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FOREWORD

Fiscal year 1994 has been an interesting time for the Atmospheric Science Program. Results are beginning to flow in from the most recent series of research awards. Ongoing research efforts to address upper-tropospheric and midlatitude issues continue to be closely coordinated with the U.S. Global Change Research Program (USGCRP) and are reported as part of the Nation’s focused global change research. The reader is advised to peruse the DOE publication Global Change Research: Summaries in FY 1994 (DOE/ER-0641T), which includes many atmospheric-science research reports on climate change.

The research of the Atmospheric Science Program will continue to focus on activities that respond to the DOE mission, national needs, and international opportunities. For example, ozone nonattainment is a problem throughout North America. To address research and policy efforts in this area, the North American Research Strategy in Tropospheric Ozone (NARSTO) was formed by a cross-section of federal, state, and local governments along with private-sector interests and the Canadian and Mexican governments. The Atmospheric Science Program has contributed in-kind support to the NARSTO program and community and will continue to do so.

The Atmospheric Dynamics Program continues its tight focus on the planetary boundary layer in complex terrain through the Atmospheric Studies of Complex Terrain (ASCOT) program. Research at the DOE Rocky Flats facility has produced data for site-specific application. However, the need for fiscal restraint has resulted in decreased budgets in this research area and the cancellation of planned field activities. Modeling and research activities are being maintained such that the programs continue to provide general scientific understanding of the lower planetary boundary layer. As the USGCRP begins to explore Earth-system models, such information will be essential in representing surface processes of the free atmosphere.

We wish to welcome Rickey Petty as a new member of our Atmospheric Science Program management team. Rickey brings with him policy and science experience gained as a participant in various meteorological programs at Brookhaven National Laboratory and in the private sector.

I would like to take this opportunity to thank Jake Hales, Science Director of the Atmospheric Chemistry Program (ACP), and Chris Doran, Scientific Director for Atmospheric Dynamics, for their inspired leadership and sustained commitment to our stakeholders. We all appreciate the efforts of Bobbi Parra for leading the compilation of the report. I would also like to thank F. M. O’Hara, Jr., for his continued assistance in this document’s compilation and publication.

Michael R. Riches, Acting Director
Environmental Sciences Division
Office of Health and Environmental Research
Office of Energy Research
This document describes the activities and products of the Atmospheric Science Program of the Environmental Sciences Division, Office of Health and Environmental Research, Office of Energy Research, in FY 1994. The report is organized into four main sections.

**SECTION**

- **Introduction**  
  - Provides a mission statement, need, and mandate for an atmospheric science program
  - Ties atmospheric-science issues to the research approach, program goals, and objectives
  - Relates each specific research area to the overall goals of the program
  - Shows the level of effort

- **Research Areas and Project Descriptions**  
  - Describe scientific questions in each research area
  - Provide descriptions of individual research projects
  - List the expected product for each project
  - Show the research approach or methodology used in each project
  - Provide results to date when applicable

- **Appendixes**  
  - Provide addresses of principal investigators and define acronyms used in the text

- **Indexes**  
  - Provide locator information on subjects, principal investigators, and research institutions for the overall report

Questions concerning the Atmospheric Science Program or specific projects may be addressed to the Environmental Sciences Division, U.S. Department of Energy, ER-74, Washington, DC, 20585. The Program’s telephone number is (301) 903-3281.
INTRODUCTION

ATMOSPHERIC SCIENCE PROGRAM

The Atmospheric Science Program of the Department of Energy (DOE) develops a comprehensive understanding of the atmospheric processes that control the transport, transformation, and fate of energy-related air pollutants. The objectives are (1) to understand the atmospheric-chemistry processes of energy-related air pollutants (e.g., sulfur and nitrogen oxides and ozone) in both gas and aqueous phases and their gas-to-particle conversion and fate, including deposition and resuspension; (2) to understand the meteorological processes that control the dispersion of material releases to the atmosphere over complex terrain (e.g., in mountainous or coastal regions); (3) to develop predictive models for the above processes and to acquire the data to validate them; and (4) to develop an understanding of the trends in midlatitude stratospheric ozone change and the heterogeneous chemistry and dynamics controlling ozone in this region.

CONTEXT: MISSION AND MISSION NEED

Meeting the nation's energy needs imposes anthropogenic demands on the environment. A basic understanding of how the atmosphere and the terrestrial and marine ecosystems transport, transform, and cycle energy-related materials and how these systems respond to natural and human-induced changes is required to ensure that future energy sources can be secured and that environmental quality is protected and enhanced.

The mission of DOE's Environmental Processes and Effects Research (EPER) Program, in which the Atmospheric Science Program resides, is to (1) quantify the physical, chemical, geological, and biological processes that transport, disperse, and transform energy-related materials in the atmosphere and in terrestrial and marine ecosystems; (2) to determine the mechanisms of degradation and detoxification of hazardous energy-related materials in the environment; and (3) to determine and manage the capacity for ecosystem adjustments to energy-related environmental stresses.

The EPER Program primarily addresses the environmental components of the National Energy Policy Act of 1992 (EPACT) that are local to regional in character (e.g., environmental restoration at contaminated DOE facilities and atmospheric modeling for emergency response). The program also complements the Office of Health and Environmental Research (OHER) Global Change Program addressed elsewhere. Research within the EPER Program provides scientific data as well as modeling, geochemical tracer, and biotechnological tools. These tools are needed to link the physical, chemical, and biological processes operating at molecular scales with the atmosphere-marine-terrestrial feedback mechanisms that operate at regional and global scales. Research on tropospheric and stratospheric ozone is also included.

PROGRAM MANDATES

Mandates to conduct research on environmental processes and effects include the Atomic Energy Act of 1954 (PL 83-703), authorizing the conduct and support of research and development related to the preservation and enhancement of a viable environment; the Energy Reorganization Act of 1974 (PL 93-438), promoting environmental research related to the development of energy sources and utilization technologies; the Federal Nonnuclear Energy Research and Development Act of 1974 (PL 93-557), authorizing a comprehensive nonnuclear energy research, development, and demon-
stration program including environmental consequences of energy technologies; and the Department of Energy Organization Act of 1977 (PL 95-91), mandating the Department to “assure incorporation of national environmental protection goals in the formulation and implementation of energy programs and to advance the goal of restoring, protecting, and enhancing environmental quality and assuring public health and safety” and to conduct “a comprehensive program of research and development on the environmental effects of energy technology.”

Research on environmental processes and effects also supports mandates in EPACT (PL 102-486), which calls for DOE to evaluate the environmental benefits and costs of technologies to improve energy efficiency and of technologies and processes for preventing pollution. EPACT also mandates DOE to evaluate and compare the environmental performance of clean coal technologies relative to that of other conventional technologies.

PROGRAM RELATIONSHIPS

At the interagency level, EPER Program components are coordinated via the subcommittees and working groups of the Committee on Environment and Natural Resources (CENR) of the White House’s National Science and Technology Council (NSTC). For example, the atmospheric-science programs are focused in the CENR Subcommittee on Global Change and contribute to the CENR Subcommittee on Air Quality. (In this sense, focused programs are those in which the research resources are accounted for once for a given CENR subcommittee effort. Contributing programs are those whose research resources can be accounted for more than once for a number of CENR subcommittee efforts.)

In addition to the CENR process, EPER programs pursue other intra-agency and interagency collaborations. The Subsurface Science Program coordinates with DOE’s Office of Environmental Restoration and Waste Management and other agencies (e.g., the Nuclear Regulatory Commission and the U.S. Geological Survey) by joint research funding and cooperative field studies; this program also sponsors research in collaboration with industry, such as oil companies, and has a formal technology-transfer program that moves basic research results to DOE sites and industry. The Ocean Margins Program has joined forces with the National Science Foundation (NSF) in the Joint Global Ocean Flux Study. The Environmental Radon Program is a part of OHER’s Radon Research Program, which coordinates with DOE’s Office of Conservation and Renewable Energy and the U.S. Environmental Protection Agency via a memorandum of understanding (MOU).

PROGRAM OBJECTIVES

The objectives of EPER are to develop the scientific tools to assess the environmental consequences of energy-related activities and to promote improvements in environmental quality. EPER currently includes five distinct components that address different problem areas of high priority to DOE. In addition to the Atmospheric Science Program, whose objectives were defined at the beginning of this section, the other four components are:

1. The Subsurface Science Program emphasizes long-term mechanistic research within the subsurface biosphere; it contributes to resolving problems of contaminant transport that result from surface and shallow land burial of defense and energy-related wastes and to environmental restoration at DOE sites.

2. The Environmental Radon Program encompasses basic research on the sources and transport of radon in the environment and on processes that can affect its entry into buildings.

3. The Ocean Margins Program addresses a quantitative understanding of the dispersal and fate of energy-related materials at the land/ocean interface. The principal focus of this program is on the fluxes of carbon in the coastal ocean and their implications to the global carbon cycle and, therefore, to global climate change.
4. The Program for Ecosystem Research examines the processes of ecosystem adjustment to environmental changes and uses this understanding to protect and restore ecosystems that are adversely impacted or threatened by energy production and use.

ATMOSPHERIC SCIENCE PROGRAM STRATEGY

The Atmospheric Science Program involves interdisciplinary field and laboratory research and modeling activities. Where appropriate, research is conducted at DOE sites. Research focuses on developing a mechanistic understanding of the processes controlling the environmental transport, transformations, and fate of energy-related materials in the atmosphere.

The ultimate goal is to develop predictive models for use in emergency preparedness and/or in assessing the environmental and human-health risks associated with energy technologies. The Atmospheric Science Program also promotes the development and transfer of results, models, and new concepts and technologies from research within DOE and industry.

The Department’s Environmental Sciences Division (ESD) implements the strategy at the Department’s national laboratories, other governmental laboratories, universities, and private industry. The research paradigm includes infrastructure development and science-team elements that provide the intellectual direction for the research and facilitate technology transfer.

PLANNED ACTIVITIES

The Atmospheric Science Program focuses on (1) field activities of the International Global Atmospheric Chemistry Program (IGAC) and the interagency Southern Oxidant Study (SOS), (2) the role of aerosols in atmospheric radiation and cloudiness modification, (3) air-surface exchange processes over land and water, (4) development of advanced boundary-layer and mid-tropospheric chemistry models for interactive chemistry and climate studies, (5) studies of areas of complex terrain to examine the interactions of local meteorology and mesoscale systems, (6) extending the understanding of scale interactions at the local and mesoscale levels, (7) providing advanced predictive capabilities for emergency preparedness at DOE sites, and (8) measurements and analysis of boundary-layer features needed for a better understanding of air-quality problems.

The Ozone Project, which is included in the Atmospheric Science Program, focuses on improving the estimates of ozone and UV-B trends and on understanding the chemical and dynamical processes that control midlatitude, lower-stratospheric ozone and free-tropospheric ozone in regions remote from major air-pollution sources. This program is conducted in close cooperation with ARM.

PLANNING ASSUMPTIONS

- The EPACT will require air-quality assessments of the reauthorized Clean Air Act.
- Safety and health considerations will require improved emergency-preparedness capabilities for accidental releases at DOE sites.
- There will be an increasing demand for changes in environmental regulations related to energy development so they will be based on a clear understanding of the relevant scientific issues, including the causes and effects of the perceived problem. Knowledge of how ecosystems respond to external stresses will contribute to sensible and cost-effective regulatory practices.

PROGRAM SCHEDULE AND PROGRESS

- Complete the summer 1995 field phase of the NARE/SOS Nashville Field Study and analyze the data (1995).
- Complete the summer 1995 field phase of the coastal ozone field study and analyze the data (1995).
Complete the reanalysis of the SAGE ozone data (1997).

Complete the comprehensive survey on UV-B measurement, occurrence, and relationships (1997).

Complete the quantitative characterization and modeling of UV/visible actinic flux in the troposphere (1997).

Conduct measurement campaigns on air-surface exchange (over land and ocean) for improved parameterizations of these processes in global chemistry models (1994–99).

Undertake atmospheric-aerosol studies to elucidate the role of aerosols in cloud and radiative-balance modification (1994–99).

Complete the analysis of data obtained from field campaigns at the Colorado Front Range, the Tennessee River Valley, and their related field studies in regions of complex terrain (1995–97).

Begin the development of a study of meteorologic factors affecting air quality in the southwestern United States, focusing on those phenomena that are poorly understood or poorly represented in existing computational models (1995–2000).


At ARM sites, initiate UV measurements in cooperation with the U.S. Department of Agriculture (1995).

Evaluate current ozone databases and incorporate recalibrations of instruments and investigate data reanalysis to include other possible impacts on the data (e.g., tropospheric aerosols and other regional factors) (1995–99).

Initiate modeling studies to understand the sensitivities and uncertainties associated with the chemical-kinetic parameterizations currently being used for simulating the chemistry of ozone formation and destruction in global models (1995–99).

PROGRAM ADMINISTRATION

The Environmental Sciences Division of the Office of Health and Environmental Research, Office of Energy Research, supports the Atmospheric Science Program to contribute to the scientific foundation for the EPACT.

LEVEL OF EFFORT

Research sponsored by the Atmospheric Science Program amounted to $12,987,000 in FY 1994. The distribution of funds to research areas is presented in Fig. 1.

ORGANIZATION OF THE PROGRAM

SUMMARY

This document provides descriptions for all projects funded by ESD under annual contracts in FY 1994. Each description contains the project’s title; three-year funding history (in thousands of dollars); the contract period over which the funding applies; the name(s) of the principal investigator(s); the institution(s) conducting the projects; and the project’s objectives, products, approach, and results to date (for most projects older than one year).

Project descriptions are categorized within the report according to program areas: atmospheric chemistry, atmospheric dynamics, and support operations. Within these categories, the descriptions are ordered alphabetically by principal investigator. Each program area is preceded by a brief text that defines the program area, states its goals and objectives, lists principal research questions, and identifies program managers. Appendixes provide
Fig. 1. Distribution of Atmospheric-Science Funding for FY 1994 in Thousands of Dollars.

the addresses and telephone numbers of the principal investigators and define the acronyms used.

This document has been indexed to aid the reader in locating research topics, participants, and research institutions in the text and the project descriptions. Comprehensive subject, principal investigator, and institution indexes are provided at the end of the text for this purpose. The comprehensive subject index includes keywords from the Introduction and chapter texts in addition to those from the project descriptions.
Atmospheric chemistry research within DOE is addressed primarily to the question of atmospheric response to emissions from energy-generation sources. As such, this program deals with the broad topic known commonly as the *atmospheric source-receptor sequence*. This sequence consists of all aspects of energy-related pollutants from the time they are emitted from their sources to the time they are redeposited at the Earth’s surface.

Because source-receptor transport distances are often large, the interaction between U.S. emissions and those from other countries becomes an important aspect of this issue. Recently, many of these aspects have become focused under the EPACT; accordingly, the DOE Atmospheric Chemistry Program (ACP) has been designed to respond directly to agency needs as defined within the EPACT.

**Research Objectives.** The principal objective of the DOE Atmospheric Chemistry Program is to provide DOE with the advanced information pertaining to the atmospheric environment that is necessary for long-range energy planning in fulfillment of the EPACT. Based on anticipated near- and intermediate-term DOE needs, initial efforts under this program will emphasize the continental and oceanic fate of energy-related air pollutants. This research activity considers all features of the atmospheric source-receptor sequence, including air-surface exchange, heterogeneous and homogeneous air chemistry, and precipitation scavenging.

**Research Questions.** Accomplishing this objective will require quantitative answers to questions on a variety of issues. Several of the more important of these issues are outlined below.

**Continental issues:** energy production’s role in
- Ozone-standard attainment or nonattainment
- Acidic deposition, both dry and wet, as related to its continental impacts
- Atmospheric aerosols: PM-10 (particulate matter less than 10 μm) standards and their attainment or nonattainment
- Effectiveness of future emission regulations in addressing the above issues

**Extracontinental and oceanic issues:** energy production’s role in
- Multinational contributions to the global pollution pool
  - The North Atlantic
  - The North Pacific
  - The Arctic
  - The significance of former Soviet Bloc and third-world contributions in the context of U.S. and western European energy development
- Long-range pollutant-transformation processes and their impact on the lifetimes and fates of pollutants
- Deposition processes and their influences on pollutant lifetimes and fates
- Natural emissions into the atmosphere and their contributions to the total mixture of atmospheric pollutants
- Aerosols and their direct influence on the atmospheric radiation balance
- Aerosols, aerosol precursors, and their effects on cloud formation
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Research Summaries

MORPHOLOGY-DEPENDENT STIMULATED RAMAN SCATTERING (MDSRS) STUDIES OF GAS-AEROSOL INTERACTION KINETICS

AKER, PAMELA M.
UNIVERSITY OF PITTSBURGH
FY 1994  73
FY 1993  74
FY 1992  83
09/15/94–09/14/95

Objective: To develop the morphology-dependent stimulated Raman scattering (MDSRS) technique to attain significantly increased sensitivity levels.

Product: A measurement technique with which to address the uncertainty in the heterogeneous reactions of pollutants on the surfaces of (or within) aerosol particles.

Approach: This project will develop the MDSRS technique to attain sensitivity levels that are much higher than those of previous measurements. The experimental measurements will apply the enhanced MDSRS technique to measure reaction and/or condensation rates of sulfur and nitrogen gases (SO₂, DMS, and HNO₃) on the surfaces of water, ice, and sulfuric acid particles. The intensity of Raman scattering is amplified appreciably when the exciting radiation's frequency is near that of an electronic transition. In essence, this sensitivity enhancement will be accomplished by applying shorter wavelength radiation, thereby approaching electronic-transition frequencies.

Results to Date: We have been developing and applying a new nonlinear Raman laser technique to study aerosol particle structure and composition called morphology-dependent stimulated Raman scattering, MDSRS. We have shown that MDSRS can be used to quantify aerosol chemical composition. We elucidated the gain equation and showed that an exponential relationship exists between the MDSRS signal and chemical species concentration. We also demonstrated that the MDSRS signal is highly stable, a result that shows that MDSRS is an extremely precise tool for determining aerosol chemical composition.

We recently discovered that MDSRS can be used to scrutinize the molecular structure of aerosol-solvent species and showed that MDSRS can be used as an imaging tool. We combined the MDSRS imaging capability with the structure-determination capacity and showed the structure changes that occur across the liquid/air interface of laboratory-produced aerosols. These results show that a detailed picture of the extent and type of chemical bonding present at the aerosol interface can be taken. These experiments open up a new path in optical imaging science and have a large impact on fields far removed from the atmospheric sciences.

MODELING IN SUPPORT OF DOE’S PARTICIPATION IN THE NORTH ATLANTIC REGIONAL EXPERIMENT

ATHERTON, CYNTHIA
LAWRENCE LIVERMORE NATIONAL LABORATORY
FY 1994  97
FY 1993  100
FY 1992  156
10/01/93–09/30/94

Objective: To use tropospheric chemistry models in support of the Pacific Northwest Laboratory G-1 aircraft measurements in the North Atlantic Regional Experiment (NARE).

Product: The ability to predict oxidant concentrations in the troposphere resulting from energy-related pollutants.

Approach: Before the experiment, model estimates of the expected abundance of NO, NO₂, HNO₃, DMS, SO₂, and aerosol will be made with meteorological fields from the LLNL-version of the NCAR general circulation model, CCM1. Within the constraints of the ability to complete the development of the nucleation
model, model estimates of the vapor phase of H₂SO₄ and aerosol number concentrations may be provided. These estimates will be focused on the region of the G-1 flights. Because these flights are close to the source regions and because the model may inaccurately portray these regions, a number of sensitivity studies will be carried out to compare the predictions of the model for this region with different numbers of parcels, different mixing coefficients, and different algorithms for mapping concentrations from the Lagrangian parcels (which are computed by the models) to concentration ratios on the Eulerian grid.

Following the G-1 experiments, predictions of the model will be compared with the data collected by the G-1 and by other groups participating in NARE. If possible, this comparison will be done with realistic meteorology that is specific to the time frame of the measurements. This model/data comparison will be used to check the consistency in understanding the sources, transport, transformation, and loss processes of the trace species involved in the formation of atmospheric oxidants and sulfate aerosol. The comparison will also be used to help plan a more comprehensive field experiment in the future.

Results to Date: Two key tropospheric chemical species are ozone (O₃), an important greenhouse gas and pollutant, and hydroxyl radical (OH), a key regulator of the atmosphere’s oxidative capacity. In the troposphere, they are formed by the oxidation of carbon monoxide (CO), methane (CH₄), and nonmethane hydrocarbons (NMHCs) in the presence of nitrogen oxides (NOₓ). The precursors have important natural and anthropogenic sources.

We developed and applied a global, 3-D chemistry-transport-deposition model to show how reducing North American fossil-fuel-combustion NOₓ emissions by 50% would affect ozone in the North Atlantic Ocean atmosphere. For the baseline scenario, predicted mixed layer and vertical profiles of O₃ compared well with observations at remote global locations. Model-predicted mixed-layer concentrations over the North Atlantic Ocean were slightly higher than observed, possibly because the measurements indicate the presence of a shallow surface layer in which O₃ is depleted. Our model simulations show that if North American fossil-fuel combustion of NOₓ is decreased 50%, mixed-layer O₃ concentrations may decrease by 4 to 12 ppbv over the North Atlantic Ocean in August.

DOE ATMOSPHERIC CHEMISTRY PROGRAM MANAGEMENT

BARCHET, W. R.

PACIFIC NORTHWEST LABORATORY

FY 1994 261
FY 1993 270
FY 1992 170
10/01/93–09/30/94

Objective: To provide scientific management and leadership of the DOE Atmospheric Chemistry Program.

Product: The atmospheric-chemistry information necessary to implement specific requirements of and for progressive modification of the National Energy Strategy (NES).

Approach: DOE’s atmospheric-chemistry program will be elevated to a point where DOE is fully recognized as a scientifically respected and equal participant in the global atmospheric-chemistry research community. The atmospheric-chemistry efforts at the DOE labs will be coordinated so that their composite contribution maximizes benefit to DOE. DOE-laboratory activity in atmospheric chemistry will be focused in such a manner that it maximizes response to the needs set forth in the National Energy Strategy. Program restructuring and implementation will address the needs for program reorientation and design to directly fulfill needs associated with the NES through:

- Conceptual and interactive design of the forthcoming scientific research plan and implementation of its management and accountability structure.
- Planning and implementation of an ACP peer review.
- Planning and implementation of a DOE university grants program in atmospheric chemistry.
- Creation and implementation of a program plan document for formal implementation of the ACP.

Scientific program monitoring and leadership will be guided by the Program Plan Document and will include:
Implementation of the programmatic adjustments necessary to ensure focused and timely performance.

Organization of external peer reviews at scheduled intervals.

Provision of basic program leadership, implementing the joint planning and decision-making process.

Coordinating major program changes, when they become necessary as a consequence of external driving forces.

Success of the DOE Atmospheric Chemistry Program will depend on its being strongly interactive with external scientific entities, both at the planning and implementation levels.

Research oversight will involve the initiation of small development projects that are critical to the evolving program but were not foreseen in the program's overall scientific design. These projects will have to respond to previously unforeseen but currently anticipated program needs.

Results to Date: During the 7-year life of this project, which ended in FY 1994, atmospheric-chemistry research within the DOE addressed major national issues, including fate of pollutants, acid rain, wet and dry deposition, and tropospheric and stratospheric ozone. Through much of this period, J. M. Hales (now with Envair) provided scientific direction and coordination among the DOE laboratories engaged in DOE-funded atmospheric chemistry research. Six major field studies in the Processing of Emissions by Clouds and Precipitation program supported the National Acid Precipitation Assessment Program and quantified the role of in-cloud oxidation of SO\textsubscript{2} to sulfate and precipitation scavenging in producing acid rain. At the close of the NAPAP era, a new Atmospheric Chemistry Program was fashioned to examine the role of energy-related chemical emissions in domestic issues (such as ozone-standard non-attainment, visibility reduction, and UV-B trends) and in global issues ranging from stratospheric-ozone trends to multinational contributions to the global pollution pool. Under the leadership of this project, five DOE laboratories, four academic institutions, three industrial firms, and one other government laboratory are now engaged in a coordinated program of atmospheric chemistry research for the DOE.

MEASUREMENTS OF RADON AND/OR RADON PROGENY WITH THE ACP RESEARCH AIRCRAFT

BECK, HAROLD; FISENNE, ISABELLE; NEGRO, VINCENT; and LEE, HSI-NA

ENVIRONMENTAL MEASUREMENTS LABORATORY, DOE

| FY 1994 | 99 |
| FY 1993 | * |

10/01/93–09/30/94

Objective: To measure radon and/or radon progeny on ACP Research Aircraft North Pacific 1994 Campaign. These data will be used to indicate or verify the presence of continental air and to make rough estimates of the transport time of that air over the Pacific.

Product: Precise measurements of radon concentrations that can be used to verify the presence of continental air masses.

Approach: For each 1994 flight, up to 12 grab samples will be obtained by flowing air at a rate of about 1 SCFM for about 10 min through cooled charcoal traps. The radon will be de-emanated into and counted in radon pulse ionization chambers (PICs). The traps will then be regenerated and reused. This procedure will allow a minimum detection limit of about 0.2 pCi and allow concentrations of less than 5 pCi/m\textsuperscript{3} to be measured with a 10-min sampling time. A new ambient-temperature charcoal trap will also be developed and phased in. It is anticipated that the new measurement capabilities will be available for the 1995 North Pacific campaigns and will allow a significantly greater sampling throughput.

Results to Date: The collection and analysis of radon and/or radon progeny samples was delayed because the Northeastern Pacific Air-Chemistry Study was indefinitely postponed. However, the precise measurements of radon gas are currently being planned for the 1995 ACP G-1 chlorine chemistry study.

*In previous years, this entry was consolidated with others under the principal investigator Merrill Heit.
CONTINENTAL AND OCEANIC FATE OF AIR POLLUTANTS

BERKOWITZ, CARL M.

PACIFIC NORTHWEST LABORATORY

Objective: To quantify atmospheric chemical species and atmospheric processes important in energy-related pollutants and to develop a model describing regional-to-hemispheric-scale atmospheric transport, deposition, and chemical reactions.

Product: The atmospheric chemistry information necessary to implement specific requirements of the DOE National Energy Strategy and to guide the progressive modification of the Strategy.

Approach: The PNL Gulfstream-1 (G-1) aircraft will be fitted with a triple quadrupole mass spectrometer to detect and quantify a number of important variables, such as NOx, NOy, DMS, acetic and formic acids, peroxyacetal nitrate (PAN), methanol, ethanol, acetaldehyde, acetone, and ammonia, in both polluted and background situations.

The Global Chemistry Model (GChM), a 3-D atmospheric-chemistry model that describes atmospheric transport, deposition, and chemical reactions on scales ranging from regional to global will be developed. Its design will allow for considerable flexibility in the addition or removal of modules describing various physical and chemical processes. The synoptic-scale meteorological fields will be defined by data from a version of the national Center for Atmospheric Research (NCAR) Community Climate Model (CCM); output fields from the CCM, at selected time intervals, will be read by the GChM and used to characterize the wind, temperature, and moisture fields. Emissions input to the model will be read from modified continental and oceanic emission inventories.

Results to Date: Representative results from this project, which came to an end in FY 1994, include:

- Wet-deposition flux of sulfur is substantially greater in most of the New England states than in many of the Ohio River Valley states.
- Approximately 20% of the precipitation events account for about 55% of the total seasonal wet deposition.
- The transport of sulfur from North America to Europe is quite small because of wet removal.
- Cloud aerosol particles are removed by scavenging more efficiently than water vapor is.
- Data collected in the 1991 Kuwait oil fire plumes was used to evaluate the diffusion and chemical-transformation rates within the plume environment.
- During the 1992 NARE field campaign, evidence of homogeneous nucleation, thought to be a source of cloud-condensation aerosol in remote regions, was collected along with measurements of dimethyl sulfide and carboxylic acids.
- Synoptic fronts were found to make a significant contribution to the import of pollutants into the Atlantic airshed, and the layering of ozone in this region was shown to be a natural phenomenon.

CHARACTERIZATIONS IN THE NORTH ATLANTIC REGIONAL EXPERIMENT

DAUM, PETER

BROOKHAVEN NATIONAL LABORATORY

Objective: To analyze data on aerosol microphysical and chemical properties collected in the North Atlantic Regional Experiment (NARE).

Product: A more specific knowledge of aerosol microphysical and chemical properties.

Approach: The horizontal and vertical distributions of aerosol number concentrations, size distributions, and chemical composition in the North Atlantic regional area will be characterized; the relationship between the sulfate mass concentration and the number concentration of cloud condensation nuclei (CCN) will be characterized as a function of supersaturation; and the relationship
between the aerosol mass and number concentrations and the number concentration and size distributions of cloud droplets formed in air containing the aerosol will be characterized in specific air masses. Analyses of data collected during the Pre-NARE program in the summer of 1992 show that these aerosol properties are a strong function of altitude and of the meteorological history of the air mass in which the measurements were conducted.

Results to Date: As expected, the composition of the aerosol samples was dominated by sulfates. The average nitrate-to-sulfate molar ratio was 0.54, and the sulfate concentration was well correlated with the accumulation-mode aerosol number density. Aerosol nitrate represented only a small fraction of the total nitrate present (28%). Although aerosol acidity was not measured, the average NH$_4^+$/SO$_4^{2-}$ molar ratio of 1.2 suggests that, on average, the aerosol was nearly as acidic as ammonium bisulfate. The generally low concentrations of sodium indicate the presence of only nominal amounts of sea salt.

FIELD STUDIES IN ATMOSPHERIC CHEMISTRY

DAUM, PETER; NEWMAN, LEONARD; and KLEINMAN, LAWRENCE

BROOKHAVEN NATIONAL LABORATORY

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10/01/93–09/30/94

Objective: To gain an understanding of the chemical and physical processes that determine the fate of energy-related air pollutants.

Product: Scientific understanding to help quantify the existing perturbation to air quality by energy-related sources and to provide information for the formulation of effective control strategies.

Approach: Field studies are conducted using a combination of aircraft and surface-based resources. Measurements typically include O$_3$, odd-nitrogen compounds, sulfur compounds, secondary photochemical pollutants, and aerosols. The strategy for use of aircraft and surface-based measurements varies. In the recently completed North Atlantic Regional Experiment (NARE), aircraft and surface sites were located at various distances downwind of the U.S. east coast to track the chemical transformations and the vertical and horizontal dispersion of pollutants as they were transported out over the North Atlantic Ocean. In a study of the Nashville urban plume to be conducted in the summer of 1995, aircraft deployment will focus on characterizing the transport and chemical transformations that occur as the Nashville plume advects downwind and mixes with the ambient atmosphere. Multiple surface sites surrounding the Nashville urban area will be arranged to provide a context within which to interpret the aircraft measurements and to estimate the effect of urban emissions on the chemistry of the background air and vice versa.

Results to Date: The first-phase analysis of the data collected during the summer 1993 NARE program has been completed. One component of this analysis was to characterize the physical and chemical properties of plumes of anthropogenic pollutants that had been transported from the intense-source regions in the northeastern United States out over the North Atlantic Ocean. Such plumes were observed as far as 1200 km from these source regions. For the most part, these plumes were observed in well-defined layers at altitudes between 0.3 and 2 km. On several days they were found to extend over distances in excess of 250 km and to be as much at 100 m thick. We have examined the synoptic meteorology and find that the presence of these plumes is associated with southwesterly flow in advance of eastward moving cyclonic systems. For this reason, the transport of pollutants from North American source regions was found to be very episodic.

Plume composition was found to be characteristic of air masses that had been mixed with substantial quantities of anthropogenic pollutants that subsequently had been extensively processed photochemically. O$_3$ concentrations as high as 150 ppbv were measured and were found to be associated with CO concentrations of up to 480 ppbv, NO$_x$ concentrations up to 20 ppbv, and accumulation-mode particle-number densities on the order of 2000 to 3000 cm$^{-3}$. Aerosol-composition measurements indicated that the aerosol was composed principally of acidic sulfate; aerosol nitrate and chloride represented only a small fraction of the mass. NO$_x$ comprised predominantly oxidation products; NO$_x$ concentrations in the plume only represented a small fraction of the total
NO3 (about 15%); t-NO3 was found to be the principal product species representing, on average, 70% of the NOy. H2O2 concentrations were quite high, averaging nearly 4 ppbv and reaching a maximum concentration in excess of 11 ppbv. The high t-NO3 and H2O2 concentrations in conjunction with high O3 concentrations indicate that the plumes had undergone extensive processing in transit from the source regions to the sampling locations and were well-aged photochemically.

MEASUREMENT OF GAS-WATER UPTAKE COEFFICIENTS FOR TRACE GASES ACTIVE IN THE MARINE ENVIRONMENT

DAVIDOVITS, PAUL
BOSTON COLLEGE
FY 1994  108
FY 1993  111
FY 1992  106
08/01/94-07/31/95

Objective: To measure air-water exchange coefficients of specific gases that are of importance in the marine atmosphere.

Product: Measurements of the interphase transport of trace gases between the gaseous and aqueous phases, which represents an important internal calculation in atmospheric chemistry models.

Approach: Measurements of uptake coefficients on aqueous droplets will be performed for reduced sulfur species, including dimethyl sulfide (DMS), hydrogen sulfide, carbon disulfide, methyl mercaptan, and their oxidation products, including carbonyl sulfide, methane sulfonic acid (MSA), dimethyl sulfoxide (DMSO), and dimethyl sulfone (DMSO2). The uptake coefficients will be measured over a sufficient range of conditions to provide both the mass-accommodation coefficient and the solubility parameters for these species. Measurements will be made as functions of temperature, pH, and ionic strength.

Results to Date: During the past three and a half years, detailed uptake studies have been completed with our droplet-train apparatus and a newly developed bubble-column apparatus for MSA, DMSO, DMSO2, DMS, H2S, CS2, CH3SH, OCS, O3, NO2, and N2O. Studies were performed as a function of temperature, pH, ionic strength (i.e., NaCl and (NH4)2SO4 concentration), and H2O2 concentration. Depending on the species studied, the results yielded mass-accommodation coefficients or Henry’s Law coefficients and the “salting out” Setchenow coefficients. The uptake of gas-phase O3, Cl2 as a function of aqueous Br+ and I- concentration and Br2 as a function of aqueous I- concentration was studied in detail. The magnitude of the measured halogen uptake and its functional dependence on ion concentration are not in accord with a simple bulk-phase reaction mechanism. The data indicate that reactions at the gas-liquid interface have a significant role in the gas-uptake process. On the other hand, the O3 uptake shows no evidence of interfacial reaction. A model for the surface reaction was developed, and the atmospheric implications of the results have been examined.
ATMOSPHERIC TRACER DEVELOPMENT AND USE

DIETZ, RUSSELL N., and SENUM, GUNNAR I.

BROOKHAVEN NATIONAL LABORATORY

FY 1994 213
FY 1993 220
FY 1992 220

10/01/93–09/30/94

Objective: To identify and use new atmospheric tracer compounds for long-range transport studies and to support these studies through the development, validation, and application of new instrumentation.

Product: The development and application of unique, highly sensitive detection methods and instrumentation to determine trace constituents in the atmosphere, specifically perfluorocarbon tracers (PFTs).

Approach: Moderate-scale (several hundreds of kilometers) atmospheric experiments using both gaseous, conservative perfluorocarbon tracers (PFTs) and the new, aerosol-particulate, water-soluble, ionic perfluorocarbon compounds (PFCs) will evaluate dry deposition of aerosol, cloud and rain scavenging, air-sea exchange (in two directions), and other processes. The technology used is also applicable to oceanic and subsurface water-transport and dispersion studies and to studies in the smaller-scale complex-terrain and very-large-scale polar-vortex and global-atmospheric regimes. Efforts will also address the development and commercialization of state-of-the-art samplers, real-time field instruments, laboratory-analysis systems, and new detector technologies.

Results to Date: Capillary-gas-chromatographic-column technology was implemented and increased the limit of detection for the PFTs by a factor of 5. This capability will significantly increase the scope of atmospheric tracer transport and dispersion experiments by decreasing the amount of PFT needed or increasing the range of deployment. The ETEx (European Tracer Experiment), an atmospheric transport and dispersion experiment with PFT release in England and sampling throughout Europe, was the first experiment to benefit from the new capillary technology.

An aerosol PFT salt has been identified and preliminarily found useful as an atmospheric aerosol tracer. This PFT salt, once sampled, can be converted to a PFT, which is then analyzed in the present laboratory GC system with a sensitivity equivalent to the gas-phase PFTs. Deployment and collection procedures have yet to be developed. The dual-trap analyzer (DTA) used in many PFT tracer atmospheric transport and dispersion experiments for semireal-time monitoring in aircraft operations is being commercially developed in cooperation with BNL.

ATMOSPHERIC CHEMICALS' SOURCES AND FATES

DOSKEY, P. V.

ARGONNE NATIONAL LABORATORY

FY 1994 194
FY 1993 200
FY 1992 200

10/01/93–09/30/94

Objective: To evaluate the fate of energy-related trace chemicals in the atmosphere in relation to their sources, chemical transformations, and processes of removal.

Product: Improved descriptions of potential changes of tropospheric pollutants, greenhouse-gas concentrations, and the tropospheric chemicals that affect the persistence and radiative properties of clouds.

Approach: Research will emphasize the assembly and analysis of data to construct mass budgets of nitrogen and sulfur species over North America and surrounding ocean areas. Data will be assembled and analyzed to integrate large-scale deposition fields in relation to both anthropogenic and natural emissions and estimation of off-continent transport. Collaboration with other participants in the ACP involves measurements with aircraft and large-scale numerical modeling. In the collaborative research, efforts by ANL centers on evaluation of air-surface-exchange measurement techniques and on parameterization of the air-sea exchange of gases.

Results to Date: Research concluded in FY 1994 on numerical modeling to develop highly efficient parameterizations of cloud microphysics processes; this effort successfully developed optimized, advanced formulas for vapor-liquid-solid interactions in cold and warm clouds. Other efforts successfully concluded in
FY 1994 included an analysis of the wet deposition of sulfur and nitrogen during 1979–1987 over the United States and Canada and modeling of atmospheric concentrations and deposition of mercury in the upper Midwest. Argonne participated in the 1993 North Atlantic Regional Experiment (NARE) and measured levels of C2−C7 nonmethane hydrocarbons (NMHCs) in canister samples collected during several aircraft flights. Total NMHC concentrations were 4 to 36 ppb of C. Olefins (mainly ethene and propene) represented 1 to 16% of the total NMHCs. Ethane, propane, i-butane, and n-butane were the predominant hydrocarbon species. These 1993 levels were similar to those measured during 1992 NARE studies, but the average olefin contribution was higher during 1993. The benzene/toluene concentration ratios were all less than unity during 1993, typical of urban air. The latter result was unexpected because the total NMHC levels and their distributions were typical of continental air that had been photochemically aged.

Results to Date: The use of cylindrical internal-reflectance spectroscopy has been evaluated for the determination of the aqueous reactions of organic oxidants and the interactions of dissolved aldehydes with peroxides, OH radical, and the HO₂ radical. During this initial investigation, we have determined that several pollutants (such as sulfate, nitrate, and a number of organics) have substantial absorbances in the infrared. As well, we have used these spectroscopic techniques to remeasure the absorption cross-sections for water. This work has led to the conclusion that longwave-radiative absorption of aerosols, fogs, and clouds cannot be ignored and is likely to counterbalance the light-scattering effects of shortwave radiation.

ACP SCIENTIFIC RESEARCH AND PROGRAM COORDINATION

HALES, JEREMY M.
ENVAIR, INC.

FY 1994 184
FY 1993 159
03/01/94–02/28/95

Objective: To provide scientific coordination for the DOE Atmospheric Chemistry Program (ACP) and to perform specific atmospheric-chemistry-modeling tasks.

Product: Coordination for the DOE ACP and atmospheric-chemistry modeling.

Approach: An annual meeting will be conducted; monthly newsletters will be issued; scientific efforts will be planned and coordinated; liaison will be provided to DOE staff, international organizations, and intragovernmental bodies; and Asian SO₂ and NOₓ emissions, transport, and transformation will be modeled.

Results to Date: ACP is now a mature program, and activities under the first objective are becoming somewhat routine. These include publication of the ACP Monthly Update newsletters, coordination of the ACP annual meeting, general planning and coordination of the composite scientific effort, and serving as a liaison resource to Headquarters staff. Tasks that have been less
routine under this objective include special activities with the World Meteorological Organization (WMO) to improve global-ozone research and, until recently, ACP coordination with the interagency CAMRAQ modeling project. An activity of particular interest under this objective is the recent development, in conjunction with other WMO participants, of a world calibration resource for ozonesonde measurements.

Progress was made on the modeling of Asian SO\textsubscript{x} and NO\textsubscript{x} emissions and their transport and transformation as they move over the Pacific Ocean toward the North American continent. During the past year, an Asian-emission inventory for SO\textsubscript{x} and NO\textsubscript{x} has been acquired, gridded meteorological fields for the spring of 1985 have been brought online, and initial simulations have been performed for air pollution over that portion of the Northern Hemisphere encompassing Eurasia, the Pacific Ocean, and western North America. Currently, these simulations are being extended to estimate the impact of observed and projected emission increases on the Asian continent.

Results to Date: The conference was held in Jackson Hole, Wyoming. Seminars were held in the following subject areas:

- Atmospheric and remote sensing
- Real-time aerosol analysis
- Trace elemental analysis
- Laser sources and spectroscopic instrumentation
- Sensors
- Novel diagnostic techniques and concepts

**PARTIAL SUPPORT FOR THE 1994 FOURTH TOPICAL MEETING ON LASER APPLICATIONS TO CHEMICAL ANALYSIS**

HENNAGE, DAVID W.

OPTICAL SOCIETY OF AMERICA

FY 1994 7


Objective: To foster communication among researchers in the broad range of fields using optical diagnostics and to provide an opportunity to commercial laser-based instrumentation companies to expand the potential applications of their products.

Product: Opportunities for scientists to discuss new developments in laser light applications in chemical analysis using new developments in optical sources, instrumentation, and spectroscopic techniques are presented.

Approach: The Department of Energy agreed to provide partial funding for the conference.

**SUPPORT FOR OPERATIONS OF THE WMO/GAW QUALITY-CONTROL/SCIENCE-ACTIVITY CENTER FOR THE AMERICAS**

HICKS, BRUCE

NATIONAL OCEANIC AND ATMOSPHERIC ADMINISTRATION

FY 1994 100

12/01/94–11/30/95

Objective: To provide, through agency collaboration, a center of excellence in the United States that would impose quality-assurance techniques on data collected by national air- and precipitation-quality networks operating in North, South, and Central America.

Product: Regional data sets compiled from observations made in different nations with a variety of sampling methods that did not share common quality guidelines.

Approach: The WMO/GAW Quality-Control/Science-Activity Center for the Americas is a formal activity of the World Meteorological Organization's Global Atmosphere Watch. The WMO has adopted a scheme in which three quality-assurance centers will deal with data intercomparability problems in three major regions of the world: North and South America, Europe and Africa, and Asia and Oceania. The QA Center for the Americas has a special focus on surface ozone, airborne radioactivity, aerosol optical depth, and precipitation chemistry. The QA center for the Americas will be funded jointly by DOE, EPA, and NOAA.

Results to Date: A major meeting of representatives of the three WMO quality-assurance centers was hosted by the QA center Europe/Africa at Garmisch-Partenkirchen,
Germany, in March 1995. At this meeting it was announced that Japan will host the third of the centers, so completing the global coverage intended by WMO.

The QA plan prepared under this project for low-level ozone was transmitted to the World Health Organization for their consideration. WHO is installing an array of ozone monitors in South America, much in line with the proposal of the WMO. The WHO stations will be mainly in populated areas; the WMO stations will be at remote locations. If present plans come to fruition, both network operations will share common quality-control and -assurance protocols, ensuring compatibility of the data obtained.

Agreement was reached to ensure the continued U.S. maintenance of the WMO Precipitation Chemistry Archive at the NOAA data center in Asheville, North Carolina.

RADON SOURCE TERM TO THE ATMOSPHERE
HUTTER, ADAM, and GOGOLAK, CARL
ENVIRONMENTAL MEASUREMENTS LABORATORY, DOE

| FY 1994 | 199 |
| FY 1993 | * |

10/01/93–09/30/94

Objective: To provide accurate radon-source-term values for global atmospheric-transport models.

Product: A global set of atmospheric radon-source-term values categorized by geographic provinces (desert, savanna, rain forest, etc.) and/or geology (clastic sedimentary, carbonaceous sedimentary, felsic igneous, etc.).

Approach: Currently, global transport models (GTM) use a single-value global-average of surface exhalation as the radon source term to the atmosphere for validation studies. We will review relevant studies appearing in the literature and compile a database. Analyses of these data will ascertain what is subsequently needed to produce a scientifically sound set of radon-source-term values defined by a dozen or so global zones. A sampling program will be implemented to fill any gaps in the existing data. Even a few such measurements would greatly improve the understanding of regional differences of the radon source term to the atmosphere.

Results to Date: A literature search and the compilation of a database of relevant studies appearing in the literature is in the final stages of completion. A field study was developed and implemented to determine the reproducibility of radon-flux measurements to the atmosphere in a homogeneous setting with the accumulation-can technique. These data will be useful in evaluating the existing data in the literature that use this technique. Additionally, these data will help direct future sampling programs attempting to determine regional differences in the radon source term to the atmosphere.

In previous years, this entry was consolidated with others under the principal investigator Merrill Heit.

TRANS-PACIFIC TRANSPORT OF COMBUSTION PRODUCTS FROM ASIA IN THE SURFACE AIR
LARSEN, RICHARD, and LEIFER, ROBERT
ENVIRONMENTAL MEASUREMENTS LABORATORY, DOE

| FY 1994 | 199 |
| FY 1993 | * |

10/01/93–09/30/94

Objective: To document current and future trends of trans-Pacific transport of combustion products from Asia and to compare surface to aircraft (G-1) measurements.

Product: A determination of the practicality of establishing pelagic-sampling sites and a database of reactive gasses (e.g., SO₂, NOₓ, and O₃); atmospheric tracers (e.g., ²²²Rn, ⁷Be, ²¹⁰Pb, nss-SO₄, and NO₃); and organic aerosols and gasses, agreed upon by ACP, at established NPACS sites.

Approach: A feasibility study will be conducted to determine if pelagic-sampling sites can be established in the North Pacific Ocean in support of NPACS. First, possible site locations will be identified at or near the existing University of Miami sites located on the Islands of Okinawa, Midway, and Shemya. Costs associated with site construction, electrical power, staffing, sam-
pling equipment, and general supplies will be evaluated. The status of current sampling and data availability at these locations will also be determined. Then, measurements of meteorological parameters and measurements (such as CN, optical absorption, and O_3) will be initiated to establish a "clean-air sector" and to determine some background aerosol and gas characteristics.

In the clean-air sector, measurements of O_3 and possibly other reactive gases (such as SO_2 and NO_x), atmospheric tracers (such as ^{222}\text{Rn}, ^{7}\text{Be}, ^{210}\text{Pb}, \text{nss-SO}_4, \text{and NO}_3), and organic aerosols and gases will be initiated at established NPACS sites, especially the Shemya site in the Aleutians, which should be close to the "footprint" of the G-1 aircraft. These measurements will complement measurements obtained with the G-1 aircraft. The long-term trends of the reactive gases and nonsea-salt sulfate, and nitrate aerosols will demonstrate the magnitude and changes of the transport of polluted Asian air masses across the North Pacific Ocean.

Results to Date: A feasibility study and literature search to determine if sampling sites can be established at islands in, or coastal areas near, the North Pacific Ocean in support of the Northeastern Pacific Air Chemistry Study (NPACS) was completed. Of nine possible sites identified, four (Cheeka Peak, Midway, Shemya, and Okinawa) may be suitable for measuring and documenting Asiatic pollutants traversing the North Pacific Ocean. It was determined that the measurements of O_3, nonsea-salt sulfate, nitrate, trace elements, and atmospheric tracers, such as CO, ^{222}\text{Rn}, ^{7}\text{Be}, and ^{210}\text{Pb}, would be suitable to accomplish program goals. Several existing programs were also identified that could provide useful information to the NPACS. Deployment of instruments and initiation of measurements was delayed because the NPACS was indefinitely postponed.

\*In previous years, this entry was consolidated with others under the principal investigator Merrill Heit.

MULTIPHASE ATMOSPHERIC CHEMISTRY

LEE, YIN NAN, and ZHOU, XIANLIANG

BROOKHAVEN NATIONAL LABORATORY

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10/01/93–09/30/94

Objective: To obtain an increased understanding of the distribution, transformation, and fate of atmospheric organic compounds and to obtain a better understanding of the involvement of organic compounds in atmospheric nitrogen and oxidant cycles.

Product: An understanding of the atmospheric photo-oxidation pathways of organic compounds to assess their importance in regional oxidant formation (and therefore acid deposition), possible health and phytotoxic effects, and global atmospheric chemistry.

Approach: Atmospheric waters, such as hydrometeors (cloud and fog) and surface water (ocean and lake), are important considerations in effecting the transformation, distribution, and removal of key atmospheric trace species. Aqueous chemical and physical properties of important gas-phase constituents will be quantitatively determined in the laboratory, including equilibrium constants, solubilities, and rate constants and their dependencies on pH and temperature. Efforts will focus initially on O_3, oxides of nitrogen (NO_x, PAN, and HNO_3), and reduced sulfur compounds (DMS and OCS). The information thus determined will be used to evaluate the contributions of atmospheric gas-liquid interactions to atmospheric ozone concentrations, the deposition velocity of O_3 to surface water, the oxidation of reduced sulfur compounds, and the transformation and distribution of nitrogen compounds.

Results to Date: Methylglyoxal (MG), a product of hydrocarbon (HC) oxidation and a precursor of PAN, is highly soluble in water. A study of the aqueous photolysis of MG showed that formic and acetic acids were produced in equimolar amounts at 1:1 stoichiometry against MG decay. The yield of pyruvic acid (PD) was <5%, and no H_2O_2 and peroxyacetic acid was found, consistent with a homolytic fission of MG followed by reaction with H_2O. Neither OH nor HO_2 was a major chain carrier. The photolysis by sunlight (summer noon, 42^\circ N) was -0.02 M min^{-1} at 10 M MG, marginally important for removing ambient MG; the rate of formation of formic and acetic acids, -0.03 ppb hr^{-1} (at LWC = 1 g/m^3), seems negligible. An aircraft technique for FA, MG, PD, glycolaldehyde (GA), glyoxal (GL), and glyoxylic acid (GD) was deployed during the 1993 NARE intensive operation in Nova Scotia, Canada. [FA] ranged between 0.6 and 5.5 ppb and correlated well with accumulation-mode particles. Although decreasing with increasing altitude, [FA] occasionally reached 2 ppb above 2 km, indicating possible HC precursors at those altitudes. Ground
measurements near the Gulf of Maine showed carbonyl patterns distinct from those aloft, consistent with the notion that the site was influenced by the cold marine inversion. Further, PD and GD were at times decoupled from FA and GA and may have major biogenic sources. Finally, FA measured during the Mauna Loa Observatory Photochemistry Experiment II was interpreted with a simple box model. The values calculated with measured CH₄, NO, and CH₃O₂H were in fair agreement with the observed medians (i.e., 0.14, 0.12, 0.16, and 0.17 ppb in autumn, winter, summer, and spring, respectively). CH₃O₂H serves as a major precursor of FA under extremely low NO concentrations.

EML SAMPLE ARCHIVES

LEIFER, ROBERT
ENVIRONMENTAL MEASUREMENTS
LABORATORY, DOE

FY 1994  
FY 1993  *

10/01/93–09/30/94

Objective: To provide a computer database of the analyses of samples from the following programs:

- Project ASHCAN (balloon) radioactivity measurements from 1957 to 1983
- Project STARDUST (aircraft) radioactivity measurements from 1957 to 1967
- Project AIRSTREAM (aircraft) radioactivity measurements from 1967 to 1983
- Project SASP (surface) radioactivity measurements from 1957 to 1993
- Strontium-90 deposition program from 1954 to 1993
- Soil samples collected from 1953 to 1990
- Diet and bone samples collected from 1969 to 1982
- Sediment samples collected from 1979 to 1991

and to provide a detailed listing of the availability of archived samples at EML for the above-mentioned programs.

Product: A complete database incorporating all the published data from the subject programs and a complete characterization of EML’s archived samples.

Approach: The state of all EML archived samples will be assessed. A method of storing and retrieving these archived samples will be established; this would include a controlled environment and easy access to the samples. A database will be developed containing information on the availability of all EML archived samples for future analysis. The best method of storing the data from EML’s various sampling programs for retrieval by the scientific community will be evaluated. The information from the upper-tropospheric and lower-stratospheric filter and gas samples collected from 1957 through 1967, presently available only in technical reports, will be entered into a computerized database. Specifically, the data from the ASHCAN and STARDUST programs from the years 1957 to 1967 must be transferred from published reports into a computer file. Since 1967, both AIRSTREAM and ASHCAN filter data are stored in a database on a VAX as is Project SASP data. The resulting database will contain all the data on radioactivity presently available at EML from the major U.S. atmospheric-sampling programs from the 1950s to the 1990s. Then, the information from EML’s soil, sediment, water, diet, and bone programs (currently available only in technical reports) will be entered into a database.

Results to Date: A series of spreadsheet templates were written to allow the efficient entry of information from reports containing radionuclide data. More than 10,000 lines of data were entered into the database. Each line can contain up to 20 variables. A quality-assurance document was written to provide a guideline for ensuring that the final database will be of the highest quality. This procedure includes the verification of all data being entered as well as a detailed review of the reports to uncover any written corrections or conflicts that may have occurred subsequent to the first publication of the data.

*In previous years, this entry was consolidated with others under the principal investigator Merrill Heit.
AEROSOL CHARACTERIZATION ON GULFSTREAM AIRCRAFT SAMPLES

LEIFER, ROBERT, and KROMIDAS, LAMBROS
ENVIRONMENTAL MEASUREMENTS LABORATORY, DOE

FY 1994 399
FY 1993 *
10/01/93–09/30/94

Objective: To characterize the continental and marine air intercepted during the Gulfstream (G-1) flights in the North Pacific Ocean campaign and to measure the $^{10}\text{Be}/^{7}\text{Be}$ ratio of air collected during G-1 flights.

Product: An independent indication of whether air sampled during the G-1 flights is truly marine or of continental origin and whether the air parcel intercepted during G-1 flights has air of stratospheric origin.

Approach: A new rotating-drum impactor designed specifically for use with scanning electron microscopy (SEM) will be tested and evaluated at close to aircraft altitudes. Samples would be analyzed with SEM and x-ray analysis that employ a special stage automation package (SAP) to provide automatically size-distribution information and elemental characterization. The use of coated impactor surfaces that would react with impacted aerosol and produce unique, stable signatures for future identification using scanning electron microscopy and x-ray detection would be explored. A similar procedure would be investigated for identifying carbonaceous material.

Filter samples from EML’s archives will be used to obtain a baseline for establishing the tropospheric and stratospheric ratios of $^{10}\text{Be}/^{7}\text{Be}$. The feasibility of installing a new filter sampler with a flow rate a factor of 10 higher than that of the current sampler will be investigated. And accelerator mass spectroscopy will be used to analyze for $^{10}\text{Be}$.

Results to Date: Changes were made in the SAP hardware and software to control the SEM stage containing our sample. Using the newly designed parallel impactor drums, we were able to move quickly from one aerosol collection to another and provide an objective analysis of the size distribution and chemical composition of the sample. Additional software needs to be written and modification of the hardware accomplished before we meet our full objectives of a fully automated system.

We have used our atmospheric filter archives to further explore the variability of the $^{10}\text{Be}/^{7}\text{Be}$ ratio in stratospheric air. A set of 20 filters were sent to LLNL for $^{10}\text{Be}$ analysis. These filters were selected to provide a vertical picture of the ratio in both the high- and low-latitude stratosphere. A couple of filters show contradictory results, and we are exploring the possible reasons for the discrepancy.

In previous years, this entry was consolidated with others under the principal investigator Merrill Heit.

ATMOSPHERE-CANOPY INTERACTIONS

LINDBERG, STEVEN E.

OAK RIDGE NATIONAL LABORATORY

FY 1994 194
FY 1993 200
FY 1992 200
10/01/93–09/30/94

Objective: To quantify rates of wet and dry removal processes and surface emission rates of both natural and pollutant constituents important to atmospheric chemistry and to biogeochemical cycling in forests.

Product: A verified model and database capable of describing the fate of both natural and pollutant constituents in forest canopies.

Approach: The rates and mechanisms of deposition to the forest canopy of trace elements involving vapor phase reactions in coal combustion emissions will be investigated to improve our understanding of the atmosphere-landscape portion of the biogeochemical cycles of Hg, Se, and As through studies of the roles of wet and dry deposition of the important chemical species of these elements. The work will initially emphasize Hg because of existing expertise, and will involve measurement of concentration gradients at the Walker Branch site, development of a dry-deposition model from canopy-resistance approaches, and analysis of wet deposition and its interaction with the forest canopy. This research will capitalize on recently developed analytical capabilities for picogram-level analysis and chemical
speciation. We have obtained cofunding from the Electric Power Research Institute to expand these studies with the application of chamber and micrometeorological methods to both deposition and emission measurements at high- and low-elevation sites. This work will be a joint effort with researchers at the National Oceanic and Atmospheric Administration (NOAA) Atmospheric Turbulence and Diffusion Division (ATDD) lab and the Swedish Environmental Research Institute in Göteborg.

An approach for scaling up air/surface exchange measurements will be developed based on surface analysis methods. A model will be developed to analyze throughfall (TF) beneath forests which separates net TF (TF flux minus rain flux) into its component parts of foliar leaching and dry-deposition washoff. The model will require single-event TF data collected over one season. The model will be applied to data from two sites: a low-elevation loblolly pine plantation near Oak Ridge, Tennessee (300-m elevation), and a high-elevation red spruce stand in the Great Smoky Mountains National Park (1740 m). Precipitation will be collected in forest clearings and beneath the canopy as wetfall—only on an event basis. Replicate TF collectors will be used to establish the spatial variability of net TF fluxes within the stands. Six permanent collectors will be established at the pine site, and two at the remote mountain site. Additional intensive collection grids of 20 collectors will be used for selected events to determine the representativeness of the permanent sampling points. The sites will include a meteorological tower extending approximately 10 m above the forest canopy and will be instrumented with standard meteorological sensors for continuous recording.

Total deposition and canopy exchange will be measured in a high-elevation spruce canopy, and edge effects and gap depictions will be quantified. This task will measure total flux of atmospheric and internal materials above and below the canopy, in the same fashion as done earlier for the deciduous canopy to address the question of the role of dry deposition in controlling the chemistry of throughfall. The methods will be applied to a newly established spruce stand at 1740-m elevation in the Great Smoky Mountains National Park. A meteorological tower has been established. The new site is considerably removed from the influence of the local DOE and Tennessee Valley Authority (TVA) coal-fired power plants and consists of a typical, heterogeneous spruce canopy consisting of numerous gaps and edges.

The chemistry of fog and cloud water will be studied in high and low elevation forests by collecting fog and cloud water at the new loblolly pine site and at the new spruce site in the Smoky Mountains for the purpose of comparing the chemistry of these solutions and, ultimately, the fluxes to each forest determined by a resistance model. Fog and cloud droplets will be sampled with a combination of passive harp-type and active impaction-type collectors operated above the canopy.

Results to Date: A new technique has been developed to measure reactive gas-phase water-soluble Hg\textsuperscript{2+} species by using a high-flow refluxing mist chamber. Extensive testing, including attempts to eliminate artifact Hg\textsuperscript{2+}, was carried out. Measurements taken at two locations (in Oak Ridge, Tennessee, and near the Ohio-Indiana border) during one year found approximately 0.055\% of the total gaseous mercury. Initial tests with ozone oxidation and cosampled aerosols to measure artifact Hg\textsuperscript{2+} production in the mist chamber suggest that the majority of Hg\textsuperscript{2+} collected exists in the ambient air in the gas phase. These measurements have significant implications for the control, transport, and fate of mercury emitted from combustion sources because the measurements represent the very first positive identification of a reactive, water-soluble, gaseous, mercury species in ambient air. This species is highly reactive and will deposit very rapidly on land and water surfaces, unlike elemental mercury vapor, which is the dominant form of mercury in air and is relatively less reactive.

ATMOSPHERIC AEROSOL MICROPHYSICS: FORMATION, CHARACTERIZATION, AND INTERACTION

MARLOW, WILLIAM H.

TEXAS ENGINEERING EXPERIMENT STATION/TEXAS A&M

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Objective: To develop the theoretical and mathematical foundations for describing the formation and metamor-
phosis of ultrafine aerosol particles (i.e., those up to 50 nm in radius).

**Product:** A reduction in the uncertainty associated with the modeling of the effects of air pollution.

**Approach:** The roles of equilibrium vapor pressure and kinetics in water condensation on an aerosol will be studied. One case that will be investigated is that of a pair of adhering spheres (as in aerosols produced by fires). Where the spheres are in contact, the water-air interface may well have a negative overall curvature (i.e., it is concave), thus enhancing condensation.

To explain the aerosol activation spectrum under transient supersaturation conditions (such as occur in cloud nucleation), the background vapor pressure will be coupled to the overall thermodynamic conditions (vapor depletion, latent heat release, etc.) and the particle equilibrium vapor pressure.

Because of the following considerations, accurate, computationally practical schemes for the determination of these interaction energies will be developed.

- The interaction energies of particles are important (1) for the determination of their collisional growth rates, (2) possibly for the determination of the shapes of aggregates, and (3) for the evolution of their shape under condensation-evaporation cycles.

- The interaction energy of a condensing fluid with a condensation substrate determines the shape of the condensed fluid.

- That shape largely determines the equilibrium vapor pressure over the particle.

Particular attention will be paid to the approach that starts from the Lifshitz–van der Waals theory for long-range intermolecular forces of a continuum or condensed matter.

**Results to Date:** A new computational approach for single-particle condensation accounting for time-dependent vapor depletion and heat of condensation has been developed. Based on this algorithm, a model has been constructed of condensational evolution of aerosols that are polydisperse in both composition and size. This model has been used to study cloud-condensation nucleation of background atmospheric aerosols. Early results of these computations explain the observed broadening of the initial, condensationally activated cloud-aerosol size distribution relative to the original aerosol prior to its entry to the cloud. In addition, calculations of stochastic condensation, arising from random variation of the cloud supersaturation, have been performed to model realistic clouds, and the results show additional broadening of the cloud-base-aerosol size distribution.

In a fundamental research effort intended to improve understanding of new-particle formation, molecular-dynamics studies of atomic cluster collisions have shown the onset of aerosol behavior as cluster size increases beyond four or five monomers per cluster. Though atmospheric clustering generally involves molecular clusters, these results are assisting the interpretation of experimental data on atmospheric nucleation and its laboratory counterparts in which the high sticking efficiency of molecular clusters of these sizes was uncertain.

**ULTRAFINE-AEROSOL SIZE DISTRIBUTIONS AND SULFURIC ACID VAPOR PRESSURES: IMPLICATIONS FOR NEW PARTICLE FORMATION IN THE ATMOSPHERE**

McMURRY, PETER H.

UNIVERSITY OF MINNESOTA

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**Objective:** To determine production rates of new sulfuric acid particles by homogeneous nucleation.

**Product:** An improved understanding of the relationship between atmospheric emissions (especially aerosol precursor gases) and atmospheric-aerosol size distributions.

**Approach:** The experimental program will focus on two key measurements. The first of these involves measurement of the vapor pressure of sulfuric acid at temperatures and relative humidities relevant to the atmosphere. Vapor pressures will be inferred from measured evaporation rates of monodisperse ultrafine (5- to 50-nm-diam) sulfuric acid droplets. The tandem differential mobility analyzer (TDMA) technique will be used for these measurements. This method is especially suitable for mea-
suring ultralow vapor pressures and has been used successfully with other species. The sulfuric acid vapor pressure is a key variable for determining nucleation and growth rates.

The second key measurement pertains to the size distribution of ultrafine aerosols. The ultrafine aerosol condensation nucleus counter (UFACNC) of Stolzenburg and McMurry will be used for these measurements. Proposed measurement techniques involve measuring the UFACNC pulse-height distribution as a function of particle size and varying the size-dependent cut point of the instrument by adjusting its condenser temperature. The ultrafine aerosol-particle distributions thus obtained will provide important information on rates of homogeneous nucleation.

Results to Date: The technique and analysis of vapor-pressure measurements have been improved, and many measurements have been made. Precise measurements of \( \text{H}_2\text{SO}_4 \) vapor pressures by the TDMA technique still show a large amount of unexplained scatter. Contamination by atmospheric ammonia is suspected of being the primary sources of error. Nevertheless, our data support the validity of the Ayers et al. values.

The MICRON aerosol-inversion method is being adapted to the ultrafine-aerosol data and has successfully inverted test data from the 1992 Mauna Loa field experiment and the 1993 field study at Idaho Hill, Colorado. An important part of size-distribution inversion from raw data is the determination of the pulse heights for a calibration aerosol. These pulse-height spectra become the kernel for the numerical inversion of experimental data, which yield estimates of the actual size distribution. We have found that the kernel spectra are dependent on the composition of the calibration aerosol. Kernel functions have been measured for ammonium sulfate and sulfuric acid aerosols (they are similar to each other) and are being used for data inversion because they should be good surrogates for particles produced by atmospheric nucleation. The dependence of kernel functions on pressure from 0.25 to 1.0 atm is also being measured.

The inversion method is fairly elaborate, so a simpler method with lower resolution has also been used to analyze the Mauna Loa and Idaho Hill data. With measurements of particles in the 2.7- to 4.0-nm-diam range and simultaneous measurements of gas-phase sulfuric acid, particle-formation rates were estimated as a function of \([\text{H}_2\text{SO}_4]\). Much higher rates of nucleation were observed than would be predicted by the conventional theory. Work is continuing to explain the results.

A new multiple-detector ultrafine-CNC (MD-UCNC) was developed that uses four condenser temperatures to obtain information about ultrafine-particle size distributions. Comparisons of the ultrafine size distributions measured with this instrument with those from the pulse-height-analysis method were disappointing because of large uncertainties in the flow through the instrument. Methods to remedy this shortcoming are under study.

DEVELOPMENT OF REAL-TIME LOW-LEVEL RADON ANALYZER

NEGRO, VINCENT, and BECK, HAROLD

ENVIRONMENTAL MEASUREMENTS LABORATORY, DOE

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10/01/93–09/30/94

Objective: To develop a compact, real-time, radon/radon progeny monitor capable of making precise measurements of atmospheric radon with a time resolution better than 10 min and a lower limit of detection of less than 10 pCi/m³.

Product: A functional, lightweight, compact, prototype instrument capable of measuring either radon concentrations directly or concentrations of \(^{218}\text{Po}\) at the required sensitivity.

Approach: Several approaches will be investigated for measuring \(^{222}\text{Rn}\) or \(^{218}\text{Po}\) directly in real time, including:

- Adaptations of present techniques which sample air at high rates through a filter, automatically move the filter to a detector, and count while a second sample is obtained, or count the sample simultaneously with sampling.
- New concepts that use electrostatic fields to collect \(^{218}\text{Po}\) directly on a solid-state detector.
Ionization chamber-filter combinations that allow larger volumes of air to be sampled while simultaneously detecting the alpha emissions of the collected radon progeny.

All approaches would use microcomputer technology for instrument control, data logging, and data analysis.

Results to Date: A prototype instrument capable of measuring charged $^{218}$Po on the G-1 aircraft has been designed, constructed, and tested. The principle of operation is to create a strong electrostatic field that collects positively charged $^{218}$Po atoms on a solid-state alpha detector. Their subsequent decay is detected, and the concentration of charged $^{218}$Po can be determined. The unit can sample air up to approximately 50 cfm, which enhances the sensitivity. Because of the short half-life of $^{218}$Po (3.05 min), its concentration should be in close equilibrium with that of its parent $^{222}$Rn, especially in the free atmosphere. Although not all of the $^{218}$Po is charged, the charged fraction in the free troposphere should be relatively high except under conditions of high absolute humidity. The analyzer appears to be extremely responsive, and the desired sensitivity should be achieved. The instrument is currently being customized for installation on the G-1 aircraft.

Results to Date: Reactions of $\text{SO}_2$ with liquid water in the dispersed and bulk states were studied in the presence and absence of gaseous ammonia. Fast oxidation occurs in the laboratory cloud in the presence of ammonia but not in the presence of ammonia with water entirely in the vapor phase. The amount of sulfate produced depends critically on the cloud liquid-water content but not on the type of aerosol used to nucleate cloud droplets. The results suggest surface-chemical processes take place at or near the liquid-gas interface. We have used the same experimental system to study the reaction of $\text{NO}_2$ with dispersed and bulk-liquid water. The reaction proceeds by an apparent first-order reaction at conditions corresponding to the phase-mixed reaction regime. A possible synergism between $\text{HNO}_3$ and the uptake and oxidation of $\text{NO}_2$ in cloud droplets was observed. Experiments were also conducted on hygroscopic properties of carbon-black and diesel-soot aerosols with and without chemical modification. The nucleation ability of chemically modified carbonaceous particles increased with increasing soluble mass fraction and was comparable to that of sulfate salts when the soluble mass fraction exceeded about 10%.

Objective: To perform laboratory simulations of aqueous and gas-particle chemical reactions relevant to the formation and removal of energy-related pollutants.

Product: An understanding of the relationships between cloud and aerosol microphysical properties and heterogeneous (aqueous and gas-particle) reactions and scavenging of carbonaceous aerosols.
PARTIAL SUPPORT OF THE FY 1994 COMMITTEE ON ENVIRONMENT AND NATURAL RESOURCES (CENR) AND SUBCOMMITTEE ON AIR QUALITY (SAQ)

PENNELL, WILLIAM
PACIFIC NORTHWEST LABORATORY
FY 1994 103
10/01/93–09/30/94

Objective: To provide technical assistance to the chairperson of the Subcommittee on Air Quality (SAQ) of the Committee on Environment and Natural Resources (CENR)/National Science and Technology Council (NSTC).

Product: Technical support to the chair and cochairs of the Air Quality Subcommittee of the CENR and the NSTC.

Approach: The Department of Energy’s Office of Health and Environmental Research is providing in-kind support of the CENR/SAQ infrastructure through detailee activities from Pacific Northwest Laboratory. This includes providing assistance to the SAQ Science Chair on an as-needed basis.

Results to Date: New project.

PARTIAL SUPPORT FOR THE INTERNATIONAL GLOBAL ATMOSPHERIC CHEMISTRY CORE PROJECT OFFICE

PRINN, R. G., and PSZENNY, A. A. P.
MASSACHUSETTS INSTITUTE OF TECHNOLOGY
FY 1994 22
06/15/94–06/14/95

Objective: To provide scientific coordination and administration of the International Global Atmospheric Chemistry (IGAC) Core Project of the International Geosphere-Biosphere Program (IGBP).

Product: Planning and the publication of research results for the IGAC.

Approach: The Scientific Steering Committee and the scientific activities of IGAC will be coordinated. Descriptions of the ongoing efforts and future plans of each of the IGAC activities will be developed and subsequently updated. An international information and communication center will be provided for IGAC. Cooperation will be fostered between IGAC’s cosponsors and between IGAC and its related IGBP Core Projects, other ICSU-sponsored global-change research programs, and other relevant international bodies.

Results to Date: New project.

1994 SEVENTH BOC PRIESTLEY CONFERENCE

ROOT, CHARLES A.
AMERICAN CHEMICAL SOCIETY
FY 1994 7
05/01/94–04/30/95

Objective: To address the technical themes of oxidative fate of atmospheric pollutants, photochemical smog and ozone, atmospheric ozone, and global tropospheric ozone.

Product: A deeper insight into the chemical processes of the Earth’s atmosphere and how these phenomena relate to the chemistry of the atmosphere with regard to oxidants and oxidation processes.

Approach: Presenters from the domestic and European chemical and/or atmospheric science communities will discuss the selected topics.

Results to Date: Four technical sessions were held during a three-day session at the Seventh BOC Priestley Conference held at Bucknell University from June 24 to June 27, 1994. They addressed, respectively, the oxidative fate of atmospheric pollutants, photochemical smog and ozone, stratospheric ozone, and global tropospheric ozone. A final session was conducted on the early history of ozone.
AEROSOL CHEMISTRY AND DYNAMICS
TANG, IGNATIUS N.

BROOKHAVEN NATIONAL LABORATORY

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Objective: To elucidate the chemical and physical processes governing the formation and dynamic behavior of ambient aerosols.

Product: A fuller knowledge about the phase transformation, droplet growth, and chemical reaction of atmospheric aerosols.

Approach: The hygroscopic and optical properties of mixed aerosol particles will be investigated with the single-particle-levitation technique developed at Brookhaven. In addition, critical supersaturations for activation will be measured in a continuous-flow cloud chamber for laboratory-generated cloud condensation nuclei (CCN) with known organic coatings. A systematic evaluation of the results and pertinent thermodynamic considerations will be made to understand heterogeneous nucleation and water-vapor condensation and evaporation on mixed aerosol particles. Gas-particle interactions of atmospheric importance will also be studied with the single-particle Raman spectroscopic technique that has been under development at Brookhaven for the past several years.

Results to Date: Experiments were carried out to measure the water activities, densities, and refractive indices of aqueous-solution droplets containing single salts of sulfates and nitrates, which are common constituents of atmospheric aerosols. The extensive data should find application in mathematical models for predicting the dynamic behavior, visibility reduction, and radiative effects of atmospheric sulfate and nitrate aerosols.

In addition, the hydration behavior of Na$_2$SO$_4$ particles was investigated in detail. The results illustrate the various phase transformations and metastable states that hygroscopic particles may undergo under the changing humidity conditions of the ambient atmosphere. The observed hydration properties, often not predicted by the bulk-phase-solution thermodynamics, are unique to micro particles. More work is in progress to elucidate the chemical and physical states of atmospheric aerosol particles.

ATMOSPHERIC PEROXIDES
WEINSTEIN-LLOYD, JUDITH B.

STATE UNIVERSITY OF NEW YORK/COLLEGE AT OLD WESTBURY

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Objective: To determine the aqueous-phase free-radical chemistry, where we currently have only a threshold understanding.

Product: Additional scientific understanding of the general area of aqueous-phase chemistry.

Approach: The occurrence and behavior of gaseous and aqueous-phase peroxides and their free-radical precursors (HO$_2$ and RO$_2$) will be investigated. A newly developed continuous detection method will be applied to speciation of organic and inorganic peroxides in the environment. High-performance liquid chromatography will be used to separate peroxide samples obtained from scrubbed air. Aqueous peroxy radicals and peroxides at representative ambient concentrations will be produced with low-intensity $^{60}$Co gamma radiolysis, enabling us to study reaction rates and formation mechanisms.

Results to Date: Peroxides play a major role in cloud-water acidification and act as a reservoir for the free radicals that are central to tropospheric ozone formation. Yet, there are still relatively few reliable determinations of ambient concentrations of the most abundant atmospheric peroxides, hydrogen peroxide, methyl hydroperoxide (MHP), and hydroxymethyl hydroperoxide (HMHP). We have developed a method for continuous analysis of hydrogen peroxide, MHP, and HMHP based on the reaction between H$_2$O$_2$ and Fe(II) (Fenton's reagent) to form hydroxyl radical. The latter is scavenged by benzoic acid to produce fluorescent hydroxybenzoic acid isomers. Varying the pH of sampling solution and using the well-known $p$-hydroxyphenylacetic acid reagent to determine total
peroxides allows us to discriminate between individual peroxides.

We deployed the Fenton method in ground-based field measurements in Georgia during the 1992 intensive of the Southern Oxidant Study and aboard DOE's G-1 aircraft during the 1993 North Atlantic Regional Experiment. The results have been used to test models of atmospheric photochemistry, and the diurnal profiles we observed have shed light on the mechanism of formation of H$_2$O$_2$ and organic peroxides.

**DRY AIR-SURFACE EXCHANGE**

**WESELY, M. L.**

**ARGONNE NATIONAL LABORATORY**

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10/01/93–09/30/94

**Objective:** To evaluate the exchange of trace chemicals in dry form between the atmosphere and natural surfaces in continental areas.

**Product:** An estimate of the role of dry deposition in the mass budgets of energy-related substances over North America.

**Approach:** This project will consist of field measurements, theoretical studies, and parameterization of dry-air-surface exchange rates. The results will be applied to estimate dry deposition at specifically designed network measurement stations with site-specific models and over large areas with regional- and global-scale models. For specific sites, an inferential technique will be used to compute weekly amounts of the dry deposition of sulfur dioxide, nitric acid vapor, sulfate and nitrate in fine particles, and ozone. The inferential approach will be used because existing reliable techniques to measure dry deposition directly are too complex for routine, continuous implementation, and the inferential approach requires data only on local meteorological and surface conditions and mean concentrations of the substances of interest. For regional- and global-scale models, a computer-coded module will provide estimates of the dry-deposition velocities for several selected inorganic and organic substances. Experimental and theoretical work will be conducted on measurement of air-surface exchange of organic substances, in addition to inorganic sulfur and nitrogen compounds and ozone because some organic substances are important in the tropospheric chemistry of energy-related trace substances. Processes of both emission and deposition of selected organic substances will be described mathematically in a form suitable for use in large-scale numerical models.

**Results to Date:** The Argonne dry-deposition module was successfully coupled with remotely sensed data on surface spectral reflectances to model dry deposition over large geographic regions. Numerical modeling of rapid in-air chemical reactions involving NO, NO$_2$, and O$_3$ has shown that NO emitted from soils can rapidly react with O$_3$ to produce NO$_2$ that can be removed moderately quickly by uptake by vegetation during the daytime before the NO$_2$ mixes to the upper portions of the atmospheric boundary layer. Analysis of data collected in 1992 and 1993 on the air-surface exchange of volatile organic compounds in a prairie ecosystem showed that a plot dominated by crown vetch and bluegrass exhibited a decrease in emissions of total non-methane organic compounds (NMOCs) from June 1992 to October 1992. Oxygenated hydrocarbons and terpenes composed about 90% and 10% of the identified NMOC emissions, respectively. Analysis of turbulence flux data collected during past experiments has shown that the usefulness of eddy accumulation with conditional sampling is limited by (1) a variation in a crucial, empirically derived numerical coefficient over deep canopies as compared to short vegetative canopies and (2) the highly demanding relative accuracies needed for the chemical analyses.
As the primary component of DOE's atmospheric-dynamics research, Atmospheric Studies in Complex Terrain (ASCOT) supports the Environmental Science Division’s goals to improve the understanding of:

- The emissions and processes that could potentially change global climate and
- The transport, dispersion, and chemistry of energy-related air pollutants.

ASCOT is a multilaboratory program investigating mesoscale processes in the atmosphere by field experiments, mathematical modeling, and theoretical research.

**Research Objectives.** ASCOT’s objectives are to:

- Improve fundamental knowledge of the atmospheric dynamics and air-pollution-dispersion processes in complex terrain,
- Incorporate this knowledge in the development of tools for air-quality assessments, and
- Apply these tools to atmospheric-dispersion problems associated with emergency response to the release of hazardous materials.

**Research Questions.** Differential heating and cooling of the boundary layer over variously exposed surfaces drive local and regional circulations that change unpredictably throughout the day and night. These circulations interact with or are protected from the overlying synoptic wind fields, depending on terrain and meteorological conditions. The description and forecasting of these evolving complex flow patterns and the dispersion of contaminants released within them are highly uncertain and constitute the current scientific focus within the ASCOT program.

A balance is maintained in ASCOT between a primary focus on basic science and a relevance to DOE mission needs. The central scientific emphasis on evolving energy transfers in flow-field development, terrain influences on mechanical and thermal forcing, and the scale interactions between circulations from local to synoptic scales must be pursued with the intent of ultimately serving the long-term needs of DOE and the country. The program maintains an awareness of needs in emergency preparedness, energy-related-air-pollution concerns, and climate-change studies and modeling.

During the past few years, ASCOT has focused its research on areas in which DOE facilities are located (i.e., in the Tennessee Valley near Oak Ridge and in Colorado’s Front Range region near the Rocky Flats Plant). Studies of the flow patterns in the Tennessee Valley have examined the relationship of local valley winds to large-scale wind patterns and have investigated methods of characterizing the spatial variability of the winds with a limited number of observation sites. Such studies are relevant to the prediction of plume transport and diffusion in areas of complex terrain and should be useful in emergency-preparedness applications.

In the Front Range, ASCOT carried out a three-month experiment during August–October of 1993. Arrays of remote-sensing instruments, including sodars and wind profilers, were used to obtain measurements of the winds in and from canyons and over the plains in the vicinity of DOE’s Rocky Flats Plant. The data are being used to study the interactions of regional and local flows and their impact on the meteorology of the Plant. Analyses
of tracer releases made in 1991 are continuing, and advanced computational tools are being developed to describe atmospheric dynamics, transport, and dispersion with improved accuracy.

ASCOT is also using radar wind profilers at other DOE laboratory sites to explore features of the local meteorology and to improve predictions of weather conditions that may affect site operations. Efforts have also begun to provide stronger links between the boundary-layer observations, analysis, and modeling activities of the ASCOT program and the research conducted by DOE’s Atmospheric Chemistry Program, with joint investigations under development for studies in the southeastern and northeastern United States.

Future studies will continue to develop closer ties with the ongoing activities of atmospheric chemists. The importance of a fuller understanding and representation of boundary-layer phenomena for a complete description of atmospheric chemical processes is being increasingly acknowledged, and expertise developed under the ASCOT program can be readily applied to this area. The ASCOT program itself will be phased out during the next two years, to be replaced by a new program focused on the meteorological processes affecting air quality in the southwestern United States. This program will build on the base of knowledge gained during ASCOT’s history while turning the emphasis of DOE’s Atmospheric Dynamics program to an emerging area of concern. Air-quality problems in the southwest are closely tied to energy development and use and clearly fall within the DOE mandate for research.

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Research Summaries

ATMOSPHERIC TRANSPORT OF CONTAMINANTS

ADDIS, ROBERT P.

WESTINGHOUSE SAVANNAH RIVER COMPANY

Objective: To evaluate and improve mesoscale modeling of atmospheric flow and contaminant transport for emergency-response applications.

Product: An understanding of the limitations and improvement in the capabilities of mesoscale atmospheric transport models in emergency-response applications.

Approach: The applicability of mesoscale atmospheric models in emergency response depends on the accuracy of contaminant transport prediction, operational functionality, reliability, and computational speed. This project will evaluate the meteorological and transport predictions made by the Colorado State University (CSU) mesoscale Regional Atmospheric Modeling System (RAMS) and a Lagrangian particle dispersion model (LPDM). These models will be used to calculate the wind field and contaminant transport for idealized topography and at both Rocky Flats and the Savannah River Site (SRS). The modeling studies at Rocky Flats will be performed in conjunction with the ASCOT 1991 winter field experiments. Model results will be compared with the predictions of several diagnostic models. The research will evaluate the effects of subgrid-scale parameterizations, surface characteristics, and 4-D data acquisition.

Results to Date: 3-D numerical simulations of drainage flows for several idealized valley-tributary systems have been performed. The valley-tributary systems examined include: (1) a valley-tributary system cut into a plateau and draining onto a broad plain, (2) a valley-tributary system cut into a plateau and draining into another valley, and (3) a valley-tributary system surrounded by a ridge top and draining onto a plain. Along-valley mass flux and tributary-flow dynamics were examined for all geometries. Tributary tracer dispersion was simulated with a Lagrangian particle model for system (1). Model tracer results were in good qualitative agreement with field measurements from the ASCOT studies in Brush Creek.

The atmosphere over the front range of the Colorado Rocky Mountains was simulated with the CSU RAMS. Simulations were performed with a nested grid model with an innermost grid spacing of 330 m centered at Rocky Flats. The model successfully predicted the mesobeta-scale drainage flows from the Front Range into the South Platte River Basin. However, significant errors occurred in the surface drainage flows adjacent to the foothills. As expected, the 4-D data-assimilation technique reduced the overall errors in the predicted flow fields and tracer transport.

ATMOSPHERIC DIFFUSION IN COMPLEX TERRAIN

ALLWINE, K. JERRY

PACIFIC NORTHWEST LABORATORY

Objective: To identify the forcing mechanisms responsible for observed wind and temperature structures in complex terrain and to develop, test, and apply numerical models that describe these mechanisms and their effects on the transport and dispersion of atmospheric pollutants.

Product: An understanding of the physical processes governing the transport and diffusion of atmospheric pollutants in complex terrain and the description of these processes with appropriate numerical or conceptual models.

Approach: Field data and numerical-modeling results will be used to identify mechanisms responsible for the
development and structure of wind and temperature fields in regions of complex terrain. The field and numerical-simulation results will be integrated to develop and test the ability to describe and predict wind and temperature structure and the behavior of pollutants in such environments. Numerical models will be refined and optimized to produce efficient, accurate, and reliable codes for research and application needs.

Results to Date: No results reported.

ATMOSPHERIC BOUNDARY LAYER TRANSPORT AND DISPERSION STUDIES
COULTER, R. L., and WESELY, M. L.
ARGONNE NATIONAL LABORATORY

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10/01/93–9/30/94

Objective: To examine the nature of atmospheric flow over complex terrain, particularly over sites where the future construction of power-production or fuel-conversion facilities is being planned and where the potential exists for release of toxic materials to the atmosphere, so that improved estimates can be made of the rates at which airborne pollutants are diluted, removed by dry and wet processes, and delivered to sensitive receptors.

Product: Improved air-pollution and emergency-response models that are required to plan and manage energy-generation and fuel-conversion facilities.

Approach: To study transport over complex and nonuniform terrain, this research group will participate in large, multiorganizational field experiments.

Experimental studies will be conducted with remote-sensing equipment, such as Doppler acoustic sounders and laser anemometers, and with direct-sensing devices deployed at the surface or carried aloft by free or tethered balloons. Advanced acoustic sounder systems will allow evaluation of the vertical structure of the mean and turbulent, 3-D wind components in the lower atmosphere without the use of instruments in towers. Transport and diffusion processes will be described mathematically, and these descriptions will be tested against experimental data.

Results to Date: Analysis of data relating drainage winds from Coal Creek Canyon to surface net radiation and external forcing found that, when external winds were less than 3 m/s, the drainage flow was markedly different between strong and moderate cooling conditions. The drainage depth and height of the wind speed maximum were maximum for external winds near 3 m/s. And in strong-cooling, light-external-wind conditions, there is a maximum in turbulence above the surface because of airflow-surface interactions.

The central hub for the ASCOT data network was established. Data were archived from the network as well as from the PROFs network and the RFP tower. Past data from the RFP tower since October 1990 has also been made available on the hub. Data from the extended network that is being installed in the area in August will be available on the hub on the Internet.

ASCOT PROGRAM SCIENTIFIC DIRECTION
DORAN, J. CHRISTOPHER
PACIFIC NORTHWEST LABORATORY

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10/01/93–09/30/94

Objective: To provide scientific coordination among the primary DOE laboratories participating in the Atmospheric Studies in Complex Terrain (ASCOT) program and with supporting NOAA laboratories and university groups.

Product: Focused, common objectives and complementary approaches to the individual research of the contributing scientists from the primary laboratories and supporting research groups.

Approach: A scientific focus will be defined for the program, representing the consolidated interests of the participants and some of the most urgent challenges to the scientific community and at the same time identifying and linking DOE’s mission-related needs with the scientific outputs. Emphasis will be directed
toward providing results important to emergency-response needs. The project will maintain this focus for ASCOT through planning with representatives of the participating laboratories, strengthening the organizational components of the program, promoting collaboration and exchange among the scientists involved, enhancing direct and frequent communications with participants and with DOE, and promoting technology transfer to user groups within DOE and elsewhere.

Results to Date: A meeting was organized for all ASCOT scientists in February 1994 in Salt Lake City, Utah. Participants presented results of their recent studies on the Front Range region of Colorado in the vicinity of the Rocky Flats Plant and related studies on other complex-terrain meteorological topics. In May, a meeting of the Science Planning Group was held in Germantown to discuss future directions for the ASCOT program or a new program. Increased participation in ozone-related research and a possible new research initiative on the meteorology of the southwestern United States and its relationship to air-quality issues there were suggested. Starting in FY 1995, funding of ASCOT research was to be based on peer-reviewed competitive proposals submitted by scientists from various federal laboratories. The solicitation for proposals had been sent out in June of 1993, and a panel of 13 reviewers drawn from universities, industry, and non-DOE federal agencies was recruited and assembled to review the proposals. The panel met in December of 1993 and evaluated the proposals; those evaluations were used as the basis for funding recommendations for future ASCOT work. Approximately $2.6 M was identified for allocation to competitively based research proposals out of requests totaling more than $7 million.

DIRECT SIMULATION OF ATMOSPHERIC TURBULENCE

DORAN, J. CHRISTOPHER
PACIFIC NORTHWEST LABORATORY

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10/01/93–09/30/94

Objective: To simulate the dynamics and turbulent properties of the planetary boundary layer (PBL) in an effort to identify and characterize the dominant mechanisms that determine the behavior of the PBL.

Product: Increased understanding of geophysical fluid turbulence and the structure and characteristics of the PBL, particularly in regions of complex terrain, and improved predictive capability for the description of transport and mixing in turbulent atmospheric layers.

Approach: Advanced computational models will be used to study the effects of nonlinear dynamics and turbulence on the properties of the PBL in complex terrain. Models will be used to describe the mean and turbulent motions responsible for PBL characteristics. New and improved models of turbulence in stably stratified layers will be developed, and improved parameterization schemes will be incorporated for processes like turbulent exchange and surface energy balances in existing mesoscale models.

- One or more numerical mesoscale models will be applied to simulate the structure and evolution of the PBL in areas of interest to DOE missions. Numerical experiments will be carried out to identify the primary physical mechanisms responsible for unusual or anomalous features of local and regional circulations and atmospheric stratification.
- The properties of the PBL will be related to the behavior of atmospheric pollutants released into the environment and compared with observations where possible.
- Through collaboration with the university researchers, a full-turbulence simulation (FTS) model of flows over an inclined surface in stably stratified flows will be developed.
- The results of the FTS model will be applied to a search for improved representation of turbulence in stable layers in mesoscale models, incorporating features such as wave-turbulence interactions.
- Modules will be developed and incorporated that improve the representation of turbulent exchange and other physical mechanisms in available numerical models.

Results to Date: FY 1994 marks the end of the Direct Simulation of Atmospheric Turbulence (DSAT) project. DSAT has supported studies (observational, analytical, and numerical-modeling) covering a variety of regions
ranging from Tennessee to Colorado to Washington. The project has provided funds for tracer studies, mass-balance studies, and measurements of wind and temperature profiles during ASCOT field campaigns. Its area of concentration, however, has been in the use of numerical models to investigate the interactions of ambient winds and local circulations generated by thermal and topographical forcing mechanisms. In this area, it has examined the development of the deep nocturnal boundary layer found above the Roan Plateau in Colorado and the role of shear (between the low-level thermally driven flows and the large-scale overlying wind field) that enhances turbulent mixing of the cold surface layer air upward to form a deep mesoscale boundary layer.

DSAT has analyzed the influence of synoptic winds above the ridgetops of the Tennessee Valley on the wind fields near the valley floor. Pressure-driven channeling, in which the winds below the ridgeline are driven by the along-valley pressure gradient component and that can result in valley winds that blow in opposition to the along-valley wind direction components above the valley, was found to be a major factor in this region. As part of a joint study with another ASCOT project, DSAT not only demonstrated the complexity of the factors influencing valley wind fields but also provided an approach toward developing practical forecasting guidelines for these difficult-to-predict winds.

DSAT obtained new insights into the mechanisms responsible for the regional drainage winds that occur regularly over the Hanford Site in Washington during the spring and summer months. Such winds become significant when a thermal low east of the Cascade Range draws the cooler air west of the Cascade Range across the mountains. The study again demonstrated the ability of advanced numerical models to contribute to both basic and applied meteorological research needs. DSAT's most recent focus on the wind fields over the RFP in Colorado and their dependence on local thermal and terrain features continued this approach.

A principal finding of the DSAT project is that it is often unwise to make definitive statements about the expected behavior of wind and temperature features and dispersion meteorology in an area of complex terrain without careful analysis and study. An early goal of the ASCOT program was to achieve transferability (i.e., the ability to transfer readily what was learned in one location to another location). The reality of the situation has turned out to be more complicated. Although much has been learned about flows in complex terrain and many useful guidelines have been developed, it is now also clear that reliance on guidelines without supplemental studies can often be badly misleading. In this sense, transferability has proved to be elusive. Nonetheless, the development of a suite of tools, including sophisticated numerical modeling techniques, has at least provided the means to carry out such supplemental investigations far more efficiently and completely than was previously possible. During its lifetime DSAT has played a significant role in demonstrating this approach in its treatment of a variety of interesting and important problems.

SUPPORT FOR COMPLEX-TERRAIN STUDIES

GUDIKSEN, PAUL H.

LAWRENCE LIVERMORE NATIONAL LABORATORY

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10/01/93–09/30/94

Objective: To measure meteorological variables in support of the Atmospheric Studies in Complex Terrain (ASCOT) Program and to develop and evaluate numerical models that are capable of faithfully simulating the physical processes responsible for pollutant dispersion in areas of complex terrain.

Product: Evaluated models of pollutant dispersion processes associated with terrain-dominated flows.

Approach: Our meteorological studies in western Colorado that involve investigating the effects of the ambient meteorology on the wind and temperature characteristics of nocturnal drainage flows in a mountain valley will be completed. Assistance will be provided in the transfer of the ASCOT field studies program to the Rocky Flats area. A network of meteorological towers will be installed and operated to support climatological studies in the Rocky Flats area. Climatological data will be analyzed to define the frequency of occurrence of nocturnal drainage flows in the Rocky Flats area and to define their temporal and spatial wind and temperature characteristics. Diagnostic model simulations will be
performed on the winter 1991 tracer experiments. ASCOT data and results of our model simulations will be exchanged with Rocky Flats personnel to assist their emergency-preparedness program.

The field measurements, data analysis, and model applications and support provided by this task will be closely integrated with the ASCOT model development activities to ensure that the field data acquired are useful for understanding the physics of the terrain-dominated flows under study as well as to provide the data needed for model-validation efforts. These activities have been well coordinated with other ASCOT program participants.

Results to Date: The continuously operating meteorological-tower network, consisting of ten 18-m dual-level instrumented towers, is situated within the Front Range of the Rocky Mountains and is over the adjacent plain. This network has produced extensive measurements of the surface wind, temperature, and net-radiation characteristics of the flows over this complex-terrain area. The resulting data, which currently reside in the ASCOT data bank, are being used by the ASCOT scientists to improve our understanding of the drainage-flow characteristics over the Rocky Flats area.

Replacement of the gradient-diffusion scheme in the ADPIC particle-dispersion model with the random-displacement (RDM) stochastic-diffusion model is currently under way in an attempt to achieve more physically meaningful simulations of pollutant dispersions and the elimination of the need for a numerical-grid mesh in the diffusion calculations.

To foster technology exchange, an international panel of experts concerned with defining the uncertainties associated with the atmospheric-dispersion aspects of accident-consequence analysis was participated in. The approach put forward involved the MATHEW/ADPIC models and a fast-running sequential-puff model. Statistical sampling of the model input parameters within their respective ranges of uncertainty, numerous calculations with the puff model, and normalization of the sequential-puff-model results to the MATHEW/ADPIC-model results led to the derivation of model uncertainties for a variety of accident conditions. The results revealed that the uncertainties of the median downwind concentrations for a range of atmospheric stabilities were typically within a factor of 3, whereas the 5% and 95% quartiles of the concentration distributions generally ranged over a factor of 30.

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**ATMOSPHERIC TURBULENCE AND DIFFUSION RESEARCH**

**HOSKER, R. P., Jr.**

**ATMOSPHERIC TURBULENCE AND DIFFUSION DIVISION, NOAA**

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**Objective:** To (1) develop better methods of describing atmospheric transport, diffusion, and air-surface exchange (including deposition) in numerical simulations; (2) extend the applicability of these techniques to realistic situations, such as nonstationary conditions, complex terrain, and dense vegetation; and (3) develop and test improved numerical models incorporating these new methods.

**Product:** The development of predictive capabilities that use the results of this research.

**Approach:** Dispersion and planetary boundary layer studies (C. J. Nappo, Jr.) will extend contemporary understanding of transport and dispersion in the lower atmosphere to nonideal conditions and will provide improved methods for measuring atmospheric-plume dispersion rates and for including realistic dispersion parameters in numerical models. Activities will focus on the roles of wave/turbulence interaction and sporadic nocturnal turbulence as mechanisms for accelerating dispersion at night and on the consequences of releases of dense and/or chemically reactive pollutants.

The research on canopy micrometeorology (D. D. Baldocchi) will identify and quantify the interactions involved in above- and within-canopy transport, dispersion, and deposition in canopy-atmosphere exchanges of materials and energy. Such information will be used to include and/or parameterize physical and biological surface effects in numerical models ranging from local to global scales.

**Results to Date:** Analysis continues of wind measurements acquired with the extensive Oak Ridge Site Survey tower network during 1989-1991. Wavelet transform theory was applied to a study of turbulent
breakdowns of the stable nocturnal boundary layer. A workshop on turbulence and diffusion in the stable planetary boundary layer was organized and held at Arizona State University. Collaborative work continued with the University of Uppsala (Sweden) on parameterizations of gravity-wave stress in mesoscale models. Surface flux data and airsonde and tower-based vertical-profile data obtained during the 1992 DOE-ARM BARFEX study in Oregon were analyzed and simulated with a modified second-order-closure model to study intrafield variability of scalar fluxes across a transition from desert to an irrigated field. Several surface-energy-balance models of increasing complexity were also compared and tested against the 1992 flux data. An analytical solution for coupled leaf-photosynthesis and stomatal-conductance models was devised, and the spatial and temporal nature of solar radiation within heterogeneous plant canopies was studied. Scaling carbon dioxide and water-vapor exchange from leaf to canopy scales in a deciduous forest was analyzed as was modeling mutual interactions of carbon dioxide, forests, and climate. Work began on organizing an international workshop on strategies for long-term studies of carbon dioxide and water-vapor fluxes over terrestrial ecosystems.

**COMPLEX TOPOGRAPHY (ASCOT)**

HOSKER, R. P., Jr.

ATMOSPHERIC TURBULENCE AND DIFFUSION DIVISION, NOAA

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Objective: (1) To design, plan, and conduct with other participating ASCOT laboratories, field experiments investigating mesoscale circulations in areas of complex terrain and the dispersion of contaminants released within them; (2) to analyze meteorological and turbulence data and tracer concentrations measured in ASCOT experiments to improve understanding of the characteristics of flow and dispersion in complex terrain; and (3) to develop and evaluate numerical models of flow and dispersion in drainage flows in complex terrain.

Product: Databases characterizing complex airflow and dispersion over nonuniform terrain under varying conditions of thermal and mechanical forcing and influences of multiscale interactions among local, regional, and synoptic flow fields; useful physical concepts and hypotheses that have been tested with these databases; and validated numerical models of flow and dispersion in complex terrain that can be used for research, parameterization studies, safety and risk assessment, and regulatory applications.

Approach: Tower networks will continuously detect near-surface variables and gradients that, together with turbulent-flux instrumentation and radiometers, provide surface energy budgets and other exchanges between the atmosphere and surface as well as their spatial and temporal variabilities. Concentration and meteorological data will be edited and analyzed to provide information to execute numerical models and to test them. The data analyses will be explained in terms of tracer release height, topography, winds, synoptic conditions, etc. Transport and dispersion models suitable for use in complex terrain will be developed and tested with the data sets. A variety of statistical techniques will be applied to quantify the ability of the models to predict winds, turbulence characteristics, and/or concentrations, as appropriate.

Results to Date: A hydrostatic mesoscale model was implemented and used to simulate wind fields in the eastern Tennessee River valley. A regional tower network was initiated in East Tennessee, to provide additional data to test the model predictions. Analysis continued of vertical tethered-balloon profiles of wind speed, direction, and temperature over Oak Ridge. Statistical analyses of the 1990 Oak Ridge ASCOT data also continued. A Lagrangian stochastic dispersion model was used to simulate tracer data from the 1991 Rocky Flats ASCOT study.

**TERRAIN-INFLUENCED METEOROLOGY**

LEE, J. T., and BOSSERT, JAMES

LOS ALAMOS NATIONAL LABORATORY

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10/01/93-09/30/94
Objective: To develop and verify improved estimation techniques for atmospheric transport and diffusion in sites of generalized topography.

Product: A fundamental understanding of atmospheric transport and diffusion in sites of generalized topography leading to improved models for atmospheric transport and diffusion calculations.

Approach: The terrain-influenced meteorology program will be formulated in a balanced approach among field experiments, laboratory simulations, and numerical modeling. The physics is vital in identifying important phenomena to be measured and modeled and in interpreting the results of the experiments. The approach will place an emphasis on the fundamentals of transport and diffusion in complex terrain. In practice, field measurements will be emphasized with the primary thrust in intensive experiments designed around clearly defined objectives. A high ratio of interpretation effort to data gathering will be maintained to explore as fully as possible the important phenomena governing the transport and dispersion at a particular site. The models that are implemented will have a similar emphasis on identifying and qualifying important complex-terrain phenomena. Once an understanding is achieved of the processes governing the transport in terrain-dominated airflow and its relation to general meteorological conditions, the parameterization of these processes for incorporation into operational air quality models will be more systematic and reliable.

To simulate pollutant diffusion and transport over complex terrain, accurate estimates must be made of the spatial distribution and temporal development of winds and turbulence. A 3-D mesoscale numerical model has demonstrated considerable ability to simulate geophysical fluid flows under quite general boundary conditions. The model equations are time dependent and three-dimensional in space so that temporal and spatial variations of temperature stratification are included. The model has been applied to obtain predictions of mesoscale wind fields for various large-scale weather conditions.

Results to Date: The Terrain-Influenced Meteorology Program at Los Alamos National Laboratory ended in FY 1994 when it was replaced by two new ASCOT programs. During the final year of this project, we concentrated on modeling and data analysis of the meteorological and tracer-release field experiments conducted in the vicinity of the Rocky Flats Plant in the winter of 1991. We have used the Regional Atmospheric Modeling System (RAMS) prognostic model and the ATMOS 1 and 2 diagnostic wind-field and plume-dispersion model to simulate the four nighttime tracer-release experiments. Both models were successful in reproducing the results of the experiments. Maximum ground-level concentrations were generally within a factor of 2 of the observed results and located radially within approximately 45° azimuth of the observed location. The prognostic-model results were obtained by using only one sounding 2 hr before the tracer release to initialize the model, whereas the diagnostic-model results used all of the meteorological data taken throughout the tracer-release experiment. The fact that the prognostic-model results are almost as good as the diagnostic model’s represents a significant advance in modeling of winds and plume dispersion in complex terrain.

MESOSCALE EMERGENCY RESPONSE
LEONE, J. M., and GUDIKSEN, P. H.
LAWRENCE LIVERMORE NATIONAL LABORATORY

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10/01/93–09/30/94

Objective: To develop and evaluate the LLNL diagnostic and finite-element-based dynamic models of the atmosphere to improve and enhance their ability to simulate flow and dispersion over complex terrain.

Product: Evaluated models of pollutant-dispersion processes associated with terrain-dominated flows.

Approach: The diffusion module will be improved in the MATHEW/ADPIC diagnostic models that currently form the core of the Atmospheric Release Advisory Capability (ARAC) emergency-response service. The initialization and turbulence-parameterization schemes will be improved in the finite-element-based boundary-layer models. FEM-PBL, the nonhydrostatic model, will be applied within the ASCOT experimental areas for improving the understanding of valley circulations. Development of SABLE, a hydrostatic mesoscale boundary-layer model that is expected to be suitable for emergency-response calculations, will be continued. The
ADPIC model will be coupled with SABLE for subsequent inclusion in the ARAC service.

Because the ASCOT program is currently focusing its research resources on problems associated with DOE's emergency-preparedness responsibilities, the plan is to continue these model-development efforts and to augment them with the development of a hydrostatic-boundary-layer model that may be suitable for emergency-response forecasting. This development effort will also provide the framework needed to define the conditions under which the simpler hydrostatic assumption may be adequate for describing the dispersion processes.

Results to Date: To overcome known deficiencies of the ARAC diagnostic wind field models, we developed the WINDGEN code, which rapidly solves the interpolation/adjustment problem via a superposition of pseudopole fields on 2-D surfaces. The resultant wind field is not only mass consistent, but has preserved the observations and is defined at every point in the domain.

Also, we completed the development and initial validation of a 3-D random displacement model (RDM) of turbulent diffusion within ADPIC. Compared to the hybrid Eulerian-Lagrangian scheme, the RDM showed improved accuracy and eliminated the need to parameterize the close-in, subgrid-resolution portion of the calculation.

In addition, we conducted simulations of drainage flow within an idealized valley with a number of tributary configurations using SABLE. These simulations revealed that although tributaries influence the details of the drainage flow within the main valley, their effects decrease rapidly with downstream distance. Also, tributaries have little effect upon total mass flux draining from the main valley unless the tributary drains a basin not available to the main valley.

Finally, we participated in the ASCOT Model comparison workshop using both the diagnostic wind field model MATHEW paired with the ADPIC dispersion model and the prognostic model SABLE paired with ADPIC.

INTERACTIONS BETWEEN SURFACE EXCHANGE PROCESSES AND ATMOSPHERIC CIRCULATIONS

SHAW, WILLIAM I.

PACIFIC NORTHWEST LABORATORY

FY 1994 145
FY 1993 150
FY 1992 150

10/01/93–09/30/94

Objective: (1) To evaluate systematically, through the use of advanced numerical models, the effects of variable surface fluxes of heat, momentum, and moisture over heterogeneous surfaces on the local and regional circulations in the planetary boundary layer; (2) to establish techniques for measuring and describing appropriate, representative values of these fluxes for use in parametric representations of their effects; (3) to examine possible feedbacks that boundary-layer properties may have on the surface characteristics; and (4) to study the diurnal and seasonal variability in these processes.

Product: An improved understanding of the interactions among surface fluxes of heat, momentum, and moisture and the characteristics of the atmospheric boundary layer.

Approach: Improved models for describing air-surface exchanges of momentum and sensible and latent heat will be acquired or developed, and these descriptions will be incorporated in existing mesoscale computer codes. Latent and sensible heat fluxes over semiarid terrain will be measured. This task will examine questions of measurement techniques; representativeness of flux values; and the use of observed, interpolated, and extrapolated fluxes as boundary conditions for mesoscale models.

Results to Date: Energy-balance and turbulence-flux data were collected in 1992 in a mature shrub-steppe environment located 2 km east of the HMS on the Hanford Site. This plant community is very patchy with distinct areas (<10 m²) of either bare soil or cheatgrass mixed with mature sagebrush. Measurements of fluxes of heat, moisture, and momentum were made at a single point in the surface layer, while measurements of canopy energetics and soil heat and moisture budgets
were made over each of the three surface elements (sagebrush, cheatgrass, and bare sandy soil). We evaluated two 1-D models: GAPS (general-purpose atmosphere-plant-soil simulator, developed at Cornell University) and SHAW (simultaneous heat and water, developed by the Agricultural Research Service). The evaluation was directed primarily at examining their algorithms for simulating the energy balance at the land surface. Our intent was to compare simulated soil temperature, soil heat flux, and net radiation with field measurements for each of the three surface types. Simulations have been completed for bare soil and sagebrush surface types. Model calibration was achieved by adjusting the soil thermal conductivity, and there is good agreement between bare soil surface observed and predicted values. The agreement is not as good between the predicted soil surface under the sagebrush canopy with near-surface thermocouples placed under the sagebrush canopy. There is a general underprediction of the lower soil temperature. This may be corrected by adjusting the leaf area index (LAI), which controls the amount of radiation reaching the soil surface. This adjustment in LAI is further supported by the current model predictions of higher leaf temperature as compared to measured leaf temperatures.

**COUPLING/DECOUPLING OF LOCAL AND SYNOPTIC CIRCULATIONS**

WHITEMAN, C. DAVID

PACIFIC NORTHWEST LABORATORY

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10/01/93–09/30/94

Objective: To develop an improved understanding of physical mechanisms leading to interactions between thermally driven, local wind systems and the ambient flows.

**Product:** Improved atmospheric transport and diffusion models capable of predictions in complex terrain for use in emergency response and environmental assessment.

**Approach:** A better understanding will be gained of the basic physics, the driving forces, and the interactions of thermally developed complex-terrain circulations of different scales. These complicated interactions represent significant challenges that will be met with an integrated program of new field investigations, data analysis from past complex-terrain experiments, and mathematical modeling of relevant physical mechanisms. Field investigations will make use of remote and in situ atmospheric-sounding devices, radiation- and energy-budget measurement systems, tracer samplers, and basic meteorological sensors. Data sets will come from ASCOT and other complex-terrain experiments, and simulations will come from a variety of meteorological models, from simple thermodynamic to full-physics numerical models.

**Results to Date:** Using climatological analyses of Doppler radar data from the western United States and a dynamical model, we discovered indications of a global, semi-diurnal, tidal, wind system in the troposphere.

A Monte Carlo technique was developed to calculate uncertainties in atmospheric heat budgets measured by triangular arrays of radar wind profilers and acoustic sounding systems. A new technique was developed to quantify atmospheric stagnations, recirculations, and ventilations for air-pollution-dispersion investigations. The horizontal variability of atmospheric turbulence in a complex-terrain area was investigated with data from a network of towers in the Colorado Plateaus Basin. And air-pollution-dispersion technology developed in the ASCOT program was transferred to the U.S. Forest Service to deal with pesticide-dispersion problems in national forests.
OZONE PROJECT

Over the past two decades, DOE ozone research has focused on the polluted regional troposphere. More recently, DOE ozone research efforts have expanded, in response to the National Energy Policy Act, to include the global free-troposphere and the midlatitude, lower stratosphere.

Research Objectives. The objectives of the DOE Ozone Project are to:

- Improve estimates of ozone and UV-B trends;
- Improve the understanding of the chemical and dynamic processes controlling ozone formation and destruction; and
- Develop improved predictions of future ozone concentrations and their climatic interactions.

These objectives follow from specific DOE information requirements [an identified subset of the research and evaluation needs delineated in the Intergovernmental Panel on Climate Change (IPCC), World Meteorological Organization (WMO), and United Nations Environment Programme (UNEP) science assessments], are being met by means of strong interaction with other federal agencies and international organizations, and are intended to maximize programmatic benefit to policy analysts.

Such activities include: (1) augmentation and quality assurance of ozone and UV-B monitoring networks (particularly by the WMO Global Atmospheric Watch); (2) systematic reevaluation and interpretation of the historical ozone measurements; (3) specific research on chemical mechanisms for ozone formation and destruction (sensitivity/uncertainty evaluation of existing kinetics parameterizations and determination of previously undiscovered and unquantified mechanisms); (4) evaluations of the joint action of transport and chemistry in determining the state of midlatitude, lower-stratospheric ozone through global models and other diagnostic tools; and (5) advanced instrumentation development for lower stratospheric chemical measurements.

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Research Summaries

OZONE PRODUCTION: A FIELD STUDY TO EVALUATE TROPOSPHERIC PHOTOCHEMICAL MECHANISMS

BERKOWITZ, C. M.
PACIFIC NORTHWEST LABORATORY
FY 1994 400
10/01/93–09/30/94

Objective: To provide measurements in support of testing postulated photochemical production mechanisms for ozone.

Product: A series of open literature publications that report on these measurements.

Approach: A combined modeling/field observation program will be pursued. The modeling effort will use a coupled mesoscale dynamic/chemistry model to investigate postulated mechanisms leading to the extensive lower tropospheric layering of trace gases detected east of North America during the NARE 1992 and 1993 field campaigns. Surface monitoring stations had not previously detected these layers of high ozone concentrations. ASCOT scientists will provide a wind profiler and analytical and modeling support to describe the process by which trace gases above and within the surface mixed layer interact as the mixed layer deepens during the day. One aircraft sampling program will make a series of predawn/sunrise measurements to evaluate the effect pollutants above and upwind of Nashville, Tennessee, have on surface ozone concentrations later in the day. To be measured are NOx, hydrocarbons, ozone, and a variety of meteorological parameters. A second campaign has three components:

- Airborne sampling for chlorine, bromine, aerosols, and radon over the western North Atlantic as part of an instrument-evaluation study related to ozone production
- Under westerly flow, a set of predawn through early-morning measurements within and above the boundary layer between New York and Boston of NOx, ozone, hydrocarbons, and related species
- Under westerly flow, airborne NOx and hydrocarbon measurements east of Boston for a pilot study to evaluate emissions inventories

This research is being done in conjunction with the Southern Oxidants Study (SOS), the North American Research Strategy on Tropospheric Ozone (NARSTO), the University of California/Irvine, Brookhaven National Laboratory, the Environmental Measurements Laboratory, Battelle Columbus Laboratories, and DOE's ASCOT Program.

Results to Date: Critical subcontracts have been put in place: one to investigate the reactions between chlorine-containing aerosols, such as seasalt, and nitrogen oxides and another to develop (1) methods for using a tandem-quadrupole mass spectrometer for measuring chloro-nitrogen oxide concentrations in the atmosphere and (2) advanced sensors for nitrogen oxides, peroxyacetyl nitrate (PAN), and volatile organic compounds. The reactions of chlorine-containing aerosols may compete with those involving hydroxyl reactions with volatile organic compounds and hence may affect the photochemical production rates of ozone. The equipment will be used in future Atmospheric Chemistry Program field studies.

GLOBAL 3-D MODELING OF ATMOSPHERIC OZONE IN THE FREE TROPOSPHERE AND THE STRATOSPHERE WITH EMPHASIS ON MIDLATITUDE REGIONS

BRASSEUR, GUY; ERICKSON, DAVID; TIE, XUEXI; and WALTERS, STACY
NATIONAL CENTER FOR ATMOSPHERIC RESEARCH
FY 1994 107
07/01/94–06/30/95

Purpose: To use global chemical-transport models to study the chemical and dynamical processes that affect midlatitude stratospheric ozone and to quantify the budget of tropospheric ozone.

Product: An estimation of (1) the dilution of air masses processed by polar stratospheric clouds inside the polar
vortex during winter and their impact on ozone at mid-latitudes; (2) the destruction of ozone via heterogeneous reactions on the surface of aerosol particles at all latitudes; (3) the photochemical production and destruction of \( O_3 \) in the free troposphere; (4) the export of tropospheric ozone from "polluted" to remote regions; and (5) the cross-tropopause exchanges of \( O_3 \) and other species.

**Approach:** Four models will be improved and used: (1) a new version of our 2-D chemical-radiative-dynamical model with coupled microphysical processes involving sulfate aerosols and polar stratospheric clouds (PSCs); (2) the stratospheric version of a 3-D chemical-transport model (CTM-stra) with relatively high-horizontal resolution coupled with a microphysical model of PSCs; (3) the tropospheric version of a 3-D chemical-transport model (CTM-TROP) with detailed surface emissions and hydrocarbon reactions; and (4) the intermediate model for the global and annual evolution of species (IMAGES) with a detailed chemical reaction scheme but with relatively low resolution. Model results will be compared with available data.

**Results to Date:** A coupled chemical-radiative-dynamical 2-D model was used to assess the response of stratospheric ozone to the injection of sulfur following the eruption of Mt. Pinatubo. We have investigated the relative importance of dynamical, radiative, and chemical perturbations in the atmosphere. Special emphasis has been put on the role of heterogeneous chemical-conversion mechanisms.

We have implemented in our 2-D model a chemical scheme describing the methane-oxidation chain in the troposphere and have estimated the change in the tropospheric ozone abundance from a doubling in the methane concentration.

We have completed the development of the IMAGES model, which describes the 3-D distribution of approximately 50 chemical compounds from the surface to 50 mb. The model has been used to assess the impact of aircraft emissions on tropospheric ozone and to investigate potential causes for the recently observed decrease in CO abundances.

We have developed a 3-D chemical transport model driven by the dynamics of the NCAR CCM2. The model includes gas-phase and heterogeneous chemical reactions and accounts for the formation of PSCs. Model results show the importance of the dilution of ozone-depleted Antarctic air for the evolution of midlatitude ozone in the Southern Hemisphere.

**EVALUATION OF ULTRAVIOLET RADIATION, OZONE, AND AEROSOL INTERACTIONS IN THE TROPOSPHERE WITH AUTOMATIC DIFFERENTIATION**

**CARMICHAEL, GREGORY R., and POTRA, FLORIAN**

**UNIVERSITY OF IOWA**

**FY 1994** 110

07/01/94–06/30/95

**Objective:** To investigate the interactions among ozone, aerosols, and ultraviolet radiation, including the flux of UV-B radiation at the Earth's surface as a function of changes in ozone and aerosols in the stratosphere and troposphere and the effects of changes in solar actinic flux on the photochemical oxidant cycle of the troposphere.

**Product:** A combined modeling system for studying these interactions that includes comprehensive sensitivity analysis and scientific insights into these interactions.

**Approach:** A detailed radiation model will be combined with a 3-D atmospheric chemistry model. These models will be joined with automatic-differentiation software to enable desired sensitivities to be calculated online with the radiation/chemistry computations.

**Results to Date:** UV-B fluxes and photolysis rates under a variety of conditions reflecting changes in stratospheric and tropospheric ozone have been calculated. A 16% reduction was found in total ozone, representative of stratospheric ozone conditions in the year 2004, and decreased ground-level ozone concentrations across the eastern United States in the summer. This finding is opposite that of previous box-model studies. The differences are attributed to transport processes and highlight the importance of using detailed models that include chemistry and transport mechanisms.

Automatic differentiation was successfully applied to the calculation of initial condition and parameter sensitivities (rate constants) for ozone production. We demonstrated that the model ADIFOR can be used for
these applications. We have also been investigating in detail the relationship between ADIFOR results and the numerical integration procedures used in the radiation-chemistry model, a key issue with ADIFOR because each atmospheric model uses different numerical schemes. Even if ADIFOR is able to calculate gradients up to machine precision, the accuracy of the sensitivities is comparable to the errors in the numerical solution of the initial problem. We have come up with some interesting results that are both of practical and theoretical importance. We have shown that applying forward automatic differentiation on a fixed-step-size QSSA chemical integrator is equivalent to solving the variational equations.

INVESTIGATING THE UMKEHR OZONE PROFILE RECORD

DELUISI, JOHN
NOAA AIR RESOURCES LABORATORY
FY 1994 108
7/1/94–6/30/95

Objective: To improve the Umkehr data record, including ancillary information on errors caused by aerosols, and to provide a well-documented climatology of the past and current state of ozone.

Product: A history of stratospheric aerosol size distribution and a record of long-term trends in stratospheric aerosols.

Approach: An observationally consistent archive of stratospheric aerosol data will be developed and extended. SAGE I and II aerosol data, ground-based lidar data, and sun-photometer data will be used to create a long-term climatology of stratospheric aerosol properties dating back to 1958. Also, an empirical time-dependent model of stratospheric aerosol characteristics will be developed.

Results to Date: The stratospheric-aerosol property parameters that have been studied include the latitudinal and temporal distribution of the aerosol optical thickness, aerosol size distribution, refractive indices, and aerosol extinction profiles. SAGE data have been received, and information from literature searches of past related work have been digitized.

Mie scattering coefficients for the Legendre polynomial expansion have been assessed, and are now in the process of being compared against the Dave's radiative-transfer computer codes. SRRB scientists are comparing the results of their complete spherical radiative-transfer models with the results from the Dave's code. Standardized ozone profiles, aerosol profiles, standardized aerosol size distribution, and refractive indices that will be used for this test have already been agreed upon.

New and old ozone profile retrieval, Mie scattering, radiative transfer, and ozone absorption calculation algorithms have been obtained. High-resolution extraterrestrial-UV flux data have been gathered, and several stratospheric aerosol data sets have been acquired, consisting of lidar and sunphotometer data.

The two ozone-profile retrieval algorithms were subjected to different levels of random error to ascertain their performance characteristics and differences. Theoretical tests of trend detection and determination of the noise effect on the average solution in the new and old algorithms were also performed.

A model of stratospheric ozone corrections was developed to perform an error analysis on Umkehr ozone retrievals with SAGE data for the period when SAGE was in operation. It will be used as a base for further checking the aerosol-correction procedures.

LABORATORY STUDIES OF THE SENSITIVITY OF TROPOSPHERIC OZONE TO THE CHEMISTRY OF SEA-SALT AEROSOL

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FY 1994 160
FY 1993 240
09/15/94–09/14/95

Objective: To examine sea salt as a possible significant source of tropospheric halogen (chlorine and bromine) radicals through heterogeneous reaction with nitrogen oxides.

Product: An improved understanding of sea salt as well as salts from other sources (such as alkaline volcanoes
and oil-well burning) as sources of halogen radicals that may affect tropospheric and stratospheric ozone.

**Approach:** Gas-solid reaction probabilities for the reactions of NO$_2$, N$_2$O$_5$, ClONO$_2$, and HNO$_3$ with NaCl, NaBr, and synthetic and real sea salt will be determined with a variety of techniques including a Knudsen cell, diffuse-reflectance infrared Fourier transform spectrometry (DRIFTS), and in collaboration with Professor Hemminger under NSF support, X-ray photoelectron spectroscopy. The possibility of other potential reactions such as that of HOCl with NaCl, analogous to the ClONO$_2$-HCl-ice reactions, will also be explored. Photochemistry on the surfaces will be investigated. A combined FTIR-MS system will be developed and applied to particles at varying relative humidities both below and above the deliquescence point to study the effects of water on these reactions. This system will also have photolysis capabilities to investigate possible photochemical enhancements of the kinetics and photochemical reactions of the products.

The fates of the products of these gas-salt reactions will be explored. Thus, the kinetics and products of reaction of gaseous water with ClINO and ClINO$_2$, formed in the NO$_2$ and N$_2$O$_5$ reactions with NaCl, respectively, will be investigated with FTIR coupled to a 40-m-long path cell. The kinetics of reaction of chlorine atoms with biogenic organics, such as isoprene and dimethyl sulfide, will be determined with relative-rate techniques by following the loss of the biogenic and a reference organic with gas chromatography. In addition, fast-flow-discharge-system studies to measure absolute rate constants for the chlorine atom-organic reactions will be carried out. The reaction products and mechanisms will be identified with FTIR combined with GC and GC-MS. The data obtained from these experiments will be used in a simple 0-D kinetic model to assess the contribution of sea-salt aerosols to the formation of chlorine and bromine atoms in the troposphere and their role in determining tropospheric ozone.

**Results to Date:** With DRIFTS, the reaction of NO$_2$ with NaCl was determined to be second order in NO$_2$, suggesting that the N$_2$O$_4$ dimer in equilibrium with NO$_2$ is the reactant. The reaction probability for this N$_2$O$_4$-NaCl reaction was determined to be $(6 \pm 2) \times 10^{-4}$; preliminary results for the NaBr-N$_2$O$_4$ reaction suggest this reaction is approximately 50% faster than the NaCl reaction. The reaction probability for the HNO$_3$-NaCl reaction has been determined in the XPS studies to be $(4 \pm 2) \times 10^{-4}$ at 298 K. Applying these results, along with our previously determined lower limit of $>2.5 \times 10^{-3}$ for the N$_2$O$_5$ reaction to typical marine conditions suggests that the N$_2$O$_5$ reaction may be competitive with, or perhaps exceed, that of HNO$_3$ at night when the N$_2$O$_5$ builds up. Thus, ClINO$_2$ production and, from its photolysis, the generation of chlorine atoms in marine atmospheres is anticipated.

Two types of relative rate systems have been developed to study chlorine-atom kinetics: a constant-volume apparatus and a constant-pressure system. These systems have been tested with well-established alkane-chlorine atom reactions and give results that are generally in good agreement with the literature. However, there are some discrepancies even for the simple alkanes, which we are resolving through the use of fast-flow-discharge studies. To date, the room-temperature rate constants determined in this system for the reactions of propane and n-butane are in excellent agreement with the literature. Finally, studies of the reaction of chlorine atoms with dimethyl sulfide are under way with FTIR to determine if this reaction could be a significant source of CH$_3$Cl in the marine troposphere.

This research has led to collaborative projects with Pacific Northwest Laboratories and Battelle Columbus, directed to planning and carrying out field studies over the North Atlantic Ocean to identify and measure the products of these "heterogeneous" sea-salt reactions, including Cl$_2$, ClINO$_2$, ClINO, HCl, and HOCl. In addition, the results are being disseminated to the modeling community for use in predicting the effects on global ozone and climate change.

**THEORETICAL STUDIES OF GLOBAL TROPOSPHERIC AND STRATOSPHERIC CHANGES OF OZONE**

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LAURENCE LIVERMORE NATIONAL LABORATORY

FY 1994 150

10/01/93–09/30/94

**Objective:** To conduct both ozone-depletion-potential (ODP) studies and global-warming-potential (GWP) studies with a 3-D chemistry-radiative-transport model.
Product: Detailed direct and indirect radiative forcing and GWP calculations for ozone using realistic atmosphere models and scenarios.

Approach: The LLNL 2-D chemical-radiative-transfer model will be used to investigate the seasonal and latitudinal changes in temperature arising from observation-consistent perturbations of the ozone distribution. Improvements will be made to modeling of tropospheric water vapor and temperatures as well as to radiative models to increase the accuracy of these studies. Atmosphere models will be used that contain cloud distributions, stratospheric-temperature-equilibrium effects, detailed chemistry models, and aerosol-layer effects to investigate the ozone balance in nonambient atmospheres both past and future and to perform radiative forcing and GWP calculations for some probable future ozone profile scenarios and expected trace gas emission scenarios. The sensitivity of a given chemical process to trace species abundances will be addressed. Indirect ozone forcing and GWP caused by a methane impulse will also be calculated. Results will be compared with those from other studies.

Results to Date: We determined the tropospheric radiative forcing of O$_3$ in the 175- to 735-nm (UV-visible) wavelength range plus the 500- to 1650-cm$^{-1}$ wavenumber range in accordance with a set of IPCC test cases that were provided to interested modeling groups. The tropospheric radiative-forcing values obtained lie in the middle of the range of values of the models used in the IPCC test calculations.

We calculated the indirect radiative forcing and GWP of O$_3$ from an impulse of CH$_4$. The O$_3$ indirect GWPs vary from about 25 after 20 years to about 4 after 500 years and are in good agreement with other estimates. The indirect O$_3$ GWP for an atmosphere containing twice the normal NO$_x$ abundance varies from about 25 after 20 years to about 9 after 500 years.

ANALYSES OF SHADOWBAND RADIOMETRIC DATA WITH EMPHASIS ON THE COUPLED EFFECTS OF PARTICULATE SCATTERING AND OZONE ABSORPTION IN THE UV-B

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UNIVERSITY OF WASHINGTON
FY 1994 58
07/01/94–06/30/95

Objective: To develop insights and software to permit the simultaneous extraction of atmospheric O$_3$, H$_2$O, and particulate scattering parameters by inversion of data from multifilter rotating shadowband radiometry (MFRSR).

Product: Software for "best possible" inversions, consistent with the data, and design of "best possible" filter selection for MFRSRs constrained by task-objectives and experimental noise.

Approach: Nonlinear inversion techniques will be applied to shadowband data with comparisons to observed O$_3$ and H$_2$O columns by independent methods and with objective criteria for determining the residual degrees of freedom that may be attributed to particulate scattering.

Results to Date: Among the techniques under development for analyses of solar radiometric data are a variety of constrained nonlinear inversions, which have been sequentially compared with Twomey's classical method. In general, all the methods must be strongly constrained to achieve stable solutions. An informative result is that the several methods achieve comparable $R^2$ but differ markedly in their derived parameters. The variance surfaces typically show multiple minima of comparable depth but quite different coordinates. This means that the information content of the data is not able to support all the demands that are being placed upon it.

In this light, I have conducted optimization studies for filter-wavelength selection, with some improvement revealed for particle-size inversions, but always at cost for establishing baseline corrections and to extract O$_3$ and H$_2$O.

An interesting question can be asked: "How much information about particle size distributions might be
extracted from solar radiometric data, with infinite spectral resolution?" The answer is unclear at present, but the results suggest that between two and three parameters may potentially be assigned to describe particulate distributions after corrections for Rayleigh scattering and absorption by \( \text{O}_3 \) and \( \text{H}_2\text{O} \). The "between" is important because a minimum of three parameters is needed to establish the particular column burden, the mean particulate radii, and their standard deviation.

PREFLIGHT AND POSTFLIGHT SPECTORADIOMETRIC CALIBRATION ERRORS FOR SBUV AND TOMS TYPE INSTRUMENTS

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RESEARCH SUPPORT INSTRUMENTS, INC.

FY 1994 88
07/01/94–06/30/95

Objective: To calibrate TOMS and SBUV-2 instruments.

Product: An accurate understanding of the instruments' calibration errors.

Approach: A combination of laboratory spectroradiometric comparison calibration data from the SBUV-2 (TIROS-N) and SSBUV (space shuttle) ozone monitoring instruments, surface-based direct sun, and zenith-sky comparison measurements will be compared with the world-standard Dobson Instrument No. 83 in Boulder and satellite overpass data from TOMS and SBUV-2 instruments.

Results to Date: A common spectroradiometric calibration scale was established for NOAA's SBUV-2, NASA's SSBUV, and ESA's GOME ozone-monitoring instruments with uncertainties of less than 2%.

Preliminary results have been obtained for surface-based direct-sun and zenith-sky comparison measurements with those from the world standard Dobson Instrument No. 83 and with those from radiative-transfer calculations using satellite-overpass ozone values.

REEVALUATION OF TOTAL AND UMKEHR OZONE DATA FROM TEN NOAA/CMDL DOBSON SPECTROPHOTOMETER OBSERVATORIES

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NOAA CLIMATE MONITORING AND DIAGNOSTICS LABORATORY

FY 1994 71
FY 1993 73
09/15/94–09/14/95

Objective: To reevaluate total and Umkehr ozone data, including corrections for instrument calibration and instrument changes.

Product: An improved column ozone data set for research on global ozone trends and ozone and climate-change research.

Approach: An international project to optimize Dobson spectrophotometer global total ozone data began in 1991 under the aegis of the NOAA National Environmental Satellite Data and Information Service (NESDIS) and the World Meteorological Organization (WMO). As part of this effort, a Handbook for Dobson Ozone Data Reevaluation was written, a section of which provided detailed instructions for reevaluating total-ozone data having well-documented calibration histories such as those available for the NOAA/CMDL data. Reevaluation of the NOAA/CMDL data has been performed in accordance with the instructions set out in the WMO manual.

Results to Date: Because of the convening of the 1994 Ozone Trends Panel, there was a demand on our
Laboratory to focus on updating, for ozone trend analyses, data from our stations having records dating back to the early-to-mid 1960s. In the past, total ozone data through 1991 through 1992 were reevaluated for Bismarck, North Dakota; Caribou, Maine; Boulder, Colorado; Wallops Island, Virginia; Nashville, Tennessee; Fresno, California; Mauna Loa, Hawaii; and Samoa, South Pacific. These records were updated through April 1994 and submitted to the Trends Panel for analysis and inclusion in the Trends Panel Report. Subsequently, the bulk of the work was completed in reevaluating 1963–1994 Huancayo, Peru, and South Pole, Antarctica, total-ozone data. Umkehr data reevaluation was completed for Boulder and for Mauna Loa Observatory.

STATISTICAL ANALYSIS AND INTERPRETATION OF STRATOSPHERIC OZONE AND TEMPERATURE TRENDS WITH SATELLITE DATA

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FY 1994 86
FY 1993 88
09/15/94–09/14/95

Objective: To understand lower stratospheric ozone and temperature distributions and trends.

Product: An improved database to analyze the trend in midlatitude ozone.

Approach: Extensive statistical analyses will be carried out on satellite ozone and temperature data sets in an attempt to determine spatial and seasonal ozone and temperature trends (and their interrelationships) as a function of latitude, longitude, and altitude. Included among the satellite ozone data sets to be studied are those obtained with the Nimbus 7 Total Ozone Mapping Spectrometer (TOMS), the Nimbus 7 Solar Backscattered Ultraviolet Spectrometer (SBUV), and the Stratospheric Aerosol and Gas Experiment II (SAGE II). Included among the stratospheric temperature data sets to be analyzed are the National Meteorological Center (NMC) geopotential height fields and measurements obtained with the Microwave Sounding Unit (MSU) on the NOAA operational satellites. Latitude, longitude, quasibiennial oscillation, and solar variability will be examined as independent variables. Although the four data sets overlap temporally, the different measurement systems yield differing ozone data sets. Further, the models and measurements do not agree. This research will assist in understanding and reducing the differences.

The physical relationship between derived lower stratospheric ozone trends at midlatitudes and observed atmospheric temperature trends will be investigated. The extent to which global mean temperature trends in the lower stratosphere can be produced by reduced radiative heating caused by the observed ozone losses will be determined. The extent to which changes in ozone transport associated with upper tropospheric and lower stratospheric dynamics can contribute to observed midlatitude ozone trends will be investigated. Model calculations and trend analyses of stratospheric transport parameters will be conducted for this purpose.

Results to Date: The altitude dependence of stratospheric ozone trends was determined based on Nimbus 7 SBUV data for the period November 1978 to June 1990. Comparisons were made of the observed trends with the predictions of several 2-D models of the stratosphere.

In the upper stratosphere, seasonally averaged SBUV-derived ozone trends agree well with 2D model predictions based on the observed ~0.1 ppb/year increase in anthropogenic chlorine. In the lower stratosphere, a major area of qualitative disagreement continues to exist in the midlatitude northern hemisphere during winter and spring where the observed SBUV-derived negative trend is about 5 times greater than the model-predicted trend.

The relationship between lower-stratospheric ozone and temperature trends was investigated with SBUV ozone data and MSU lower-stratospheric temperature data. A 1-D radiative-transfer model with fixed dynamical heating was used to calculate the approximate latitude and seasonal dependences of temperature changes to be expected from the observed ozone trends. The resulting calculated temperature changes agreed approximately in amplitude and seasonal dependence with those derived from the MSU data. Thus, the observed stratospheric cooling trends can be attributed primarily to reduced radiative heating associated with ozone depletion.
NEW UV-B SPECTRAL RADIOMETER (UVB-RSS)

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PACIFIC NORTHWEST LABORATORY and STATE UNIVERSITY OF NEW YORK AT ALBANY
FY 1994 150
10/01/93-09/30/94

Objective: To develop and test a new, low-cost, ultraviolet-B (UV-B) rotating shadowband spectroradiometer (UVB-RSS) with performance characteristics to detect trends and to provide status monitoring of surface UV-B radiation.

Product: An intermediate resolution (2- to 5-nm spectral bandpass) rotating shadowband radiometer, ready for commercialization, suitable detecting trends in UV radiation on the order of 0.1% per year over periods of one decade or more and able to operate unattended in distributed field networks with remote access to the data.

Approach: Using the experience gained from the development of the multifilter rotating shadowband radiometer (MFRSR) and the rotating shadowband spectroradiometer (RSS) developed under the DOE Quantitative Links and Atmospheric Radiation Measurement (ARM) programs for the measurement of solar irradiance, we will design, fabricate, and field test a UV version of the rotating shadowband radiometer. To maximize throughput and minimize the effects of out-of-band light, the UVB-RSS uses all-quartz optics, a photomultiplier-tube detector that is blind to solar radiation at wavelengths greater than about 330 nm, and a rotating-slit wheel for selecting 3 to 5 spectral bandpasses between 300 nm and 330 nm.

After the prototype is completed, it will undergo rigorous bench tests in the laboratory before deployment in the field to evaluate operational performance characteristics. After the initial field and bench tests are completed, shortcomings of the prototype unit will be corrected, and a final hardened version will be readied for extensive field testing. The final field tests will be conducted at the ARM Cloud and Radiation Testbed (CART) site in the Southern Great Plains (SGP), and results will be compared to other high-resolution UV-B instruments, such as the Dobson spectrophotometer.

Results to Date: Funding was provided late in FY 1994, so no significant technical results were available. A subcontract to the State University of New York at Albany (SUNY-Albany) was initiated and placed.

ULTRAVIOLET RADIATION CLIMATOLOGY OF THE EARTH'S SURFACE AND LOWER ATMOSPHERE

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NATIONAL CENTER FOR ATMOSPHERIC RESEARCH and UNIVERSITY OF ALASKA
FY 1994 88
07/01/94-06/30/95

Objective: (1) To develop a climatology of tropospheric UV amounts (averages and variability) as functions of wavelength, altitude, geographical location, time of the year, and secular change (e.g., from stratospheric ozone depletion). (2) To develop fast but accurate methods for calculating tropospheric photodissociation coefficients (J-values) and surface biologically weighted UV doses.

Product: (1) A tabulation of climatological values for tropospheric J-values and surface biologically weighted UV doses and (2) codes for calculating tropospheric UV radiation and the corresponding J-values and biologically effective doses.

Approach: Radiative transfer schemes will be optimized to the UV region; coupled to satellite and other observations of atmospheric optical parameters; and coupled with critically evaluated molecular absorption, quantum yield, and photobiological spectral response data. The model's results will be evaluated with measurements from recently deployed UV networks.

Result to Date: New project.
SAGE II–UMKEHR OZONE COMPARISONS AND AEROSOL EFFECTS: AN EMPIRICAL AND THEORETICAL STUDY

NEWCHURCH, MICHAEL J.

UNIVERSITY OF ALABAMA

FY 1994 236
FY 1993 190
09/15/94–09/14/95

Objective: To examine the differences between the satellite-borne SAGE I and II ozone and aerosol profiles and the ground-based Umkehr ozone profiles for aerosol effect and trend information. The SAGE-derived trends have been less negative than the Umkehr trends in the upper stratosphere where the chlorine-catalyzed ozone depletion is expected to be most severe.

Product: An improved understanding of the impact of aerosols on stratospheric-ozone-measurement techniques and on stratospheric-ozone trends.

Approach: The interference of stratospheric aerosols with the detection and measurement of vertical ozone profiles will be investigated by comparing SAGE II data with Umkehr data. This comparison will include statistical analysis and hypothesis testing of potential relationships between Umkehr and SAGE II measurements. The comparison will allow the data to be corrected for interference and the assessment of ozone concentration trends in the stratosphere.

A modeling study will be conducted that will include how the aerosols influence the radiative transfer and the wavelengths in the Umkehr inversion to determine ozone profiles. Background aerosols as well as the Mt. Pinatubo events will be considered. A number of additional parameters will be also be included in the analysis such as climatology, season, latitude, longitude, and particular station characteristics. The model study will account for multiple scattering and anisotropic scattering and will use two separate radiative transfer models: the JPL/Cal Tech (Jet Propulsion Laboratory/California Institute of Technology) model and the University of Arizona model.

Results to Date: We compiled an exhaustive set of available SAGE/Umkehr coincidence pairs comprising 540 SAGE I profiles at 24 Umkehr stations and 7400 SAGE II profiles at 34 Umkehr stations with the Umkehr ozone profile derived from both the old (1964) and new (1992) algorithm. Analysis of this dataset revealed that:

- The Umkehr aerosol effect from 1984 to 1989 is similar to previous estimates in layers 7 to 9, but not in 4 to 6.
- SAGE I altitude registration is low by approximately 1/2 km; adjustment for this offset reconciles SAGE I/II and Umkehr trends in the upper stratosphere.
- SAGE/Umkehr differences vary among stations, indicating significant station differences.
- SAGE II total ozone is 5 to 10% higher than new (1992 algorithm) Umkehr.
- New Umkehr significantly redistributes ozone in nearly all layers compared to the old algorithm.

Because of the paucity of Umkehr data after 1990 (post-Pinatubo), we have established a collaboration with NOAA/CMDL to process the Umkehr data from the NOAA stations so that it will be available for our analyses. We have also established a collaboration with the World Ozone Data Center to obtain the Umkehr data from other global stations (post-Pinatubo) and with Carl Mateer (coauthor with John DeLuisi of the new Umkehr algorithm) to investigate the scientific implications of the new ozone reductions.

MEASUREMENT OF AEROSOL PROPERTIES

NOVAKOV, TIHOMIR

LAWRENCE BERKELEY LABORATORY

FY 1994 150
10/01/93–09/30/94

Objective: To experimentally obtain the relationships between aerosol extinction, scattering, and absorption and size-resolved chemical composition, mass, and number-size distribution.

Product: Closure with calculated optical properties based on Mie theory and size-resolved chemical composition of anthropogenic aerosols.

Approach: The experimental techniques originally developed for measuring the scattering and absorption coefficients in the visible to UV region will be
modified. These techniques will be applied to both laboratory-generated and ambient aerosols.

Results to Date: We have completed a conceptual design of an apparatus to measure on-axis and off-axis filter-sample transmittance of a light beam (in the 280- to 315-nm and 530- to 570-nm spectral regions). It consists of a light-beam source (Cermax Xenon lamp with UV output), UV and visible interference filters, the sample section, and on-axis and off-axis light detection systems (optical fibers and a PMT detector). This optical setup should be less prone to surface internal-reflection effects occurring between the sample and the light-diffusing surface. The components of the system are being assembled. In parallel, we have started to develop a data-analysis procedure based on radiative-transfer models for both particle layers and for the particle-filter interaction.

3-D GLOBAL ATMOSPHERIC CHEMISTRY MODELING

PENNER, JOYCE

LAWRENCE LIVERMORE NATIONAL LABORATORY

FY 1994 200
10/01/93–9/30/94

Objective: To develop a 3-D, global tropospheric model with realistic treatment of atmospheric chemistry.

Product: An understanding of the role of energy-related pollutant sources on the chemical climatology of the global troposphere.

Approach: We have developed a global tropospheric chemical model that is capable of treating the chemistry of CO, CH₄, NOₓ, and a full suite of nonmethane hydrocarbons. The development of this model has been possible because of partial funding from the DOE Atmospheric Chemistry Program in the past. The model is computationally fast, while providing much better resolution than presently available in other tropospheric chemistry models, and the model is configured so that it may also be run interactively with a climate model. In this work, we propose to exercise this model, examining the role of nonmethane hydrocarbons in the chemical climatology of the global troposphere. We plan to develop increasingly more sophisticated versions of the model so that increasingly realistic treatments of the chemistry and transport in the troposphere will be possible. Throughout the project, careful comparison of model results with data and separate tracer transport tests will help build confidence in the quality of the simulations, allowing us to discern the impact of energy-related emissions on the global troposphere.

Results to Date: We have begun to use our model to understand the impact that anthropogenic emissions from North America have on downwind areas, such as the North Atlantic Ocean and Western Europe. Our model results show that ozone is transported from the Northeastern coast of the United States in "events," with a frequency of 5 to 7 days, similar to observations. In a simulation in which North American fossil-fuel emissions of NOₓ were decreased by 50%, ozone concentrations decreased by more than 20 ppbv near North America, by 8 to 10 ppbv at a location midway across the North Atlantic Ocean, and by 4 to 5 ppbv near Mace Head, Ireland. Further model improvements are currently under way.

TRENDS AND EVOLUTION OF MINIMAL OZONE AMOUNTS, 1979 TO 1992

RECK, RUTH A.

ARGONNE NATIONAL LABORATORY

FY 1994 140
10/01/93–09/30/94

Objective: To describe long-term changes and variability in total ozone (TOZ).

Product: A new approach for describing long-term changes and variability in TOZ.

Approach: Long-term changes and variability in total ozone will be described by using frequency distributions of daily ozone and analyzing the extreme values. Using the database of column ozone provided by the Total Ozone Mapping Spectrometer aboard Nimbus 7 from 1979 to 1991, we will analyze a region of the midwestern United States extending from 35.0°N to 42.0°N and from 81.25°W to 87.50°W, which includes much of Indiana, Ohio, Kentucky, Tennessee, and the western portion of West Virginia.
Results to Date: Substantial variability of the maximum and minimum daily TOZ values, as much as 50%, during the spring and fall seasons were found. The largest change between consecutive daily differences was also shown to be substantial; for example, 137 DU for spring and 69 DU for fall.

The spring and fall frequency distributions of TOZ were also developed. Based on a significance level of 0.01, the resulting $c^2$ are consistent with Gaussian distribution functions. An interesting finding of this work is a digression from Gaussian behavior in the wings. On the high-ozone side, a positive deviation occurs, while on the low-ozone side a negative deviation is seen. The positive deviation is much more pronounced in the spring than in the fall, which we postulate may be caused by increased concentrations of tropospheric ozone during the spring. On the low-ozone side, the observed negative deviation may result from stratospheric ozone depletion. If the time rate of depletion of stratospheric ozone is proportional to the amount of stratospheric ozone present, the effect would be to change the shape of the Gaussian curve where smaller values of TOZ are affected less than larger TOZ values. The negative trend in TOZ is known to be more pronounced in the spring than in the fall.

Extremes of TOZ were also analyzed. The 100 lowest values for spring occurred in two events during 1985. The first event began on March 3 and persisted until March 8; the second lasted from March 29 until March 31. The 100 lowest values for fall occurred in nine events between 1982 and 1991 and ranged in duration from one to four days, mainly in the month of November. Our analysis of the extremes for fall and spring demonstrated that the frequency of low-TOZ events has increased from the first six-year period to the second six-year period during the fall season, while the frequency of low-TOZ events during the spring season has decreased.

CONFIRMATORY ANALYSIS OF SAGE I AND SAGE II DATA

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FY 1994 217

07/01/94–06/30/95

Objective: To re-invert the SAGE I and SAGE II transmission data to confirm or modify ozone densities in the stratosphere derived from SAGE data.

Product: A database of the retrieved ozone densities as a function of altitude, latitude, season, and year.

Approach: A simulation code, or forward model, will be used to test the inversion algorithms and to investigate effects of atmospheric perturbations, such as volcanic aerosol emissions, on the retrieval of ozone densities. The inversion problem will be approached in two ways: (1) inverting both SAGE I and SAGE II data with only the channels present on both instruments and (2) inverting SAGE II data with all available channels. Inversions of the data will be compared to correlative data from ROKOZ and other sources as deemed necessary. Once the inverted data have been satisfactorily validated, a trend analysis will be performed. A significant part of the inversion will include assignment of error bars as a function of altitude. Results from the trend analysis will be compared to other measurements, such as SBUV. The final database will also be compared to the currently available SAGE data, and any discrepancies between these data sets will be explained.

Results to Date: The data simulation algorithm (the forward model) has been made operational; the SAGE database has been brought online at LASP; and the inversion code is being designed. Instrumental characteristics (such as viewing geometry, filter sensitivity, and wavelength passbands) have been included in the data simulation.

To simulate atmospheric transmission, Rayleigh scattering, aerosol scattering, and ozone absorption are included. Extinctions are calculated with total densities derived from NMC data, ozone densities obtained from a climatological model, and several different values for aerosol densities. By using the forward model to simulate data at times of high and low atmospheric aerosol densities, we have shown that the SAGE transmissions are predictably sensitive to aerosol emissions. Nevertheless, during most of the time covered by the SAGE observations, the forward-model results indicate that the retrieval of ozone densities from the SAGE data will be successful.
INVESTIGATION OF THE EFFECT OF NATURAL PHENOMENA AND INDUSTRIAL ACTIVITY ON STRATOSPHERIC OZONE TRENDS

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FY 1994 164
FY 1993 168
09/15/94–09/14/95

Objective: To gain a better understanding of the effects of natural variability vs man-made perturbations on ozone.

Product: Analysis of ozonesonde data and a model capable of investigating interannual changes in ozone.

Approach: The ozone data will be critically analyzed, and different data sets will be compared. A 2.5-D model of the stratospheric dynamics and chemistry will be used to isolate the effects of different perturbations on ozone.

Results to Date: We have completed an analysis of trends in ozonesonde data. Ozonesondes provide the only information on trends in tropospheric ozone above the surface and on trends in the stratosphere below 90 mbar; they also provide unique information on stratospheric trends below 10 mbar prior to 1979, when satellite measurements commenced.

It was found that significant spatial variations occur in the trends in the troposphere, with the largest increases over Europe (about 2% per year throughout the troposphere) and no long-term trend over Canada; there is a small (<1% per year) increase over the east coast of the United States in summer. The Japanese stations show increases in ozone only below 5.5 km.

Stratospheric ozone decreases are found from about 24 km to near the tropopause. Ozone losses below 17 km appear to be responsible for the 20% difference between trends in column ozone derived from SAGE and TOMS. Ozone changes in the troposphere make an important contribution to the column ozone change for some stations. The stratospheric decreases are larger in winter than in summer over Europe and the midlatitude stations of North America; they are larger in summer than in winter over the high-latitude (>53° N) stations of North America. These seasonal losses are consistent with the patterns reported by Stolarski et al. using TOMS data. Losses are found year round over Syowa, Antarctica, although they are largest in spring.

To separate effects of dynamical variability (i.e., changes in the transport circulation caused by variations in the tropospheric wave driving) from chemical effects on the observed ozone distribution and trends, we have developed a 2.5-D model of the stratosphere. The dynamics module is a low-resolution 3-D model from which we derive 2-D transport parameters from explicitly calculated planetary waves that are forced in the troposphere. Chemistry and transport are computed in two dimensions, latitude and altitude. The model is interactive (i.e., heating rates are computed from model-calculated ozone). The diabatic heating drives the dynamics, which, via transport, feeds back on tracer distributions.

An important issue in understanding total-ozone data is the mass transport between tropics and midlatitudes in the stratosphere. As a first application of the model, we have tried to reproduce recent UARS measurements of species like HF and CH₃, which exhibit large gradients between tropics and midlatitudes, as well as observations of CO₂ obtained during the SPADE and ASHOE/MAESA missions. Our model is able to provide a consistent picture of the mechanisms responsible for these tracer distributions with a minimum of assumptions.

APPLICATION OF SENSITIVITY ANALYSIS TO REDUCE UNCERTAINTIES IN STRATOSPHERIC-OZONE CHEMICAL-DYNAMICS MODELS

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MISSION RESEARCH CORPORATION and PRINCETON UNIVERSITY

FY 1994 151
07/01/94–06/30/95

Objective: To define key mechanistic pathways in models, determine the importance of reactions with large uncertainties, provide a better understanding of the ozone chemical dynamics models, and provide a quan-
titative assessment of the uncertainties in the model predictions.

**Product:** An ordered list of important chemical mechanisms and reactions with large uncertainties and estimates of model-prediction “error bars” for total ozone concentration, change in ozone concentration over time (trends), and ozone predictions affected by various trace gas deposition rates (freon, NO₃, CO₂, etc.).

**Approach:** State-of-the-science, systematic, and rigorous sensitivity-analysis tools will be applied to stratospheric/tropospheric ozone chemical-dynamics models. A gradient-based Green’s-function approach will be used to calculate the model-prediction sensitivities to each of the model parameters. These sensitivities will provide a foundation for a full statistical analysis of model predictions by “guiding” Monte Carlo calculations.

**Results to Date:** A sensitivity-analysis code was validated by reproducing early work by Dickinson and Galinas. Approaches were outlined to address the following concerns: Which reaction rates are critical to the model? Which species are critical to the model? What are the important pathways? How important are the missing reactions? How does the functional form of k(T) affect the species predictions? How does the shape of a driving function control the species predictions? How good are the model predictions? An initial guided Monte Carlo technique was developed to propagate the effect of model-parameter uncertainties to model-prediction uncertainties.

**SENSITIVITY AND UNCERTAINTY ANALYSIS OF ATMOSPHERIC-OZONE PHOTOCHEMISTRY MODELS**

**Objective:** To determine the sensitivities and uncertainties associated with chemical-kinetics models of the global atmospheric ozone.

**Product:** An assessment of the uncertainty in the models and thus a basis for model improvement.

**SENSITIVITY AND UNCERTAINTY ANALYSIS OF ATMOSPHERIC-OZONE PHOTOCHEMISTRY MODELS**

**Objective:** To determine the sensitivities and uncertainties associated with chemical-kinetics models of the global atmospheric ozone.

**Product:** An assessment of the uncertainty in the models and thus a basis for model improvement.
Approach: The current chemical-kinetics models of the global atmospheric ozone couple photolysis, chemical rate constants, transport, and dynamics. The effects of elementary rate constants and in the atmospheric kinetics on the performance of these models will be investigated by compiling and developing information on the kinetics parameterizations, the basic atmospheric conditions used in reduced-form models (0-D models), and the direct sensitivity calculation codes Chemkin/Senkin. A well-tested framework will be used for the analysis. The Chemkin code provides gas-phase and gas-surface chemical reactions and thermodynamics and a first-order sensitivity analysis. The uncertainties associated with the rate constants and the kinetics data will be analyzed directly with the Chemkin/Senkin codes. To test for possible higher-order interaction, a series of repeat calculations will be conducted varying key rate constants individually and in pairs. The results will be compared with the first-order sensitivity calculations. Collective sensitivities and uncertainties will be calculated for ozone and other selected species with the rate constant and kinetic-sensitivity information. Optimization techniques and 1-D atmospheric modeling codes will be used to evaluate the net uncertainty in ozone modeling.

Results to Date: The LLNL photochemistry mechanism has been successfully integrated with the Senkin code, and ozone sensitivities have been computed for selected zero dimensional boxes from the LLNL 2-D atmospheric model. Time scales appropriate to local ozone photochemical lifetimes must be applied to obtain correct, converged sensitivity coefficients. Transport terms must be considered at lower and polar stratospheric locales to obtain a steady solution. When convolved with the basic rate-data uncertainties from the NASA-JPL compilation, the initial results show the basic photochemistry uncertainties in ozone prediction are equally divided between photolysis and kinetics rate constants, mostly in the few key steps of the mechanism.

Objective: To improve understanding of the chemical and dynamic processes that control midlatitude O$_3$ in the lower stratosphere and free troposphere and to develop improved predictions of future O$_3$ changes in these regions and their influence on (and response to) future climate changes.

Product: A quantitative evaluation of the physical and chemical processes important to free tropospheric ozone at the middle latitudes of the northern hemisphere.

Approach: The observed O$_3$ climatology, together with a radiation model, will be used to evaluate the importance of atmospheric O$_3$ to radiation balance and to study how the O$_3$ changes observed between 1980 and 1990 affect the total radiation and UV fluxes as well as how they compare with increases in other greenhouse gases. This study of the climate-chemistry interactions will be based on the NCAR-GENESIS global climate model and the University of Oslo's multidimensional photochemical-transport model. The photochemical-transport model will be used to calculate the tropospheric O$_3$ distribution with a variety of emission scenarios for NO$_x$, HC, and CO and compare the distributions of these gases with observed values. Next, the O$_3$ and other gases consistently calculated from the photochemical-transport model will be used as input to the climate model to study the effect on climate and on the UV flux reaching the surface. These experiments will be conducted for both the current climate state and the global-warming climate state. Exploratory studies to incorporate the chemical code into the climate model with a focus on the computational efficiency and consistency will also be conducted.

Results to Date: A 2-D chemistry-transport model of the global troposphere is being used to study (1) the chemical response to increased UV radiation caused by stratospheric ozone depletion and (2) increased temperature and water-vapor density caused by global warming as simulated from the global-climate model. Increased UV radiation increases the photolysis rates for several tropospheric gases, in particular ozone. This increase leads to enhanced levels of odd hydrogen and reduced concentrations of tropospheric ozone. Increases in temperature and water-vapor density reduce the levels of tropospheric ozone through temperature-dependent reaction rates and increased production of odd hydrogen. In both cases, the methane levels are also reduced. Thus, the results indicate that the considered mechanisms constitute damping effects on global warming.

ATMOSPHERIC OZONE AS A CLIMATE GAS

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STATE UNIVERSITY OF NEW YORK AT ALBANY

FY 1994 111

07/01/94–06/30/95
TREND ANALYSIS OF OZONE AND ULTRAVIOLET-B RADIATION

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UNIVERSITY OF COLORADO

FY 1994 194
07/01/94–06/30/95

Objective: To develop the techniques for analyzing surface UV data for trends and to continue the analysis of total ozone and ozone profile data for trends.

Product: Well formulated and documented techniques for producing defensible trend estimates from surface UV radiation data.

Approach: A team of atmospheric scientists and statisticians will assess the quality and reliability of past UV data. Both broadband and spectral UV data will be analyzed for quality assurance and trend estimates. Advanced radiative-transfer programs will be used to look for consistencies between ozone and UV data for a variety of locations at midlatitudes.

Results to Date: A survey of existing UV monitoring activities was completed to assess what information is available for analysis. The longest running UV network, the Robertson-Berger meter network, has been carefully examined for problems in the data. Significant trends in the data from this network are unexplained by ozone changes. These data were compared with atmospheric reflectivity data from the TOMS satellite. The surface reflectivity gives an indication of cloud cover. For the one station examined, it was determined that changes in cloud cover do not explain the unexpected trends in the UV data.

From the initial analysis of the UV data, it is likely that level shifts and other forms of intervention take place within the time period of analysis. These level shifts are caused by changes in instrumentation that are unavoidable in a 20-year monitoring effort. These changes will be accounted for in the statistical analysis so that the trend results will not be affected by instrumentation changes. Newly reevaluated Dobson total-ozone data from eight U.S. stations were obtained, and trend analysis has been performed to examine the seasonal trend features. For the five long-running stations in the contiguous United States, during the past 16 years (1979 to 1994), an average downward trend of about −5% per decade has been observed during the winter and spring seasons, and an average downward trend of about −2.5% per decade has been observed in the summer and fall seasons. These Dobson total ozone data will also be useful in the comparisons of total ozone and UV data and their trends during the past 15 to 20 years.

HETEROGENEOUS CHEMISTRY AFFECTING UPPER TROPOSPHERIC AND STRATOSPHERIC OZONE

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AERODYNE RESEARCH, INC., and BOSTON COLLEGE

FY 1994 140
07/01/94–06/30/95

Objective: To study heterogeneous gas-liquid interactions related to atmospheric ozone formation and destruction.

Product: An understanding of what possible heterogeneous processes may covert less-reactive NO species (e.g. HNO₃) to the more-reactive NOₓ (e.g. HNO₂) species.

Approach: Laboratory experiments will measure gas/liquid mass transport rates in bubble-column and droplet apparatuses that can measure both fast and slow heterogeneous processes. Gas-phase species to be studied include NO, NO₂, NOₓ, HONO, H₂CO, and PAN. Heterogeneous-chemistry studies will be conducted with aqueous solutions, sulfuric acid solutions, and nitroxylic sulfuric acid solutions for the species of interest. The effects of photochemistry, ionic additives, and species codeposition will be studied.

Results to Date: Initial work has centered on apparatus upgrades and improvements related to the precision in the experimental control of gas/liquid interactions and gas-phase detection sensitivity.
SUPPORT OPERATIONS

The objective is to provide an instrumented, twin-turboprop atmospheric-research aircraft for direct access by DOE research projects with the possibility of secondary access by non-DOE clients.

The aircraft chosen for this purpose, a Gulfstream-1 (G-1), has been selected on the basis of its performance characteristics (range, speed, altitude, carrying capacity, and economy) in reflection of DOE programmatic needs. The aircraft has been extensively instrumented for air-chemistry, cloud-chemistry, and cloud-physics measurements.

The following example applications illustrate DOE uses of this facility.

The Kuwait Oil-Fire Evaluation: During FY 1991 (July–August 1991), DOE contributed to a multinational effort, coordinated under the World Meteorological Organization, to characterize the atmospheric, climatological, and biological effects of the Kuwait oil fires. As a part of this effort, DOE dispatched the G-1 to the Arabian Gulf region for extensive plume measurements at close and extended distances downwind from the source region. The G-1’s instrument ensemble included equipment for measuring aerosol physical and chemical properties, concentrations of organic and inorganic pollutant gases, cloud-nucleating properties, and solar/terrestrial radiation. Approximately 15 research flights were conducted during this series, and the resulting data are being pooled with the international effort for joint interpretation.

The G-1 research aircraft was developed with the expectation that DOE would play a major role in future global-scale atmospheric research as a part of its implementation and development of the EPACT. This process is now beginning to occur, and as a result, extensive DOE application of the G-1 is anticipated under its Atmospheric Chemistry Program (ACP; programmatic emphasis: continental and oceanic fate of energy-related air pollutants) and its Atmospheric Radiation Measurement (ARM) Program.

The current G-1 mission is an ACP flight series over the North Atlantic in coordination with the international North Atlantic Regional Experiment (NARE) Program.

Research instrumentation for the G-1 is continually being developed and upgraded as these applications occur. The most recent and ambitious instrument-development effort to date has been the modification of a tandem mass spectrometer for G-1 use. This instrument will provide the aircraft with part-per-trillion measurement capability for a large number of important pollutant species and the possibility of emission/deposition flux determination.

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Research Summary

RESEARCH AIRCRAFT OPERATIONS
BUSINESS, KENNETH M.

PACIFIC NORTHWEST LABORATORY

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10/01/93–09/30/94

Objective: To conduct airborne sampling with enhanced high-altitude and in-storm operation capability.

Product: Airborne measurement capabilities necessary to supply the diagnostic environmental measurements demanded by modern interpretive modeling techniques.

Approach: A twin-turboprop research aircraft will be provided under the direction of the Atmospheric Sciences Department, PNL. Operations will be designed to provide direct access to the aircraft for DOE projects that have the possibility for secondary access by non-DOE clients.

Results to Date: No major field programs were planned in FY 1994 to allow researchers the opportunity to analyze and publish from the extensive data sets acquired during the field programs conducted in the western North Atlantic during 1992 and 1993. Activities undertaken in 1994 relate primarily to aircraft and equipment improvements and processing of prior data sets.

Definition and operation of the restructured Research Aircraft Facility (RAF) continued with the development of methods to respond to DOE researcher requests for aircraft support. The RAF is expected to provide aircraft operations, logistics support, and data-acquisition service for at least two field campaigns in 1995.

To provide and maintain an instrument ensemble appropriate to the needs of researchers, an Atmospheric Chemistry Program Instrument Development Team was convened. The team, composed of members of the national laboratories involved in ACP studies, assesses immediate and future aircraft-instrument requirements and guides the development and/or acquisition of suitable capabilities. The resulting instrumentation will be available to users of the G-1 aircraft as "ACP community property."

Improvements were initiated (1) to modify the G-1 cabin to reduce internal heating and to provide additional space and (2) to acquire a compact navigation system capable of providing aircraft attitude parameters. Significant improvements were made to the aircraft data-acquisition system and to the postcollection-analysis workstation to enhance its speed and expand its capacity.
APPENDIX A:
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Applied Sciences Division  
Brookhaven National Laboratory  
Upton, NY 11973  
516-282-5855
APPENDIX B: ACRONYMS

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