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Transport and Photochemical Modeling Studies of Atmospheric Species

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Abstract

This report describes a program of research studies related to the photochemistry, radiative transfer, and dynamics of the stratosphere. Investigations were conducted in two broad areas: (1) studies of the stratospheric processes and their response to external perturbations, and (2) analysis of satellite measurements in conjunction with theoretical models. Contemporary one-dimensional photochemical, radiative-convective model has been used to assess the impact of perturbations such as solar flux variability, increases in atmospheric carbon dioxide, chlorofluoromethanes and other greenhouse gases. Data from satellite experiments such as LIMS and SBUV, have been used along with theoretical models to develop a climatology of trace species in the stratosphere. The consistency of contemporary ozone photochemistry has been examined in the light of LIMS data. Research work, described in this report, also includes analysis of stratospheric nitrogen dioxide distributions from different satellite experiments, investigation of the wintertime latitudinal gradients in NO_2, estimation of the stratospheric odd nitrogen level and its variability, and studies related to the changes in ozone in the antarctic, and mid latitude southern hemisphere.

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SECTION 1 - INTRODUCTION

During the past 6 years scientists from ST Systems Corporation (STX), formerly known as Systems and Applied Sciences Corporation and SASC Technologies, have been engaged in performing a variety of theoretical studies related to the atmospheric processes. This work has been performed under a contract from NASA Langley Research Center (Contract No. NAS1-16456). The period of performance of this work is from January 1981 to March 1987. A description of the research activities and the accomplishments is given in this document.

The main contract included various tasks concerned with tropospheric and stratospheric studies. This document describes only the tasks related to the stratospheric studies. The work involved two major efforts, namely the study of the stratospheric processes using one-dimensional photochemical, radiative-convective models, and the analysis of satellite data using theoretical models. Other tasks included the maintenance of the Langley radiative-convective, photochemical model and preliminary work on the development of a two-dimensional model.

The following sections describe the work done under the various tasks and the results that were obtained. A list of publications and presentations is given at the end.

SECTION 2 - MAINTENANCE OF ONE-DIMENSIONAL MODEL

The work described under Task 6 of the contract schedule requires the STX personnel to maintain and update the Langley radiative-convective, photochemical model. The one-dimensional model makes use of a variety of input parameters such as kinetic rate constants, solar flux and eddy diffusion coefficients. As new and more accurate data become available, there is a need to incorporate these data in the model in a timely manner. As is evident, this task has to be performed periodically. Many major changes in the input data occurred during the course of the present contract. A brief description of some of the important modifications is given below.

Kinetic rate constants and absorption cross sections were updated during 1981, 1982, 1983 and 1985. The recommended data were obtained from the publications of the NASA panel for data evaluation. The photochemical model is designed in such a way as to facilitate addition or deletion of reactions and modification of rate constants. The treatment of the Schumann-Runge band system in the calculation of the atmospheric opacity and the photodissociation rate constant of molecular oxygen was modified to include the parameterizations given by Allen and Frederick (1982). The absorption cross sections of molecular oxygen in the Herzberg continuum have been updated to reflect the values given by Herman and Mentall (1982). The effects of self absorption by NO have been included in the calculation of the photodissociation rate constants for NO. The vertical eddy diffusion coefficients have also been updated. Sensitivity studies involving variations in the diffusion coefficients have been performed. Solar uv flux values measured by the SBUV instrument have been incorporated in the model.

SECTION 3 - ONE DIMENSIONAL MODEL STUDIES

Research work described in this section refers to Task 10 of the contract schedule. The main objective is to study the effects on atmospheric thermal structure, chemical composition and dynamical properties of perturbations in the solar uv flux, level of carbon dioxide and other trace species in the atmosphere. The basic tool used in all these assessment studies is the Langley one dimensional photochemical, radiative-convective model. Steady state and time-dependent versions of the model have been used in both thermally coupled and uncoupled modes. Specific problems studied and the results are summarized in the following paragraphs.

3.1 Natural Variability of Upper Stratospheric Ozone

The time-dependent radiative, photochemical model has been used to simulate the variations in the tropical upper stratospheric ozone and temperature over the course of one year. Changes in the solar zenith angle and the earth-sun distance were incorporated as a function of time. Model results have been compared with the ozone data from Nimbus-4 BUV experiment and the temperature data from the SCR experiment. At 2 millibar level and near the equator both the model and data indicate semi-annual cycles in ozone and temperature. The correlation between ozone and temperature is negative near 2 mbar, and positive near 10 mbar. At 15 degrees north latitude the model predicted ozone variation is not in complete agreement with the data. Analysis of the results was presented at the AGU Fall Meeting at San Francisco in 1981.

3.2 Solar UV Flux Perturbations

The effect of long-term (11-year solar cycle) solar uv variability on stratospheric chemical and thermal structure has been studied by various investigators (Callis et al., 1979; Natarajan et al., 1981; Penner and Chang, 1980; Brasseur and Simon 1981). A source of uncertainty in all these theoretical studies is the assumed magnitude of the solar flux variability. Recent studies of the solar flux variation do not indicate much variability for wavelengths around 300 nm. We have repeated the one dimensional model calculations using the recent estimates of solar flux variations. These calculations also made use of the updated cross sections for molecular oxygen in the Herzberg continuum. The lower cross sections make the model middle stratosphere more sensitive to the solar flux variations in the 200 nm region. It is noted that the production of odd oxygen in the stratosphere depends critically on the photolysis of molecular oxygen in the Herzberg continuum. Any variation in the solar flux in this spectral range directly affects the ozone concentration. The ozone perturbation profiles obtained from the above model calculations have been used in an assessment of the temperature and wind changes in the middle stratosphere (Callis et al., 1985b).

3.3 Studies of the Effects of Increases in Atmospheric CO_2, Chlorofluoromethanes, CO and N_2O

The atmosphere is a highly interactive system characterized by strong coupling between chemical, thermal and dynamical processes. Thus a perturbation in any of these processes is bound to have important feedback effects. The climatic effects of increased levels of CO_2 have received worldwide attention. Also the effects of increases in chlorofluoromethanes on the atmospheric ozone and the thermal balance of the earth-atmosphere system have been reported in various publications. As a part of the present contract (Task 10) we have conducted detailed studies of the coupling that exists between the atmospheric photochemistry and the thermal structure. We have examined the feedback effects of increases in CO_2, CFM, CO and N_2O.

A fully coupled one-dimensional, radiative-convective, photo-chemical model has been used for this study. The model has the capability of calculating the changes in the surface and atmospheric temperatures due to both chemical and radiative perturbations. Contemporary atmospheric chemistry involving the O_x, HO_x, NO_x, and Cl_x species is included in the model. The calculations were carried out for two reference atmospheres, one with high and the other with low levels of NO_x in the troposphere. It is found that an unrealistically large tropospheric source of NO_x is required to maintain the high level of NO_x used in one of the reference cases. With the reference atmosphere as the base, the following perturbation studies have been conducted, using the steady state model.

(a) $2.0 * CO_2$

(b) $4.0 * CO_2$

(c) CFM (1975 release rate, fixed)

(d) $2.0 * CO_2 + CFM$

(e) $2.0 * CO_{(flux)}$

(f) $CFM + 2.0 * CO_{(flux)}$

(g) $2.0 * N_2O_{(flux)}$

(h) $2.0 * CO_{(flux)} + 2.0 * N_2O_{(flux)}$

The time-dependent version of the model has been used to simulate the response of the atmosphere to a given CO_2 and CFM injection scenario. Results from these studies suggest that: (1) infrared opacity changes due to CO_2 and CFM increases can cause significant changes in tropospheric O_3, OH, CH_4, CO and other trace species through changes in the tropospheric H_2O; (2) perturbation or sensitivity studies should consider both the stratosphere and troposphere because of the modulating effects of one over the other; (3) model response to perturbation depends upon the level and distribution of tropospheric NO_x; and (4) the thermal or chemical perturbations introduce feedback effects of importance. A full description of this study is presented in Callis, Natarajan and Boughner, 1983a.

SECTION 4 - THEORETICAL STUDIES IN CONJUNCTION WITH SATELLITE DATA

This section describes the various studies involving the use of satellite measurements (Tasks 7, 8, and 9). These measurements were provided by the LIMS, SBUV and SAMS experiments aboard the Nimbus-7 satellite. The major emphasis in these studies was the use of satellite data to check in a general way the consistency of the currently accepted stratospheric photochemistry. This necessitated the development of a climatology of the concentrations of trace species which were not measured by the spacecraft. In addition we have also examined certain features in the stratospheric structure such as the wintertime latitudinal gradient in stratospheric NO_2, and the diurnal variation in stratospheric ozone.

4.1 Stratospheric Photochemical Studies: Development of Inferred Trace Species Distribution

In this study the LIMS, SAMS and SBUV measurements of stratospheric temperature, O_3, H_2O, NO_2, HNO_3, CH_4 and solar uv flux are used in a zero-dimensional photochemical model to infer the distributions of the other trace species. Latitude-altitude distributions of OH, HO_2, H_2O_2, NO, NO_3, N_2O_5, O(1D), O(3P), HCl, Cl, ClO, $ClNO_3$, HOCl and HNO_4 have been inferred for periods in October, December, March and May. The calculations have been done in 10 degree latitude increments from 60^oS to 80^oN, and for an altitude range extending from 20.5 km to 53.5 km, with a vertical grid of 1.5 km. The input data included zonal averages of temperature, O_3, H_2O, HNO_3 and NO_2, developed from the radiance averaged profiles of LIMS data. Nimbus 7 satellite did not include any instrument for measuring chlorine species, and hence it was necessary to assume the level of total odd chlorine based on current model predictions.

The zero-dimensional model makes use of photochemical equilibrium relations to calculate the concentrations of species with short lifetimes, for example, O(1D), O(3P), NO, NO_3, OH, HO_2, Cl, and ClO. Equilibrium relations are also used for species such as $ClNO_3$, H_2O_2 and HNO_4, knowing fully well that certain error is introduced in regions where the lifetimes are of the order of days. Explicit integration of the nighttime NO_x chemistry is performed in order to take into account the production of N_2O_5. This method also yields a correction factor for the inferred $ClNO_3$. The entire procedure is iterated until convergence is reached. The results obtained refer to the trace species concentrations at the time of measurement during the day. A complete description of the model, method of analysis and the results is given in Callis et al. (1986a). A significant finding of this study is the level of maximum odd nitrogen in the stratosphere. This ranges around 24-27 ppbv, much higher than predicted by most models. A paper based on this study was presented at the International Ozone Symposium held in Greece in 1984. In an effort to determine the sensitivity of the results to possible errors in the input data, a Monte Carlo analysis has been performed for 3 altitude levels at 30 degree north latitude for March conditions. The uncertainties in the derived quantities are given in the publication by Callis et al. (1986a). In general there is good agreement between the inferred species distribution and available data. These distributions coupled with the satellite data should prove to be extremely useful in model studies of the stratosphere.

4.2 Studies of the Ozone Photochemistry

The data from Nimbus 7 satellite have been used to check the consistency of the currently accepted stratospheric ozone photochemistry. Model calculations have been performed for 30 degree north latitude, for March conditions and for the altitude range from 31 km to 52 km. Under these conditions stratospheric ozone is expected to be under photochemical control, and thus the consistency of the photochemical reaction scheme and the input parameters could be checked. The procedure adopted for this study consists of two stages. The first stage is the inference of the concentrations of the trace species which have not been measured. This was described in the earlier section. In the second stage a full diurnal calculation is done using the inferred concentrations as initial data. This method removes most of the errors introduced by the assumption of photochemical equilibrium. Ozone is treated as a variable in these calculations, and any major drift from the measured concentration could be attributed to inconsistencies in the photochemical model. It is also possible to fix the ozone concentration at the measured level and compare the photochemical production and loss rates for odd oxygen.

Results from this study were presented at the International Ozone Symposium held in Greece, in 1984. A complete description of the study is also given in Natarajan et al., (1986). A major conclusion from this study is that current photochemical models using recommended reaction rate parameters underestimate the upper stratospheric ozone by 15-30%. In order to study the sensitivity of the model results to uncertainties in the input parameters, a Monte Carlo analysis has been performed using the equilibrium version of the model. It is found that uncertainties in key photochemical rate constants could partly explain the discrepancy between the calculated and measured ozone concentration. Better agreement between the model results and the measurement is realized when selected key rate constants are changed to the recommended limits of uncertainty. It is to be emphasized that this is only a model sensitivity study, and that it does not rule out the possibility of hitherto unidentified processes influencing the stratospheric ozone structure. It is significant that the reaction rate modifications discussed in this study, while reducing the discrepancy in the ozone mixing ratio, also seem to improve the agreement in other photochemical parameters such as daytime levels of NO_2.

We have examined the ozone-temperature relationship in the upper stratosphere using the LIMS data and the model. It is found that the variability in the ozone-temperature sensitivity parameter, obtained from the satellite data, is large, and in many instances, this may be due to the non-equilibrium conditions existing in different parcels of air. Thus it is difficult to check the photochemical consistency using equilibrium values of ozone-temperature sensitivity parameter.

Diurnal variations in ozone provide a means of validating the ozone photochemistry. Our studies indicate only a partial agreement between model results and the LIMS data regarding the day-night difference in ozone. Especially, the current photochemical models do not indicate large day-night differences in ozone near 10 mbar. The LIMS data shows nearly a 10% difference in ozone near 10 mbar, with daytime values being larger than the nighttime values.

In summary, our study describes a method of checking the consistency of the contemporary stratospheric ozone chemistry using satellite data. It indicates a discrepancy in the upper stratospheric ozone between model calculations and the measurements. This is in concurrence with the findings of other model studies. Further research into possible causes of this disagreement is under progress.

4.3 Examination of Wintertime Gradients in Stratospheric NO_2

Ground-based measurements made by Noxon (1975, 1979) have indicated the existence of a sharp decrease in the column abundance of NO_2 in the winter, poleward of $50^{\circ}N$. This feature, popularly known as the 'Noxon cliff', has not been reproduced by most modeling studies. An examination of the northern hemisphere, wintertime LIMS data reveal the presence of a sharp drop in the NO_2 column density. We have identified the location of such sharp gradients in NO_2, and have investigated their formation through the interaction between transport and photochemistry. This study considers parcel trajectories on isobaric surfaces. The period January 7-15, 1979, was selected for our study because during this time the geopotential height field was relatively constant. The geopotential contour on an isobaric surface gives a reasonable representation of the parcel trajectory. Zonal and meridional wind velocities were determined along the trajectory, assuming geostrophic balance. These data sets were used in a time-dependent, zero-dimensional photochemical model to calculate the variations of species mixing ratios along the trajectory. These calculations were repeated for different trajectories and different pressure levels.

Results of the trajectory calculations at 30 mbar for an averaged latitude of 62.5°N show that N_2O_5 represents nearly 50% of the total odd nitrogen, while NO_2 ranges between 5% and 20%. At high latitudes, in wintertime, HNO_3 and N_2O_5 act as important reservoirs for odd nitrogen. The lifetime of N_2O_5 is not very long, and its photodissociation plays an important role in the formation of latitudinal gradients in NO_2. The steepness of the gradient is affected profoundly by the shape of the geopotential contours. A Noxon cliff type feature is seen, whenever the air parcel south of the cliff originates from lower latitudes and the air parcel north of the cliff is predominantly of polar origin. The conversion of NO_2 to N_2O_5 occurs in the high latitude polar night region. A strong interaction between the dynamics and photochemistry is, therefore, believed to be the mechanism responsible for the sharp latitudinal gradients in the stratospheric NO_2 during the northern hemisphere winter. A paper describing this study has been published in the Geophysical Research Letters (Callis et al., 1983b).

4.4 Studies on Stratospheric Odd Nitrogen Levels

LIMS experiment aboard the Nimbus 7 satellite provides the first global scale measurements of HNO_3 and NO_2 mixing ratios. Both daytime and nighttime measurements are available for about 7 months. We have collaborated with NASA scientists in estimating the latitude-altitude distribution of the stratospheric odd nitrogen using the LIMS data. It is to be noted that the sum of nighttime measurements of HNO_3 and NO_2 represents a reasonable lower limit estimate of the stratospheric odd nitrogen. This is because, at sunset NO is rapidly converted to NO_2, and then the conversion of NO_2 to NO_3 and subsequently to N_2O_5 takes place. Over most of the globe, the nighttime levels of NO, NO_3, N_2O_5, HNO_4 and $ClNO_3$ are relatively small. Latitudinally averaged LIMS nighttime HNO_3 + NO_2 for March shows a maximum value of about 19.6 ppbv. Based on model calculations, this represents a maximum odd nitrogen of about 23.5 ppbv. This is significantly larger than the odd nitrogen levels predicted by one-dimensional models. The distribution of NO_x in the stratosphere is important to the balance between the odd oxygen production and destruction. The larger odd nitrogen levels estimated in the present work will have ramifications in studies of the ozone depletion by odd chlorine. A paper describing the odd nitrogen study has been published in the Geophysical Research Letters (Callis et al., 1985a).

SECTION 5 - PHOTOCHEMICAL STUDIES FOR HIGH LATITUDE SOUTHERN HEMISPHERE

Research activities described in this section are related to the work under Tasks 19 and 20(G). The main objectives are: (1) use satellite data and radiation models to develop diabatic circulation fields, (2) construct diabatic trajectories based on the circulation fields, and (3) perform photochemical calculations along the diabatic trajectories and compare the changes in chemical composition with available data. The focus on southern hemisphere high latitude was mainly a result of the recent reports on the springtime ozone depletion in the antarctic. In addition to changes in ozone, a comparison of the satellite data reveal an increase in the stratospheric NO_2 from 1979 to 1984. The following paragraphs describe briefly the research work done under this contract on changes in stratospheric ozone and NO_2.

5.1 NO_2 Changes in the Stratosphere During 1979-84

The availability of satellite measurements in recent years has greatly facilitated the analysis of the variability in stratospheric composition. During 1979-84 measurements of stratospheric NO_2 have been made by four different satellite experiments, namely LIMS, SAGE, SME, and SAGE II. Direct comparison of the different data sets is often hindered by the differences in the local time of measurement. This is a critical factor since NO_2 has a profound diurnal variation. It is however possible to compare the LIMS daytime data for high southern latitudes with the SME data for the same region, the measurement times being nearly the same. This problem doesn't arise when one compares the SAGE and SAGE II data, since both are solar occultation measurements. However, the orbital characteristics of the two satellites impose certain restrictions in the use of SAGE and SAGE II data.

The available data in different periods have been analyzed and the results indicate definite increases in the stratospheric NO_2 levels from 1979 to 1984. For example, near 60^oS, the SME NO_2 peak values for March 1982 are about 60% larger than the LIMS NO_2 data for March 1979. The SAGE II NO_2 profile for high southern latitude in early December, 1984 shows nearly a 60% increase compared to SAGE data for the same latitude range in November, 1979. The identification of the source of this increase in NO_2 is clearly an important issue. Callis and Natarajan (1986a) suggest the possibility of enhanced upper atmospheric formation of odd nitrogen during solar cycle 21. High levels of odd nitrogen could be transported downward into the stratosphere during polar night at high latitudes. The path and timing of their advective descent have been confirmed with diabatically diagnosed transport and trajectory calculations. A full description of this mechanism, and of the stratospheric effects of the odd nitrogen increases is given in Callis and Natarajan (1986a).

5.2 Springtime Minimum in the Antarctic Ozone

Recent reports have shown a large decrease in total ozone during the months of September and October in the antarctic region. Satellite and ground-based measurements have confirmed the deepening of this ozone hole from 1979 to 1985. Several theories based on photochemistry and dynamics have been put forward to explain this phenomenon. At present the cause of this ozone depletion remains an unresolved issue. We have collaborated with NASA scientists to study the possibility that the antarctic ozone minimum is related to the increases in the stratospheric odd nitrogen, discussed in the previous section.

The proposed mechanism suggests that during solar maximum high levels of odd nitrogen are transported downward to 25-35 km region within the polar vortex in the southern hemisphere. This odd nitrogen gets mixed with the stratospheric odd nitrogen, and within the vortex, during polar night, the odd nitrogen is mainly in the form of N_2O_5. When the sun comes up in late August, the photolysis of N_2O_5 begins and the resulting high levels of NO and NO_2 lead to the catalytic destruction of ozone. In order to support this hypothesis, we have performed photochemical calculations along diabatic trajectories for appropriate conditions. Since under the very cold conditions the vertical descent of the parcels is almost absent, we have also done calculations for fixed parcels. The sensitivity of the change in ozone to different levels of odd nitrogen in the altitude range 16-31 km has been studied. The results from these studies and discussions of various related questions have been published.

(Callis and Natarajan 1986b).

5.3 Diurnal Model Studies for Southern Hemisphere, High Latitude Conditions

The objective of this study is to derive information on the levels of reactive nitrogen species within the polar vortex during springtime in the southern hemisphere. Available data on NO_2 and ozone from SAGE II experiment are used in this study. It is to be noted that the level of reactive nitrogen within the polar vortex is a critical factor in different photochemical theories on the antarctic ozone minimum. SAGE II experiment made measurements of ozone and NO_2 within the polar vortex near $73^{\circ}S$ latitude during early October, 1985. Averages of about 5 scans from the data for October 5, 1985, covering the longitudinal extent of the vortex have been used an input data in the photochemical calculations. This study considers the altitude range 22-34 km. The results indicate that the levels of reactive nitrogen ($NO + NO_2$) within the vortex, based on the SAGE II data, are not very high. We have also done diurnal model calculations for the same altitude range and for the 15th of September. The model results predict a difference between the sunrise and sunset NO_2 vertical column. It has been reported that ground-based NO_2 column measurements within the vortex show very little difference between the sunrise and sunset values. Photochemical data appropriate for the very cold conditions within the vortex are required in order to make valid comparisons between model studies and the data. A paper based on this study was presented at the

AMS conference on the Dynamics and Chemistry of the Middle Atmosphere, Baltimore, 1987.

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APPENDICES

REPRINTS OF ARTICLES

SECTION 7 - PRESENTATIONS

1. Natarajan, M., and L. B. Callis, Stratospheric NO_2 in the high latitude southern hemisphere: A model sensitivity study, Sixth Conference on the Dynamics and Chemistry of the Middle Atmosphere, AMS, Baltimore, MD, 1987.

2. Callis, L. B., and M. Natarajan, Explanation of the spring minimum in antarctic ozone, AGU Spring Meeting, Baltimore, MD, 1986.

3. Natarajan, M., L. B. Callis, J. M. Russell III, and R. E. Boughner, A study of the ozone photochemistry in the upper stratosphere using LIMS data, Quadrennial Ozone Symposium, Halkidiki, Greece, 1984.

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