, O_3, and HNO_3 were derived. Efforts are now focused on correlating the variation of these gases with date in the season, potential vorticity (an indicator of depth of the observations in the polar vortex), and thermal history of the air mass as indicated by back trajectories calculated from meteorological observations. The observations are being used to compute synthesized two-dimensional fields of constituents which will help trace the evolution of the chemistry, and in particular correlation with PSC's. The natural clearing of the volcanic aerosols from Mt. Pinatubo makes these observations interesting to compare with ones from earlier years.

Network for Detection of Stratospheric Change (NDSC)

The NDSC is a group of five primary stations and numerous complementary observation sites for enabling the earliest possible detection of stratospheric changes that will affect ozone, by comparing carefully calibrated groundbased data with satellite measurements. The Optical Techniques project will provide the high resolution infrared spectroscopic observations from Thule, Greenland, at a latitude of 76 degrees north.

The spectrometer, a Bruker Model 120M, has been acquired. This spectrometer has sixteen times better resolution than the spectrometer previously used at Søndre Strømfjord. Initial testing of the instrument is underway, in preparation for an intercomparison of the measurements from this instrument and several other NDSC instruments in spring 1996, prior to deployment to Thule. Methods are being developed for remote operation of the instrument by transmitting commands and data over the Internet between Boulder and Thule. A second instrument optimized for airborne use is under construction at NCAR to be used for intercomparing instruments located at different sites.

Substantial progress has been made, in collaboration with Aaron Goldman (University of Denver) in automating the analysis of spectra using least squares fitting methods. These methods have been used for analysis of the SESAME spectra, as well as certain species measured in the long term airborne spectroscopy data base.

Temporal Trends of Atmospheric Gases

From the data base of over 40,000 high resolution infrared spectra collected by aircraft operating at high altitude (typically 12 km) between 1978 and 1992, the temporal changes in various stratospheric compounds may be determined. Work has been accomplished recently by Hannigan, Coffey, and Mankin on carbonyl sulfide and carbon tetrafluoride.

Carbonyl sulfide is the principal source of sulfur in the stratosphere during volcanically quiet times. In the stratosphere, it is oxidized to sulfuric acid which forms sulfate aerosol. To explain increasing levels of background statospheric aerosol, it has been hypothesized that OCS is increasing. The analysis of infrared spectra recorded between 1978 and 1992 indicates no increase in carbonyl sulfide, with an upper limit of ±0.2% per year. It is thus necessary to look elsewhere for the origin of the increase in stratospheric aerosols.

Carbon tetrafluoride is produced primarily as a byproduct of the aluminum industry. It is extremely inert in the atmosphere with a lifetime of hundreds to thousands of years. Thus, the present burden represents substantially all the CF_4 ever released

into the atmosphere. Its temporal trend indicates current release rates, and they determined that the stratospheric CF_4 column is increasing at about 2% per year. Current concentrations amount to approximately 78 pptv in the stratosphere.

Volcanic Effects

Volcanoes are an important source of gases and particles to the stratosphere. The direct injection of certain gases, such as HCl, can modify the catalytic chemistry which controls ozone in the lower stratosphere. Other, larger inputs, such as SO_2, serve as precursors for aerosol particles which become the sites for heterogeneous reactions which can further affect the delicately balanced ozone chemistry. Using long-term and volcanic plume observations, Coffey and his colleagues have described the impact of volcanic injections on stratospheric HCl, SO_2 and O_3 amounts, and on the repartitioning of nitrogen oxides, such as NO, NO_2 and HNO_3, due to heterogeneous reactions on the surfaces of volcanically initiated sulfate aerosols.

Instrument Development

Key to the interpretation of many chemical measurements are dynamical tracer species to characterize the air mass in which measurements are made. N_2O has been extensively used for this purpose in the lower stratosphere, and CO is particularly valuable in the upper troposphere. The OT project has undertaken the development of an instrument based on absorption of radiation from a tunable infrared diode laser. The instrument will fly on the WB-57 measuring both gases simultaneously with a time resolution of about a second. Most of the design has been completed, and detailed engineering, parts acquisition, and testing is underway. The available funds will not permit having the instrument developed to NCAR specifications by an outside contractor as originially planned, but portions of the instrument will be built by a contractor.

Other Collaborations

The OT project is also collaborating with Robert MacQueen and John Streete (Rhodes College) on the design of an atmospheric research spectrometer for Rhodes College. Streete has been a visitor here during a sabbatical, assisting in the development of both the Bruker interferometer and the TDL instrument for the WB-57, as well as working on design of the Rhodes spectrometer.

Service Activities

Mankin serves on the international Steering Committee for the NDSC and has been a member of this group since its inception. He organized the 1995 meeting of the NDSC Infrared Working Group at NCAR. Coffey serves on the NSF/UCAR Observing Facilities Advisory Panel, which he will chair in 1996. Mankin is a member of the NCAR Appointments Review Group.

IX. Tunable Diode Laser (TDL) Spectroscopy

Idaho Hill Photochemistry Experiment Analysis

The hydrocarbon intermediate formaldehyde (HCHO) plays an important role in understanding tropospheric oxidation mechanisms and the partitioning among odd hydrogen radicals. Previous HCHO measurement comparisons as well as measurement-modeling relationships have at times exhibited large and unexplained discrepancies. These discrepancies raise significant questions regarding the accuracy of ambient HCHO measurements as well as potential deficiencies in current photochemical oxidation mechanisms. The 1993 Fritz Peak/Idaho Hill HCHO measurement intercomparison, between the NCAR field tunable diode laser absorption spectrometer (TDLAS) and the NOAA long path uv-visible spectrometer, provided an opportunity to address these issues. Detailed wind vector analyses indicated that during times of uniform and well mixed clean continental background air, both instruments measured equivalent HCHO concentrations to within 5 pptv ± 28%. This is quite remarkable considering the differences in sampling, measurement, and calibration approaches, and supports the accuracy determined in both techniques.

The HCHO measurements generated by both instruments provided a high quality data set to further investigate the HCHO measurement-modeling discrepancy as well as the relationships among various reactive intermediates. Detailed modeling studies were carried out in collaboration with other NCAR scientists as well as scientists at Pennsylvania State University (William Brune), and the NOAA Aeronomy Laboratory. Some of these studies employed the NOAA box model constrained by measurements of hydrocarbons, oxides of nitrogen, odd hydrogen radicals, photolysis frequencies, ozone, and water vapor. Mid-day HCHO model-measurement comparisons resulted in agreement to within ± 30%. This is a significant improvement over the previous 300% discrepancy reported during the first Mauna Loa experiment. The calculated HCHO concentrations were strongly dependent upon the input OH values and the particular oxidation mechanisms employed for pinene reactions. The current level of agreement places bounds on the yields for these pinene reactions and provides information regarding the nature of potential missing hydrocarbons which are needed to balance measured and modeled OH levels. The measurement and modeling results also suggest the presence of nighttime biogenic HCHO sources. If further verified, such sources may represent an important early-morning source of odd hydrogen radicals from HCHO photolysis.

Five papers authored or co-authored by the TDL group, including the HCHO intercomparison and modeling studies above, were recently submitted to a special issue of the *Journal of Geophysical Research* on the 1993 Idaho Hill Photochemistry Experiment. One of the papers describes the newly developed field TDLAS, while two additional papers discuss the role of HCHO and other species in measurement-modeling relationships for OH, HO_2/OH, and RO_2/HO_2 ratios.

Field TDLAS System

The Idaho Hill Photochemistry Experiment pointed to the importance of carrying out 24-hour unattended field measurements. This activity, which was initiated last year, has been completed and successfully employed during the 1995 Southern Oxidant Study (SOS) HCHO Intercomparison. A host program, based on the Lab View Graphical System, was developed for computer-controlled on-line spectral shifting, automated valve sequencing, on-line spectral fitting, and application of laser wavelength correction signals. In addition, significant performance improvement (factor of 4 to 5) was attained by employing the technique of rapid background subtraction which was made possible by the small volume sampling cell used. In this method, sample and background spectra were acquired in a time period fast (20 - 30 seconds) compared to changes in background structure. Subtraction of the averaged-background (immediately before and after the sample) very effectively removes performance-degrading optical noise. Employing this technique along with Fourier filtering, signal demeaning, and digital signal processing, the TDL group demonstrated routine HCHO detection sensitivities approaching 20 parts-per-trillion (pptv) in 4 minutes of integration. This, which corresponds to minimum detectable absorbances around 6×10^{-7}, is one of the lowest HCHO detection limits reported by an actual field TDLAS instrument on this time scale. Bryan Wert (graduate student, University of Colorado) working in the TDL group, was instrumental in helping to implement many software improvements. The ground-based TDLAS instrument is presently being modified for

aircraft operation on future NCAR and NOAA missions.

SOS Formaldehyde Intercomparison

A formal blind HCHO intercomparison experiment was hosted by NCAR including the TDL group, ACD scientists Tim Gilpin, Eric Apel, and retired member Jack Calvert. Reference HCHO calibration standards in the 0.3 to 6 ppbv concentration range, a range spanning levels encountered at Mauna Loa as well as urban areas, were developed and verified by TDLAS using independent approaches. Various interferences, including water, ozone, isoprene, sulfur dioxide, and nitrogen dioxide, were systematically added to these standards to identify interferences and artifact problems. Scientists from Brookhaven National Laboratory (Yin-Nan Lee), the University of Rhode Island (Brian Heikes), Texas Technical University (Purnendu Dasgupta), Washington State University (Halvore Westberg), and Mantech Environmental Laboratories (Taddeous Kleindienst) were involved in this study. The results were submitted in a blind fashion to Calvert who served as the referee. Upon completion of this phase, continuous ambient samples were acquired by each instrument. These results, including the TDLAS measurements, were also submitted blind to the referee. The results of this study, which will be described in a *Journal of Geophysical Research* publication, will provide important guidelines and recommendations for future ambient measurements of HCHO.

Laboratory Heterogeneous Studies on Sulfuric Acid Aerosols

In collaboration with Michael Mozurkewich (York University, Toronto), the TDL group continued the second phase of heterogeneous studies focusing on chlorine nitrate reaction on sulfuric acid aerosols. This reaction, and its temperature, composition, and size dependencies, is extremely important in effecting the depletion of stratospheric ozone on a global scale. The work will be completed in the 1996 fiscal year.

Kinetics Group TDL System

Fried has been active in collaborating with scientists in the kinetics group in setting up a TDL system to measure the HCl product yield from the reaction of ClO with OH. Small differences in this product yield can have large effects in modeling stratospheric ozone concentrations. This system has been completed and the studies will commence this fiscal year.

Involvement with the International Community of TDL Users

As in past years, Fried has been actively involved with the international community of TDL users in an effort to advance this technique through collaborations and international symposia. Fried has served on the organizing committee for the 1995 Russian High Resolution Spectroscopy Conference and is on the organizing committee for the 1996 Optical Society's Laser Applications to Chemical Analysis Symposium, and the 1996 International Society of Photo-Optical Instrumentation Engineers (SPIE) conference in Denver.

X. Regional Modeling and Processes

Episodic Regional Chemistry Transport Model

Peter Hess, Sasha Madronich, Siri Flocke, and Mary Barth continued the development of a regional model for the interpretation of atmospheric field measurements. This is a photochemistry-transport model using off-line episodic meteorology from the NCAR-Penn State Mesoscale Model (MM5). It currently includes advective transport, parameterization of convective and boundary layer transport, emissions, wet and dry deposition, and gas phase chemistry with diurnally variable photodissociation coefficients. The convective scheme, shallow convective scheme, and boundary layer scheme have been improved, and post-processing capability has been added. Cloud heterogeneous chemistry is now being implemented. The model is scale-flexible and is transportable. A variable-grid modeling system to allow connection of global, regional, and local scales is also being explored by Paul Ginoux (visitor, University of Brussels), through the development of a finite-difference based transport model with realistic chemistry. Preliminary simulations suggest realistic distributions of gas phase chemicals over the central Pacific region. Versions of the regional model have been used in studies of stratosphere-troposphere exchange, and of the effects of model resolution on chemistry of aircraft emissions in the upper troposphere and lower stratosphere.

Barth, Hess, and Madronich have identified errors induced by common numerical integration methods when both gas and aqueous (cloud) processes are involved. A new implicit Euler-backwards scheme, that reduces these errors with minimal additional computational cost, has been developed and implemented in the regional model.

Modeling and Interpretation of MLOPEX II Measurements

Didier Hauglustaine (ASP) and Madronich modeled the fast photochemistry during the MLOPEX II experiment and found that model-predicted peroxy radicals are systematically higher than direct measurements obtained by the chemical amplifier technique. OH predictions are in good agreement for the spring 1992 intensive, but overestimate the measurements by about a factor of two for the summer intensive. Measurements of the NO/NO_2 ratio allowed an independent estimation of the peroxy radical concentrations that was found to agree well with model predictions. The results of the model-measurement comparisons suggest that additional HO_x losses, currently not included in the model, may be occurring in the MLOPEX II environment. Work in progress focuses on intermediate-lived species (e.g., CH_2O, HNO_3, PAN) with special interest in the possible role of heterogeneous reactions.

Hess and Flocke computed ten day back-trajectories for the MLOPEX II campaign. Correlations with chemical measurements provided some insight on the origin and chemistry of free tropospheric air observed during MLOPEX II.

Patrice Gregoire (University of Blaise-Pascal, France) and Madronich updated the NCAR Master Mechanism (a comprehensive, explicit representation of the gas phase chemistry with about 5,000 reactions among ca. 2,000 species) with new kinetic data for the oxidation of methane and its subsequent oxidation intermediates. The model was then used to show that predicted CH_2O concentrations are in good agreement with the measurements obtained during the MLOPEX II campaign, if the experimental values of CH_3OOH are used to constrain model simulations. This demonstrates that model-measurements discrepancies in CH_2O previously found for MLOPEX I and by other workers in different experiments may actually be traced to the discrepancies of one of its principal precursors, CH_3OOH, and indicate the need for further study of CH_3OOH chemistry.

Kathy Lantz (ASP), Madronich, and Flocke computed photodissociation coefficients for comparison with actinometric and radiometric measurements at Mauna Loa, with special emphasis on the radiative perturbations due to Mt. Pinatubo aerosols. Comparisons with the MLOPEX II measurements has lead to elucidation of the quantum yield for the critically important reaction, O + hn Æ O + O(^{1}D). Large model-measurements discrepancies were identified for the NO photolysis rate

3 2 2
coefficients.

Ultraviolet and Visible Atmospheric Radiation

Madronich and Flocke completed the development of the Tropospheric Ultraviolet-Visible (TUV, version 3.3) model for the computation of photochemical rate coefficients as well as biologically and radiometrically pertinent radiation. Model results have generally compared favorably to measurements, and have been used in a number of theoretical studies and in international assessments of UV changes associated with stratospheric ozone depletion. The model is in use at a number of institutions around the world.

Madronich and Barbara Prezelin (University of California, Santa Barbara) combined theoretical UV calculations with measurements of phytoplankton primary productivity in the Antarctic to estimate the spectral sensitivity to UV radiation under ozone-depleted conditions.

Madronich and Elisabeth Weatherhead (CIRES/NOAA) developed a technique for reconstructing the detailed UV spectrum (280-400 nm) from measurements made at only a few wavelengths. This allows the use of relatively inexpensive filter radiometers in lieu of more expensive spectral radiometers under most conditions.

Jun Zeng and Madronich began development of a global climatology model of tropospheric photodissociation rate coefficients and biologically effective UV radiation. Initial results have shown that more accurate photodissociation coefficients can be obtained by applying the delta-scaling transformation in fast two-stream models.

Madronich collaborated with Andreas Voltz and other researchers from the Kernforschungzentrum (Jülich, Germany) to compare theoretical NO_2 photodissociation coefficients with measurements obtained as a function of altitude in the troposphere from an aircraft platform.

Theoretical Studies

Madronich and Hess continued the exploration of the non-linearities of the tropospheric oxidizing capacity system. Self-generating chemical oscillations were identified for a number of different mechanisms, indicating that they are a robust feature of the tropospheric oxidizing system. Analytical solutions were derived for various characteristics such as regimes and periods of oscillation.

Madronich, Flocke, Hess, and Chris Fischer have also carried out a theoretical study of the response of ozone to CO changes at low NO_x conditions, and showed that the direction and magnitude of the effect of CO on ozone depends critically on the HO_x loss processes.

In another study, Madronich and Bernard Aumont (University of Paris, France) used the Master Mechanism to show the possible importance of aerosols in scavenging oxygenated hydrocarbons from the lower atmosphere. This has potential implications for estimates of the contributions of non-methane hydrocarbons to regional and global scale ozone formation.

Claire Granier, Guy Brasseur, Jean-François Müller (Belgian Institute for Space Aeronomy) and Madronich examined recent trends in CO and CH_4, and carried out a modeling study to explain recent trends in terms of decreases in tropospheric temperatures (following the Mt. Pinatubo eruption), increases in UV radiation (due to stratospheric ozone depletion), and changes in emissions from sources such as biomass burning.

Madronich and John Herring and Dan Jaffe (University of Alaska) carried out a study of the springtime tropospheric ozone maximum observed in the Arctic. The results of measurements and modeling suggest that strong net photochemical ozone production results from the wintertime build-up of precursors such as NO_x and hydrocarbons.

XI. Global and Stratospheric Studies

Global Chemical Transport Models of the Troposphere

Guy Brasseur, Stacy Walters, Didier Hauglustaine (ASP), and Claire Granier completed version 0 of MOZART (Model for OZone And Related species in the Troposphere), a three-dimensional chemical transport model that determines the global distribution of approximately 40 chemical species in the troposphere and lower stratosphere. The model is driven by winds, temperature, and convective fluxes provided by the NCAR Community Climate Model at a spatial resolution of 2.8 degrees in longitude and latitude. Surface sources, as well as the chemical scheme, are similar to those used in IMAGES (Intermediate Model for the Global and Annual Evolution of Species), a simpler three-dimensional model developed earlier by Jean-François Müller (Belgian Institute for Space Aeronomy) and Brasseur, which provides monthly averaged distributions of chemical species in the troposphere. These two models are currently being validated with available observations (provided primarily by airborne field campaigns). A paper describing IMAGES appeared in *Journal of Geophysical Research*. The chemical code developed in ACD was also implemented be John Caron and Granier in the GENESIS (Global ENvironmental and Ecological Simulation of Interactive Systems) model. A simple version of MOZART is being developed by David Erickson to be included in the NCAR Climate System Model (CSM).

Three-dimensional chemical transport models developed in ACD were used by Brasseur to assess the potential impact of subsonic aircraft on tropospheric ozone, by Erickson to determine the response of the climate system to increasing concentrations of anthropogenic sulfate in the atmosphere, by Granier to quantify the role of biomass burning on the chemical composition of the troposphere, and by Hauglustaine and Brasseur to analyze observations made during the MLOPEX II experiment in Hawaii. A scheme describing aqueous phase processes is currently being implemented in MOZART by XueXi Tie.

Global Chemical Transport Model of the Stratosphere

Chemical-Transport Model Driven by CCM-2 Dynamics

Tie and Brasseur have completed the development of a middle atmospheric version of a chemical transport model driven by the dynamics of CCM2. This model includes specifically the effects of chlorine and bromine on stratospheric ozone plus the impact of the nitrogen and hydrogen families. Heterogeneous chemical processes on the surface of solid and liquid particles (polar stratospheric clouds, sulfate aerosols) are explicitly represented in the model. The model was used to simulate the formation and fate of the Antarctic ozone hole, and its impact on mid-latitude ozone. It was shown, for example, that, during the austral spring and summer, ozone in the southern hemisphere upper troposphere could be dramatically reduced after the break up of the polar vortex. The model was also used to assess the role of heterogeneous chemistry related to bromine compounds, which seem to dramatically amplify the role catalytic destruction of ozone by chlorine radicals.

Mechanistic Model of the Stratosphere

Anne Smith completed a study in which the three-dimensional dynamical-chemical model was used for assessing the impact of diurnal averaging methods used in two- and three-dimensional models. The results give a quantitative measure of the impact on the composition of using daylight average and 24-hour average photolysis rates.

The new dynamical components of the three-dimensional model, with interactive radiation and the Fritts gravity wave parameterization, are being coupled with the improved chemistry to make a more accurate mechanistic model. Several university collaborators (Kunihiko Kodera, Meteorological Research Institute, Japan; Joseph Sabutis, UCLA; Yonghua Wang, Georgia Institute of Technology) are now using part or all of this new model. The new mechanistic model is also being used for a study of the interaction of tides and gravity waves in the mesosphere.

New Two-dimensional Model of the Middle Atmosphere

Theresa Huang, Brasseur, Walters, and Simon Chabrillat (Belgian Institute for Space Aeronomy) have further developed a new two-dimensional model of the middle atmosphere which extends up to 120 km altitude and interactively treats chemical, dynamical, and radiative processes. Compared to the previous version of this model, the new code includes a more detailed chemical scheme including several heterogeneous reactions, a new radiative formulation (Delta-Eddington method), and a representation of non-local thermodynamical equilibrium for CO_2 cooling in the upper atmosphere. The dynamical scheme includes a relatively explicit representation of Rossby waves established by Wanli Wu (University of Michigan), as well as the new formulation of gravity wave breaking developed by David Fritts (University of Colorado). A tide model as well as a simple representation of the quasi-biennal oscillation are also included in the model. Currently, the model is being tested extensively.

Smith and Wu are using the wave model that has been added to the two-dimensional model to study transport. The "three-dimensional" winds are reconstructed from the model two-dimensional winds and the winds associated with the planetary wave. Comparisons of the transport by these winds and the net (parameterized) transport used in the two-dimensional model indicate problems with the flux-gradient relationship that is the basis of the transport parameterization. These differences are being used to improve the representation of net wave transport.

Impact of Sulfate Aerosol on Stratospheric Ozone

Tie and Brasseur have used their two-dimensional model coupled with microphysics to determine the relationship between ozone changes caused by volcanic eruptions and chlorine loading of the stratosphere. They showed that for low chlorine loading, an eruption such as that of Mt. Pinatubo would have caused an increase (rather than a decrease) in the ozone column abundance.

Rolando Garcia, with Susan Solomon and Robert Portmann (NOAA), has submitted two papers to *Journal of Geophysical Research* showing that the observed trend in ozone column since the 1970's --both at midlatitudes and in the polar vortex-- can be explained if the precise amount of aerosol loading is taken into account in the calculations.

Dynamics of the Middle Atmosphere

Working with Joseph Prusa (Iowa State University), Garcia used a high-resolution, semi-Lagrangian code developed by Piotr Smolarkiewicz (MMM) to model the forcing of small-scale gravity wave's by tropospheric sources of variable spatial dimensions. The work may have important implications for gravity wave parameterizations, and for understanding the formation and location of the turbopause. A paper has been submitted to *Journal of Atmospheric Science*. Garcia and Fabrizzio Sassi have shown that high-frequency convective forcing over the tropical continents forces "intermediate scale" waves ($k>6$) that may contribute significantly to the momentum budget of the semi-annual oscillation in the stratosphere and mesosphere.

Climate and Global Dynamics Division

CGD Director's Message

CGD Mission

CGD ASR Roadmap

- Significant Accomplishments
- Staff, Visitors, and Collaborators
- Publications

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Division Director's Message

Maurice Blackmon, Climate and Global Dynamics Divison Director

For many years, individuals have attempted to understand how the climate system works and, from that understanding, how to forecast climate. Better predictions of future climate could warn of droughts, floods, the El Nino-Southern Oscillation (ENSO) and its midlatitude effects, as well as possible harm to the environment from human activities, such as deforestation of the landscape and industrial emission of carbon dioxide ($CO_2$and other trace gases into the earth's atmosphere. Climate models have projected that increased concentrations of carbon dioxide and other trace gases in the atmosphere could create global climate change, the so-called enhanced greenhouse effect. Climate predictions, whether for naturally occurring or human-induced enhanced climate change, can only be accomplished through the use of physically based models.

Climate and Global Dynamics (CGD) Division scientists have a long history of development and use of models in the atmosphere and ocean. Progress in modeling and increases in computer power led to the conclusion that the time was ripe for development of a comprehensive climate system model to facilitate investigations into the understanding of the earth's climate. To this end the Climate System Model (CSM) project was begun in fiscal year 1994 (FY94). A plan was prepared in late FY94, and distributed, peer reviewed, revised, and approved by the National Science Foundation (NSF) early in FY95. The plan cited early model development by the National Center for Atmospheric Research (NCAR) scientists, with university participation increasing in the near future. This approach allowed for meeting the time table of the Intergovernmental Panel on Climate Change (IPCC) and the United States Global Change Research Program (USGCRP). A small workshop in late 1995 reviewed the configuration of the CSM and initiated involvement of the scientific community. Another workshop in May 1996 will bring in many users and students for training and discussion of the CSM.

Our intent is to expand current climate models to include all aspects of the climate system-- atmosphere, ocean, cryosphere (including snow, sea ice, glaciers, and ice caps), biosphere and terrestrial ecosystems, other land surface processes, and additional parts of the hydrosphere, including rivers--and all the complex interactions of these components, (schematic diagram, 5K) emphasizing the importance of the flux coupler concept. We support climate model development with observed data analysis, model evaluation and diagnosis, regional model development, and climate change assessment.

NSF-sponsored research projects in which CGD is the NCAR lead division are the Climate Modeling, Analysis, and Prediction (CMAP) program, Geosystems Databases, and CLImate VARiability and Predictability (CLIVAR)/Tropical Ocean and Global Atmosphere (TOGA). CGD also participates in the Global Tropospheric Chemistry Program (GTCP) and the High Performance Computing and Communications (HPCC) program.

There is a strong relationship among CSM, CMAP, the Climate System Modeling Program (CSMP), and the Climate Simulation Laboratory (CSL) (diagram, 7K). CMAP is an NSF-funded program (to study the physical processes involved in the climate system). Research areas at NCAR and within the university community include climate model development, simulation and prediction validation, error estimation, and assessment of predictability. CSM is NCAR's effort to computationally model the climate system. CSMP supports linkages to universities and national laboratories to study the physical and chemical aspects of the climate system, particularly hydrology, ecology, and glaciology. CSL is the computer

hardware system needed to facilitate the model(s) developments and uses.

CGD plans and carries out research activities by collaborating within the division, within NCAR, with university scientists, and with other institution scientists. As we expand our knowledge of the climate system, we communicate our results through scientific journal publications (109 refereed papers in FY95, 68 in FY94, 71 in FY93), scientific seminars, workshops, positions at universities, public presentations, and cataloging information on the World Wide Web (http://www.cgd.ucar.edu). We also involve students in our research, including postdoctorates, graduate research assistants, undergraduates, and Summer Employment Program (SEP) students. CGD is divided into six research sections and a computer support group with 90 regular and term employees (full- and part-time), including 32 Ph.D. scientists. We have about 35 long-term visitors (greater than six-month stay) annually, as well as 120 short-term visitors, whose stays are from one day to six months. The FY95 NSF budget was $5.1 million out of $57.9 million for all of NCAR.

Navigating

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