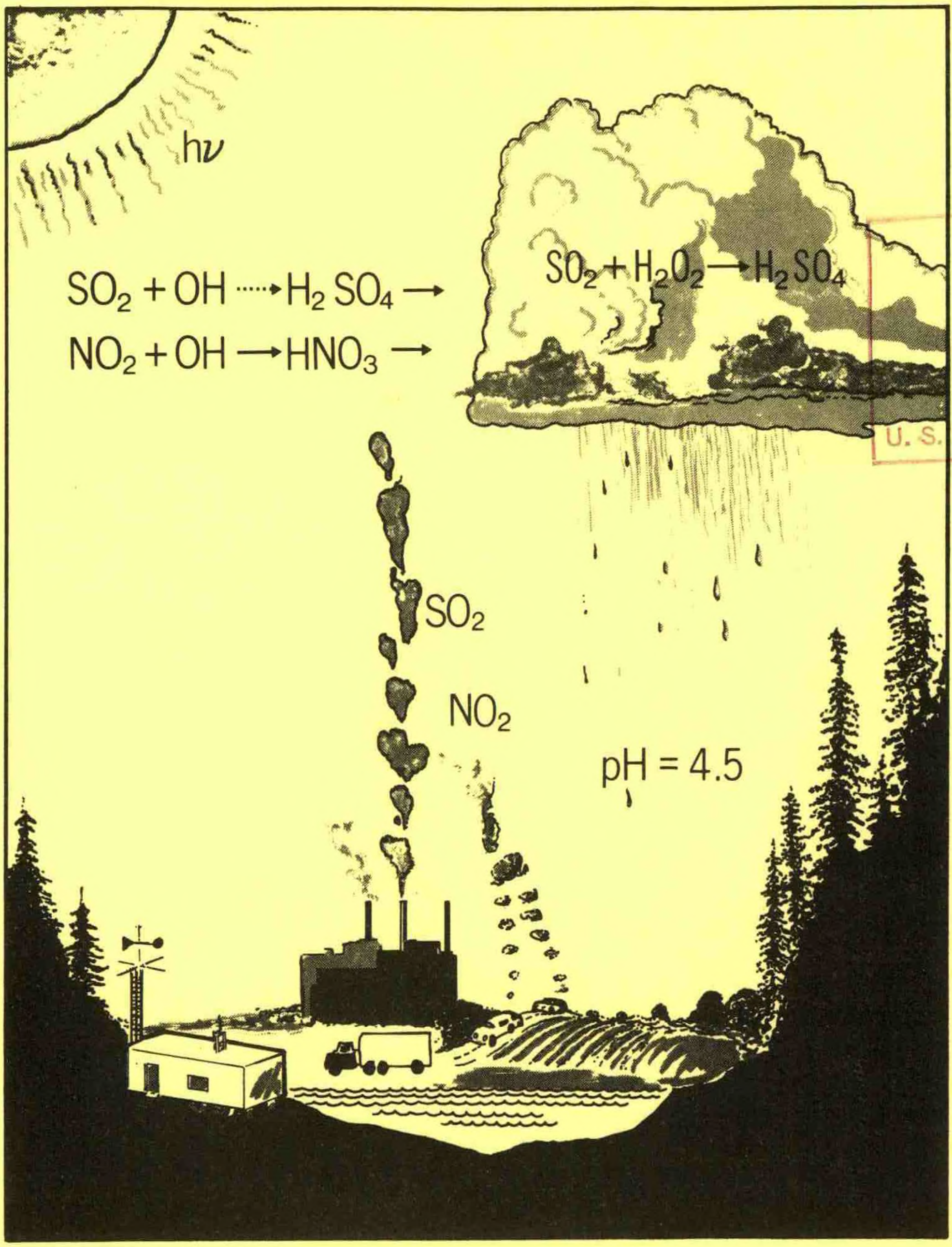
Aeronomy Laboratory

Environmental Research Laboratories

ANNUAL REPORT FY84



AERONOMY LABORATORY

ANNUAL REPORT - FISCAL YEAR 1984

October 1, 1983 - September 30, 1984

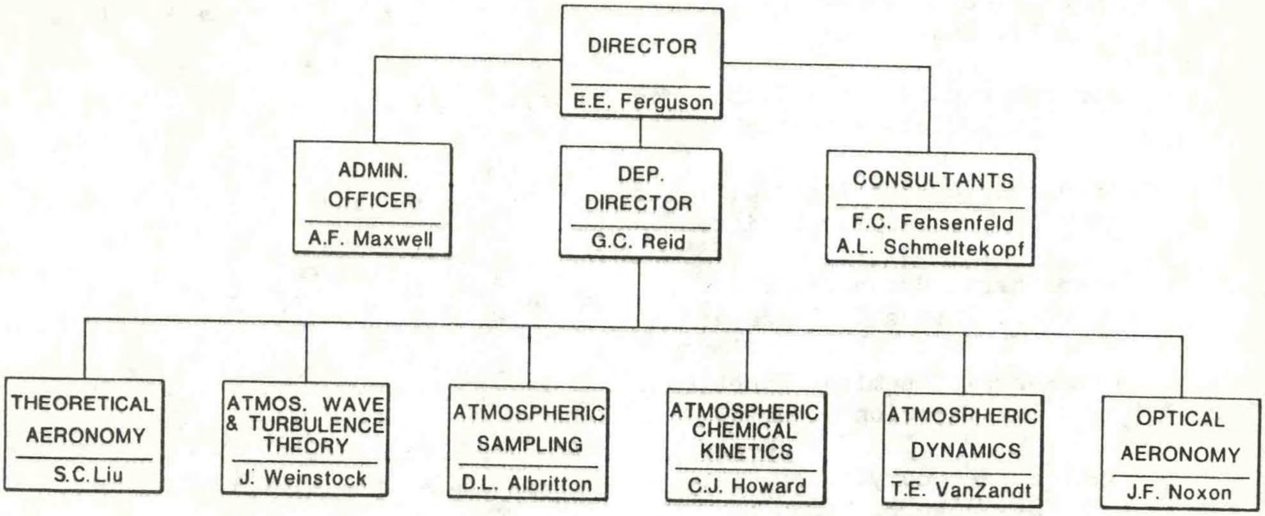
Aeronomy Laboratory
325 Broadway
Boulder, Colorado 80303
October 1984

CONTENTS

COVER

The drawing depicts a highly simplified picture of the processes leading to the formation of acid rain. Acid deposition is a serious environmental problem in the eastern regions of both the United States and Canada. Recent measurements indicate that populated areas in the western U.S. are also affected. The Atmospheric Sampling, Atmospheric Chemical Kinetics, and Theoretical Aeronomy Groups are carrying out experimental and modeling research aimed at understanding the atmospheric processes that convert acid precursors, such as SO_2 and NO_2, to acids, the natural sources of the acid precursors, and, finally, the deposition processes that remove the acids from the atmosphere. The foreground depicts one of the research trailers operated by the Atmospheric Sampling Group at the Niwot Ridge research station, a site located in the Colorado mountains about 80 northwest of Denver, Colorado.

AERONOMY LABORATORY •
ORGANIZATION CHART

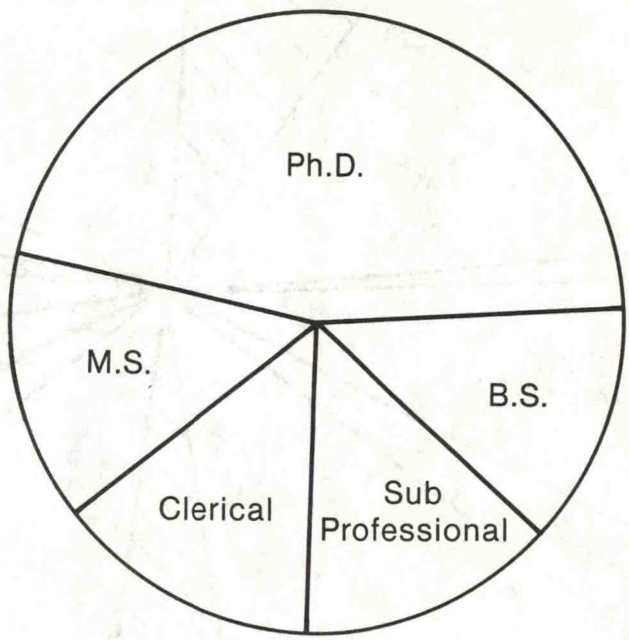


AERONOMY LABORATORY—PERSONNEL STATISTICS

FY 84 Oct. 1, 1983-Sept. 30, 1984

TOTAL FULL-TIME PERSONNEL

Professional	33
Sub Professional	6
Clerical	4
Total Full-Time Staff	43*



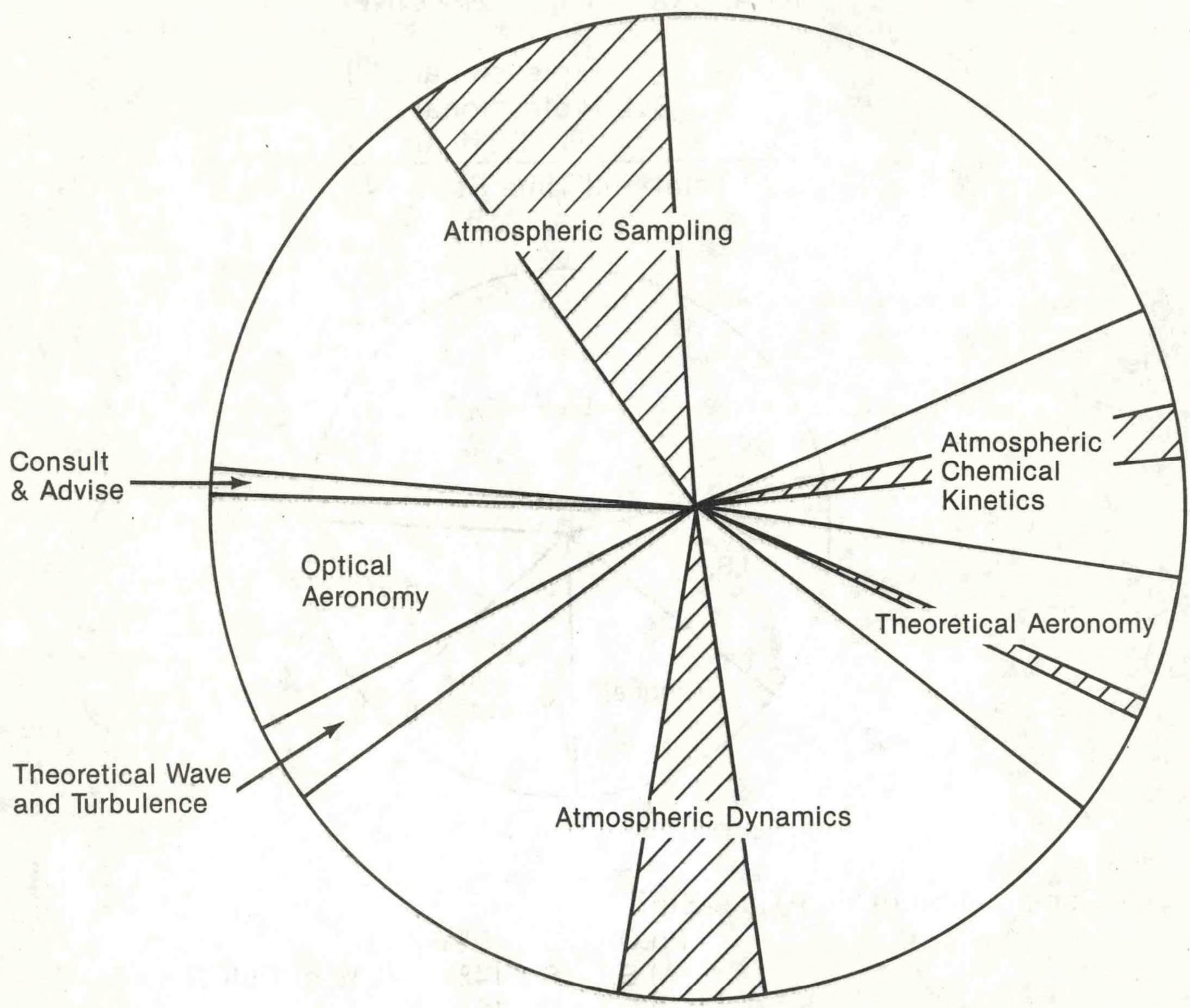
Composition of Full-Time Staff

Ph.D.	20	46%	72% of Full-Time Staff
M.S.	6	14%	
B.S.	5	12%	
Sub Professional	6	14%	28% of Full-Time Staff
(No Degree)	6	14%	

*Does not include visiting personnel

AERONOMY LABORATORY—1984 FUNDING

FY 84 Oct. 1, 1983-Sept. 30, 1984



OR and F	3,646,000	☐
Other Agencies	756,000	▨
Total Funding	4,402,000	

AERONOMY LABORATORY

Laboratory Director's Office

Permanent Staff

Eldon E. Ferguson	Director
George C. Reid	Deputy Director
DeeAnna Doerfler	Secretary
Alton F. Maxwell	Administrative Officer
Sharon R. Dorland	Administrative Assistant

Introduction

This document is an annual report on the research activities of the Aeronomy Laboratory, one of NOAA's Environmental Research Laboratories located in Boulder, Colorado. Descriptions of the various program activities are given, including some background to put the research programs into perspective as well as discussions of recent results and plans for the near future. While there is an administrative division of personnel into Program Areas, much of the research is carried out in groups by members from various Program Areas and is not compartmentalized.

The Aeronomy Laboratory has historical antecedents extending back into the National Bureau of Standards Central Radio Propagation Laboratory. The laboratory research effort in that earlier period is best described as ionospheric physics. The mission of the Aeronomy Laboratory has changed over the years due to a combination of factors: the changing nature and mission of our parent organization in the Commerce Department (NBS → ESSA → NOAA), the changing emphasis in atmospheric research due to the normal advance of science and technology, and the ever changing priorities for scientific knowledge in response to national needs. The program that has evolved encompasses research in several critical areas of atmospheric physics and chemistry in which the Laboratory has unique expertise through the skills and experience of its personnel.

The Laboratory's long tradition of excellence in the field of radar studies of the ionosphere has led to an extension of VHF coherent radar techniques to studies of winds, waves and turbulence in the neutral atmosphere, from the troposphere through the stratosphere into the mesosphere. The Atmospheric Dynamics Program has played a pioneering role in this area, and continues at the forefront of research.

The Optical Aeronomy Program is founded on the long record of leadership in the traditional areas of airglow and auroral studies, and has used its unique competence to develop powerful new techniques for remote measurement of important atmospheric constituents at all heights, and to obtain unique information on high-altitude atmospheric dynamics.

The Atmospheric Sampling and Atmospheric Chemical Kinetics Programs have evolved over a period of years out of an Atmospheric Collision Processes Program that has a long tradition of international leadership in the field of Laboratory reaction-rate measurements. The Atmospheric Chemical Kinetics Program represents an extension of the Laboratory's pioneering work in ionospheric ion chemistry to atmospheric neutral chemistry and the program has reached a level of international prominence in stratospheric and tropospheric photochemistry.

The Atmospheric Sampling Program has developed as an outgrowth of the measurement expertise and experimental technology acquired in laboratory reaction studies of *in situ* atmospheric composition measurements. The program has a position of world leadership in stratospheric composition measurements using balloon technology. This has been extended by the use of the U-2 aircraft as a stratospheric and tropospheric platform and by the use of research aircraft and ships to make global tropospheric measurements. This program has been recently augmented by a very comprehensively instrumented research site in the mountains near Boulder in which detailed measurements are being carried out with advanced new technology developed for the purpose.

The experimental and observational Programs are supported by strong theoretical programs in both photochemistry and dynamics, employing sophisticated computer models as the principal tool. These theoretical programs also carry out wide-ranging studies of problems that are of critical importance, but lie beyond the domain of the Laboratory's field programs.

The driving force behind many of the programs in recent years has been society's concern with the ozone layer, and its vulnerability to man's activities. The Aeronomy Laboratory's programs have had a major impact on our understanding of this vitally important problem of modern society. More recently, the problem of acid rain has provided the basic motivation for the tropospheric chemistry programs. The laboratory has established a leading role in research on this problem and has provided key data and manpower to the National Acid Precipitation Assessment Program.

The following pages contain detailed descriptions of the various Programs, and listings of the Laboratory's publications in calendar year 1983 and 1984.

THEORETICAL AERONOMY PROGRAM

Permanent Technical Staff

Shaw C. Liu, Program Leader	Physicist
George C. Reid, Consultant	Physicist
Carl H. Love	Mathematician
John R. McAfee	Physicist
Susan Solomon	Chemist
DeeAnna Doerfler	Secretary

Temporary Staff

Joseph M. Coughlin	CIRES Research Assistant
Eirh-yu Hsie	CIRES Research Associate
Stuart A. McKeen	CIRES Research Associate
Tallamraju, Raja	CIRES Research Associate
Michael K. Trainer	NCR Research Associate

Introduction

The objective of the Theoretical Aeronomy Program is to undertake theoretical studies of important atmospheric problems, to construct and utilize computer models of the chemistry and dynamics of the atmosphere, and to analyze atmospheric data collected within the laboratory or by collaborative experiments. In recent years the principal concern has been with problems related to the minor-constituent composition of the stratosphere and mesosphere (the middle atmosphere), deriving largely from the widespread practical interest in stratospheric ozone and its potential depletion by artificial pollutants. Recently, however, the interests of the group have expanded both downward in altitude to the complexities of tropospheric chemistry and outward in discipline toward the problems of radiative effects in the atmosphere and of the dynamics of climate. Future years should show a further expansion of these new and exciting areas.

Although the chief concern is with the middle and lower atmosphere, there is some continuing interest in the unsolved problems of the thermosphere. The ultimate goal of the program as a whole can best be described as that of attaining a sufficiently detailed understanding of the composition and energy budget of the atmosphere that accurate predictions of future trends can be made. Many aspects of the thermosphere are better understood than the corresponding aspects of the lower regions of the atmosphere since they have been explored on a global basis by such satellite missions as the Atmosphere Explorer series. To the extent that important problems of thermospheric composition or energy budget remain unsolved, and are of potential importance to the atmosphere as a whole, a continuing involvement is expected. A similar philosophy applies to problems of the atmospheres of other planets and to problems connected with the evolution of the earth's atmosphere, since studies of these topics can yield important insight into the processes operating in our own atmosphere.

In addition to these internal studies, an important function is that of providing assistance to other Laboratory programs on problems that require advanced computer programming techniques. In addition to this direct service function, strong scientific coupling exists in several areas, and the objectives of the Program are continually developed and approached in collaboration and consultation with the experimental and observational Programs.

Recent Results

Troposphere

Research in the tropospheric photochemistry centers around two major subjects: acid deposition and tropospheric ozone.

Acid deposition is a serious problem in the north-eastern U.S. and eastern Canada. Precipitation with pH in the range of 4.0 to 4.5 is quite common in these areas downwind of the mid-western heavy industrial states. Most of the anions contributing to the high acidity are $SO_4^=$ and NO_3^-.

Tropospheric ozone has been one of the major research subjects of this laboratory in recent years. Ozone plays a central role in the photochemistry that controls the abundance and interaction of most of the important trace species (e.g. CO, CH_4, H_2S, NO_2 and SO_2) in the troposphere. There is increasing evidence that tropospheric ozone may have been perturbed by the authropogenic emissions of hydrocarbons and NO_x ($NO + NO_2$). Perturbation of tropospheric ozone may cause a chain reaction that could change the distributions of the above mentioned trace gases. Since ozone and some of these trace gases absorb IR radiation in the window of CO_2 and H_2O absorption, the radiation budget in the troposphere and thus the climate may be altered. In addition, high surface ozone may damage plants and may be a health hazard.

The photochemistry and transport of acid material and ozone are closely related. For instance, the hydroxyl radical OH is produced by photolysis of O_3

$$O_3 + h\nu \rightarrow O(^1D) + O_2$$

followed by

$$O(^1D) + H_2O \rightarrow 2\ OH.$$

The gas phase oxidation of NO_2 and SO_2 are both initiated by OH,

$$NO_2 + OH \overset{M}{\rightarrow} HNO_3$$

$$SO_2 + OH \overset{M}{\rightarrow} HSO_3.$$

Furthermore, OH, HO_2, and NO_x are catalysts that control the photochemical production of ozone.

The Theoretical Aeronomy group is involved in several topics of research in the areas of tropospheric ozone and acid deposition:

(a) collaboration with the Atmospheric Sampling group on planning and interpreting measurements of NO_x, O_3, HNO_3, SO_2, particulate NO_3^- and $SO_4^=$, with emphasis on measurements made at Niwot Ridge, Colorado;

(b) collaboration with scientists at GFDL on modeling the tropospheric ozone and NO_x distributions with a 3-dimensional general circulation model;

(c) Studies of the detailed photochemistry of O_3, NO_x, OH, and hydrocarbons in a 1-dimensional model;

(d) collaboration with scientists at NCAR on developing a mesoscale air quality model for the Colorado Front Range;

(e) development of a combined liquid phase and gas phase photochemical model to study the oxidation of NO_x and SO_2;

(f) model studies of the distribution of NO_x and SO_2 that are produced from natural sources.

Collaboration with the Atmospheric Sampling Group has resulted in some very important contributions to the understanding of atmospheric processes of tropospheric ozone and acid rain. Parrish et al. (1984) investigated the ratios of HNO_3/NO_x and NO_3^-/NO_x and concluded that the lifetimes of HNO_3 and NO_3^- are both shorter than 24 hrs in the planetary boundary layer and that the model calculated OH concentration is probably too high by about 50%. Fehsenfeld et al. (1984) took advantage of the high sensitivity of the NO, NO_2 detector and the extensive data of NO_x and O_3 to deduce the background O_3 level. They proposed that the natural background O_3 level is the asymptotic value of O_3 when NO_x mixing ratio is less than 0.5 ppbv. This proposal provides an objective method to determine the background O_3 level at a rural station by measuring O_3 and NO_x simultaneously. In addition, Fehsenfeld et al. discovered that the seasonal variation of O_3 mixing ratio may be strongly influenced by anthropogenic NO_x and hydrocarbon emissions when O_3 measured in the afternoon is analyzed. However this is not the case for O_3 observed at night or in the morning because of low photochemical ozone production. This finding will help the analysis of ozonesonde data to evaluate the anthropogenic impact on the vertical distribution of ozone in the industrialized regions.

Nitrate deposition over remote oceanic regions and in the pre-industrial polar ice cores is studied in order to quantify natural background budget of NO_x, NO_3^- and HNO_3. It is shown that total nitrate deposition lies between $5x10^8$ to $20x10^8$ cm^{-2} s^{-1} which corresponds to 2 to 8 Tg(N) Yr^{-1} (Kley et al., 1984). Of particular interest is the nitrate deposition in the ice core in the polar regions, post-industrial values in Greenland are about a factor 2 greater than the pre-1900 values while there has been no detectable change in Antarctica. These data can be used to evaluate the long range transport of nitrate. Seasonal variation in the polar ice core nitrate deposition shows a clear strong summer maximum. The nitrate deposition fluxes over remote oceanic regions are not correlated with either ^{222}Rn or dust particles, indicating a non-continental NO_x source. It is concluded that the most likely source is NO produced by lightning in the upper troposphere.

The total lightning source is estimated to be about 10 to 20 Tg(N) Yr^{-1}. Based on the satellite lightning frequency, nitrate deposition flux due to lightning in North America can be estimated to be about 5 to 10 per cent of the anthropogenic value.

The atmospheric-biospheric fixed nitrogen budget and chemical transformation is discussed by Liu and Cicerone (1984) and Davis et al. (1984). Major uncertainties in the source of NO_x identified are emissions from soil biogenic activities, lightning, and biomass burning. Chemistry involving organic nitrate, NO_3, and N_2O_5 in both homogeneous and heterogeneous phase is poorly understood. Long range transport of organic nitrate may be important for NO_x budget in the remote areas. Modeling the global impact of anthropogenic emissions of relatively short lived species such as NO_x and some of the hydrocarbons needs to interface models in the global scale (i.e. GCM) and mesoscale (Dickinson and Liu, 1984).

Development of the mesoscale air quality model has progressed as planned. A 2-day mesoscale meterological model has been run for the case of July 26, 1983. There was an upslope wind during that day and the Niwot Ridge station was in full operation. The domain contains the whole western US with 60 x 60 km resolution with finer resolution (20 x 20 Km) for Colorado. Tracer experiments have been run with a simple mass conservation scheme with satisfactory results.

The transport of tropospheric ozone has been investigated by a 3-dimentional GCM simulation in a collaborative effort with scientists at GFDL (Levy et al., 1984). The model assumes that ozone is transported from the stratosphere and deposited at the surface. Many of the observed spatial and temporal variations of ozone have been successfully simulated by the model, including seasonal and latitudinal variations in both absolute concentration and relative changes. There are also many disagreements between modeled results and observed values. Latitudinal distribution above 50^{o}N indicates that there is a major defect in the model meridional transport. Continental surface ozone in the industrialized areas is too low, suggesting the need of including photochemical production of ozone.

The effect of condensation on the energetics of the frontogenesis is studied by evaluation of the kinetic energy budget. The latent heat of condensation generates available potential energy on the mesoscale and enhances the conversion of zonal available potential energy to eddy available potential energy by enhancing the north-south component of flow across the front. It also enhances the conversion of eddy available potential energy to eddy kinetic energy by producing a stronger direct secondary circulation in the moist simulation.

The diagnostic Sawyer-Eliassen equation is used to partition the forcing responsible for the generation of the ageostrophic secondary circulation. The processes that produce this circulation are geostrophic shearing deformation, vertical exchange of heat and momentum, and latent heat release.

The Sawyer-Eliassen diagnostic studies revealed a number of facts concerning the generation of the secondary circulation. The major findings are: (1) The large-scale features of both the moist and dry ageostrophic circulations are generated by geostrophic deformational forcing. (2) A jet of vertical velocity observed ahead of the surface cold front in the dry simulation is the result of forcing by both deformational and frictional processes. (3) The magnitude and structure of the vertical motion field in the moist case are produced primarily by latent heat release. (4) The increase in the horizontal ageostrophic circulation observed in the moist case is due to combined effects of deformation and latent heating. (5) Because of a thermally insulated lower boundary, the turbulent exchange of heat had very little influence on the generation of the secondary circulation. (Hsie et al., 1984a; 1984b; Baldwin et al., 1984)

Middle Atmosphere

Chemical-dynamical studies of the middle atmosphere have continued, with an increased focus on the photochemistry and transport of ozone in both the stratosphere and mesosphere, and on the chemistry of stratospheric chlorine compounds. These studies have been performed in collaboration with Rolando Garcia of the National Center for Atmospheric Research, using a two-dimensional residual Eulerian model that extends from 16 to 116 km altitude, from pole to pole. The advantage of the residual Eulerian framework can be briefly summarized as follows: It can be shown that when the classical Eulerian mean and eddy transports are computed self-consistently, a large cancellation occurs such that the remaining net transport in the stratosphere is a small residual. Problems can arise in photochemical modeling because often neither of the two terms is computed at all; rather the eddy transports are parameterized by eddy diffusion coefficients and the mean circulation is taken from a dynamical model study. Thus, the eddy coefficients may not be consistent with the adopted mean circulation, and the appropriate cancellation between the two may then not be achieved. Recent dynamical studies have shown that the cancellation problem can be alleviated by using the residual Eulerian or diabatic circulations, which represent the desired net transport in the stratosphere without the need for eddy-mean flow cancellation, provided that the eddies are approximately steady and conservative.

A shortcoming of this approach, however, is the question of the role of transient, dispersive eddies in the transport of chemical constituents. While steady conservative eddies do not appear when the dynamical equations are cast in the residual Eulerian representation, some degree of eddy transience/mixing must occur in the stratosphere. A great deal of recent work in the field has focused on the elucidation of the importance of these mixing effects. One approach to the problem is to examine the computed and observed distributions of chemical tracers such as N_2O, CH_4, $CFCl_3$, etc. N_2O and CH_4 are particularly attractive as tracers because satellite data on their global distributions has just become available. We have therefore added chlorine chemistry to our model and have compared results for the chlorofluorocarbons, N_2O, CH_4 to available data. We find that relatively small vertical and horizontal mixing coefficients (about $3x10^9$ cm^2s^{-1} in the horizontal and $1x10^3cm^2s^{-1}$ in the vertical) provide the best calculated distributions of these diverse tracers as compared to the ensemble of observations (Solomon and Garcia, 1984b).

In agreement with available satellite and in-situ data, our model results indicate that a substantial latitude gradient in atmospheric methane occurs near 40 km, with tropical values that are about two to three times greater than those obtained at mid-latitudes. This is a result of upward transport from the methane-rich troposphere in the tropics, and downward, poleward transport at higher latitudes, by the computed mean meridional circulation. These spatial variations in methane influence the partitioning of chlorine between HCl (an inert reservoir) and ClO (a free radical that catalytically destroys ozone). Thus, the distribution of ClO depends in turn on the methane distribution (particularly near 35-40 km), with associated effects upon the chlorine catalyzed destruction of ozone. Further, observed local variability in methane at middle latitudes is consistent with much of the observed variation in stratospheric ClO near 40 km. We have shown that spatial and short-term temporal variability in methane has potentially important consequences for the HCl and ClO distributions in the stratosphere, and their local variability, as well as for ozone densities (Solomon and Garcia, 1984b).

A particularly fruitful application of our two-dimensional dynamical chemical model has been in the interpretation of satellite data, both from the Solar Mesosphere Explorer (SME) satellite, and from the Limb Infrared Monitor of the Stratosphere (LIMS) experiment onboard NIMBUS 7. The latter experiment revealed the presence of extremely large mixing ratios of NO_x in the polar night mesosphere, and a gradual accumulation of polar mesospheric NO_x throughout the winter. Garcia and Solomon (1983) and Solomon and Garcia (1984a) previously suggested that downward transport of thermospheric NO_x could lead to such an enhancement in NO_x at mesospheric and perhaps even stratospheric levels. If the thermospheric NO_x could reach the stratosphere, it might even influence stratospheric ozone abundances, providing a mechanism for long-range thermosphere-stratosphere coupling. The observations obtained by the LIMS experiment provide striking evidence of downward transport of thermospheric NO_x to mesospheric levels, and suggest that the upper stratosphere is also affected by downward transport poleward of about 60° in winter. We have discussed and interpreted these satellite data in detail (Russell et al., 1984).

Collaboration with the Solar Mesosphere Explorer (SME) satellite team has continued. We have used the observed distribution of NO_2 to infer the N_2O_5 distribution in the stratosphere (Solomon et al., 1984), with the hope of providing theoretical information about N_2O_5 to aid experimental efforts to detect it in the stratosphere. We have also continued our study of mesospheric ozone data from SME. A particularly puzzling aspect of the SME data was the observation of pronounced seasonal oscillations near 80 km, with maxima at equinox that are about twice as large as the observed abundances at summer and winter solstice. Purely photochemical and temperature effects would tend to produce maxima at solstice, not equinox, so we were led to pursue a dynamical explanation for the observed features. Recent work has suggested that breaking small scale gravity waves play an important role in the dynamics of the mesosphere. We have incorporated a parameterization of the propagation and dissipation of gravity waves into our dynamical-chemical model. This parameterization is used to compute both the momentum forcing and turbulent diffusion induced by the waves at mesospheric altitudes, providing the needed transport parameters for the photochemical constituents in the model. We find that the structure of the observed equinox maximum in ozone near 80 km is consistent with our theoretical results when the seasonal and latitudinal variations in turbulent diffusion induced by such waves are considered (see, Thomas et al., 1984). This work suggests that observations of mesospheric ozone may have important applications in furthuring our understanding of mesospheric dynamics.

Atmospheric Dynamics and Climate

Collaborative studies with the Atmospheric Dynamics Program of the properties and variability of the tropical tropopause have continued using the existing data base of radiosonde measurements made at tropical stations over the past 30 years or more. The principal results are as follows:

(1) The correlation in the interannual variations of tropopause heights at different stations is excellent for stations within about 10° of the equator, but falls off substantially between 10° and 20° latitude.

(2) There is a significant correspondence between the height of the tropopause and the phase of the quasi-biennial oscillation in the winds of the tropical lower stratosphere; the phase relationship is qualitatively consistent with the existence of the vertical motions needed to maintain geostrophic balance in the time-varying winds.

(3) The height of the tropopause is positively correlated with the sea-surface temperature anomalies of the eastern tropical Pacific Ocean and hence with the phase of the Southern Oscillation; in particular, tropical tropopauses tend to be high all over the world during El Nino years.

(4) Interannual variations in average tropical tropopause height are positively correlated with interannual variations in the total global angular momentum of the atmosphere (Reid and Gage, 1984); such a correlation could have been predicted from current theories of the general circulation of the atmosphere and angular momentum transport, but had not been observed before.

(5) A pronounced periodicity of about 20 days has been found in the height of the tropopause in the western Pacific at certain times of year; the same period appears to be present in the winds near the tropopause, and its relationship to the periodicities reported by others is under active investigation.

(6) The relationship between deep cumulus convection, tropopause height, and troposphere-stratosphere exchange is being studied; hopefully this will help to shed light on some basic aspects of atmospheric dynamics, and will lead to a sounder basis for estimating the response of the global atmosphere to such external influences as changes in solar radiation or changes in radiative heating brought about by changing concentrations of carbon dioxide and other trace species.

Future Plans

Troposphere

Tropospheric ozone and its possible perturbation by anthropogenic activities will continue to be one of the major subjects of our research. Important problems in this area are the photochemical production and destruction of O_3, transport of O_3, the distribution of tropospheric NO_x, OH, and RO_2 radicals, and the effects of nonmethane hydrocarbons. We will continue to study these problems by working closely with the Atmospheric Sampling group and the Atmospheric Chemical Kinetics group. Collaboration with scientists at GFDL on 3-dimensional modeling will be strengthened in both stratospheric and tropospheric modeling.

Studies of the acid deposition problem will be expanded. Emphasis will be on atmospheric transformations of SO_2 and NO_x, heterogeneous processes, and natural emissions of sulfur and nitrogen compounds. Developing a regional acid deposition model for the Colorado Front Range is a long-range goal for this group. The model will be very useful for interpreting the data at Niwot Ridge and for designing other measurement strategies. It will consist of a mesoscale meteorological model and a photochemical model. It is clear that such a model can be readily applied to study regional oxidant problems such as that of rural O_3. This model will be developed in collaboration with scientists at NCAR.

Middle Atmosphere

The interaction of dynamics and chemistry in the middle atmosphere represents an important element in our understanding of aeronomy. We plan to continue to pursue our studies of the natural and perturbed stratosphere and mesosphere. We hope to concentrate on the effects of future chlorine perturbations on stratospheric ozone, and to begin to include a more detailed treatment of infrared radiation in both the mesosphere and stratosphere. The latter goal should eventually lead to a coupled radiative/dynamical/chemical model, and we anticipate that such studies will lead to a more detailed understanding of the middle atmosphere and its response to perturbations.

Atmospheric Dynamics and Climate

The study of the tropical tropopause region using radiosonde data will continue. Emphasis will be on (1) further development of the conceptual picture of troposphere-stratosphere interaction in the tropics, (2) a thorough investigation of the 20-day periodicity in tropopause heights and winds in the western tropical Pacific, and (3) a refined and more complete study of the correlation between tropopause height and global atmospheric angular momentum, aimed at exploring the cause-and-effect relationship. The connection between tropopause properties and the wind fields of the tropical lower stratosphere and upper troposphere will be investigated.

PUBLICATIONS 1983

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ATMOSPHERIC WAVE AND TURBULENCE THEORY PROGRAM

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Introduction

This program is devoted to theoretical studies of turbulence, waves, and transport in the atmosphere, with an emphasis on flexibility and with a direction toward problems that are challenging and timely. Although small in terms of staff and funding, the program has had a very high output, and as a result it has attracted a number of visiting scientists and post-doctoral research associates over the years.

The program grew out of an earlier program aimed at understanding and predicting turbulence and wave fluctuations in the plasma environment of the earth's ionosphere and upper atmosphere. This original program was developed in response to the mission of the Aeronomy Laboratory as part of the Central Radio Propagation Laboratory of NBS, and was designed to provide the necessary theoretical background for understanding and predicting the behavior of the ionosphere, and its influence on radio propagation. The more recent development of the program has taken place in response to the changing mission of the Laboratory as part of NOAA, and the basic theme has been the application of the theoretical expertise and techniques developed in the studies of plasma turbulence to the problems of turbulence and wave fluctuations in the lower and middle atmosphere.

Turbulence is widespread in geophysical fluids, and it often has a dominating influence on transport. The conceptual and mathematical difficulties associated with turbulence theory, however, are notorious, and until recently there have been no satisfactory theories available to determine the strength of the fluctuations produced by nonlinear wave interaction and turbulence. In the case of the ionosphere, this program provided the first, and often the only, formulas for predicting the strengths of irregularities in plasma density, temperature, electric field, and other important parameters. Confirmation of the theory has been provided in many cases by optical and radar measurements and by direct rocket observations.

Specific accomplishments of the original ionospheric turbulence program included the development of the Dupree-Weinstock theory of strong plasma turbulence, and of a theory of strong turbulence in fully ionized gases in a magnetic field that is valid at all frequencies. Theories were developed to allow the prediction of the strength of irregularities produced by plasma instabilities in the ionsophere and of the modification of the ionosphere by strong radio transmitters. Contributions were also made to the theory of laser heating of laboratory plasmas.

As mentioned above, the program in recent years has been devoted to the investigation of turbulence and waves in the neutral atmosphere, applying the novel approaches that were so successful in the area of plasma turbulence. Among the accomplishments have been the development of a comprehensive nonlinear theory of atmospheric gravity waves, which are important contributors to transport and dynamics throughout the atmosphere, a theory of turbulence in the buoyancy subrange of stably stratified shear flow, and a theory of the influence of gravity waves on airglow emission from the atmosphere. Observations made by the Optical Aeronomy program confirmed the theoretically predicted oscillations in the $O_2(^1\Sigma)$ airglow, showing that gravity waves were indeed responsible through the oscillations in temperature that they produce (See Figure 1). The theory can now be extended to other airglow emission, such as those of OH and O.

Other major investigations have included studies of the vertical propagation of a broad spectrum of gravity waves from the troposphere to the mesosphere, the development of a relationship between turbulence energy dissipation rate and turbulent diffusivity under conditions of stable stratification, and development of a theory that relates radar measurements of the refractive index structure constant C_n^2 to the energy dissipation rate (See Figure 2). The observed correlation between wind shears and turbulence layers was also explained quantatively (See Figure 3).

Recent Results

The following are the principal recent results of the program:

(1) It has been proven that diffusion, friction, momentum deposition, and heat flux are all interrelated in a simple way. All of these transport parameters are needed for dynamical models of the middle atmosphere.

(2) A simple algorithm for nonlinear gravity waves has been developed for use by non-experts.

(3) A theoretical calculation of the pressure-strain relationship in shear flows has been made for the first time. The theory is fundamental in modeling the planetary boundary layer.

(4) Diffusion due to gravity waves in the 30-100 km height range was modeled, tending to establish that gravity waves are the major cause of diffusion in the mesosphere (See Figure 4).

(5) Theory of the interaction of gravity waves with turbulence in the ocean and atmosphere is being developed. The theory explains the observations of a numerical experiment carried out recently at GFDL to study this interaction.

(6) A theoretical calculation was made of the heat flux caused by nonlinear gravity waves in the middle atmosphere. This flux was shown to actually cause cooling, and has potentially important implications for the circulation of the middle atmosphere.

(7) A theoretical formula was developed to predict the kinetic energy of clear-air turbulence (CAT) from a knowledge of dissipation rates, which can be obtained from radar measurements.

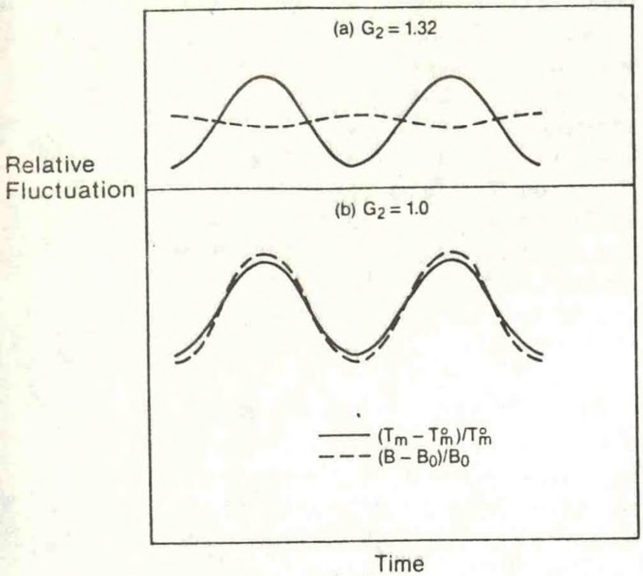


Figure 1. Theoretical ratio of $O_2(^1\Sigma)$ brightness fluctuations $B - B_o$ to temperature fluctuations $T_m - T_m^o$ for (a) large amplitude gravity waves and (b) small amplitude gravity waves. Both kinds of ratios are observed (Noxon, 1978).

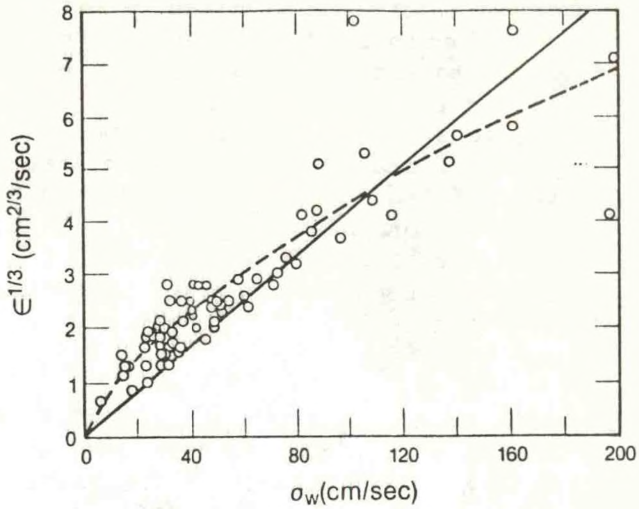
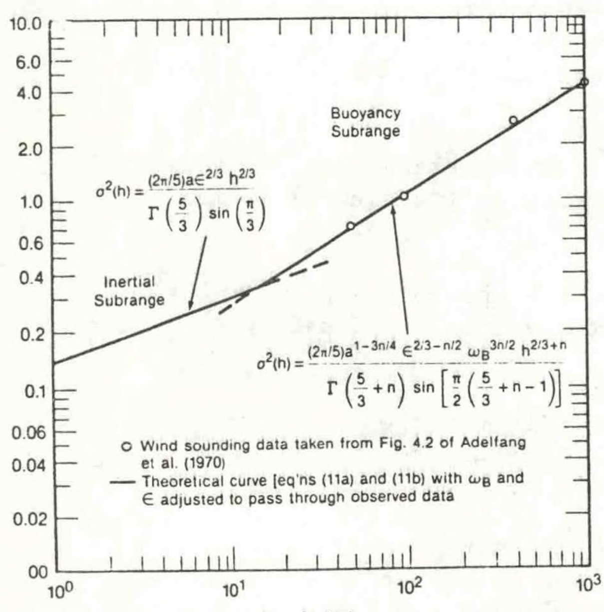


Figure 2. Turbulence dissipation rate ε versus vertical RMS velocity σ_w. A preliminary theory is given by the dashed line -----. The observations in the stratosphere are given by circles O.



ure 3. Turbulence RMS energy $\sigma(h)$ versus layer thickness h. Our theoretical prediction is given by the solid lines ——→ and the observations from rockets and balloons are given by the circles O. Layer thickness less than 50m are not adequately resolved by balloons or rockets, and their turbulence properties must be determined by theory (i.e., theoretical extrapolation).

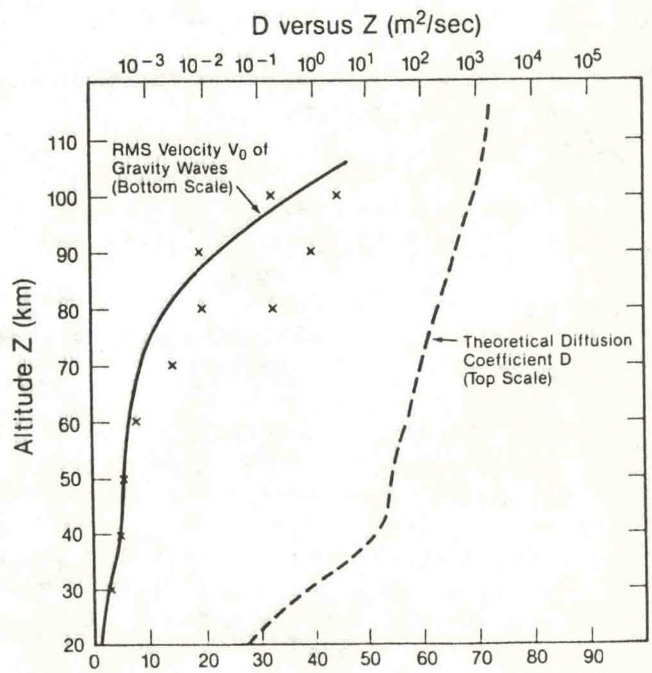


Figure 4. Theoretical curves of RMS gravity wave velocity V_o and diffusion coefficient D versus altitude Z. The small crosses are observed values of V_o.

(8) The major program to develop turbulence models of the planetary boundary layer has been strengthened and extended by financial support from and personal collaboration with the Naval Environmental Prediction Research Facility (NEPRF) at Monterey, California. Use of their boundary layer model codes and of their computing facilities is allowed under this arrangement.

Recent research progress was aimed at determining the influence of pressure correlations on the mean flow. It was discovered that such correlations could decrease the development of turbulence along the direction of the main flow, a result that was unexpected and that could explain some observations by Wyngaard (1979).

(9) Proved that gravity waves "break" in a manner resembling the surfing of ocean waves, and that this "breaking" is the principal process by which waves cause transport in the atmosphere.

(10) Determined the buoyancy subrange spectrum of temperature fluctuations in atmosphere and oceans, and corrected a commonly quoted twenty year-old error in the literature concerning such spectra.

(11) Proved that "return to isotropy" -- the principal hypothesis of turbulence model -- is unvalid and developed a theory to determined realistic deviations from isotropy.

(12) Theoretically determined how observed height variations of gravity wave amplitudes can be used to infer eddy diffusivities in the middle atmosphere.

(13) Predicted that gravity waves cause diffusion to be anisotropic (with horizontal diffusivities greatly exceeding vertical diffusivities).

(14) Explained why vertically towed grid turbulence experiments differ from horizontally towed experiments, and how each are related to atmospheric turbulence phenomena.

(15) Discovered a new mechanism by which gravity waves generate thin layer of turbulence observed in oceans and atmosphere, namely a dynamical instability caused by nonlinearly steepened wave shear. This mechanism might provide the widely sought-after "sink" of fluctuation energy in oceans.

Future Plans

Future plans are divided into two major program areas: (1) theory of turbulence, wave enhanced diffusion, friction, and heat transport in the troposphere, stratosphere and mesosphere; and (2) a rigorous theoretical model of the atmospheric boundary layer; this also has important, practical engineering applications because of the great present need of reliable turbulence models in so many areas of our industrial complex.

Future research goals of these programs areas are as follows:

1. Theoretical investigations on fluid turbulence, waves, and enhanced transport in the atmosphere. Research activities (not in order of priority) will be

(a) Development of a comprehensive theory of turbulence in stably stratified flows, with relevance to both the atmosphere and the oceans. A goal is to explain the velocity spectra observed in the oceans and atmosphere.

(b) Development of a nonlinear theory of wave breaking in atmosphere and oceans.

(c) Studies of turbulent and wave diffusion in the stratosphere and troposphere.

(d) Deduction of the turbulence state in the lower atmosphere from Radar measurements and from measurements of temperature fluctuations.

(e) Determination of the role of tidal waves in atmospheric eddy diffusion. Derivations of a nonlinear theory of tidal waves for this purpose.

(f) Determination of the contribution of gravity waves to atmospheric diffusion from 20 km to 100 km and modeling of the upward propagation of gravity waves from the stratosphere to the theormosphere.

(g) Modeling the profile of momentum flux and friction needed to predict the mean circulation in the middle atmosphere. Determine if the Rayleigh friction assumed by mean circulation models is theoretically justifiable.

(h) Fundamental study of inertial range turbulence in incompressible fluids (Kolmogoroff turbulence). The recently developed Three-Point Method will be used to obtain a rigorous self-contained Test Field Model.

(i) Modeling the cooling of part of the middle atmosphere by gravity waves.

(j) Explanation of the universal vertical spectra of velocity and temperature found in atmospheres and oceans.

(k) Determine the formation of steady winds by gravity waves.

(l) A study of gravity waves and transport in the mesosphere and thermosphere based on Noxon's observations of OH and $O_2(^1\Sigma)$ airglow--see Figure 1 for example of previous work.

2. Major comprehensive program to develop a theoretical model of the earth's boundary layer (BL) (this model also has industrial applications). Until now boundary layer models have relied on ad hoc, heuristic considerations to determine the influence of pressure fluctuations and turbulent diffusion on boundary layer flows. These models are often not satisfactory, and there is no way of knowing when their forecast will be accurate or not. The theoretical model will determine the influence of fluctuations of pressure, temperature, and humidity from first principles, and the resulting forecasts are expected to be more accurate and reliable than existing models. This research is a new direction in modeling. It is based on contemporary methods of statistical physics and turbulence theory. Our goal is to supply the theory needed to make turbulence modeling succeed.

Specific research goals are:

(a) To model the pressure-strain rate in the neutral boundary layer, correct the Rotta model, and to calculate the influence of terms nonlinear in the velocity shear;

(b) To extend the pressure-strain rate theory (developed last year) to stable and unstable atmospheric conditions. This means including the effects of buoyancy on the pressure-strain terms needed for modeling. A theory of pressure fluctuations (the pressure-strain rate) is vital for modeling because such fluctuations cannot be measured directly (except near a wall) and yet have a profound influence on the mean flow circulation;

(c) To theoretically model buoyancy (heat) flux, kinetic energy flux, pressure flux, momentum flux, humidity flux, mechanical energy dissipation, and heat dissipation from first principles -- for neutral, stable and unstable conditions. These flux and dissipation quantities are all required for a model of the boundary layer, although, as of now, they are not known in other than an ad hoc, unsatisfactory, empirical way -- analogous to eddy transport terms.

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ATMOSPHERIC SAMPLING PROGRAM

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ATMOSPHERIC SAMPLING

Introduction

The origins of the present Atmospheric Sampling Program lie in the recognition that man's activities may inadvertently pose a threat to the earth's stratospheric ozone layer, which serves as a protective shield from harmful solar radiation. This potential threat arose because of the possibility that the chlorofluoromethanes released from spray cans would diffuse upward into the stratosphere and would there be photolyzed into chlorine, which could then catalytically destroy ozone at these altitudes. Since the loss of even a fraction of the stratospheric ozone may produce disastrous consequences in the biosphere, this potential environmental threat attracted immediate and widespread attention. However, despite the seriousness of the possible problem, the severe economic dislocations that would follow an immediate cessation of the large chlorofluoromethane industry prohibited taking such an action on the basis of hypothesis alone.

The Atmospheric Sampling Group was formed to address this critical problem. The research effort mounted by the Group led to the first successful measurements of the chlorofluoromethanes at the altitudes in the stratosphere where these compounds are significantly photodissociated. The findings supported the predictions from theoretical models concerning the photochemistry of these compounds and, hence, the predictions of the potential adverse consequences to stratospheric ozone. Subsequent measurements by this group provided a comprehensive set of data describing the distribution of the chlorofluoromethanes in the stratosphere, which has proven useful not only to policy formation regarding these pollutants but also to serve as tracer information for atmospheric dynamics studies.

The approach used in these stratospheric chlorofluoromethane measurements has guided the scientific efforts of the Group since that time. The problems that are selected are those that combine significant new scientific research with important national or global atmospheric environmental questions. The instruments and techniques required in the studies are generally conceived, designed, and developed within the Group and are subjected to rigorous laboratory and field validations. The subsequent field application of these instruments and techniques employ a variety of platforms: balloons, stratospheric and tropospheric aircraft, ships, vans, and semipermanent ground stations. The field campaigns are conducted by the Group's engineers and scientists who developed the instruments. These research efforts are undertaken in close collaboration with other groups in the Aeronomy Laboratory, such as Theoretical Aeronomy and Optical Aeronomy, and with other laboratories, such as the National Center for Atmospheric Research and the National Aeronautics and Space Administration.

The experience, skills, and interests of the Group have expanded considerably since the initial stratospheric chlorofluoromethane studies and now encompass a broad range of topics in atmospheric chemistry; e.g.:

the natural emissions that contribute to atmospheric acidity and alkalinity,

the transport, transformation, and deposition processes involved in acid deposition,

the tropospheric/stratospheric exchange processes that are a factor in regulating stratospheric and tropospheric chemistry and climate, and

the tropospheric and stratospheric photochemical cycles responsible for the production and destruction of global ozone.

Several key environmental issues are being addressed. All of them involve man's potential inadvertent deleterious alteration of the earth's atmosphere: stratospheric ozone depletion, acid deposition, tropospheric ozone production, and the greenhouse effect. As such, this research figures strongly in the NOAA/ERL Air Quality and Climate Programs.

Recent investigations undertaken by the Group and its future plans are summarized below.

Recent Results

1. Total Reactive Nitrogen Measurements

During the summer of 1984, the abundance of the sum of all of the reactive nitrogen species, NO_y, at a tropospheric site was determined for the first time. The reactive nitrogen species play important roles in tropospheric chemistry and climate, and the capability to detect their overall abundance opens up attractive prospects for determining global budgets of this key chemical family. The measurement site was located near Niwot Ridge, Colorado, at an altitude of 3000 m. NO_y was detected with a new technique developed recently in the Group for that purpose. It uses the reduction of these reactive nitrogen compounds by carbon monoxide, CO, at a heated gold catalyst to yield NO, which is then detected by the Group's sensitive chemiluminescence detectors. The air quality at the site varied from clean to moderately polluted due to transport of air from the Denver metropolitan area, thus the NO_y chemistry could be examined over a range of conditions. Correlations involving NO_y with other species measured at the same time demonstrated the basic relations of the chemistry and transport, such as the age of the air mass and the role of the nocturnal boundary layer, in determining the production and deposition of NO_y.

In addition to the sum of the reactive nitrogen species, the summer investigation at Niwot Ridge also included separate measurements of a number of the reactive nitrogen species, many measured in concert for the first time. Knowing both the sum of all of the reactive nitrogen species,

NO_y, and the sum of many of the individual components, $\Sigma(NO_y)_i$, provided new insight into the partitioning among the members of this chemical family. Nitric oxide, NO; nitrogen dioxide, NO_2; and nitric acid, HNO_3 were measured by the Group's instruments, and peroxyacetyl nitrate, PAN, concentrations were determined by SRI International and the National Center for Atmospheric Research. It was observed that these four species constitute a large fraction of NO_y, but not 100%, indicating that additional reactive nitrogen species are significant. This shortfall is shown in Fig. 1. Over a wide range of NO_y values, the fraction represented by NO, NO_2, PAN, HNO_3, and NO_3^- was typically only 0.4 to 0.7.

PAN proved to be comparable to NO and NO_2, an important fact established for the first time by this series of measurements. Diurnal and seasonal behavior of the ratios showed the photochemical behavior predicted for the formation and loss of HNO_3. For example, nitric acid and NO_x (NO + NO_2) have been measured now at all times of day during all seasons. The ratio of HNO_3 to NO_x concentrations is observed to rise during the day and to decrease at night. For each season, this diurnal pattern can be well fit by modeling the production of HNO_3 from NO_2 by combination with hydroxyl radicals and the heterogeneous removal of HNO_3. The conclusion is that HNO_3 has a very short lifetime in the troposphere (~12 hours in summer and ~24 hours in winter) and that surface deposition is the primary removal process.

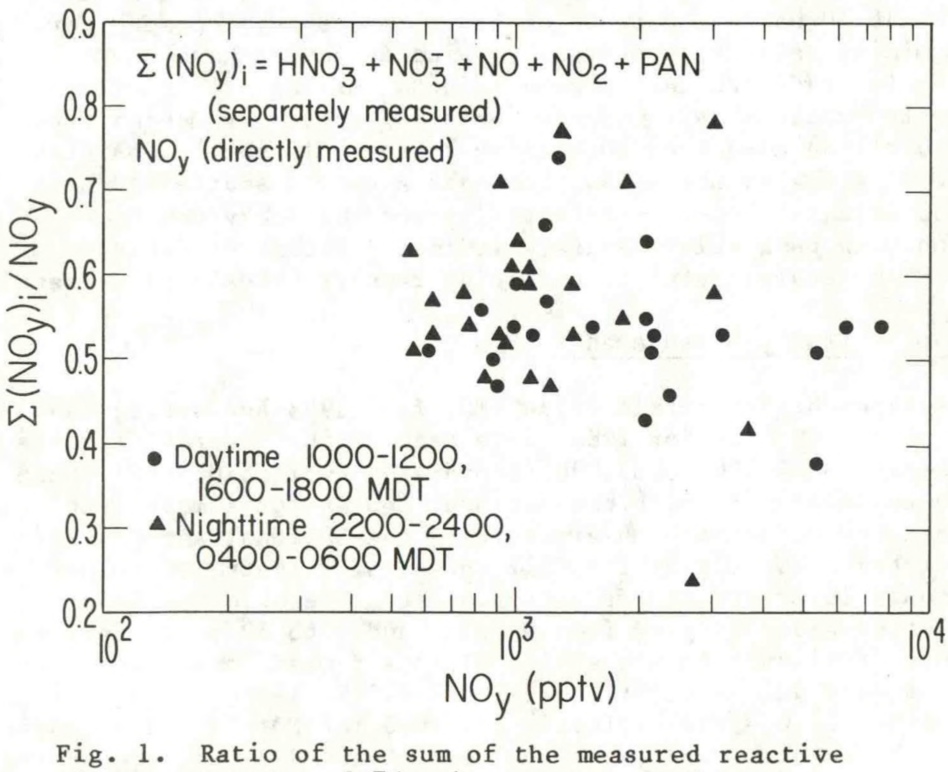

Fig. 1. Ratio of the sum of the measured reactive nitrogen species, [$\Sigma(NO_y)_i = HNO_3 + NO_3^- + NO + NO_2 + PAN$], to the measured total reactive nitrogen, NO_y. The data were taken at Niwot Ridge, Colorado, during the summer of 1984.

2. Organic Acids in Precipitation

Formate and acetate, as well as other organic and inorganic anions, have been measured in precipitation collected at Niwot Ridge (a remote site) and Boulder (an urban site). The organic anion concentration is usually at least 20% of the nitrate concentration and occasionally is equal to the nitrate. Formate is the dominant organic anion measured with concentrations as large as 9×10^{-5} M occurring in summer rain showers. A variety of dicarboxylic anions are observed also, but their concentration is generally much less than formate. The total ion concentration is usually less at Niwot Ridge than at Boulder. However, ionic balance often leads to a somewhat lower pH at Niwot Ridge. Organic acids have been observed in precipitation previously in remote oceanic areas. The present observations show that they can be significant contributions to the acidity in urban and rural continental areas.

3. Background Sulfur Studies

Accurate assessment of the contribution of sulfur bearing species to acid rain in "clean" continental air requires measurement below 0.1 parts per billion by volume (ppbv) in locations where local anthropogenic contributions are usually absent and clearly recognizable. Measurements of surface tropospheric mixing ratios of sulfur dioxide, SO_2, have been carried out as part of the measurements at Niwot Ridge. The investigation was conducted using an automated portable gas chromatograph with a detection limit of about 10 parts per trillion by volume (pptv). Strong correlations of SO_2 mixing ratio with prevailing wind direction have been observed. Westerly winds frequently result in SO_2 mixing ratios below 10 pptv. Easterly "upslope" air movement bringing urban air masses from the Denver metropolitan area show SO_2 mixing ratios > 1 ppbv. Positive correlation of SO_2 with gas phase HNO_3 bespeaks a common source and has implications for acid rain on the eastern slope of the Rocky Mountains. Weak correlation with particulate sulfate indicates either differing sources, a slow interconversion rate, or mediating factors not always present.

4. Free-Tropospheric NO_x Measurements

In-situ measurements of nitric oxide (NO, fall 1983 and spring 1984) and nitrogen dioxide (NO_2, spring 1984) were made during aircraft flights at altitudes ranging from 500 to 33,000 ft over the Pacific Ocean. These studies were in collaboration with the National Center for Atmospheric Research and were conducted with an aircraft of the National Aeronautics and Space Administration. During the fall series of flights, NO values in the marine boundary layer and the free troposphere were observed to be extremely low, with values ranging from 0 to 10 and 0 to 50 pptv, respectively. Altitude profiles within a single clean air mass were constructed from measurements made during constant-altitude flight legs, and a positive gradient with altitude was typically observed for NO. A strong positive correlation between NO_x and ozone, O_3, was observed during the spring series of flights. Typical free tropospheric NO_x values ranged from 10 to 100 pptv, with NO_2/NO ratios exceeding by an average factor of 2.5 those that would be expected during conditions of photochemical steady state.

During the fall flights, elevated NO values were observed in the free troposphere during periods of subsiding stratospheric air. Furthermore, evidence for the production of NO in electrically active clouds was also observed, as is shown in Fig. 2. The results are from a flight near Hawaii, during which clouds with considerable lightning activity were encountered. As the data show, NO was normally very low in this remote area. However, within the cloud, exceptionally large concentrations were sampled. At other times, clouds with no electrical activity were sampled and the NO values were at background levels. These 1983 and 1984 aircraft flights have provided the most extensive look thus far at the budget and chemistry of NO and NO_2 in the remote global troposphere.

5. Tropopause Fold Investigation

The Group's Lyman-alpha detector for water vapor was flown on board the U-2 research aircraft of the National Aeronautics and Space Administration in the spring of 1984. The goal was to examine the structure of a tropopause fold with fast-response instruments measuring stratospheric and tropospheric trace species. The data elucidate the exchange processes induced by the folding event. The negative correlations between water vapor and temperature, which is opposite to those expected in stratospheric air, suggest that these air parcels originated from near the tropical tropopause. Since the folding event was at midlatitudes, this may be evidence of rapid long-range transport. This flight series was the first in the Stratospheric Tropospheric Exchange Program, in which the Atmospheric Sampling Group will be involved.

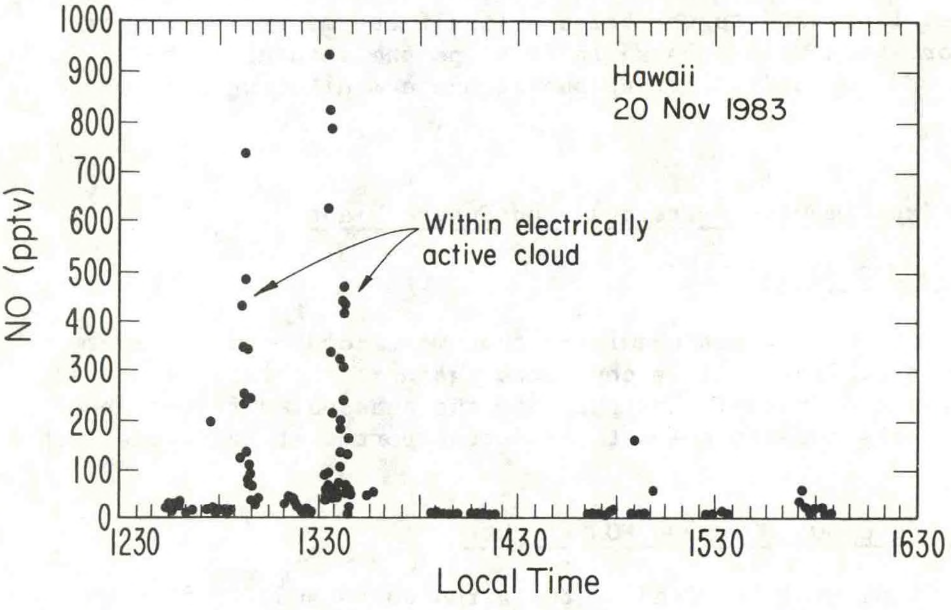

Fig. 2. Evidence for the production of NO by lightning. The data were gathered using an airborne NO instrument of a flight near Hawaii during the fall of 1983.

6. Stratospheric Water Instrument Intercomparison

The Group participated in the second and last of the balloon flights conducted to intercompare the results of stratospheric water vapor instruments, sponsored by the National Aeronautics and Space Administration. These intercomparisons demonstrated that stratospheric water vapor can be measured with an accuracy of about ±25%. The most consistent results were obtained by the Lyman-alpha detector of this Group and the frostpoint instrument of the Geophysical Monitoring for Climatic Change Division of the Air Resources Laboratory. The difference, about 10 to 15%, was the same throughout all of the flights. This consistency affords the opportunity to discover the cause of the difference, thereby allowing a homogeneous data set from these two instruments, which are the only two making regular stratospheric water vapor measurements.

7. Stratospheric Ozone Instrument Intercomparison

The Group also participated in the third and last balloon flight of the ozone-intercomparison campaign sponsored by the National Aeronautics and Space Administration. The goal was to obtain insight into how well stratospheric ozone can be measured with current instruments. While much of the data are still being examined and compared, the initial results demonstrate that it is formidably difficult to measure ozone reliably at 40 km, where good data are critically needed to evaluate the potential alteration of the ozone layer due to man's activities. The Group had two ultraviolet dual-beam ozone photometers on the last balloon flight and configured the pair such that tests could be made of the main sources of ozone measurement uncertainty. These data show that losses of ozone to the walls of inlet lines and to the balloon itself are major sources of error. Such information will substantially shape the strategies being formulated as to how to conduct stratospheric ozone monitoring over the coming decades.

Experiments-in-Progress and Future Plans

1. Fall/Winter NO_y Studies

The studies of the nitrogen chemistry that were conducted at Niwot Ridge in the summer of 1984 will be conducted again in the fall or winter. This should provide considerable insight into the seasonal differences of NO_y and its components and the chemistry and transport that introduce such differences.

2. Free-Tropospheric NO, NO_2, and NO_y Flights

The NO_y technique will be added to the airborne NO and NO_2 instruments and used, in collaboration with the National Center for Atmospheric Research, on a series of aircraft flights in the late summer of 1985. The National Aeronautics and Space Administration's aircraft will carry a suite of instruments that will focus on the reactive nitrogen chemistry of the troposphere: the distributions, reactions, and instrument reliability.

3. NO Soil-Emission Investigation

A newly instrumented research van will be used to explore two aspects of the tropospheric nitrogen species. First, in California, the constituents of Pacific air masses will be examined. This measurement series will provide the opportunity to test the models of the chemistry of maritime air, a system where a few fundamental processes are thought to be dominant. Secondly, the research van will make a direct study of the emission of reactive nitrogen species from the soil, which are thought to be one of the major natural sources that leads to nitrate in precipitation.

4. Measurement of Natural Sulfur Emissions

In conjunction with the above nitrogen emission studies, a newly developed gas-chromatographic apparatus will measure the emissions of natural sulfur compounds from the soils. Two University groups will be making simultaneous studies, so that the comparison will provide insight into the reliability of such measurements. Calibration standards will have been intercompared earlier. These nitrogen and sulfur studies will be conducted in the southeastern United States, where such emissions are thought to be the largest.

5. Long-path Absorption Measurements of Tropospheric Nitrogen Species

A state-of-the-art diode-array spectrometer was incorporated into a spectrometer/computer absorption spectroscopy apparatus. The device has been used to obtain spectra of stratospheric NO_2 of heretofore unobtainable quality. A key part of the procedure is a least-squares data reduction procedure that employs standard spectral relative intensities obtained from laboratory measurements. The system obtained total-column NO_2 data when it accompanied the stratospheric NO/NO_2 experiment into the field. Now, the system is at Fritz Peak Observatory, Colorado, and will use long-path (approximately 10 km) absorption techniques to examine the photochemistry of several tropospheric species. The initial measurements suggest a role of aerosols in the removal of NO_3 and these studies will be expanded.

6. Development of Nitric Acid and Ammonia Instruments

The tungstic acid denuder tube will be tested as a viable method for the measurement of nitric acid and ammonia. Tests will be conducted at Niwot Ridge and comparisons will be made with the measurements of the filter-collection technique.

7. Development of a Tropospheric Ozone Lidar Instrument

The differential absorption lidar technique will be explored as a means for measuring ozone in the free troposphere. There is currently no fully acceptable method to do such, despite the need to assess man's potential alteration of this important climatic and chemical species.

8. Stratospheric/Tropospheric Exchange Investigation

The test flights will be conducted for the new instruments that will take part in the Stratospheric Tropospheric Exchange Program. The aircraft will be the ER-2 and the tests will be of the Group's water vapor, water vapor and ice, and ozone instruments. These species are tropospheric and stratospheric tracers, respectively. The Group will later add NO_y, a stratospheric tracer, to the set. The first experiment will address small scale exchange processes in the vicinity of jet streams. It is planned to concentrate on the lower side of the jet stream where flow of tropospheric mass into the stratosphere has been postulated to occur.

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Introduction

The objective of the Atmospheric Dynamics Program Area is to further our understanding of the dynamics of the atmosphere below 100 km by taking advantage of the unique experimental and analytical capabilities of the group. The experimental research of the Program is based on the study of the atmosphere by analysis of radar echoes from irregularities in the atmosphere. Backscattered radio waves are preferentially scattered by radio refractive index irregularities whose scale is half the radar wavelength. In the ionosphere the scatter is by irregularities in electron density; in the troposphere and stratosphere, by irregularities in the atmospheric density or humidity (Balsley and Gage, 1980). The frequency spectrum of the radar echoes can be interpreted as the Doppler spectrum of the motions of the irregularities in the volume observed by the radar.

Since below about 90 km the irregularities are carried by the atmosphere, the mean Doppler shift is proportional to the mean motion, or wind, in the sampled volume, and the width of the spectrum is determined partly by the spectrum of turbulent velocities in the volume. Also, the radar reflectivity is proportional to the amplitude of the spectrum of turbulent irregularities of radio refractive index. An example of the echo power spectra is shown in Fig. 1. The variation with altitude of the horizontal wind is clearly visible. Many comparisons with balloon wind profiles have shown that the mean Doppler shift is indeed a good measure of the wind (Gage and Balsley, 1978). Other, more sensitive, Doppler radars can measure over a much larger altitude range, including the troposphere, at least the lower half of the stratosphere, and, in the daytime, the mesosphere.

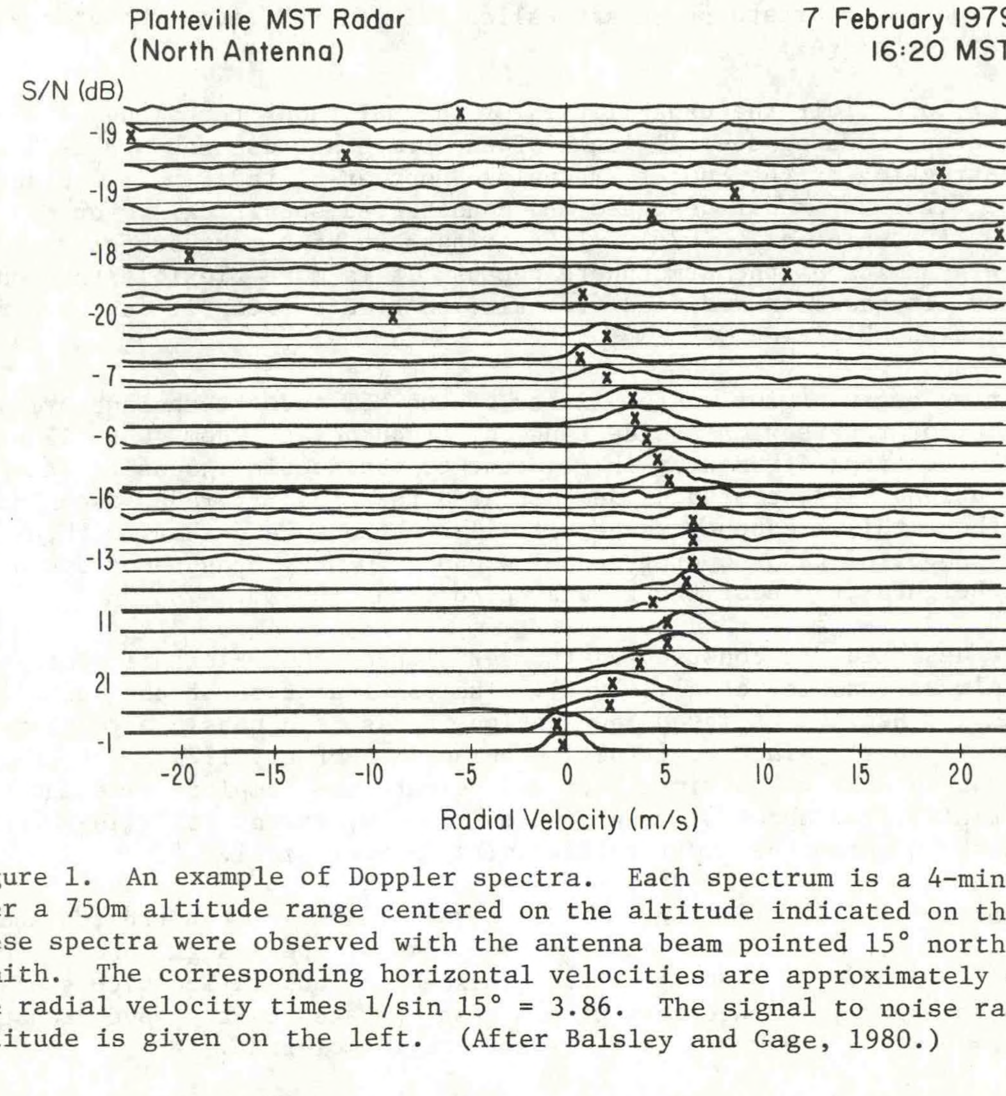

Figure 1. An example of Doppler spectra. Each spectrum is a 4-minute average over a 750m altitude range centered on the altitude indicated on the right. These spectra were observed with the antenna beam pointed 15° north of the zenith. The corresponding horizontal velocities are approximately equal to the radial velocity times 1/sin 15° = 3.86. The signal to noise ratio at each altitude is given on the left. (After Balsley and Gage, 1980.)

Doppler radars can routinely make measurements about 1000 times more frequently than routine balloon or rocket observations in the same altitude range. Their much faster cadence of measurement together with their great altitude range make Doppler radar uniquely suited for studying phenomena that vary rapidly in time, such as buoyancy waves (internal gravity waves) and turbulence. They can, of course, also study slowly varying phenomena such as planetary waves and tides.

These capabilities can be realized, however, only by very sensitive coherent radars with on-line computer analysis of the received data. Because the Doppler radar technique and the radar systems that make it possible are so different from conventional meteorological radars (which depend on echoes from hydrometeors and/or do not do spectral analysis of the echoes) it is convenient to have a concise term to describe this technique. It has been generally agreed to call it the "MST radar technique", for "Mesosphere-Stratosphere-Troposphere". Radars that are sensitive enough to make measurements in all three regions are called "MST radars"; those that can observe only in the troposphere and lower stratosphere are called "ST radars", even though they use the MST radar technique.

In order to exploit the capabilities of the MST radar technique, the Program has been pursuing several lines of experimental research: In 1973 we started construction of the Sunset VHF pulsed Doppler ST radar near Boulder. This was the first VHF radar designed and constructed specifically for MST radar studies (Green et al., 1975). It continues to be a very useful tool for study of the dynamics of the atmosphere because it is more flexible than our other systems. It has been modified from time to time to keep it in the forefront of radar capabilities.

In 1976 we began exploring the limits of the MST radar technique by using a wide variety of radars with a wide range of frequencies (from 40 to 1300 MHz), geographical locations (from near the equator to the arctic), sensitivities, and configurations. These studies demonstrated that the MST radar technique works throughout this frequency range, at all locations, and that, with a sufficiently powerful radar operating below about 70 MHz, measurements can be made at all heights from near the ground up to about 100 km.

We have designed and constructed the very large Poker Flat MST radar near Fairbanks, Alaska (Balsley et al., 1980). The configuration of the radar is shown in Fig. 2. Because of its unique design it has been possible to operate it almost continuously since construction began in February 1979. The Poker Flat MST radar is able to obtain echoes and measure the complete wind and turbulence fields from about 3 km up to about 100 km, except for a 20-km gap centered on 45 km where the radar reflectivity is very small.

The Platteville, CO, (Ecklund et al., 1979) radar was constructed and operated for a month in 1978 in order to test some of the design concepts of the Poker Flat radar. In 1980 it was refurbished in cooperation with the Wave Propagation Laboratory. Since then it has been operated by the Wave Propagation Laboratory as part of the PROFS Profiler network in Colorado.

We have developed a portable ST radar system three of which were used in France during the ALPEX campaign in 1982 to study buoyancy waves (Ecklund et al., 1984).

Recently we have become interested in the use of ST radars in the tropics. In May 1984 we installed an ST radar on Ponape, Federated States of Micronesia, in order to assess the operational characteristics in the tropics. We have also been funded to install three radars on equatorial islands as part of the TOGA (Tropical Oceans, Global Atmosphere) program.

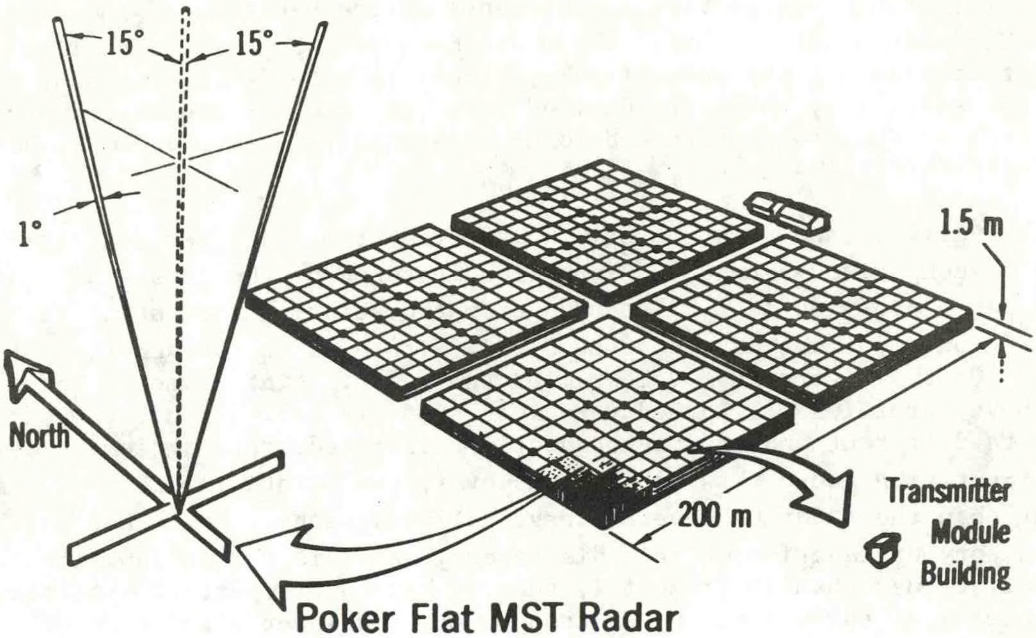

Figure 2. Artist's sketch of the Poker Flat system. (After Balsley et al., 1980.) The other Aeronomy Laboratory ST radars are similar in design but with only one or two transmitters.

Recent Results

MST Radar Studies

1. Atmospheric Turbulence. Parameters of turbulence can be inferred from MST radar observations in two ways.

(a) The variance of the Doppler spectrum involves the variance of the turbulence velocity fluctuations, which is directly related to the turbulence energy dissipation rate ε. ε is the most important parameter of turbulence, since it is proportional to the intensity of turbulence and since it plays an important role in atmospheric dynamics. Unfortunately, because of instrumental and geophysical limitations, thus far it has been impossible to infer ε from spectral widths except when ε and the spectral width are unusually large.

(b) The radar reflectivity, which can be inferred from the observed signal-to-noise ratio, is usually proportional to the turbulence structure constant C_n^2 of the radio refractive index. This quantity is very important in electromagnetic wave propagation in the atmosphere.

The availability of radar profiles of C_n^2 has enabled us to develop a statistical model for the occurrence of turbulence that permits the calculation of profiles of C_n^2 and $\bar{\varepsilon}$ from measurements of the background wind and temperature (VanZandt et al., 1981). The model depends upon the probability distribution functions for the unobservable, fine-structure fluctuations of vertical shear and stability, which are derived from our model of the spectrum of buoyancy waves in the troposphere and lower stratosphere (see Section 3, Buoyancy (Internal Gravity) Waves).

The validity and accuracy of C_n^2 inferred from radar reflectivities was tested by comparison between C_n^2 profiles measured by the Sunset ST radar and simultaneous profiles of C_n^2 inferred from the scintillation of stars. The optical experiment was developed by Dr. J. Vernin of the University of Nice, France. On two nights in November 1983 the radar, scintillometer and model (see above) profiles all agreed very well (see Fig. 3). Since the experiments are quite different and are independently calibrated, this agreement is very significant. On two nights in June, however, the scintillometer C_n^2 was much smaller than the radar C_n^2 over a large altitude range. We do not yet have a satisfactory explanation of this discrepancy. But if the explanation is physical, rather than instrumental, then it may have important implications on the dynamics of turbulence in the troposphere and lower stratosphere.

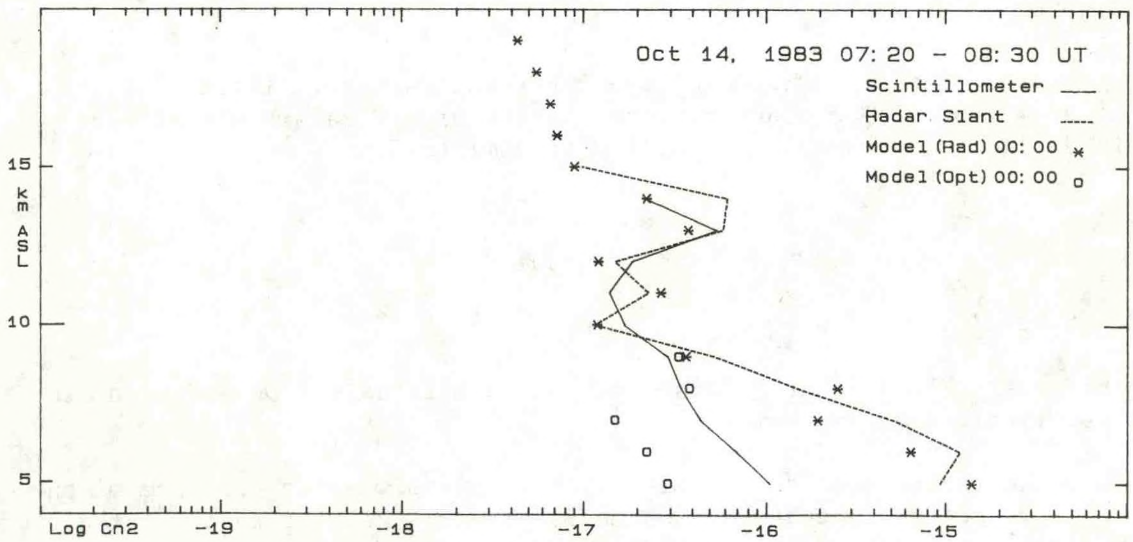

Figure 3. Comparison of profiles of C_n^2 observed by the Sunset radar and by the optical stellar scintillometer and calculated from our model. (After Green et al., 1984.)

2. Fresnel Reflectivity and Atmospheric Stability. When a radar operating at lower VHF frequencies is pointed toward the zenith the reflectivity is greatly enhanced in stable regions of the atmosphere (Gage and Green, 1978). This enhancement is attributed to partial specular or Fresnel reflection from horizontal stratification of the radar refractive index.

We have developed a model, called the Fresnel scattering model, that predicts the profile of Fresnel reflectivity as a function of the atmospheric lapse rate (dT/dz) (Gage et al., 1981). The model parameterizes the scattering from the half-wavelength Fourier component of the horizontally stratified refractivity (Gage and Balsley, 1980). An adjustable constant in the model was determined by normalization to data from the Sunset ST radar. But it is found that the model calculations also agree very well with observations from the Poker Flat MST radar.

There has been controversy over the dependence of the Fresnel reflectivity on the length of the radar range gates. Careful measurements of the range gate dependence were made by the Sunset ST and Poker Flat MST radars (Green et al., 1983; Green, 1984). The result was that in most cases the reflectivity was linearly proportional to the length of the range gate as is predicted by simple theory, while in a small fraction of cases the reflectivity varied as a larger power (up to 2) of the range gate length.

The Fresnel scattering model explains the echo strength given the horizontal stratification of refractivity, but it does not attempt to explain the origin of such structure. VanZandt and Vincent (1983) showed that the required structure might be due to oblique displacements by buoyancy (internal gravity) waves (see the section on Buoyancy Waves) acting on the background gradient of refractive index. Their model is not inconsistent with any of the observed features of Fresnel reflectivity. Moreover, their model is subject to test by further appropriately designed experiments.

The Fresnel scattering model leads naturally to an objective method for determining the tropopause height from radar observations of the Fresnel reflectivity (Gage and Green, 1982). The model is used to construct a profile of Fresnel reflectivity for the minimum stratospheric lapse rate (2K/km). The tropopause is determined as the height where the observed reflectivity first exceeds the model reflectivity. Time series of radar and radiosonde tropopause heights are compared in Fig. 4. The radar can measure the tropopause height every few minutes in contrast to the 12-hr radiosonde determinations.

The existence of a quantitative relationship between the atmospheric lapse rate and the magnitude of the Fresnel reflectivity observed by VHF radar permits the determination of temperature gradients in stable regions of the atmosphere (Gage and Green, 1980). The significance of this development is that if the temperature is known at some reference level, a temperature profile can be constructed from the inferred gradients through the stable region. Also, radar determinations of tropopause heights or large temperature gradients can be used to improve the retrieval of temperature profiles from satellite or ground-based radiometer measurements (Westwater et al., 1983), by as much as 1 K rms.

3. Buoyancy (Internal Gravity) Waves. The MST radar technique is particularly well suited for the study of rapid variations of wind, such as buoyancy waves. Such waves are observed as nearly monochromatic wave trains. But discrete waves are rather rare, not occurring more than 5% of the time at a given height. During the rest of the time the wind is observed to fluctuate almost randomly. These fluctuations may be due to a random superposition of buoyancy waves with a range of frequencies, wavelengths, and azimuths, but

there may also be contributions from other processes. For this reason, these fluctuations are treated in Section 4, Mesoscale Fluctuations of Atmospheric Winds.

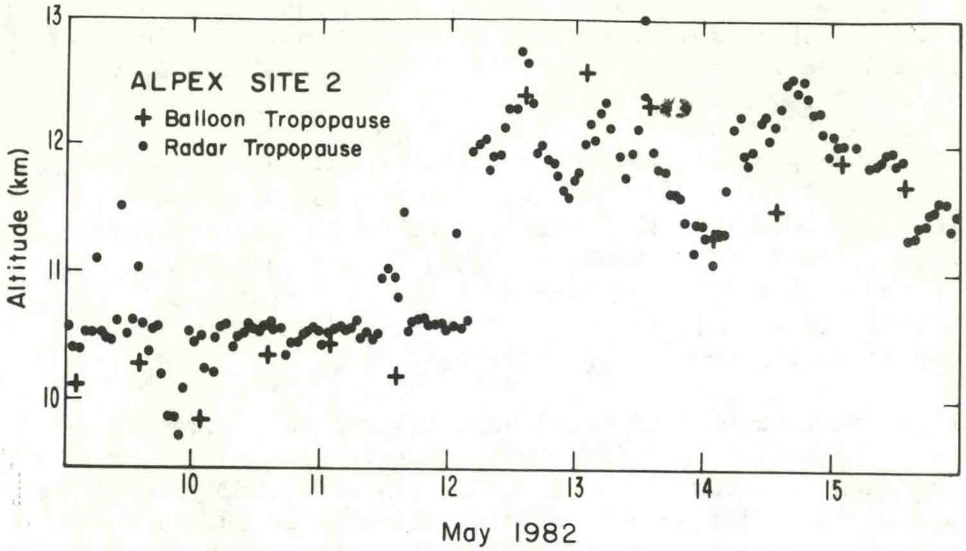

Figure 4. Comparison of radar derived tropopause heights (•) with tropopause heights derived from routine balloons launched from Nimes, France, 85 km NW of the radar site (+). (After Riddle et al., 1983.)

Buoyancy waves can be generated by various processes, including jet streams, convective storms, flow over mountains, etc. But because wave motions oscillate rapidly (except for mountain lee waves), the generation processes have been very difficult to study. With the advent of the MST radar technique we were able to show, by comparison of detailed observations of discrete wave trains with a theoretical model, that in a jet stream the buoyancy waves are generated by shear instability (VanZandt et al., 1979).

We have also observed wave trains associated with convective storms. In Fig. 5 the large, quasi-sinusoidal wave train between 17 and 24h is thought to be associated with a large thunderstorm that was active from 15 to 18h about 100 to 200 km from the Platteville ST radar (Lu et al., 1984). In addition to such discrete wave trains, we found that the amplitude of the wave spectra was about an order of magnitude larger during periods of thunderstorm activity than during quiet periods.

Because these observations were made by only a single radar, the wavelengths or phase velocities that are essential for a complete understanding of wave dynamics could be determined only by the use of simultaneous microbarograph observations, which are not always available.

In order to remedy this deficiency, in July and August 1983 an array of three existing ST radars in an $\sim$ 60 km triangle in Colorado together with over 20 barographs was used to observe further the buoyancy wave activity associated with thunderstorms. These observations, which were made in cooperation with PROFS and the Wave Propagation Laboratory of NOAA, constitute the most complete set of data for the study of buoyancy waves that has ever been taken. Several buoyancy wave events observed during the experiment are being analyzed in cooperation with F. Einaudi, Georgia Institute of Technology, and D. Fua, Istituto per la Fisica dell' Atmosfera, Rome, Italy.

Our first use of an array of ST radars--indeed, the first used anywhere--consisted of a temporary array of three radars in a 5-6 km triangle in southern France, in April and May 1982. This experiment was done in cooperation with M. Crochet of the Laboratoire de Sondages Électromagnetiques de l'Environment Terrestre (LSEET), Toulon, France, as part of the ALPine EXperiment (ALPEX) of the Global Atmospheric Research Program (GARP). The radars obtained continuous vertical wind data during nearly two months of operation.

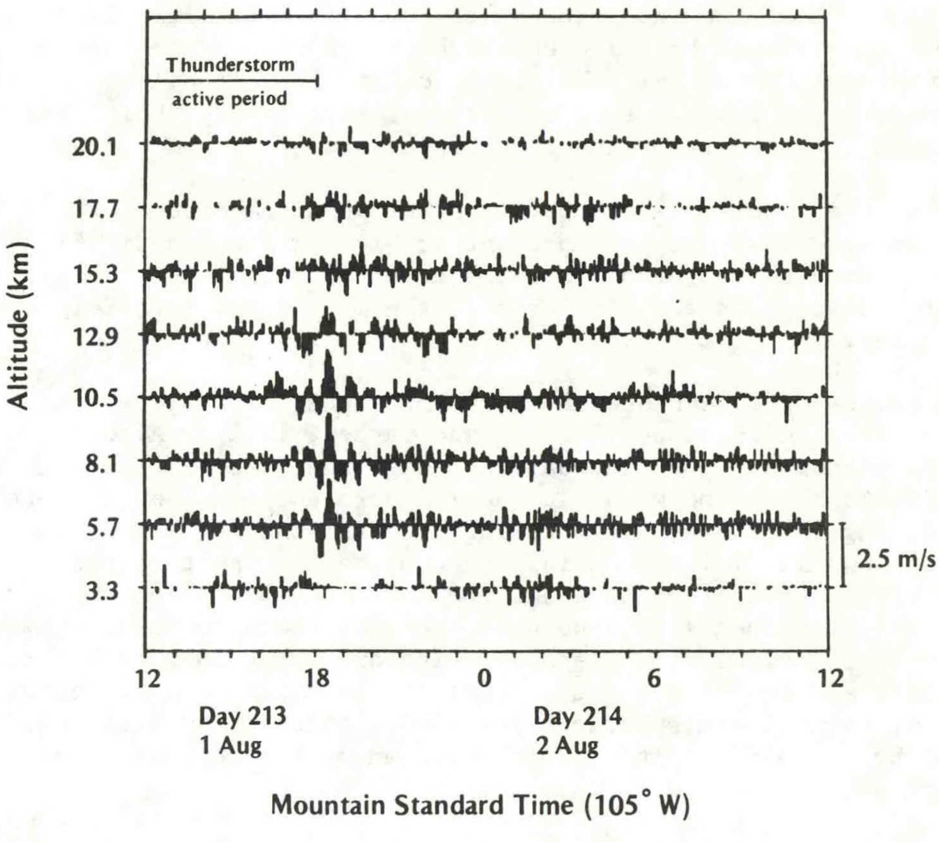

Figure 5. Buoyancy waves associated with thunderstorm activity. (After Lu et al., 1984.)

Final analysis of the data has been completed (Carter et al., 1984). By cross-spectral analysis the parameters of many waves were determined, with periods ranging from 15 to 90 min, horizontal wavelengths from 7 to 40 km, and phase speeds from 5 to 20 m/s. This experiment thus demonstrates that arrays of ST radars can determine the properties of buoyancy waves in the free atmosphere, which is difficult to do by any other technique.

During the period 23 January - 10 February 1984 the Sunset Radar was used to measure the wind, particularly the vertical component, over the Front Range of Colorado in an FAA sponsored experiment to assess the reliability of aircraft altimeters over mountains. As part of this experiment many other meteorological sensors were operated by the FAA, NCAR, WPL, and NWS. This data base should be be the best ever taken for the study of mountain lee waves and associated phenomena such as downslope winds and turbulence.

The program has installed an array of four microbarographs in the Boulder area to support our studies of buoyancy waves.

4. Mesoscale Fluctuations of Winds. The capability of MST radars to measure rapidly atmospheric winds over a wide altitude range has provided important new insight into the variability of atmospheric winds. This subject is of considerable practical importance since the variability of the wind field is the meteorological "noise" background against which synoptic scale measurements must be made. The wind and temperature input to initialize numerical weather prediction models must be representative of synoptic scale motions. One of the major advantages of the MST radar technique is that it can average the observed winds over time to provide such a representative input, while balloon observations are a snapshot that cannot be averaged.

It is also important to understand the physical nature of the fluctuations, not only because they are an important problem in atmospheric flud dynamics, but also because they probably are the cause of all of the energy dissipation in the free atmosphere and also part of the mixing and vertical transport of trace species.

MST radars have been used to obtain the frequency and vertical wavenumber spectra of the fluctuations (Balsley and Carter, 1982). Also, in order to gain a complete picture of the fluctuations, Gage and Nastrom (1984) have completed a definitive analysis of wind fluctuation data collected on commercial aircraft by NASA during the Global Air Sampling Program (GASP). The results of this study are shown in Fig. 6. At low wavenumbers the spectrum has a slope of about -3 with a transition at about 500 km to a slope of about -5/3 at high wavenumbers. VanZandt (1982) pointed out that the spectra of atmospheric mesoscale fluctuations of atmospheric wind are remarkably universal, that is, their slopes and amplitudes are insensitive to variations in geophysical parameters, such as atmospheric stability, altitude, and latitude. This is confirmed by the GASP spectra, which have an overall standard deviation of 2718 spectra of about a factor of 2.5.

Two quite different explanations have been put forward to explain the physical nature of these mesoscale fluctuations. Gage (1979) suggested that they are due to quasi-two-dimensional turbulence; VanZandt (1982) suggested that they are due to a saturated spectrum of buoyancy waves. The GASP spectra are consistent with either hypothesis. However, we are currently analyzing MST radar data in a manner designed to test critically between these competing hypotheses.

5. Energy and Momentum Transport in the Troposphere, Stratosphere and Mesosphere. One of the major unknown areas in middle atmospheric dynamics lies in the coupling of energy and momentum between the troposphere, stratosphere and mesosphere, as well as between low and high latitudes. Such coupling not only must occur, but in some instances it is the controlling factor in ultimately determining the global atmospheric circulation pattern. The process

involves the deposition of buoyancy wave energy and momentum in a region of the atmosphere far removed from the buoyancy wave source. For example, mesospheric circulation models require large deposition of wave momentum to limit the meridional wind magnitude to reasonable (i.e., measured) values.

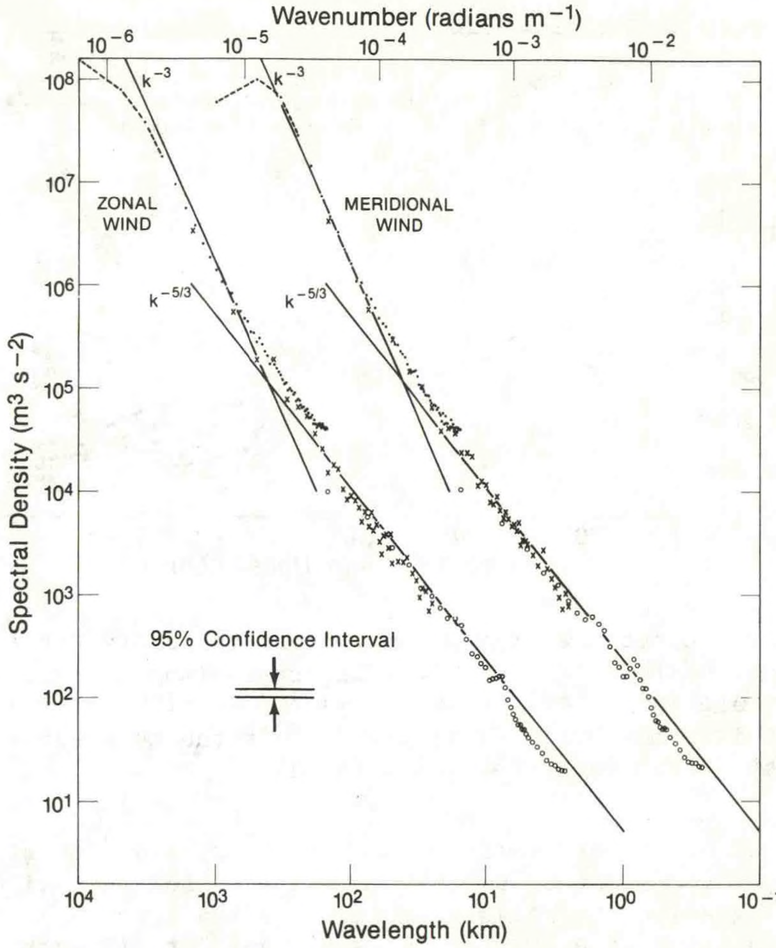

Figure 6. Kinetic energy spectrum near the tropopause from GASP aircraft data. The spectrum for meridional winds is shifted one decade to the right. (After Nastrom et al., 1984.)

The problem of vertical energy and momentum transport is being studied in several different ways. First, the distribution of total kinetic energy density as a function of frequency and horizontal and vertical wavelength, geographical location, altitude, season, etc. is being studied, as described in Section 4. Mesoscale Fluctuations of Winds. Second, we are measuring the vertical flux of horizontal momentum as a function of height by an adaptation of the technique introduced by Vincent and Reid (1983).

The extensive high resolution data base at Poker Flat over the past five years has been employed to measure the kinetic energy density of the atmosphere (Balsley and Garello, 1984). Results are shown in Fig. 7 as a function of season and frequency. In all instances, the data show that the kinetic energy density decreases steadily with increasing height. A more extensive analysis of these and similar results is currently underway.

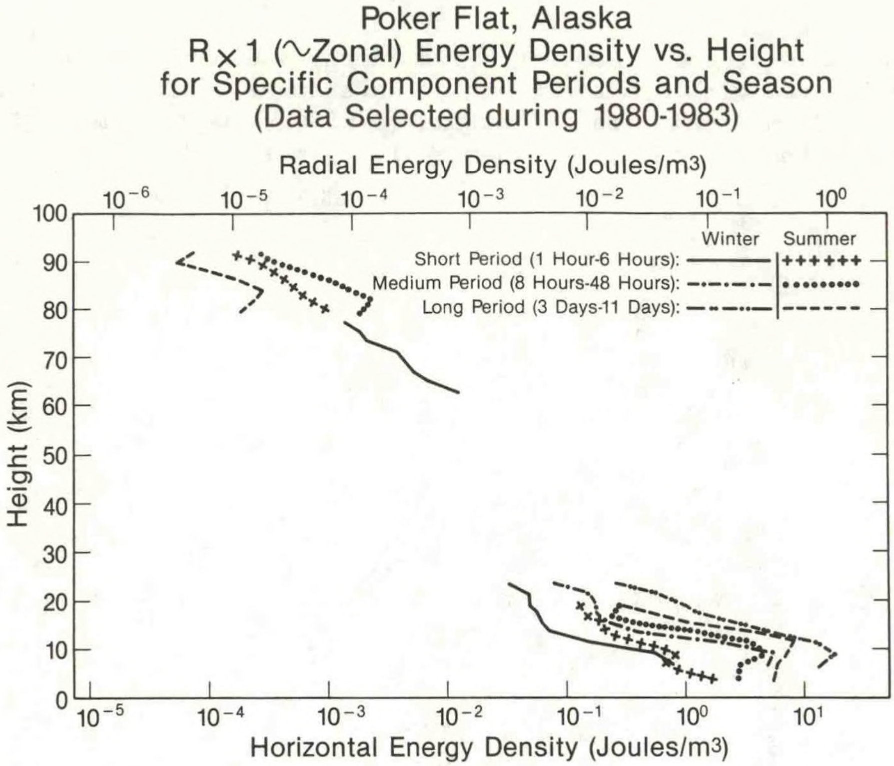

Figure 7. Kinetic energy density profiles of atmospheric wind fluctuations for three spectral intervals (short, medium and long periods are indicated). Kinetic energy density is defined as $\rho\overline{u^2}/2$, where ρ is atmospheric density and $\overline{u^2}$ is the mean square (∿ zonal) velocity within each spectral interval.

Dr. S. K. Avery has completed a preliminary measurement of $\overline{u'w'}$ in the troposphere, lower stratosphere, and mesosphere for both winter and summer. The vertical gradients of this quantity is a measure of the gravity wave momentum deposition as a function of altitude. Preliminary estimates of this deposition agree with similar measurements at other locations.

6. Mean Vertical Velocity. The mean vertical velocity plays an important role in the dynamics of atmospheric circulation. It is particularly important in the vertical transport of energy (including latent heat) and momentum and in the initiation of precipitation. Yet until the advent of the MST radar technique, there had been no method of measuring the vertical velocity continuously. As a result, our present knowledge of large-scale vertical velocity has been based upon inferences from routine radiosonde data in the lower atmosphere and from satellite temperature data in the upper atmosphere. The accuracy of these analyses is not well known, and, in any case, they can be performed only every 12 hours.

Therefore, considerable attention has been given to the possibility of direct measurement of large-scale vertical motion by averaging vertical velocities measured by the MST radar technique at a single station. The fundamental difficulty in doing this is that the large-scale vertical velocities are small compared with the small-scale vertical wind variability, i.e., the mesoscale fluctuations discussed in the previous section. Even under quiet conditions the rms vertical velocity is about 10 cm/s (Nastrom and Gage, 1984), whereas large-scale vertical velocities seldom exceed several cm/s. As a consequence, it is necessary to average several hours of vertical velocities to reduce the vertical velocity variance for small-scale processes to the level where large-scale vertical motion can be detected. Nastrom et al. (1984) have shown that this can be done with existing radars under certain circumstances. Figure 8 shows a comparison of directly measured vertical velocities observed using the Platteville radar with vertical velocities inferred from NMC analyses using the kinematic, omega and adiabatic methods. Radar vertical velocities are typically a factor of 2 or so larger than the corresponding analyzed vertical velocities. This may be a result of the sub-synoptic scale motions influencing the radar results or it may be due to the overall slope of the land in the region around the Platteville radar.

In a separate study (Gage and Nastrom, 1984) vertical motions observed by the Platteville radar have been compared with rainfall rates observed nearby during an upslope storm. Figure 9 shows the location of the two recording rain gauges and the radar relative to the topography of the region. Comparison in Fig. 10 of the rainfall rates observed at the two stations with vertical motion observed by the radar shows that upward vertical motion is well correlated with rainfall rates observed at the surface.

Results such as these are invariably influenced by topography. In order to make further progress in measurement of large-scale vertical motion it is imperative to establish a radar in flat terrain away from the influence of mountains. This will be discussed further in the Future Plans section.

The mean vertical velocity also plays an important role in the mesosphere. Observations using the Poker Flat MST radar show the monthly mean vertical velocity (the solid curve in Fig. 11) is roughly sinusoidal during the year, with extrema of about 25 cm/s in winter and summer (Balsley and Riddle, 1984). These results may be contrasted with current theoretical models, which predict vertical velocities that are an order of magnitude smaller and in the opposite sense to the observations. Since the vertical motion is intimately related to the horizontal flow by means of the continuity equation, it is essential to resolve this discrepancy in order to have a complete understanding of middle atmospheric dynamics. Possible complicating factors include nonuniform zonal flow, multicellular flow in the meridional plane, and the presence of an appreciable Stokes drift (see below).

Dr. L. Coy, an NRC fellow currently working with our group, has suggested that this discrepancy could arise from the effects of intense buoyancy wave action known to occur in the mesosphere. These effects could result in a different value for drift motions in an Eulerian coordinate system (fixed to the earth) and in a Lagrangian coordinate system (fixed to the moving medium). The differential drift values between these coordinate systems (the so-called Stokes drift) is normally insignificant. The existence of an appreciable Stokes drift in the mesosphere, if it can be proven, will have a large impact on the comparison between theory and observations, and will also effect our interpretation of current model calculations.

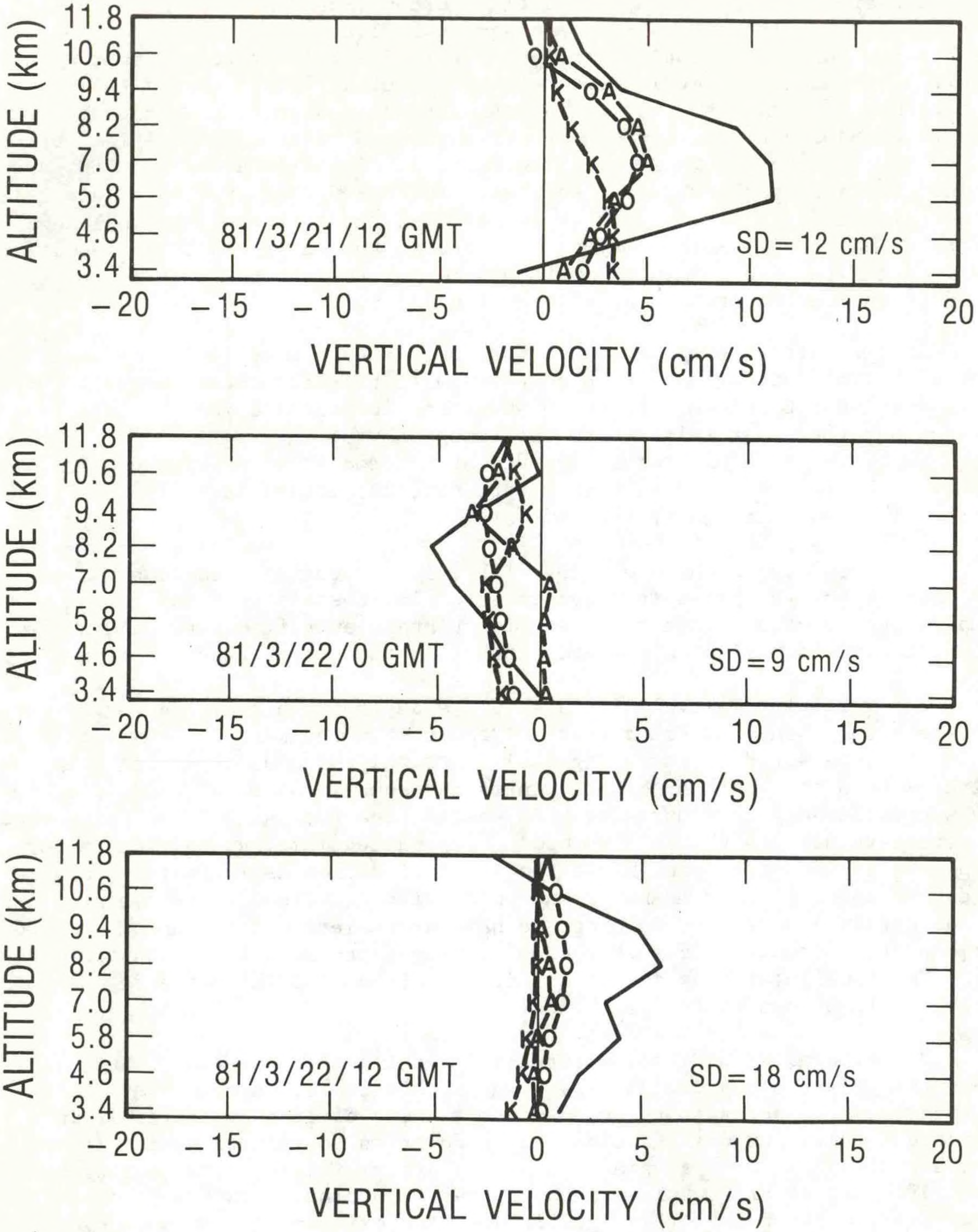

Figure 8. Profiles of 9-hour average vertical velocity observed by the Platteville radar (solid line). Vertical velocity profiles calculated from NMC data using the adiabatic (A), kinematic (K) and omega methods are shown for comparison. (After Nastrom et al., 1984.)

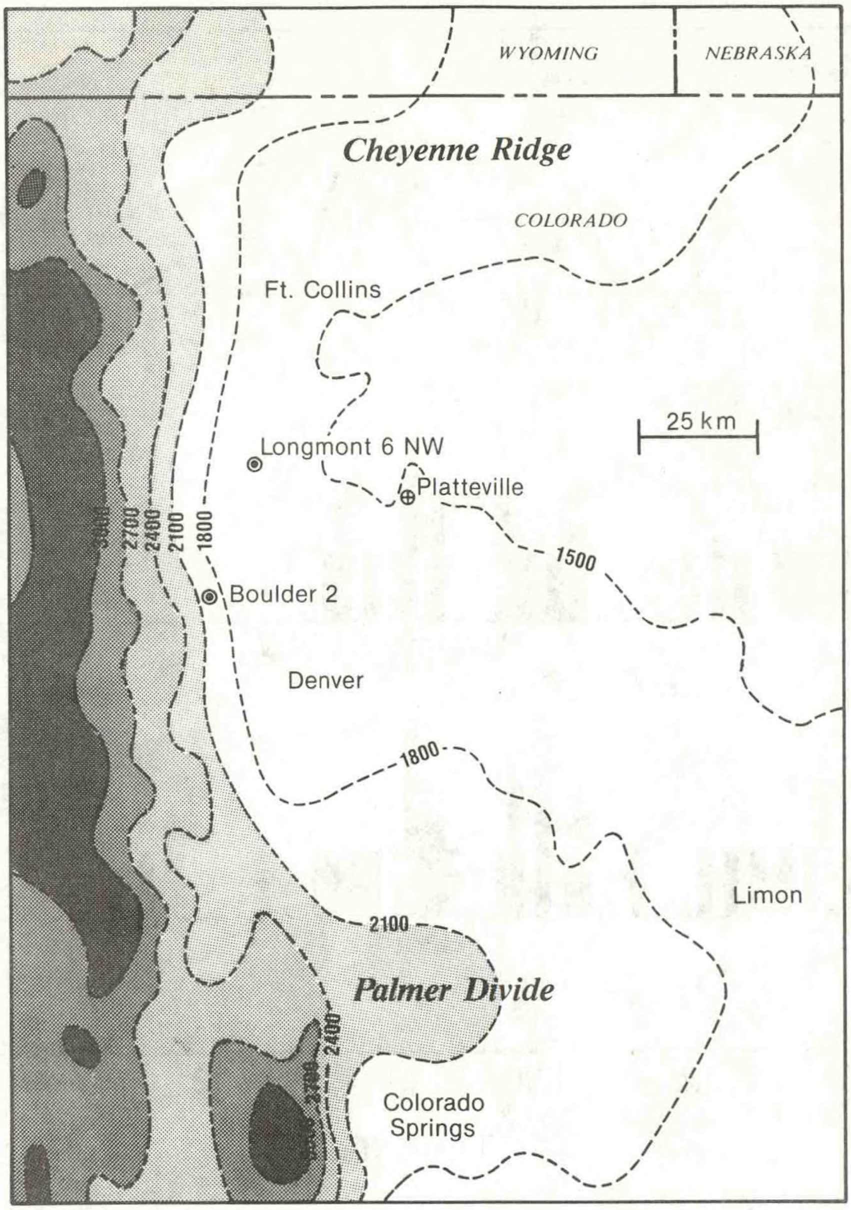

Figure 9. A map of northeastern Colorado showing broad-scale features of topography. (After Gage and Nastrom, 1984.)

7. Mesospheric Studies. The theory of nonlinear wave breaking in the mesosphere was refined, in collaboration with Prof. D. C. Fritts of the University of Alaska, to explain the strong seasonal variability of the mesospheric echo structure observed at Poker Flat (Balsley et al., 1984).

The STATE series of experimental rockets launched at the Poker Flat Rocket Facility by the Air Force Geophysical Laboratory described in last years' AL Report is still under active analysis. Preliminary results reveal the summertime arctic mesosphere is extremely cold and supports regions of intense small-scale electron density fluctuations. These fluctuations are responsible for the extremely strong summertime radar echoes observed at Poker Flat. There is

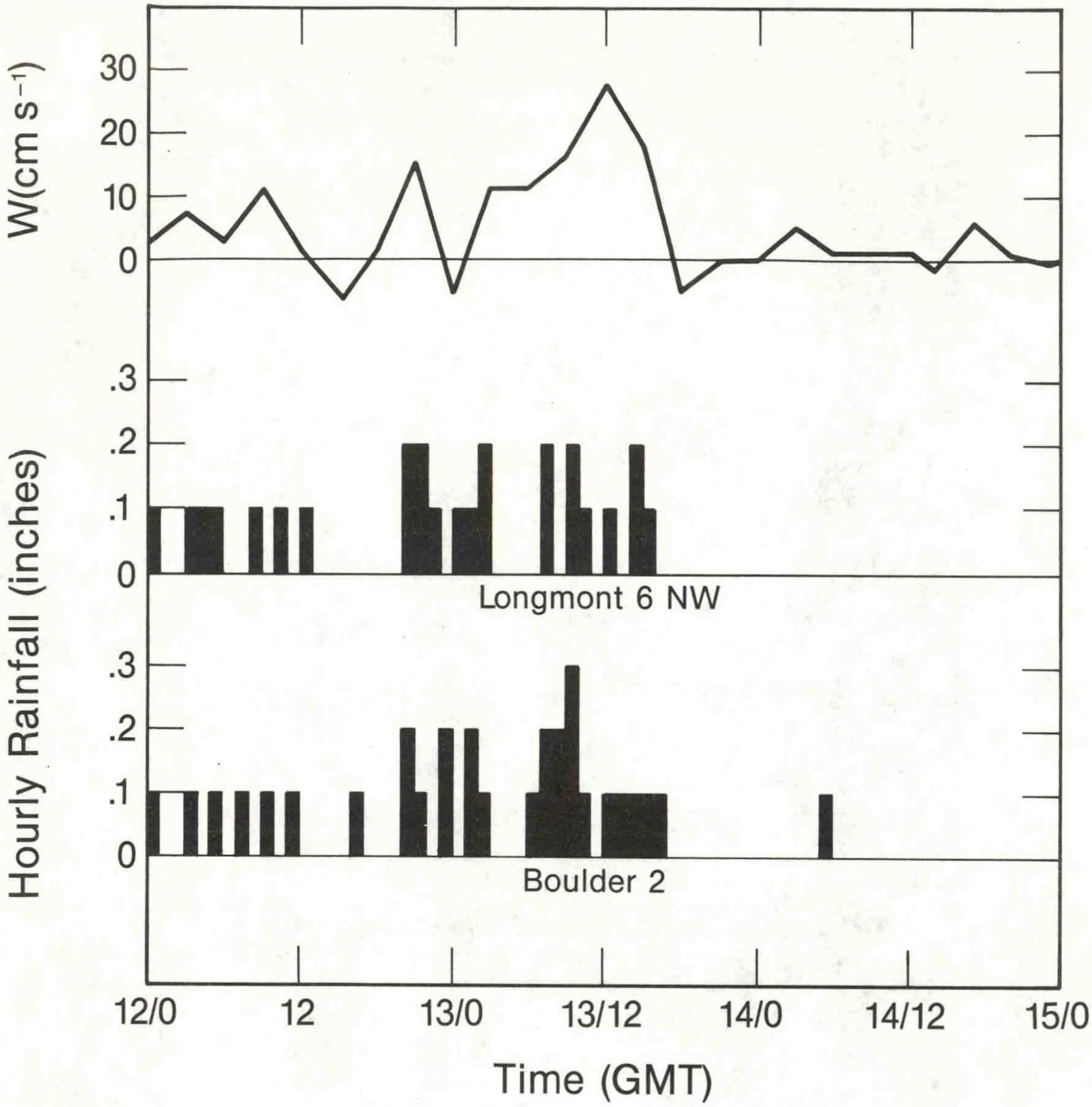

Figure 10. A comparison of hourly rainfall data with three-hour averages of vertical velocity observed by the Platteville radar at 5.9 km MSL. (After Gage and Nastrom, 1984.)

good agreement between the vertical profile of electron density fluctuations and the concurrent radar echo profile at Poker Flat. An important conclusion of this experiment is that the minimum scale of turbulence in the arctic summer mesosphere is much smaller than at lower latitudes.

A movie of high-resolution radar echo spectra obtained during STATE has been completed using NCAR's CRAY computer by Dr. Coy. The short-term dynamics of mesospheric motions revealed by this movie are impressive and give an excellent first-look into the violent processes that are active in this region.

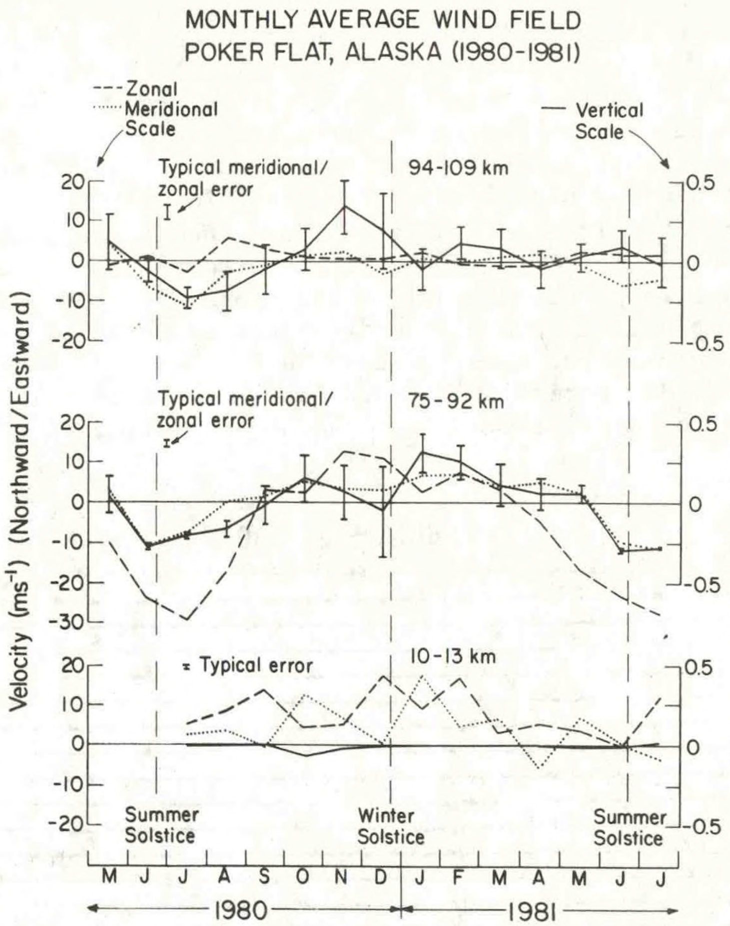

Figure 11. Monthly-averaged values of the wind field over Poker Flat, Alaska. Zonal, meridional, and vertical winds (dashed, dotted, and solid curves, respectively) are both larger than and in the opposite sense to theoretical values. (After Balsley and Riddle, 1984.)

The capability of the Poker Flat MST radar as a meteor radar has been further examined by Dr. S. K. Avery of CIRES (Avery et al., 1983). This work, which was initiated several years ago through a NOAA Grant, has yielded atmospheric tidal amplitudes and phases and mean zonal and meridional winds as a function of season. Dr. Avery has received a two-year NSF grant to do a full analysis of the Poker Flat meteor winds during a five year period. This study will support at least one Ph.D. candidate.

The dependence on magnetic activity of the summer mesospheric zonal winds at Poker Flat continues to be studied by Dr. J. G. Luhmann and R. Johnson of UCLA in order to verify the results of an earlier AL investigation (Luhmann et al., 1983). Their results to date indeed verify the existence of a partial control of the mesospheric winds by magnetic activity. This study, which should soon be completed, will result in a Ph.D. dissertation for Ms. Johnson.

8. Study of the Tropical Atmosphere with ST Radars. It has recently been recognized that much of the climate variability in middle latitudes is related to variations in the tropical Pacific. As a result, we anticipated receiving major funding for the installation and operation of ST radars in the tropics (see Future Plans). Before such radars could be properly designed, however, data on operational conditions in the tropics needed to be obtained, particularly data on the radar reflectivity, which could be quite different in the tropics from middle and high latitudes. For this purpose, in 1983 we received funding from the ERL Director's Discretionary Fund to install an ST radar on Ponape, Federated States of Micronesia. This radar began operation in May 1984. The Ponape radar has not only already provided invaluable design data, but it has also revealed several new dynamical features of the tropical atmosphere. An example of the data is shown in Fig. 12, which is a plot of vertical velocity for a 23-day period in May and June. The intervals of large vertical velocity appear to correlate with periods of precipitation and associated convention.

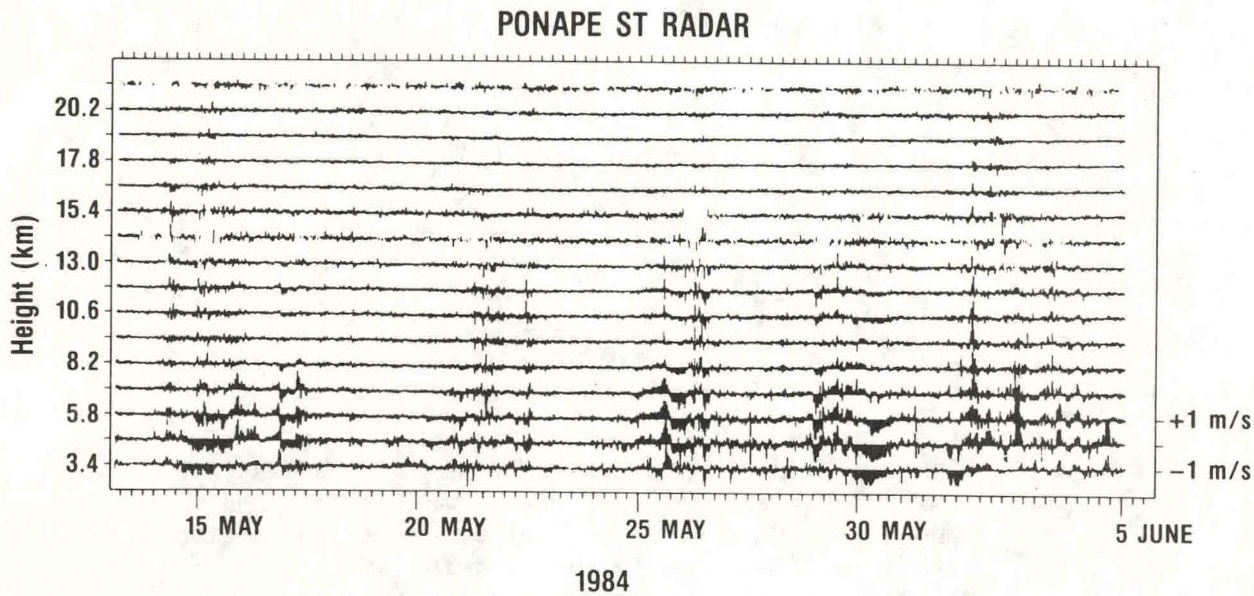

Figure 12. Fifteen-minute averaged values of the vertical winds over Ponape for May-June 1984. Note periods of alternating quiet and active vertical wind fluctuations. Enhanced fluctuations tend to be associated with periods of enhanced precipitation.

9. Application of VHF Radar to Short-Range Forecasting and Nowcasting. In collaboration with Dr. T. Schlatter of PROFS, we have used data from the WPL network of wind profilers to explore their impact on Nowcasting and short-range forecasting (Gage and Schlatter, 1984). Two storms were selected for analysis during PROFS winter campaign of 1984. The first storm of 17-18 February illustrated a situation where heavy snow was forecast but did not materialize. Profiler data showed rapid wind shifts associated with the passage of a sharp trough. Later, a new storm formed too far east to effect Denver. A second storm 25-26 February did produce heavy snow. The profiler winds showed a deep upslope circulation which persisted for many hours. In both cases the profiler data helped forecasters understand the weather and

explain short-term changes. Their high temporal frequency and vertical resolution provided much needed information never before available.

Climate Variability and Solar Radiation. We have continued a major effort in the description and modelling of seasonal and secular changes in the height of the tropical tropopause (Reid and Gage, 1981; Gage and Reid, 1982a,b). This work is a cooperative effort with the Theoretical Aeronomy Program, and a detailed description of it will be found in that chapter of this Annual Report.

Future Plans

MST Radar Studies

1. Atmospheric Turbulence and 2. Fresnel Reflection and Atmospheric Stability. Work in these areas will be at a low level and will consist of completing present studies.

3. Buoyancy (Internal Gravity) Waves. The generation of gravity waves by thunderstorms will be studied further by completing the analysis of the radar and microbarograph data taken in the summer of 1983.

4. Mesoscale Fluctuations of Winds. We will continue study of mesoscale fluctuations by examining additional power spectra of atmospheric parameters (including both wind and temperature) from various sources and by comparing the spectra with models based on the theories of buoyancy waves and two-dimensional turbulence. Comparison of models with power spectra from MST radar data taken simultaneously at the vertical and at a slant will be particularly critical. Analysis of the GASP data will continue in order to determine the climatology of mesoscale spectra as a function of latitude and underlying topography.

5. Energy and Momentum Transport in the Troposphere, Stratosphere, and Mesosphere. The preliminary study of kinetic energy density profiles will be expanded by further, more detailed analyses. Initial calculations of rotary spectra will be done. The ratio of the power in the clockwise to counterclockwise spectra is a measure of the ratio of downgoing to upgoing buoyancy wave energy. A more detailed study of the vertical flux of horizontal momentum will be undertaken at Poker Flat by using antenna beam switching to make measurements symmetrical about the zenith, which is a better geometry than the present vertical/slant configuration. If the fluxes measured with the new and the present configurations agree, then fluxes can be determined from the entire 5-year data base at Poker Flat.

In combination with theoretical developments in the Atmospheric Waves and Turbulence program and elsewhere, these studies should lead to a greatly improved understanding of waves energy transport and deposition in the middle atmosphere.

6. Mean Vertical Velocity. The mean vertical velocity will continue to be studied by extensions of the methods described in Recent Results. However, before the capability of the MST radar technique can be fully assessed it will be essential to have extensive data from a radar sited in flat terrain. For this reason, we have proposed to the National Science Foundation to construct and operate a state-of-the-art ST radar near Urbana, Ill. Although this

"Flatland" radar was motivated by the need to measure vertical velocities, the absence of terrain effects will also make the data uniquely valuable for studying buoyancy waves and energy and momentum transport in the lower atmosphere. If funded, the Flatland radar will constitute a major new direction for the program.

7. Mesospheric Studies. We will continue analysis of the STATE data base in order to understand better the relationship between neutral turbulence, echo power, and wave breaking processes in the mesosphere and lower thermosphere. This effort will involve collaboration with a number of scientists outside the Aeronomy Laboratory.

8. Study of the Tropical Atmosphere with ST Radars. The second major new direction of the program is the study of the tropical atmosphere using ST radars.

The Ponape radar described in the Recent Results will be improved to include a beam-swinging capability so that the horizontal wind as well as the vertical wind can be measured. These measurements should reveal the character of wind variability in the oceanic tropics.

The relation between the enhanced echoes and rainfall will be studied quantitatively in order to understand the nature of the enhancements and their relation to convective cloud dynamics.

The program has been funded by the Office of Oceanic and Atmospheric Research to establish three ST radars in the equatorial Pacific for a ten-year period as part of the international Tropical Oceans, Global Atmosphere (TOGA) program. Site surveys for two of the radars have been completed and the radars should be installed during FY 1985. The data from the TOGA and Ponape radars will be used by the Aeronomy Laboratory and other scientists to study vertical motions in large cumulo-nimbus clouds, the vertical transport of minor constituents, waves and turbulence properties of the tropical atmosphere, and equatorial (large-scale) waves, which are thought to be important in controlling climate variability in middle latitudes.

Climate Variability and Solar Radiation. The study of the tropical tropopause region using radiosonde data will continue. Emphasis will be on 1) further development of the conceptual picture of troposphere-stratosphere interaction in the tropics, 2) a thorough investigation of the 20-day periodicity in tropopause heights and winds in the western tropical Pacific, and 3) a refined and more complete study of the correlation between tropopause height and global atmospheric angular momentum, aimed at exploring the cause-and-effect relationship. The connection between tropopause properties and the wind fields of the tropical lower stratosphere and upper troposphere will be investigated.

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ATMOSPHERIC CHEMICAL KINETICS

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Introduction

The major goal of the Atmospheric Chemical Kinetics program is to develop and apply laboratory techniques for the study of important atmospheric chemical reactions. The products of these studies include reaction rate coefficients, reaction product identities, and thermochemical data. The information obtained is used to develop an understanding of how natural and man-made chemicals released into the atmosphere affect our environment. The evaluation of current environmental issues, such as stratospheric ozone depletion, photochemical air pollution, and acid rain require an understanding of the basic chemical reactions that occur in the atmosphere.

The Atmospheric Chemical Kinetics program has evolved from the Atmospheric Collision Processes program which was dedicated to the study of ion-molecule reactions which dominate the chemistry of the ionosphere, i.e., the upper regions of the atmosphere. The laboratory continues to play a role in atmospheric ion chemistry measurements. These measurements have important applications in communication and in atmosphere composition studies.

In the Aeronomy Laboratory, two principal experimental methods have been developed to study ion chemistry. The first is the flowing afterglow technique, which has now been reproduced in numerous laboratories around the world and which has become the standard technique for studying ion-molecule reactions at near-thermal energies. Its success is due, in part, to the unique capability of studying the reactions of an enormous variety of ion and neutral chemical species. The second device is the flow-drift tube which combines the chemical versatility of a flowing afterglow with the energy variability of a drift tube. This combination permits the study of the kinetic energy dependence of an extensive variety of ion-neutral reactions, ground state or excited state ions with stable or unstable neutrals. The accessible energy range is from 300 K to several electron volts, a range that bridges the experimentally difficult gap between room temperature and beam energies.

With these techniques, the Aeronomy Laboratory has provided a large fraction of the available data on the ion-neutral reactions that control the ion composition in the earth's atmosphere. Some examples include the development of a detailed D-region negative ion reaction scheme that correctly predicted the dominant ion species before composition measurements were available, the development of a positive ion reaction mechanism to explain rocket observations of water vapor ions, and the discovery of associative detachment reactions, a new class of ion-neutral interaction with important physical and atmospheric applications.

Recent interest in atmospheric ion chemistry has concentrated on the application of ion-chemistry as a sensitive analytical tool for the determination of critical trace atmospheric species, on the role of ion chemistry in the production and destruction of critical atmospheric species in the troposphere and stratosphere, and on the production and reactions of vibrationally excited ions. For example, the first stratospheric measurements of sulfuric acid vapor have been obtained from the combination of ion kinetic data from AL and *in situ* negative ion composition measurements by a research group in Germany. The latest analytical application of the flowing afterglow experiment has been to use it as a reactor and detector for chemical ionization detection of reactants and products of neutral reactions. Ions, noble gas metastables, and uv photons have been used to achieve the sensitive detection of trace gases at concentrations down to about 10^9 molecule cm^{-3} in a flow tube experiment.

In the 1970's the laboratory extended its research in reaction kinetics to neutral processes which dominate the chemistry of the lower regions of the atmosphere. This shift was in response to the increasing interest of the atmospheric sciences community in environmental issues.

The laboratory was the first to adapt the technique of laser magnetic resonance (LMR) for kinetic studies of the reactions of certain critical atmospheric radicals such as OH, HO_2, and ClO. LMR was invented in the Boulder National Bureau of Standards Laboratories for the study of the Zeeman spectroscopy of radicals. In an early study it was used to make a thorough investigation of the reactions of man-made chlorocarbons with atmospheric OH radicals providing estimates of the expected atmospheric lifetimes of these molecules. Subsequent legislation and regulations on the uses and emissions of these materials were based in part on these data.

Using the LMR apparatus the laboratory has made measurements that have lead to major revisions in the kinetic data base for HO_2 radical reactions. These new data have resulted in major changes in the predictions of stratospheric ozone depletion by man-made nitrogen oxides and chlorocarbons. These studies have also provided evidence that new chemical species such as HOCl and HO_2NO_2 must be included in stratospheric models.

The LMR technique has been improved in collaboration with NBS scientists. The latest instrumentation permits a larger variety of radical molecules to be detected and, therefore, more complex reactions to be studied. For example, recent work has concentrated on measuring rate constants for reactions involving two unstable radicals. These studies not only improve our knowledge of environmental chemistry, but their unusual and unpredicted temperature and pressure behavior has generated interest and attention beyond the atmospheric science community.

A tunable diode laser, was recently put into operation. The first study was a measurement of the N_2O concentration in the air. This work has resolved a significant discrepancy in the previous measurements of atmospheric N_2O.

Much of the research in this program is carried out with collaboration and support from other organizations. NBS physicists from the Time and Frequency Division provide a valuable service as consultants in the area of laser technology. The program provides opportunities for young scientists to be trained in atmospheric and environmental chemistry. This includes both Postdoctoral Research Associates from CIRES and the National Research Council and graduate students from the University of Colorado, Department of Chemistry and CIRES. Substantial financial support has been obtained through research contracts with the Chemical Manufacturers Association, the Defense Nuclear Agency, National Aeronautics and Space Administration, and the Federal Aviation Administration.

Recent Results

Collisional quenching of vibrationally excited NO^+ ions by a number of atmospheric and other neutral molecules has been carried out in a collaboration with former Aeronomy Laboratory Post Docs at the University of Innsbruck. A technique that was developed in the Aeronomy Laboratory was employed in these studies. NO^+ is a dominant ion in certain regions of the ionosphere ($\sim$ 100 km) when it is formed in the reactions of O^+ with N_2, N^+ with O_2, and N_2^+ with O. Vibrationally excited NO^+ is an infrared emitter so that its concentration is of concern in atmospheric spectroscopy. It is found that NO^+ is rapidly quenched in the atmosphere by N_2,

$$NO^+(v=1) + N_2 \rightarrow NO^+(v=0) + N_2, \; k = 7 \times 10^{-12} \; cm^3 s^{-1} \text{ at } 300 \text{ K}.$$

The rate coefficient decreases very slightly with increased temperature. On the other hand O_2 does not vibrationally relax NO^+ efficiently. A theoretical model has been developed which explains these observations. The model involves complex formation followed by vibrational predissociation. It also satisfactorily explains most of the vibrationally excited O_2^+ and other NO^+ quenching measurements. This work is the first systematic and detailed study of vibrational relaxation in ions that have been made to date.

The first measurements of vibrational energy transfer rates from excited neutral molecules to molecular ions at thermal energies were carried out in collaboration with colleagues in the Department of Space Science at Birmingham University in England. These results provide a very important insight into the molecular interaction process.

In collaboration with another former Post Doc at Meudon, France, ion-molecule reaction studies have been carried out to extremely low temperatures (8 K) and interpreted in terms of basic reaction mechanisms. This work has application to developing an understanding of fundamental reaction processes and to planetary and interstellar chemistry.

Two experiments, a laser magnetic resonance spectrometer (LMR) and a chemical ionization flowing afterglow (CI-FA), have been used to study the mechanism by which sulfur dioxide, SO_2, is converted to sulfuric acid, H_2SO_4, in the atmosphere. This process is a major source of uncertainty in modelling acid precipitation chemistry. The central issue is whether odd hydrogen radicals, OH or HO_2, are consumed in the conversion process. If radicals are consumed, a reduction in SO_2 emissions would not necessarily produce a proportionate reduction in the amount of H_2SO_4 deposited in critical areas. This follows because the present rate of H_2SO_4 production may be limited by the number of odd hydrogen radicals produced and not by the amount of SO_2 released into the atmosphere. Recent experiments in other laboratories have provided indirect evidence that the gas phase SO_2 oxidation process may not consume radicals. Our direct LMR study confirms these experiments and shows that the OH radical which reacts with SO_2 in the primary process is regenerated as an HO_2 radical, when oxygen is present. The CI-FA experiment has directly established that the second product of the critical reaction is sulfur trioxide, SO_3. The SO_3 product is probably rapidly converted to H_2SO_4 by water and on the surface of droplets and aerosols. The proposed mechanism is described by the following scheme:

$$OH + SO_2 + M \rightarrow HOSO_2 + M$$

$$HOSO_2 + O_2 \rightarrow HO_2 + SO_3$$

$$SO_3 + H_2O \rightarrow H_2SO_4.$$

In this mechanism the odd hydrogen radicals are oxidized from OH to HO_2, but OH is rapidly regenerated from HO_2 by reactions with NO and ozone. The overall effect is that the radicals act to catalytically oxidize SO_2 to SO_3 so that one may expect a nearly linear relationship between SO_2 emission rates and H_2SO_4 deposition rates. Also the production of SO_3 in this mechanism indicates that the chemistry of this molecule must be investigated. There was previously no significant atmospheric source of this molecule.

The reaction of nitrate radicals, NO_3, with nitric oxide has been studied

$$NO_3 + NO \rightarrow 2NO_2$$

using laser induced fluorescence detection of NO_3. This reaction is used for laboratory calibrations of NO_3 concentrations and is important in nighttime urban chemistry. A sample of the data obtained in this experiment is shown in Figure 1. The rate coefficient at room temperature was found to be $(3.0 \pm 0.5) \times 10^{-11}$ cm^3 molecule^{-1} s^{-1} at 298 K which is about 50% larger than the accepted published value. Preliminary results at low temperatures show that the reaction has a negative temperature dependence.

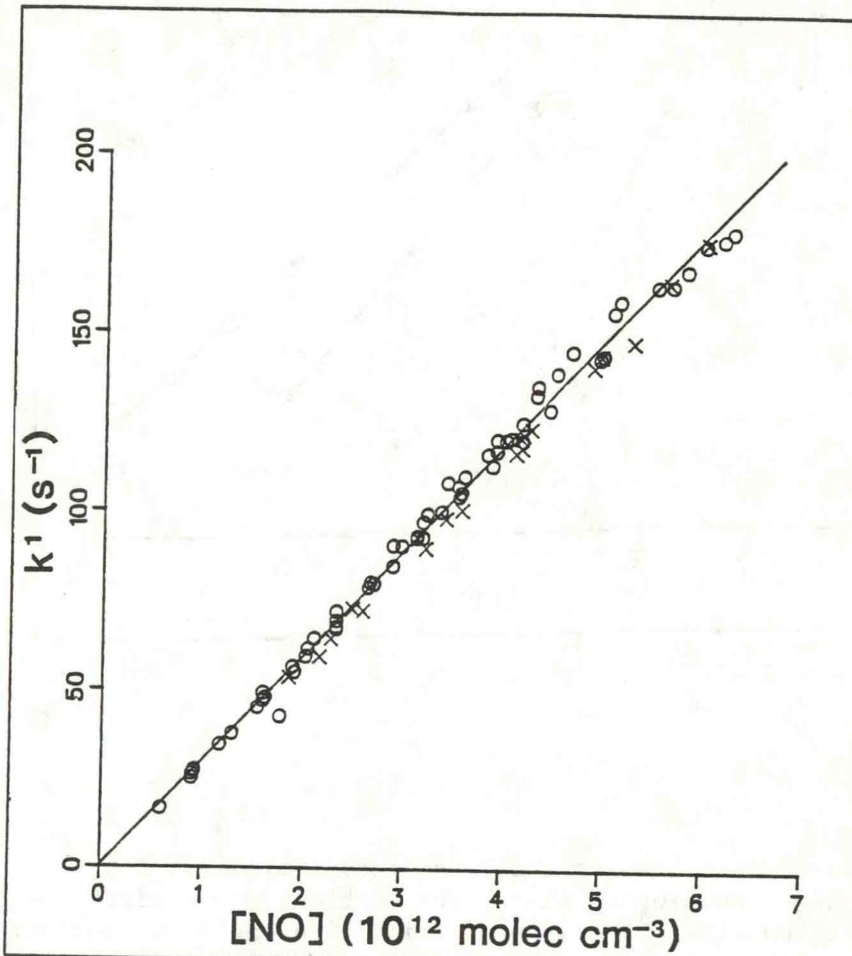

Figure 1. Summary of data on NO_3 + NO rate constant measurements. T = 298 K P = 1 to 3 torr, and initial $[NO_3] = (0.3 \text{ to } 5) \times 10^{10}$ molecule cm^{-3}. The data points indicate two different sources of NO_3 were used: (o) Thermal dissociation of N_2O_5 and (x) the reaction of fluorine atoms with nitric acid. The slope of the line gives the bimolecular rate coefficient, $k = (3.0 \pm 0.5) \times 10^{-11}$ cm^3 molecule^{-1} s^{-1}.

The kinetics and transport properties of gaseous sodium, Na, have been studied in a fast-flow reactor with resonant fluorescence detection of Na. Samples of the data from this experiment are shown in Figures 2 and 3.

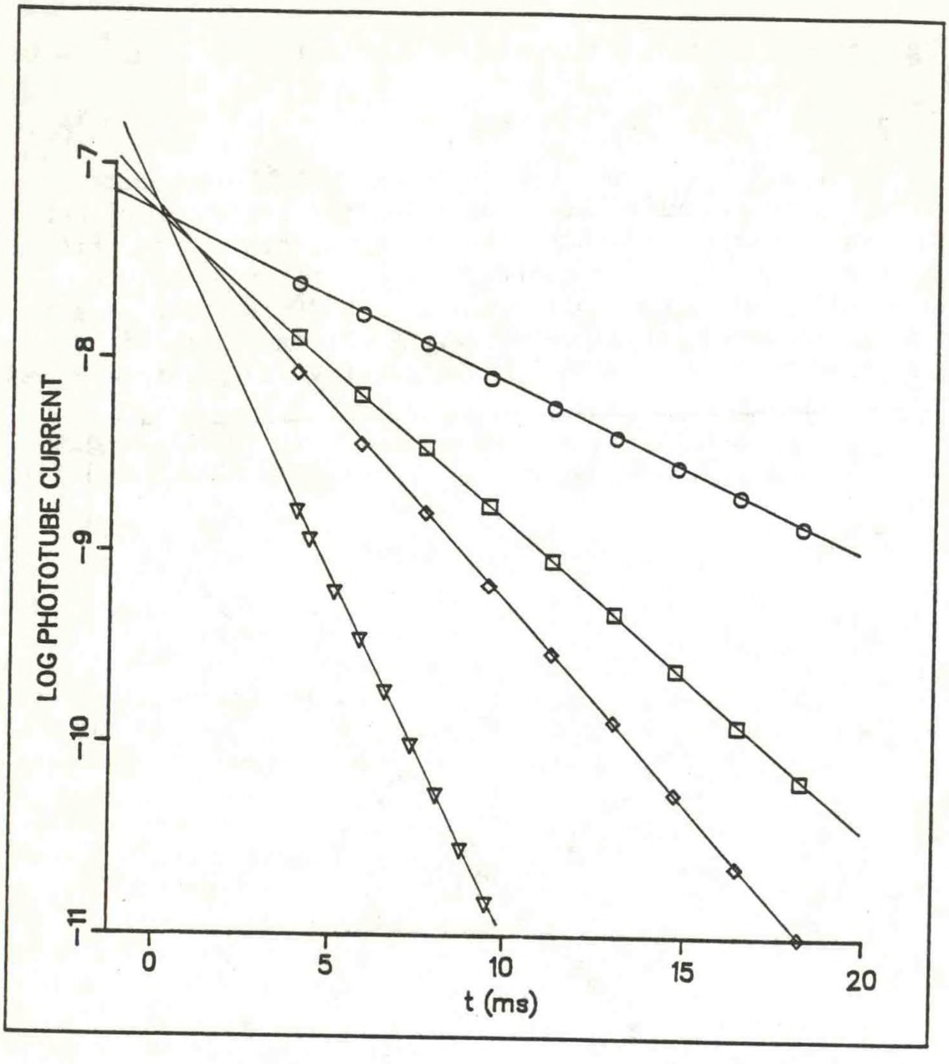

Figure 2. Sample data showing the loss of sodium atoms in a flow tube reactor as a function of time. The vertical scale gives the fluorescence intensity, which is proportional to the atom concentration, falling over several decades. The different symbols indicate different concentrations of ozone: (o) $[O_3] = 0$, (□) $[O_3] = 3.72 \times 10^{11}$ molecule cm^{-3}, (◇) $[O_3] = 5.68 \times 10^{11}$, and (▽) $[O_3] = 1.45 \times 10^{12}$. When $[O_3] = 0$, the loss is caused by diffusion alone. T = 290 K, P = 0.916 torr helium, and $\bar{v} = 2740$ cm s^{-1}.

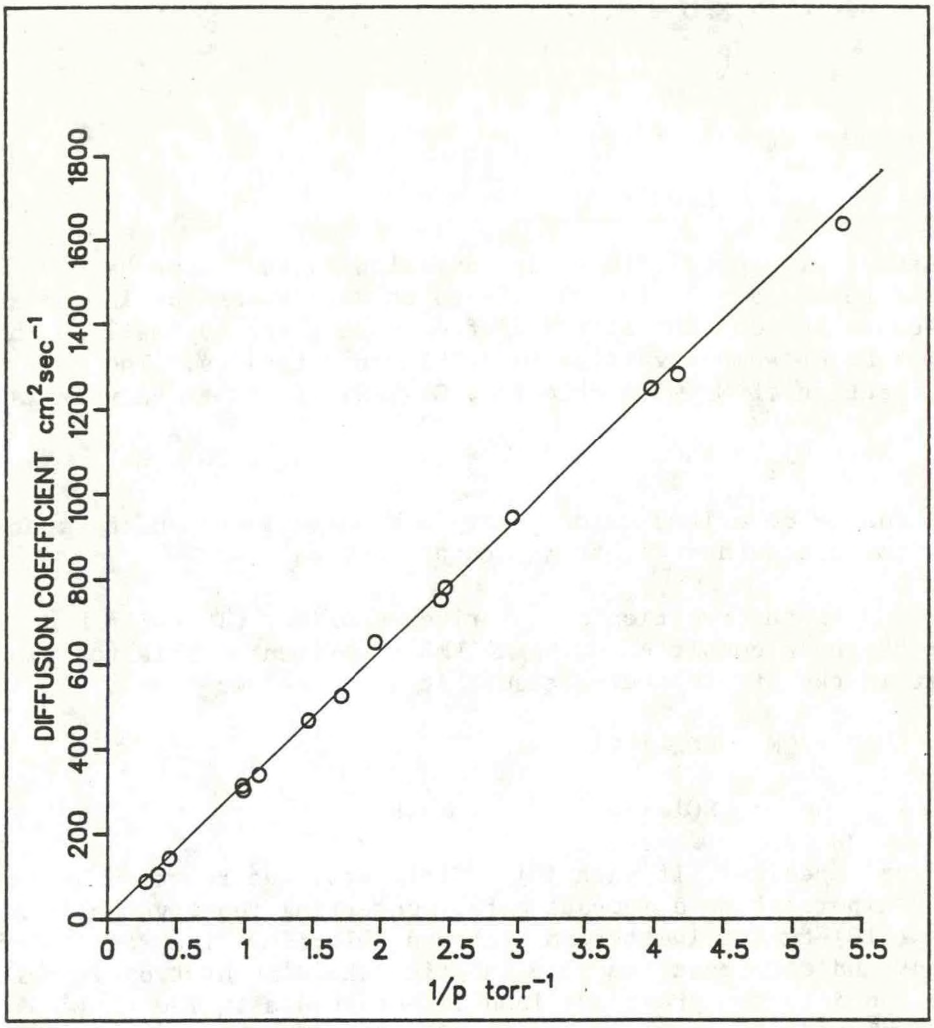

Figure 3. Plot of diffusion coefficient for sodium in helium versus the reciprocal of pressure. Gas kinetic theory predicts that the gaseous diffusion coefficient is proportional to 1/P. The slope of the line gives the pressure independent coefficient $D_p = 315 \pm 30$ torr cm^2 s^{-1}.

Sodium is deposited in the upper atmosphere by meteors. It was recently proposed that the presence of Na in the stratosphere could modify the chemistry of chlorine species which have been shown to be effective ozone destruction catalysts. This study has shown that the reaction of Na with ozone is very rapid, $k = (7.0 \pm 1.5) \times 10^{-10}$ cm^3 molecule^{-1} and that the NaO product undergoes a second rapid reaction with ozone to regenerate the Na.

$$Na + O_3 \rightarrow NaO + O_2$$

$$\rightarrow NaO_2 + O \quad (?)$$

$$NaO + O_3 \rightarrow Na + 2O_2$$

$$\rightarrow NaO_2 + O_2$$

The existence of a catalytic cycle involving Na and ozone has not been previously reported. It is unlikely to be very important in the stratosphere because the concentrations of free sodium are so small in that region but it may be of some significance at higher altitudes. The rate coefficient for the reaction of Na with chlorine, Cl_2, was found to be very large,

$$Na + Cl_2 \rightarrow NaCl + Cl \; ; \quad k = (1.1 \pm 0.3) \times 10^{-9} \; cm^3 \; molecule^{-1} s^{-1}$$

but this can be described using a simple kinetic model and parameters derived from the Na and Cl_2 transport properties.

A study of the reaction of chlorine monoxide, ClO, with hydroxyl radicals has been completed using an LMR experiment. This reaction is important in the stratosphere because it involves two

$$ClO + OH \rightarrow HO_2 + Cl \qquad (a)$$

$$HCl + O_2 \qquad (b)$$

key chemical species. If path (b) is followed, the reaction becomes extremely important as a mechanism for converting reactive chlorine radicals (ClO) to the inert form hydrogen chloride. The results of this study indicate that the rate coefficient is about two times larger than was found in two previous, less direct studies. The yield of hydrogen chloride is small, ≤ 15% at 298 K, but the uncertainty limits in the present result does not place it at the insignificant level. The temperature dependence of the reaction was also measured as shown in Figure 4. The kinetic data on path (a) was used to establish the heat of formation of the hydroperoxyl radical, $\Delta H^{\circ}_{f_{298}}$ $(HO_2) = 3.0 \pm 0.4$ kcal mol^{-1}.

The HS radical has been detected in an LMR experiment. This development makes it possible to study the kinetics of HS which is important as a precursor to sulfuric acid. HS is known to be an intermediate in the atmospheric oxidation of H_2S and is thought to be involved in the oxidation of other sulfur compounds such as COS and CS_2. The detection limit for HS in this experiment is approximately 2×10^9 molecule cm^{-3}.

A new experiment employing a high resolution Fourier transform spectrometer has been developed to evaluate the products of atmospheric radical reactions. The experiment consists of discharge and hot wire radical sources, a reactor, a 1.6 meter long multipass absorption cell with a high speed pump, and the Fourier transform interferometer.

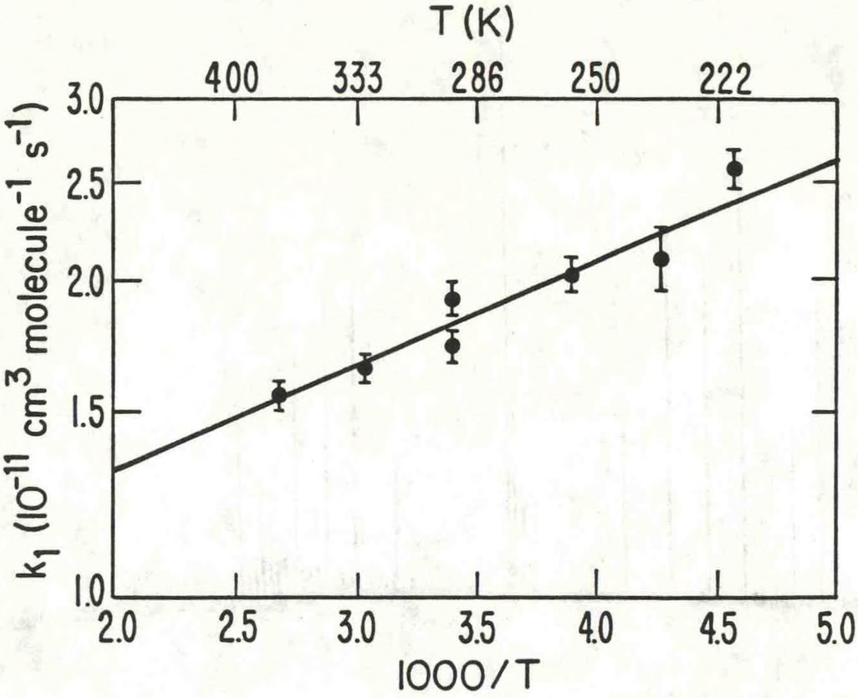

Figure 4. Arrhenius plot for data on OH + ClO reaction. Rate coefficients were measured at temperatures between 219 and 373 K. The slope of the line gives the temperature dependence. $k(T) = (8.0 \pm 1.4) \times 10^{-12} \exp[(235 \pm 46)/T]$ cm^3 $molecule^{-1}$ s^{-1}.

Using multiple reflections the multipass cell has produced optical paths up to 100 m. The first tests with this system have demonstrated that product molecules are detectable in the concentration range 10^9 to 10^{11} molecule cm^{-3}. A sample of the data from this experiment is shown in Figure 5. This spectrum was obtained for the highly reactive OH radical by infrared absorption. It demonstrates the potential of the FTS experiment to detect radical and unstable species.

A series of tests were performed using hot metal wires with the objective of developing new sources of atoms and small radicals. The metals that were tested were platinum, iridium, tungsten, nickel, and nichrome. The species that were generated included atomic hydrogen, oxygen, fluorine, chlorine and hydroxyl. All were detected by resonant fluorescence except fluorine which was measured by titration with hydrogen. Both hydrogen and oxygen atoms were generated in large amounts ($\sim 5 \times 10^{14}$ atoms s^{-1}) on about 0.1 cm^2 platinum and iridium surfaces and hydrogen was also produced on hot tungsten. No favorable source was found for fluorine, chlorine, and hydroxyl radicals.

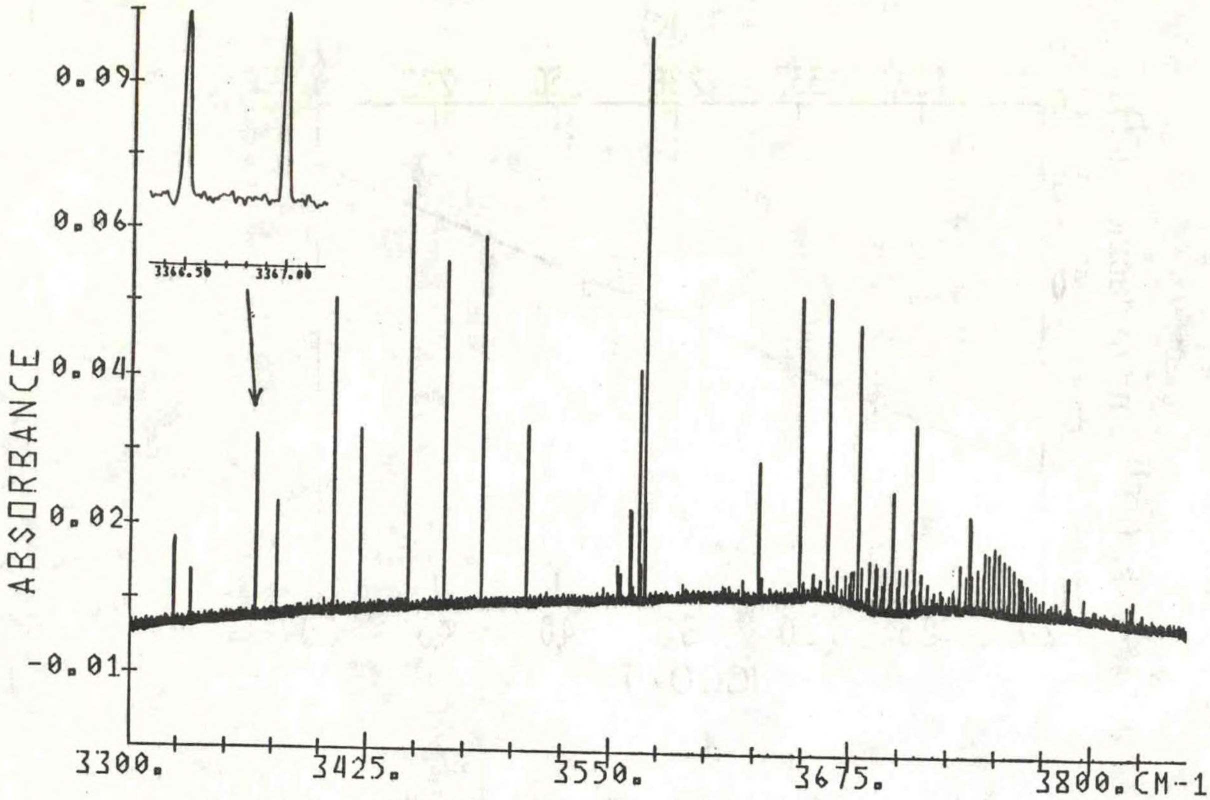

Figure 5. High resolution absorption spectrum of OH radicals. The strongest line in the middle is part of the Q branch and represents about 10% absorption in a 100m path at 298 K and 0.35 torr. The P branch is on the left and the R branch is on the right. The concentration of OH in the absorption cell was about 10^{12} molecule cm^{-3}. The weak band of lines in the R branch is thought to be from NO_2. The detail in the upper left shows a resolved Λ doublet on an expanded scale. This spectrum was taken at a resolution of about 0.02 cm^{-1}.

Future Plans

The Aeronomy Laboratory Ion Chemistry program has been terminated except as an adjunct to the neutral atmospheric kinetics program. The ion-molecule studies are now being continued in collaborative efforts at Innsbruck, Birmingham, and Meudon. Detailed NO^+ vibrational relaxation measurements from Innsbruck are being analyzed, low temperature reactions are being measured in Meudon and kinetic energy dependencies are being measured in Birmingham. In addition there is a collaboration with groups at Heidelberg and Dallas on the interpretation of <u>in situ</u> ion composition measurements.

A new experiment to study gas phase reactions at atmospheric pressure will be initiated. Free radicals for these studies will be generated by pulsed laser photolysis of stable molecules. Radical detection will be achieved using resonant fluorescence, laser induced fluorescence, or long path absorption techniques. This experiment will be directed toward investigating reactions which are thought to exhibit a pressure dependence and which are related to the formation of acid species in the troposphere.

An experiment will be developed to study the products of atmospheric photochemical processes. Although a great deal is known regarding the rates of such reactions, there is often a major uncertainty associated with the product yields. The product identities are critically important in determining the role of a reaction in the atmosphere. The objectives of this experiment will be to quantitatively evaluate the products of photochemical processes such as the photolysis of NO_3 and the reaction of $O(^1D)$ with N_2O. Various optical techniques and mass spectrometry will be used to measure the product yields.

Further studies will be carried out on the SO_2 oxidation mechanism. First an effort will be made to measure the efficiency of the conversion of $HOSO_2$ to HO_2 and SO_3. Then a study of SO_3 kinetics will be initiated using chemical ionization detection to see if the gas phase reaction with water proceeds with a significant rate.

The kinetic studies of NO_3 reactions will be continued, the temperature dependence study of the $NO + NO_3$ reaction will be completed first, then the reaction of NO_2 with NO_3 will be investigated as a function of temperature and pressure.

The reaction of Na with ozone will be studied further with the objective of measuring the rate coefficients associated with the catalytic ozone destruction cycle. The temperature dependence of these processes and the Na diffusion coefficient will also be studied.

The laser magnetic resonance detection of HS radicals will be pursued with the objective of studying the reactions of HS with atmospheric gases such as O_2, O_3, and NO_2. A search for the HSO radical, which is expected to be the product of the O_3 and NO_2 reactions will also be made.

Product detection studies will be continued with the Fourier transform spectrometer experiment. The current experiments on unstable molecules such as OH will be extended to include searches for HOONO, $HOONO_2$, and HO_2 radicals.

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Introduction

This program utilizes optical measurements of the atmosphere as a tool for studying fundamental atmospheric processes such as energy balance, composition and dynamics. The center for the observational program continues to be the Fritz Peak Observatory in the mountains west of Boulder. For more than a decade after its establishment the observational program was concentrated on measurements of light emitted in the upper atmosphere (> 70 km) either from chemiluminescent airglow reactions or the aurora. This was at a time when the Commerce Laboratories in Boulder were principally concerned with ionospheric radio propagation and so with the physical state of this same high region of the atmosphere. With time the program moved towards more current concerns such as the upper portion of the ozone layer and the temperature and winds in the upper atmosphere, the latter being of importance to the perturbation of satellite orbits.

The major change in the program has been an expansion into studies of the lower atmosphere to include both the stratosphere and troposphere. The composition and chemistry of the lower atmosphere had begun to receive major attention as the fragility of the ozone layer and problems of pollution became apparent. The lower atmospheric studies have generally utilized measurements of absorption by molecules rather than the measurements of emission formerly employed for upper atmospheric studies. But almost all of the expertise and optical equipment previously developed could be immediately applied to these new areas and so the program was able to move swiftly into a leading role in them. In so doing we have exploited the extraordinary sensitivity of optical absorption for the detection and quantitative measurement of minute quantities of chemically important species in both the stratosphere and the troposphere. The location of the Fritz Peak Observatory has proven to be a very favorable one in the new program, particularly for troposphere studies, since the wind patterns allow us to both study the occasional downwind pollution from Denver as well as the extremely clean air experienced during the normally westerly wind flow. We have also carried out measurements at a large number of other locations using platforms ranging from vans, ships, and aircraft to the presently operational Solar Mesosphere Explorer satellite built and operated by the University of Colorado in Boulder. In all of this work we have taken advantage of the flexibility of a small group to quickly adapt broad and powerful optical techniques to new problems in the atmospheric as they arise.

Major Areas of Investigation

Stratospheric and Tropospheric Composition

A technique was developed for separately determining the stratospheric and tropospheric abundance of NO_2 using ground based absorption spectroscopy. Changes in the abundance and altitude distribution of NO_2 in the stratosphere can be followed on a routine basis; at the same time the variations in tropospheric NO_2 with changing weather patterns can be studied. In the stratosphere NO_x ($NO + NO_2$) is the principal naturally occurring species controlling the ozone abundance. In the troposphere it plays a similar important role.

The natural clean air background of NO_2 has been found to be very much lower than previously reported and a major enhancement was discovered in thunderstorms. A method has been developed for measuring NO_2 in the free troposphere from mountain tops or aircraft. The results from Mauna Loa, Hawaii, confirm that NO_x is extremely low in mid-Pacific air. Measurements made on board a jet aircraft have revealed a large and unexpected decrease in stratospheric NO_2 at high latitude in winter with a return to high values in the summer. Only now is the interpretation of this beginning to emerge, largely as a consequence of studies by Dr. Solomon of the Aeronomy Laboratory and her collaborators. Major perturbations in the structure of stratospheric NO_2 accompany the formation of a jet stream and others have been shown to be associated with a change in the large-scale stratospheric circulation pattern which accompanies a stratospheric warming.

Efforts to understand these new observations are proving to be of value in the larger problem of the effect of nitrogen oxides in controlling the ozone layer. Automated instruments for monitoring NO_2 by this technique were installed at a number of NOAA GMCC stations at both high and low latitude. The results from Barrow, Alaska, confirm earlier airborne observations at high latitude and show changes at stratospheric warmings. These changes have been identified with major alterations in the quasi-horizontal flow patterns in the stratosphere. Under certain circumstances the bahavior of NO_2 can thus be used to study stratospheric circulation patterns.

The technique for NO_2 was extended to give the first measurements of NO_3 in the atmosphere; NO_3 is an important intermediate in the nitrogen chemical cycle. The method distinguishes between stratospheric and tropospheric NO_3 when the moon is used as the light source. We have also studied NO_3 in the troposphere over a horizontal 15 km path. The NO_3 behavior exhibits a number of unexpected facets. In the troposphere NO_3 is considerably below the amount expected and the evidence is strong that the molecule is attached by an as yet unknown scavenger. The unraveling of the NO_3 anomalies is likely to lead to changes in our understanding of atmospheric odd nitrogen.

We have now completed a 5-year study of stratospheric NO_3 which shows it to be controlled by large circulation in the stratosphere; an unknown species appears to be created in air at high altitude which has the ability to destroy NO_3 (See Figure 1). We have begun a new series of observations of stratospheric NO_3 with the diode array spectrometer which has already given a wealth of new information.

To further improve the ability to study chemically important tropospheric gases, such an NO_2 and NO_3, a powerful well collimated light source has been installed 10 km from the Observatory. We expect this long path absorption facility to become one of our principal centers of activity in the next few years with the use of new instrumentation such as the sensitive diode-array spectrograph developed in the Aeronomy Laboratory by Dr. Schmeltekopf. In 1983 and 1984 a new series of NO_3 observations has been carried out using this facility.

The Florida Atlantic University "PEPSIOS" interferometer has been used successfully to measure stratospheric OH. This important and highly reactive species has now been studied for several years by the Burnetts at Fritz Peak and their work continues to yield almost all of the information presently available on its behavior (See Figure 2).

The Solar Mesosphere Explorer Satellite was launched successfully in October 1981 and continues to return data on stratospheric ozone and a number of other species important in its creation and destruction. As a result of our suggestion in the planning stages the satellite measures stratospheric NO_2 on a global scale, (see figure 3). We are actively participating in the analysis of these measurements as are others from the Aeronomy Laboratory. The results for NO_2 confirm the global picture obtained earlier by us but will naturally lead to a far better and more detailed one than previously possible. One of the most interesting aspects is the ability to study global circulation patterns in the stratosphere using NO_2 as a tracer of the motion. The satellite is also used to measure tropospheric NO_2 by observing the light backscattered from the earth's surface. This permits observation of pollution, and its dispersal from space on a global scale. This unplanned extra output from SME is being actively pursued and has already revealed a large contribution from lightning to the tropospheric NO_x budget.

The eruption of the El Chichon volcano in Mexico drastically altered the aerosol composition of the stratosphere. The dust cloud was observed by the SME satellite and we have studied the unexpected effects of the aerosol upon stratospheric NO_2. To do so we had to revert to the older ground based methods since the dust itself temporarily rendered SME incapable of detecting NO_2 itself. A remarkably large decrease in stratospheric NO_2 is associated with certain regions of the cloud; no explanation for this yet exists. Whenever the NO_2 is reduced there is a corresponding small increase in the total column abundance of stratospheric ozone. This appears to be the first direct evidence that the total column of ozone does exhibit the theoretically expected dependence upon a measurable change in NO_2 in the stratosphere. The Burnetts have also discovered a sizable effect of the dust cloud upon stratospheric OH.

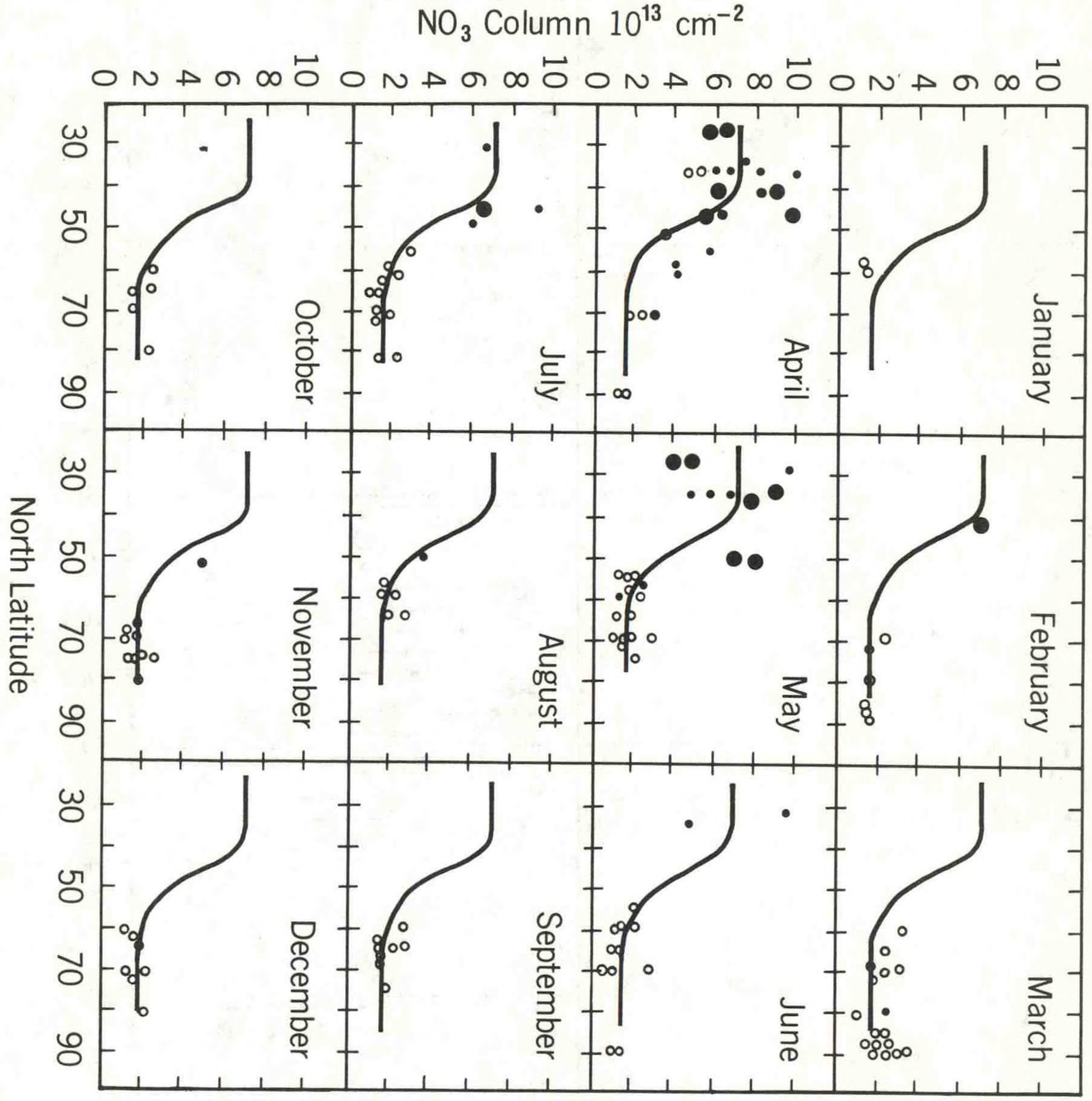

Figure 1. Measurements of stratospheric NO_3 at Fritz Peak, 1978-1983, plotted against the highest latitude experienced by stratospheric air at NO_3 altitude (~35 km) prior to arrival over Colorado. The high correlation suggests production of an NO_3 scavenger at high latitude.

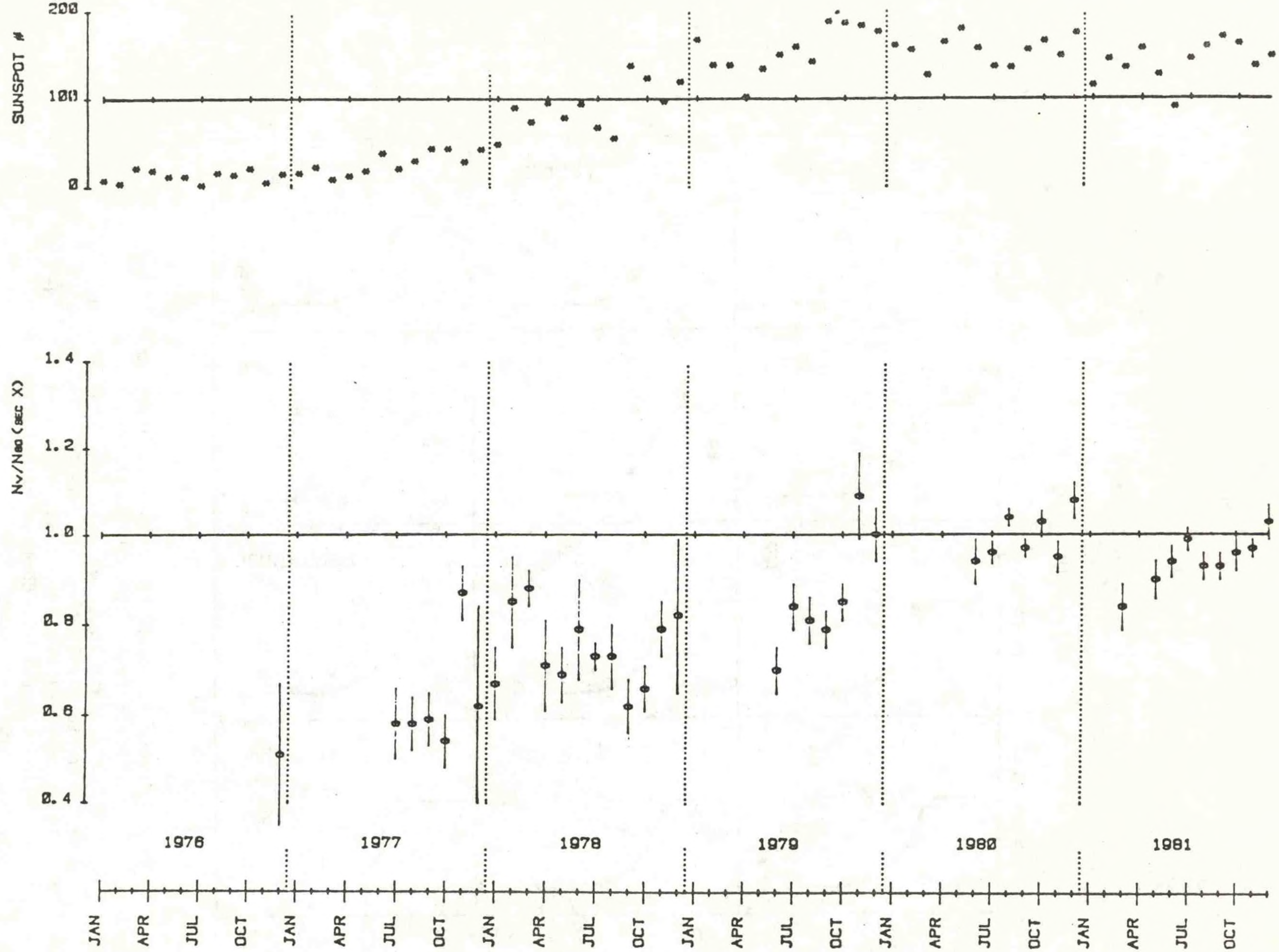

Figure 2. Normalized monthly stratospheric OH abundance and sunspot numbers: 1976 - 1981. The similarity of the long-term trends of the OH abundances and sunspot numbers is apparent.

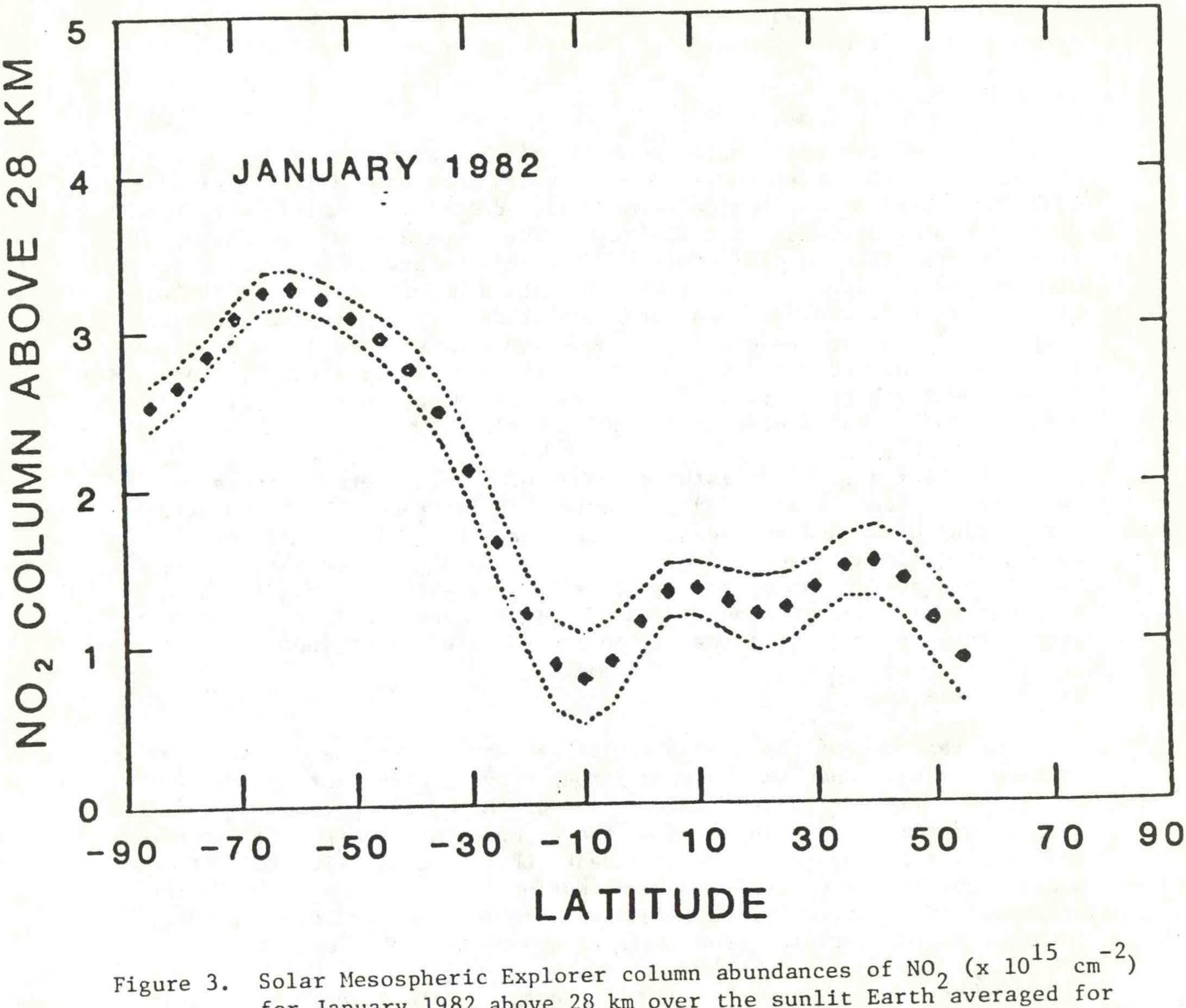

Figure 3. Solar Mesospheric Explorer column abundances of NO_2 ($\times 10^{15}$ cm^{-2}) for January 1982 above 28 km over the sunlit Earth averaged for longitudes between $40^{o}W$ and $130^{o}W$. The dashed lines show the $\pm 1\sigma$ standard deviation of the data at each latitude, including both geophysical "noise" and instrumental noise.

Mesospheric and Thermospheric Studies

1. Dynamics and thermal structure in the thermosphere

High resolution interferometric techniques have been applied to the study of atmospheric emission lines at Fritz Peak Observatory in order to determine both temperature and wind fields in the thermosphere, fundamental quantities which cannot be directly determined by other means. The observational program, coupled with theoretical modeling, provides insights on the dynamics and thermal structure of the atmosphere above ~ 70 km.

a) Solar minimum

The geomagnetically quiet behavior of the thermosphere during solar minimum has been determined, and the results show the existence of strong equatorward winds during the summer which decrease to fairly weak winds, sometimes poleward, during the winter. The temperatures show lower values in the winter than in the summer. These results are in general agreement with the predictions of a General Circulation Model (GCM) where the winds are primarily driven by global pressure gradients established by solar heating and high-latitude heat and momentum sources at a large distance from Fritz Peak Observatory. The present observations showed the necessity to include the high latitude forcing as a permanent feature of the global circulation, in particular during quiet periods.

The geomagnetically disturbed behavior of the thermosphere shows enhanced temperature and wind gradients with latitude. The wind structure during disturbed periods tends to have a very strong equatorward component reaching speeds of 640 m/s. Large scale thermospheric waves have been observed during some of these geomagnetic storms. The high latitude energy source required to explain the observations has been successfully parameterized using the auroral electrojet index AE.

b) Solar maximum

The response of the atmosphere to geomagnetic storms during solar maximum is quite complex, showing strong convergences lasting for several hours, as well as elevated temperatures of the order of 2000 K. This has been interpreted again in terms of the expanded magnetospheric pattern reaching the neighborhood of Fritz Peak, thus placing Fritz Peak at the boundary of two circulation patterns during part of the storm. Further measurements at other latitudes are now in progress, or being planned, in order to further understand this thermospheric behavior.

The need for wider latitudinal coverage of atmospheric circulation has been partially fulfilled by making observations at high latitudes near Fairbanks, AK (64^{o} N), in collaboration with the Geophysical Institute of the University of Alaska, and at the University of Michigan field site at Calgary, Alberta (51^{o} N). The observations show considerable meridional divergences in the winds over the length of the chain of these stations, in keeping with the localized heating effects at high latitudes. Some of the recently initiated investigations at high latitude have revealed the presence of large vertical winds in both the lower and upper thermosphere. These vertical motions appear both as short-term (minutes) and long-term effects (hours). The short-term effects are interpreted as being caused by the localized heating at those latitudes but the long term effects require further observations.

Investigations of the upper thermospheric temperature at 40^{o}N latitude over an 12-year period have provided the ability to study solar, geomagnetic and long-term effects on the thermosphere. An unexpected finding is the existence of a semi-annual variation of the temperature in addition to the known semi-annual density variation. The present (one station) results do not provide confirmation for the hypothesis of a semi-annual Joule heating effect or of planetary wave energy leakage into the upper atmosphere that are normally invoked to explain the semi-annual variation in the densities.

Global studies of the dynamical and thermal behavior of the atmosphere by means of high resolution optical techniques are to be undertaken using the Space Shuttle as a platform. These studies will be carried out in collaboration with colleagues at the University of Michigan, University College (London), York University, and the National Center for Atmospheric Research.

Measurements of vertical motions of the atmosphere have been made with the prototype high luminosity TESS device. The results show large amplitude (~ 40 m/s) oscillations with a periodicity of about 40 minutes, coupled with small emission rate changes. This has been interpreted to be the atmosphere's resonse to the passage of gravity waves since the observed periodicity is within the narrow range of periodicities possible at that atmospheric height, and the observed ratio of horizontal to vertical velocities is that expected for gravity waves. The measured vertical winds also show that individual (high-time-resolution) zenith measurements of neutral winds cannot be used as (zero) reference winds with any degree of certainty.

The first measurements of Doppler shifts (winds) from molecular, rather than atomic, species have been successfully made. This new technique opens a new vista into atmospheric dynamics, since there exist a larger variety of molecules than atoms in the atmosphere.

2. Twilight Airglow

In line with our policy of continuing a few selected studies of the upper atmosphere we observe infrared molecular oxygen emission at twilight which permit a measurement of ozone at altitudes where it cannot otherwise be determined. The dramatic seasonal and short term changes observed indicate a corresponding change in the upflow of hydrogen compounds from the underlying stratosphere. In effect this permits a study of the upper boundary conditions on the stratosphere which are necessary for realistic modelling of its behavior.

Another twilight program involves measurement of emission from O^{+} ions in the upper thermosphere created by the absorption of solar ultraviolet light by O atoms. We can thus determine directly the density of the atmosphere at high altitudes where the majority of satellite instruments can no longer make in-situ observations. It is thus possible to follow the changes in upper thermospheric composition with season and geomagnetic activity.

3. Mesospheric Gravity Waves

Both the O_2 and OH emission in the nightglow can be analyzed to yield a relatively direct measurement of the atmospheric temperature at 95 and 85 km respectively. The most important discovery has been that large periodic fluctuations exist at both altitudes and show a phase coherence similar to that expected from internal gravity waves originating in the troposphere as well as in the high latitude auroral mesosphere. An instrument to monitor mesospheric gravity waves is now in automatic operation. When analyzed in conjunction with the nonlinear theory of gravity waves developed by Weinstock these observations permit a study of the deposition of heat and turbulent energy in the lower thermosphere by internal gravity waves. Correlations are evident between these waves and other mesospheric quantities, such as ozone.

Spectroscopic Instrumentation

A theoretical study of high resolution spectrometers shows that the ultimate precision and accuracy of measurements is limited by the characteristic noise spectrum of the nearly incoherent radiation sources under measurements, the instrumental spectrum, as well as the noise spectrum of unwanted radiation. The results of this study are applicable to any instrument which depends on the interference of light for its operation.

The conventional high-resolution Fabry-Perot spectrometer is light-limited in its operation. This limitation exists because only in interference ring is utilized out of the 50,000 or more available. Recently we have broken this barrier and we find we are able to use (theoretically) all the available light from this instrument. A practical version of this multiplexed Fabry-Perot spectrometer has been built where we find that gains of 100 in the throughput of the instrument are easily attainable. This high luminosity and high resolution spectrometer has been dubbed TESS, which is the acronym for a Twin Etalon Scanned Spectrometer.

The observations of atmospheric emissions in the auroral zone have, because of the large changes in the source irradiance over short periods of time, led into a new method of measurement. This method is called an equal noise technique, because the measurements are limited to a fixed signal to noise ratio. The basic method consists of measuring for each spectral element only long enough to reach a preset signal to noise ratio, recording this time and then moving on to the next spectral element and so on. This new method is of general applicability to spectroscopic problems, such as hyperfine structure investigatious, since the signal-to-noise ratio is fixed for all features present in the spectrum, regardless of their intensity.

Future Plans

The outline of our current areas of study is intended to indicate our concern with using optical methods to open up new areas in atmospheric studies and to pursue them as long as important results continue to emerge or until a clear pattern of change with time is evident in the species measured. Major emphasis will of course remain upon studies of the troposphere and stratosphere, their composition and the exhange between them. The stratosphere is both a source and sink for minor species in the troposphere and so one must consider both regions jointly in many cases.

Optical methods lend themselves well to many remote sensing problems, particularly those involving the measurement of minor but chemically important species in both the troposphere and stratosphere. The long path absorption facility at Fritz Peak has only begun to be exploited and we expect the coming years to see a program develop in which a number of important tropospheric species can be studied. One great advantage of the long path absorption technique is that it provides an unambiguous quantitative spectral identification with perturbing the atmosphere itself.

While the great outflow of data from satellites, in particular the Solar Mesosphere Explorer, will receive our active study, the recent experience with the volcanic dust cloud shows once again that carefully planned ground based studies of the stratosphere must continue to be carried through. Thus we shall maintain a strong activity in such studies.

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AERONOMY LABORATORY SEMINARS

1983 - 1984

October 17, 1983	Measurements of Nitric Oxide Fluxes from Various Soils, Dr. Franz Slemr, Max-Planck Institute for Atmospheric Chemistry, Mainz, West Germany
November 9, 1983	The Measurement of Atmospheric Free Radicals, Prof. Donald Stedman, Chemistry Department, University of Denver, Denver, Colorado
November 30, 1983	Radon and Tropospheric Vertical Transport, Dr. John McAfee, Aeronomy Laboratory, Boulder, Colorado
December 7, 1983	Optical Remote Sensing of Atmospheric Turbulence by Stellar Scintillation: A Comparison with Simultaneous Sunset Radar Measurements, Jean Vernin, Department d' Astrophysique, Université de Nice, France
December 21, 1983	Fine Structure Within an Elevated Inversion and Its Implication for Ground-based Profiling Systems, E. E. Gossard, CIRES, University of Colorado, Boulder, Colorado W. Neff and R. Zamora, Wave Propagation Laboratory, Boulder, Colorado
January 11, 1984	Stratospheric Bromine Reconsidered, Dr. Tony Delany, Atmospheric Chemistry and Aeronomy Division, National Center for Atmospheric Research, Boulder, Colorado
January 25, 1984	The Aeronomy Lab Ion Chemistry Program, 1983, Dr. Eldon Ferguson, Aeronomy Laboratory, Boulder, Colorado
February 8, 1984	New Developments in Tunable Diode Laser Spectrometry for Tropospheric Measurements, Prof. Harold Schiff, York University, Downsview, Ontario, Canada
February 22, 1984	The Role of NO_3 in Day and Nighttime Chemistry of Nitrogen Oxides, Dr. Dieter Perner, Max Plank Institute für Chemie, Mainz, Germany

February 29, 1984	The Objectives and Plans for the Stratospheric-Tropospheric Exchange Program (Including the Micronesia Cumulonimbus Experiment), Dr. E. F. Danielsen, NASA/Ames Research Center, Moffitt Field, California
March 21, 1984	Termites: A Potential Source of Tropospheric Methane, Dr. W. Seiler, Max Planck Institute for Chemistry, Mainz, Germany
April 4, 1984	Tropospheric Ozone: Observed and Modeled, Dr. Hiram Levy, II, Geophysical Fluid Dynamics Laboratory, NOAA, Princeton, New Jersey
April 11, 1984	Recent Laboratory Studies of Electronic Recombination, Electron Attachment and Ion-Molecule Reactions of Atmospheric Significance, Dr. Nigel Adams, Dept. of Space Research, University of Birmingham, Birmingham, England
April 12, 1984	Atmospheric Background Measurements of NO_x and PAN, Dr. Peter Warneck, Max-Plank Institute für Chemie, Mainz, Germany
May 18, 1984	Studies of Middle Atmosphere Dynamics Using VHF and HF Radars at Adelaide, Australia, Prof. Robert A. Vincent, Dept. of Physics, University of Adelaide, Adelaide, Australia
May 24, 1984	Review of NASA's Global Habitability Program, Dr. Robert T. Watson, Jet Propulsion Laboratory, Pasadena, California, and NASA Headquarters, Washington, D.C.
May 30, 1984	Eiscat Observations of the Mesosphere, Stratosphere, and Troposphere, Dr. Jürgen Röttger, Eiscat Scientific Association, Kiruna, Sweden
June 6, 1984	Some Considerations for Modelling the Dynamics of the Mesopause Region, Dr. Adolf Ebel, Institüt für Geophysik und Meteorologie, Universität zu Köln, Federal Republic of Germany
June 13, 1984	Internal Gravity Waves in the Ocean: A Review of Their Spectral Distribution and Dynamics, Dr. Peter Müller, Department of Oceanography, University of Hawaii, Honolulu, Hawaii

June 27, 1984	On the Interaction of Gravity Waves and Convective Storms, Prof. Franco Einaudi, School of Geophysical Sciences, Georgia Institute of Technology, Atlanta, Georgia
July 18, 1984	Using Radioactive Nuclei to Study Boundary Layer Free Atmosphere Exchange, Dr. Mark Kritz, NASA/AMES Research Center, Moffitt Field, California
August 1, 1984	MASS Transfer of Trace Gases to Cloud and Rain Drops, Dr. Chris J. Walcek, Acid Deposition Modeling Project, National Center for Atmospheric Research, Boulder, Colorado
August 15, 1984	Measurements of Hydroxyl Radicals by Long Path Absorption Spectroscopy, Dr. Gerhard Hübler and Dr. Uli Platt, Kern Forschungs Anlage, Jülich, Federal Republic of Germany
September 19, 1984	The Tropical Tropopause and Global Climate, Dr. George Reid, NOAA, Aeronomy Laboratory, Boulder, Colorado
September 26, 1984	A Theoretical and Experimental Study of a Universal Gravity Wave Spectrum in the Atmosphere, Dr. Edmund M. Dewan, Air Force Geophysics Laboratory, Hanscom AFB, Massachusetts