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RECENT RESEARCH RESULTS AND ONGOING AND  
PLANNED RESEARCH PROGRAMMES

submitted by the

United States of America

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Current Program

The United States of America has developed and is implementing a comprehensive program of research, technology, and monitoring of the Earth's upper atmosphere, particularly the stratosphere. This program is focused on expanding our understanding of this important region of our atmospheric environment and on developing our ability to assess potential perturbations, particularly to the ozone layer. The long-term objectives of the present program are to perform research to:

- a. understand the physics, chemistry and transport processes of the upper atmosphere, and
- b. accurately assess possible perturbations of the upper atmosphere caused by man's activities.

The program involves analysis of data obtained by large satellite missions such as Nimbus-4, SAGE, Nimbus-7, and the Solar Mesospheric Explorer, as well as a variety of research tasks proposed by scientists from the university, government, and industrial research communities. In addition, periodic assessments of the state of our knowledge of the upper atmosphere and of the effects of specific perturbations such as chlorofluoromethane (CFM) releases, aircraft effluents, and other potential pollutants are sponsored. Of greatest urgency at present is an assessment of the combined effects of continued increases in the atmospheric concentrations of  $\text{CFCl}_3$ ,  $\text{CF}_2\text{Cl}_2$ ,  $\text{CH}_3\text{CCl}_3$ ,  $\text{CO}_2$ ,  $\text{NO}_x$  possibly, and other gases such as  $\text{N}_2\text{O}$  and  $\text{CH}_4$ . Another goal is to understand the role of stratospheric ozone in the radiative heating and thus the dynamics of the atmosphere, and to thereby assess the importance of chemical-radiative-dynamical feedbacks on the meteorology and climatology of the stratosphere and troposphere. In the non-satellite portion of the program, activities generally fall into four broad categories: (1) field measurements (in-situ and remote sensing techniques using ground-based instruments, aircraft, balloon, and rocket platforms); (2) laboratory studies (gas kinetics, photochemistry, spectroscopy, and the development of calibration standards); (3) theoretical studies (1-D, 2-D and 3-D models); and (4) data analysis (especially analysis of large satellite data sets). Specific investigations include:

- a. Determination of the distribution of trace gases in the upper atmosphere, with emphasis on those which influence the ozone balance.
- b. Observations of the global distribution of ozone, its vertical profile, and temporal variations.
- c. Improvements in the theory of upper atmospheric photochemical processes, and validation of theory by comparison with measurements.

- d. Improvements in the understanding of atmospheric dynamics and transport processes by both theory and measurement.
- e. Determination of the characteristics of motions responsible for the exchange of air between the troposphere and stratosphere.
- f. Determination of the geographic distribution and strengths of sources and sinks for stratospheric compounds.
- g. Measurements of the ultraviolet solar irradiance and its temporal variations.
- h. Laboratory studies in spectroscopy and chemical kinetics relevant to the interpretation of atmospheric measurements and to theoretical simulations of the atmosphere.
- i. Development of new technological ideas, techniques, and instruments for use in upper atmospheric research.

During the last two years substantial advances in our knowledge of the upper atmosphere have been made in each of the four major research categories. Laboratory studies have strengthened our knowledge of the chemical kinetics of stratospheric components, and have provided improved data on the spectroscopy of atmospheric gases for applications to atmospheric measurements of trace species. Many of the trace gases in the hydrogen, nitrogen, and chlorine chemical families which participate in the chemistry of ozone have now been detected in the stratosphere, while investigation of their spatial and temporal variations is underway.

In order to judge our understanding of the important chemical and physical processes that occur in the stratosphere, the theoretically predicted trace gas distributions are compared to experimental field observations. To date, the accuracy of the reported field measurement data has been difficult to assess and the reported data on the concentrations of most species show large scatter that can either be due to atmospheric variability or instrumental differences. Consequently, the comparison of observational data with theory is currently limited in scope. In order to be able to constrain or test the theoretical models, the reason for the scatter in the data must be understood. Therefore, it has been essential to perform a series of instrument intercomparisons under field conditions.

A major effort has been expended during the last few years to determine the accuracy and precision of atmospheric chemical composition measurements by conducting a series of international intercomparison balloon campaigns where chemical constituents were measured using a variety of observational techniques simultaneously in time and space. The intercomparison campaigns performed to date include:

- a. A series of three campaigns, employing both in-situ and remote sensing balloon and rocket-borne instruments, to measure ozone.

- b. Two campaigns utilizing remote sensing balloon-borne instruments (13 on the first, and 18 on the second), using eight different techniques including grating spectrometers, radiometers, and Fourier transform interferometers, to measure several key atmospheric constituents including  $\text{HNO}_3$ ,  $\text{NO}_2$ ,  $\text{NO}$ ,  $\text{HCl}$ ,  $\text{HF}$ ,  $\text{O}_3$ ,  $\text{H}_2\text{O}$ ,  $\text{CH}_4$ , and  $\text{OH}$ . These sensors utilized the visible, infrared, far-infrared, and microwave region of the electromagnetic spectrum in both the absorption and emission modes.
- c. Three campaigns employing in-situ and remote sensing balloon-borne instruments to measure water vapor.

The data from these intercomparison campaigns is currently in the final stages of analysis. The results have shown that some of our current measurements, such as those for  $\text{O}_3$ , are accurate to better than ten percent. Others are accurate to no better than a factor of two, e.g., those for  $\text{NO}_2$ .

Within the last few years, several newly developed in-situ and remote sensing techniques have been demonstrated for species for which there had been inadequate measurements, e.g.,  $\text{OH}$ . In addition, some existing techniques have been improved which will result in greater sensitivity for a number of species, e.g.,  $\text{ClO}$  and  $\text{O}_3$  by balloon-borne microwave emission. This newly developed and improved instrumentation is required to measure nearly all key atmospheric species over a significant altitude range with the accuracy and precision sufficient to critically test our understanding of atmospheric photochemistry. Techniques that have been demonstrated within the last four years include:

- a. A balloon-borne far-infrared emission interferometer for remote sensing detection of  $\text{OH}$ ;
- b. Balloon-borne laser-induced fluorescence systems for in-situ and range-resolved detection of  $\text{OH}$ ;
- c. A balloon-borne laser diode absorption system for in-situ detection of  $\text{NO}$  and  $\text{NO}_2$ ;
- d. A shuttle-borne high resolution infrared absorption interferometer (ATMOS) for detection of a wide range of chemical constituents including several of the temporary reservoir species, e.g.,  $\text{ClONO}_2$ ,  $\text{N}_2\text{O}_5$ , and  $\text{HO}_2\text{NO}_2$ .
- e. A ground-based microwave emission system for remote detection of species including  $\text{ClO}$ ,  $\text{HO}_2$ , and  $\text{O}_3$ .

Atmospheric dynamics studies are continuing on (1) the exchange of air between the troposphere and stratosphere using aircraft, and (2) turbulence and gravity wave phenomena using ground-based radar.

A number of laboratory kinetics and photochemical studies are being pursued. Key new data on  $\text{NO}_3$ ,  $\text{HO}_2$  and  $\text{ClONO}_2$  reactions have been obtained. Recent significant revisions in rate coefficients for the key

reactions  $O+ClO$  and  $OH+HCl$  have been confirmed. Reaction rates which strongly affect the hydroxyl radical concentration, such as  $OH+HNO_3$ , and  $OH+HO_2$ , are now better known under stratospheric conditions. A comprehensive program of spectroscopic studies in the ultraviolet, infrared, and microwave regions is being carried out to support the field measurement program and to improve the accuracy of our calculations of the penetration of solar radiation through the atmosphere.

A solar UV-B radiation computer-developed almanac has been completed. The almanac contains data on solar UV-B radiation fluxes at the earth's surface for combinations of radiation wavelengths, total-column ozone, latitude, season, and time of day. The almanac's intended users are researchers, such as biologists studying health, terrestrial or aquatic effects of UV-B radiation, who need to simulate the solar UV-B radiation fluxes as they correlate to various amounts of total-column ozone change at their laboratory locations.

Development of general circulation models for both troposphere and stratosphere continues. Theoretical studies using one- and two-dimensional models continue to investigate the effects of adding chlorofluoromethanes,  $N_2O$ ,  $CO_2$ , and other species to the atmosphere. The key effects are on abundance and distribution of ozone, and consequent changes in the thermal structure and dynamics of the upper atmosphere. It is now well recognized that the chemical effects of these trace substances on atmospheric ozone are strongly coupled and should not be considered in isolation, and that their direct radiative effects on atmospheric temperature are approximately additive. The most recent assessment report (Present State of Knowledge of the Upper Atmosphere: An Assessment Report) estimates that the release of fluorocarbons 11 and 12, at the 1980 rates in an otherwise unchanged atmosphere, will result if an ozone column decreases in the range of 4.9-7%. The calculated effect would be increased to 6.1-9.6% by inclusion of effects of temperature feedback. The models continue to predict fluorocarbon-related depletions near 40 km, which are comparable in magnitude to those in previous reports ( 50%).

Two-dimensional models give similar results for the global average as the one-dimensional models for the case of CFC-only release. The two-dimensional model predicts a significant latitudinal variation in the ozone column decrease. More depletion is predicted near the poles than at the equator, with the ratio ranging from 2:1 to 4:1 depending upon the model transport formulation. Seasonal effects are predicted but are somewhat less pronounced than the latitudinal effects. In the upper stratosphere, significant ozone decreases are predicted at all latitudes for all seasons. In the lower stratosphere, the predictions show an increase in ozone below about 25 km within 40 degrees of the equator. From 40 degrees to the poles, ozone decreases are predicted at all altitudes.

Predictions of ozone change from the combined effects of changes in CFC's,  $N_2O$ ,  $CH_4$ , and  $CO_2$  demonstrate that their effects are not additive. For instance, one model predicted the following column ozone changes for a series of idealized scenarios: (1) increase of total

chlorine to 8 ppbv, -5.7%; (2) doubling of  $\text{CH}_4$ , +2.9%; (3) 1.2 times  $\text{N}_2\text{O}$ , -1.7%; and (4) doubling of  $\text{CO}_2$ , +3.5%. The same model predicted the following column ozone changes<sup>2</sup> for combinations of the isolated scenarios: (1) 8 ppbv  $\text{Cl}_x + 2 \times \text{CH}_4 + 1.2 \times \text{N}_2\text{O}$ , -2.8%; and (2) 8 ppbv  $\text{Cl}_x + 2 \times \text{CH}_4 + 1.2 \times \text{N}_2\text{O} + 2 \times \text{CO}_2$ , +0.2%. The calculated change in 40 km<sup>x</sup> ozone remains large for all of these cases (e.g., -35 to -55% for 8 ppbv  $\text{Cl}_x + 2 \times \text{CH}_4 + 1.2 \times \text{N}_2\text{O} + 2 \times \text{CO}_2$ ).

Time-dependent scenarios were run in a one-dimensional model assuming  $\text{CO}_2$ ,  $\text{CH}_4$ , and  $\text{N}_2\text{O}$  annual atmospheric concentration growth rates of 0.5%, 1.0%, and 0.2%,<sup>2</sup> respectively, in conjunction with CFC emission growth rates of 1.0%, 1.5%, and 3% per year. The ozone column changes were calculated to be relatively small (<3% over the next 70 years) for CFC emission increases of  $\leq 1.5\%$  per year, but with a CFC growth rate of 3% per year, the predicted ozone depletion is 10% after 70 years and rapidly increasing. In this case the CFC effects were temporarily masked by other gases, but eventually chlorine dominates ozone loss.

Satellite measurements provide a global-scale picture of the state of the upper atmosphere and its variations which is not feasible from measurement platforms such as balloons and aircraft. In view of this, a variety of missions conducted from earth orbit are supported. The most important development in our knowledge of  $\text{O}_3$ ,  $\text{HO}_x$ , and  $\text{NO}_x$  in the stratosphere has been the recent availability of several large validated satellite data sets (the LIMS, SAMS, and SBUV/TOMS instruments on the Nimbus-7 satellite, and the SAGE instrument on the AEM-2 satellite). They have greatly improved our knowledge of the spatial and temporal distributions of  $\text{O}_3$ ,  $\text{H}_2\text{O}$ ,  $\text{CH}_4$ ,  $\text{N}_2\text{O}$ ,  $\text{NO}_2$ , and  $\text{HNO}_3$  on a global scale. In particular, these data sets have been used to:

- a. Reconfirm that air is transported upward and poleward from the tropics, consistent with the Brewer-Dobson hypothesis for  $\text{H}_2\text{O}$ .
- b. Show that the total hydrogen budget of the stratosphere, principally  $\text{H}_2\text{O} + 2 \times \text{CH}_4$ , is relatively constant with values ranging from six to seven ppmv.
- c. Measure the total budget for odd nitrogen in the stratosphere yielding values of 20-25 ppbv, depending somewhat on the inferred concentrations of unmeasured species.
- d. Show that the thermospheric source of total odd nitrogen to the stratosphere is significant on a local, but not global, scale.
- e. Verify the ground-based observations of the British Antarctic Survey that there has been a 30-40% decrease in total column ozone over Antarctica during the spring period since the mid-1970's.
- f. Show the spatial structure and contribute to the understanding of the dynamical and photochemical processes responsible for regional phenomena, such as the  $\text{NO}_2$  (Noxon) cliff, at high latitudes in winter.

A second SAGE was launched in 1984. The analyzed observations of trace species and temperature are proving extremely useful as input to global models and for testing certain facets of our theories regarding the coupling between chemistry, dynamics and energetics.

These satellite missions each focus on a specific part of the upper atmospheric system and on measurements of a limited set of species in the different chemical cycles. The proposed Upper Atmosphere Research Satellite (UARS) mission, planned for flight in the late 1980's, will provide the first simultaneous measurements of stratospheric chlorine, nitrogen, hydrogen, and oxygen species, coupled with measurements of dynamical parameters (e.g., winds), and energy inputs and losses. The complement of UARS experiments will be focused on global budgets of gases important in the ozone balance, on the dynamics and energetics of the upper atmosphere, and on couplings among these processes. The extensive UARS data base will allow extensive study of the mechanisms of atmospheric variability and the response of the upper atmosphere to changes in external factors such as solar activity.

#### Future Program

The balanced research program of field measurements, laboratory studies, theoretical studies and data interpretation will continue at the same level in the near future. Specific thrusts will include:

- a. continuation of a balanced program of in-situ and remote measurements of atmospheric constituents from balloon-borne platforms with an increased emphasis on using multi-sensor platforms in order to perform intercomparisons and to obtain comprehensive data sets;
- b. studies of the chemical composition of the lower stratosphere, with increased effort to measure the reservoir species ( $\text{ClONO}_2$ ,  $\text{HO}_2\text{NO}_2$ ,  $\text{N}_2\text{O}_5$ , etc.), which have only recently been detected by the Shuttle-borne ATMOS instrument;
- c. an accurate assessment of the budget and partitioning of the odd hydrogen, nitrogen and chlorine families;
- d. an enhanced ground-based program to measure tropospheric source gas concentrations;
- e. flux studies of source gases from key ecosystems;
- f. application of ground-based lidar and microwave techniques to determine stratospheric structure and composition;
- g. a series of experiments in the tropics and extra-tropics with multiple instrumented aircraft to understand stratospheric/tropospheric exchange mechanisms;
- h. utilization of MST radar systems to study the dynamical processes in the stratosphere;

- i. the high resolution infrared interferometer (ATMOS) will be reflown on the Shuttle in 1986/1987;
- j. passive and active remote sensor development for future satellite (e.g., the proposed Upper Atmosphere Research Satellite program) and Space Shuttle applications;
- k. development of an SBUV instrument for flight on a Space Shuttle to calibrate satellite SBUV instruments;
- l. increased emphasis on analysis and interpretation of existing satellite data sets;
- m. enhanced development of 2-D models for assessment studies;
- n. pursuit of multiple approaches to the interactive 3-dimensional modeling of coupled chemical, radiative, and dynamic processes;
- o. the Solar Backscatter Ultraviolet Ozone Monitor (SBUV) operational program will continue to provide an SBUV instrument for the USA polar orbiter in an afternoon polar orbit. This will provide near-global coverage;
- p. a ground-based program of regular Dobson and automated Umkehr Dobson measurements, together with a three-station balloon-based sampling program, will be operated for a satellite ground truth program.