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Transport Processes and Trace Constituents
in the Stratosphere

Final Report

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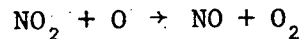
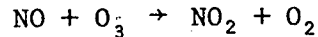
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Abstract

A three-dimensional dynamical-chemical model of the stratosphere has been developed. The model includes predictions not only of the dynamical structure of the stratosphere but also of atmospheric ozone. A three year integration of this model has produced seasonal and latitudinal variations of the zonal wind and of columnar ozone which are similar to those observed. Additional model calculations have included the steady state nitrogen dioxide distributions resulting from a source of 1.8×10^6 tons/year inserted at various locations in the lower stratosphere. It is thus concluded that the most significant effect of a large fleet of SST's producing such an injection of NO_2 is to deplete total atmospheric ozone by of order 10%. The latitudinal structure of this ozone depletion for various source locations is compared.

Background

One of the most environmentally serious objections raised against supersonic transport (SST's) in the early seventies was that of Johnston (1971) and Crutzen (1971) that nitrogen oxides would be introduced into the stratosphere and would destroy ozone by the catalytic reactions



Initial estimates of the resulting ozone destruction were based upon one dimensional (ie. globally-averaged) models of the atmosphere in which questionable approximations included the use of typical photodissociation rates at a particular latitude to represent globally averaged values and the use of a height-dependent vertical diffusion coefficient to represent vertical transport on a global scale by all scales of motion. Moreover flight patterns of SST's were projected to be concentrated at mid-latitudes of the Northern Hemisphere which might result in substantial latitudinal variation in the resulting ozone depletion; this effect of course could not be resolved with a 1-dimensional model.

In principle some of these questions could be resolved with the use of 2-dimensional models of the stratosphere. Such models had previously been used to study the (unperturbed) ozone distribution in the atmosphere (eg. Prabhakara, 1963). However such a model depends upon a diffusion-like parameterisation to represent both vertical and horizontal transports. This is an ad-hoc non-physical assumption and recent studies with a general circulation model (Mahlman, 1975) have tended to confirm the suspicion that the transport of a species is dependent on the chemical nature of the species.

Moreover the studies with a general circulation model by Hunt and Manabe (1968) had indicated the complex nature of the transport of an ozone-like tracer in the lower stratosphere with the net transport being the result of the small difference between the transports by the mean circulation and by the eddy motions. We therefore decided to develop a three dimensional model of the stratosphere in which the motions would be predicted and which would include a limited number of chemical reactions believed to be important in controlling the atmospheric ozone distribution.

Several three dimensional models had previously been developed and applied to studies of stratospheric dynamics and of the latitudinal variations of ozone. For example Manabe and Hunt (1968) performed a simulation for an annual mean insolation with an 18 level hemispheric general circulation model extending from the ground to 37.5 km with a vertical resolution of approximately 3 km and a horizontal resolution of approximately 500 km. This simulation produced many of the observed gross features of the dynamics of the lower stratosphere although it was in better agreement with observed January rather than equinoctial conditions. Two other models with which we were particularly impressed were constructed by Clark (1970) and Trenberth (1972, 1973) both of whom expressed the dependent variables in terms of a limited number of spherical harmonics and invoked the quasi-geostrophic approximation. Trenberth used a 9 level model extending from the ground to 70 km and retained only planetary wave numbers 0,2,4, and 6. He was able to reproduce realistic stratospheric warmings in late winter. While Trenberth's model included no chemistry, Clark included the Chapman scheme of chemical reactions and was able to reproduce numerically in a qualitative sense the observed tendency

for the large scale motion field to shift the maximum of vertically integrated ozone poleward from the low latitude source region. Thus the time seemed propitious for the development of a similar type of model with increased horizontal resolution which would include the recently suggested destruction of ozone by nitrogen oxides (Crutzen, 1971) and which would permit several years integration with varying solar position. With this model it was hoped to be able to adequately simulate the observed seasonal and latitudinal variations of ozone so that ozone destruction produced by an additional source of NO_x (from SST's) could be estimated.

Current status of modeling work

During the first two years of this three year project a numerical model of the global stratosphere was coded. The model contains 26 levels equally spaced in log(pressure) between the ground and approximately 70 km. In the horizontal domain the model extends from pole to pole and spherical harmonics are used (up to and including planetary wave number 6) to represent variations with latitude and longitude. The basic dynamical set of equations used is one of the simpler forms of the "balance" equations (Lorenz, 1960) and incorporates the quasi-geostrophic approximation. This permits time steps of 1 hour to be used during the model integration so that the computation time on the 360/95 at Goddard Institute for Space Studies is approximately 4 hours/model year or approximately 12 hours for the 3 year integration required to reach a state of quasi-equilibrium. The dependent variables in this integration are the stream function, vertical velocity, temperature and ozone mixing ratio.

The first successful 3 year integration of this model was made early in 1974 and this was followed several months later by a simulation of the effect of SST's on the atmospheric ozone distribution. Both of these calculations have since been reported in the open literature and are the most significant results of this contract; copies of these papers are included in the appendix to this report. During the third year of this contract additional calculations of the effect of SST's on ozone have been made and investigations have begun into techniques of eliminating certain undesirable features of the model.

The previously reported model results produced a successful simulation of the ozone distribution and of the large-scale motions of the stratosphere. In particular it was the first model to have simulated the seasonal variations of ozone without any ad-hoc parameterisation of the horizontal motion field. Moreover the variability of ozone was approximately similar to that observed. It appeared that the model was primarily governed by the same physical laws as is the atmosphere. The model produced a westerly wind maximum of the observed amplitude and location in the winter stratosphere which changed over to easterlies in summer. Moreover the three cell structure of the troposphere diminished to a two cell circulation in the stratosphere with substantial cross-equatorial transport from the summer to the winter hemisphere. Ozone was being transported polewards and downwards from the equatorial sources region thus producing the ozone peak at high latitudes. The standing wave patterns of columnar ozone, while showing some agreement with observation, were not particularly satisfactory. To correct this deficiency we are considering other sources of tropospheric heating information. We believe that this type of model improvement is also needed in order to produce more realistic stratospheric warmings.

In order to perform our calculations within the time limit prescribed and to reduce computation time, certain approximations have been made. Detailed testing of the adequacy of these approximations has already begun and the continuing sponsorship of this modeling work by the National Aeronautics and Space Administration will permit this work to continue and will allow a more careful study of the physical and dynamical behavior of this model.

One particular area of investigation related to the fact that although no "sub-grid scale" diffusion has been used in the horizontal directions - that is to say all horizontal motions have been explicitly predicted - vertical motions have included a diffusion-like transport process. This diffusion term was used to represent vertical transport by those scales of motion which were beyond the truncation limits of the model.* In the first reported run of the model rather large values of k_z were used with a minimum value of $4 \times 10^3 \text{ cm}^2/\text{sec}$ at the tropopause. It was found that the vertical transport of ozone between 20 and 30 km was primarily the result of diffusion as opposed to transport by the large-scale motions. This was clearly an undesirable feature of the model since we had hoped that most of the transport would be the result of the explicitly predicted motions and furthermore small scale k 's in this region had been measured to be only $10^2 \text{ cm}^2/\text{sec}$. An attempt was made to eliminate k_z entirely above 10 km but we found that very large oscillations of ozone occurred as a result of the vertical finite differencing and which resulted in negative ozone concentrations. Consequently, in subsequent model runs we have used an intermediate k_z profile which assumes a value of $10^2 \text{ cm}^2/\text{sec}$ at the tropopause. Such a value of k_z still allows undesirably large amplitude oscillations in ozone at the tropopause level and we have thus spent considerable time recently on developing alternate representations for the smaller scale oscillations associated with our vertical finite differencing scheme. Assuming that we are successful in

* It should be noted that the vertical diffusion profile assumed could be quite different from that used by one-dimensional modelers who seek to parameterise vertical transport by all scales of motion.

this regard it seems possible that we may still find that 6 planetary waves are not alone capable of providing sufficient downward transport through the tropopause to yield observed tropospheric ozone concentrations. Furthermore certain dynamical deficiencies are also evident in our model in the neighborhood of the tropopause as for example in the unrealistically high temperatures at high latitudes in this region. The jet stream also is located too close to the equator. We are therefore including additional diffusion in the tropics to account for cumulus convection there and which according to the G.I.S.S. model contributes to a more realistic zonal wind system (Stone et al, 1974). We are also constructing a triangular truncation as opposed to the rhomboidal truncation currently in use, and over the next year or two we are hoping to be able to increase the resolution of the model by including more planetary waves.

The orography included in the model is that of the Northern Hemisphere. This orography has been reflected into the Southern Hemisphere. This procedure does have some statistical advantages. However it will be interesting to study the differences between the two hemispheres. This means not only including a more realistic Southern Hemisphere orography (a trivial task) but also a Southern Hemisphere tropospheric heating distribution. We are planning to run the model using a predicted global heating distribution from the general circulation model of the Geophysical Fluid Dynamics Laboratory. It is thus hoped to simulate the important differences between the ozone distributions of the two hemispheres and in particular the observation that the Southern Hemisphere columnar ozone peak is found at approximately 50° latitude instead of at the pole as in the Northern Hemisphere. This calculation will be performed within the next 6 months.

The chemistry of our three-dimensional model as originally proposed and as incorporated in the reported runs of the model consists of just seven reactions. Thus distributions of NO_2 , OH , and HO_2 are required as input data to the model; the distributions of O and NO are then solved for diagnostically at each time step while the distribution of O_3 is predicted. In the first reported run of the model NO_2 , OH , and HO_2 were assumed to be dependent only on altitude. In more recent model runs however a latitudinal and simple seasonal variation of NO_2 has been included. This necessitated a two dimensional calculation of NO_2 and since a satisfactory distribution of NO_2 was not available at the time we wished to run the three-dimensional model, we assembled a two-dimensional model. The two-dimensional model contains a prediction of total odd nitrogen, HNO_3 , and N_2O , while OH , HO_2 , H , H_2O_2 , O , and $\text{O}(\text{^1D})$ are solved for diagnostically. Since the latitudinal distribution of ozone is fairly well defined we chose to input values of ozone based on observation. Horizontal and vertical transports were parameterised and based upon the experience of other scientists it seemed unlikely that an adequate ozone distribution could be predicted without "fitting" of the transport parameters. Thus using an observed ozone distribution removed one uncertainty from the prediction of nitrogen species. The derived distributions of nitrogen species are in good agreement with the limited observations available. In particular without prescribing nitric acid at the lower boundary, latitudinal gradients of nitric acid of similar magnitude to those observed were obtained. Moreover Evans (private communication, 1975) indicates that profiles of NO , NO_2 , and HNO_3 at 60°N agree well with the recent simultaneous measurements from a balloon. Additional details of the two-dimensional

time-dependent model are given in the appendix which includes a copy of a paper by Prinn, Alyea, and Cunnold, accepted for publication in the Journal of Geophysical Research. Because of the current interest in chlorine chemistry additional reactions involving chlorine have recently been added to this model.

Because of the questions that always arise about describing the transport of several chemically different species by a single set of diffusion coefficients, and for physical consistency, it is desirable that the two-dimensional model be eliminated and that additional chemical species be included in the three-dimensional model. A start on this task has been made with the three-dimensional model now being capable of making a prediction of N_2O . We anticipate that the addition of the chemistry currently used in our two-dimensional model will approximately double the computation time for a three year run of the three-dimensional model. While using the two-dimensional model it was found that approximately 25 years of integration were required to arrive at a steady state odd nitrogen distribution. We cannot of course run our three-dimensional model that long. We are currently investigating two approaches to shortening the computation time. One approach involves making an updated estimate of the equilibrium NO_y value by calculating the flux loss out of the model and the total source rate and then applying a linear correction to update the estimate. The other approach would store 1 year of dynamical variables of a tape and then perform a separate calculation of NO_y over many years using this set of dynamical variables repeated each year. This approach depends upon the fact that NO_y is essentially a passive tracer. Thus although the inclusion of a more complete chemistry in the three-dimensional model has not yet been accomplished, we expect this work to be completed shortly.

The dynamics of the three dimensional model although apparently adequate to simulate many of the gross features of the stratosphere are as already stated unsatisfactory in certain respects, particularly in the neighborhood of the tropopause. In order to provide a basis against which to assess the dynamical behavior of our model we undertook an analysis of geopotential height and temperature at 100, 50, 30, and 10 mb. and between 20° and 90° N. We first derived horizontal winds using the geostrophic approximation. The monthly average mean zonal winds were presented for each month of the 5 year period 1963-1967; the five year average values are contained in a report which was partially funded under this contract and which was presented at the IAMP Conference in Melbourne, Australia. The report is entitled "Diagnostic studies of the general circulation of the stratosphere" and is authored by Newell et al. Statistical correlations between meridional velocity and temperature as well as individual standard deviations have also been calculated. These have been broken down into contributions from planetary waves 0 through 12 at each latitude and mid-latitude averages have been obtained for comparison with the model results. Although the model results exhibit the same gross features at the observations with the higher planetary waves becoming of lesser importance with increasing height with considerably less activity in Summer than in Winter, a detailed comparison between model results and the dynamically related observations have not yet been performed. Part of the reason for this is that it is not clear to what extent the smoothing of the individual station data has limited our ability to resolve the higher order planetary waves in the observational data. Other dynamical investigations supported by this contract include a study of the efficiency of ozone transport in the lower stratosphere by waves of various length scales by Klein (1974).

In our three-dimensional model absorption of ultraviolet and visible radiation by ozone is treated explicitly. However radiative cooling in the 9.6μ and 15μ bands are approximated by Newtonian cooling. For this reason we originally proposed that Dopplick's (1970) computation of infrared cooling be extended to higher altitudes to provide a basis against which to test the adequacy of the Newtonian cooling approximation. However, the thorough calculations by Dickinson (1973) and the apparent success of the model in simulating the temperature structure of the atmosphere have made this task postponable.

SST results

The simulation of the effect of SST's on ozone was accomplished by comparing two three year integrations of the three-dimensional model. The only difference between the two runs was that different distributions of NO₂ were used. For the unperturbed atmosphere we used a two-dimensional distribution derived by Hesstvedt (1974). It was then discovered that a distribution of NO₂ in the presence of a fleet of SST's was unobtainable. We therefore assembled a two-dimensional model and obtained distributions of NO₂ both in the unperturbed atmosphere and in the presence of an additional source of NO₂ equal to 1.8×10^6 tons/year and centered at 45°N and 20 km. This source was intended to represent the emission of 500 of the now defunct and no longer planned Boeing (2707) SST's flying 8 hours per day. Unfortunately time did not permit us to make another 12 hour computation with the unperturbed NO₂ distribution derived from our two-dimensional model. Fortunately it was noted that although our unperturbed NO₂ distribution possessed stronger horizontal gradients than did Hesstvedt's, the two distributions were fairly similar in vertical structure. We therefore chose to take the difference between our two NO₂ distributions and to thus derive the NO₂ perturbation resulting from SST's. This perturbation was then added to Hesstvedt's distribution and the resulting distribution was used as input to the three-dimensional model in order to calculate the ozone distribution in the perturbed atmosphere. We feel this procedure to be justified because it is expected that the ozone perturbation is sensitive to the perturbation in NO₂ than to the absolute level of NO₂ and ozone concentrations.

With this calculation procedure certain feedbacks have been neglected. In particular NO₂ distributions were not recalculated to reflect the predicted ozone depletion. However the ozone depletion was sufficiently small (12% globally) that we noted no significant changes in stratospheric

dynamics and we believe that the chemical feedbacks neglected have no substantial effects on the total ozone depletion.

Two three year integrations have been reported in the literature and we include a copy of the Science article as an appendix to this report. More recently we have completed two similar simulations in which the location of the source of NO_2 was varied. Thus the calculations were

- run 17: unperturbed stratosphere
- run 18: NO_2 source located at 20 km and 45°N
- run 19: NO_2 source located at 17 km and 45°N
- run 20: NO_2 source located at 20 km and 10°N

In all cases the source magnitude was 1.8×10^6 tons/year. The latitudinal variation of ozone depletion for each of four seasons is compared in Fig 1. The total global ozone depletion was found to be approximately 12% in run 18, 6% in run 19, and 13% in run 20. A more detailed analysis of these results is in progress.

Perturbations in the atmospheric heating rate due to SST associated aerosols have also been considered. Malchow (1974) found that the absorption of direct, visible radiation was the dominant term in the net aerosol heating. For SST soot particles maximum heating rates of approximately 10^{-3} $^\circ\text{K}/\text{day}$ were calculated for the North Atlantic region. For sulfate aerosols there was some uncertainty about the net heating rate because of the large uncertainty in the absorptive index of refraction. Assuming a representative value of .01 for this index, the SST sulfate aerosol heating at 15 km was calculated to be approximately .05 $^\circ\text{K}/\text{day}$. Thus it seems clear that the perturbations in atmospheric heating due to SST aerosols are much smaller than the gaseous heating and can be neglected.

After the above aerosol results were obtained it was decided that emphasis in this area be shifted to defining the aerosol distribution and

physical properties in the unperturbed atmosphere. Horizon data from Skylab, EPN-587, was to have been inverted to yield aerosol number density, size distribution, and real and complex index of refraction.

However, unforeseen errors in the processed NASA Skylab data required diverting the level of effort in order to be able to begin to process these data correctly. In particular, there were three areas where significant errors were determined to exist in the S-191 instrument that were not known to NASA to have existed during the design, construction, prelaunch testing and calibration, data acquisition and post flight processing. These three areas are as follow.

1. Short Wavelength intensity correction (wavelength dependent)
2. Straylight (Skylab local environment which is wavelength dependent)
3. Line-of-Sight offsets for variable mirror angles

In each of these areas the determination that the processed Skylab data was incorrect was based upon prior knowledge of the theoretical horizon characteristics and experimental solar and horizon data. However, once there was an acknowledgement of questionable data, procedures had to be defined and verified that would determine these corrections and establish the magnitude of the residual uncertainties. Since each correction was initially unknown to NASA there was a period of time where there was extensive communication to obtain NASA's own acceptance and verification of these errors which led to correction tables and reproducing of these data. The present status of the skylab EPN-587 data is that to the best of our knowledge all intensity and altitude corrections are known. The flight data has been processed to the point where the data can now be inverted and analysed.

Summary

A stratospheric circulation model has been developed which is remarkably successful in simulating the gross features of the atmospheric ozone distribution. The model has been used to estimate ozone depletions resulting from an intensive level of operation of SST's. A digestion period is now required which will allow us to study in more detail the extent to which the model simulates the real atmosphere. This will require additional comparisons with ozone and dynamical data as well as modifications of the model to permit more extensive chemistry and increased resolution. Fortunately these studies are expected to continue under the sponsorship of the National Aeronautics and Space Administration.

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Figure Caption

Figure 1, (a), (b), (c), and (d). A comparison of the seasonally-averaged zonal-mean total ozone distributions for the unperturbed stratosphere and for sources of 1.8×10^6 tons/year of NO_2 at three different locations.

Run 17: the unperturbed stratosphere.

Run 18: source at 45°N and 20 km altitude.

Run 19: source at 45°N and 17 km altitude.

Run 20: source at 10°N and 20 km altitude.

Results are shown for four seasons.

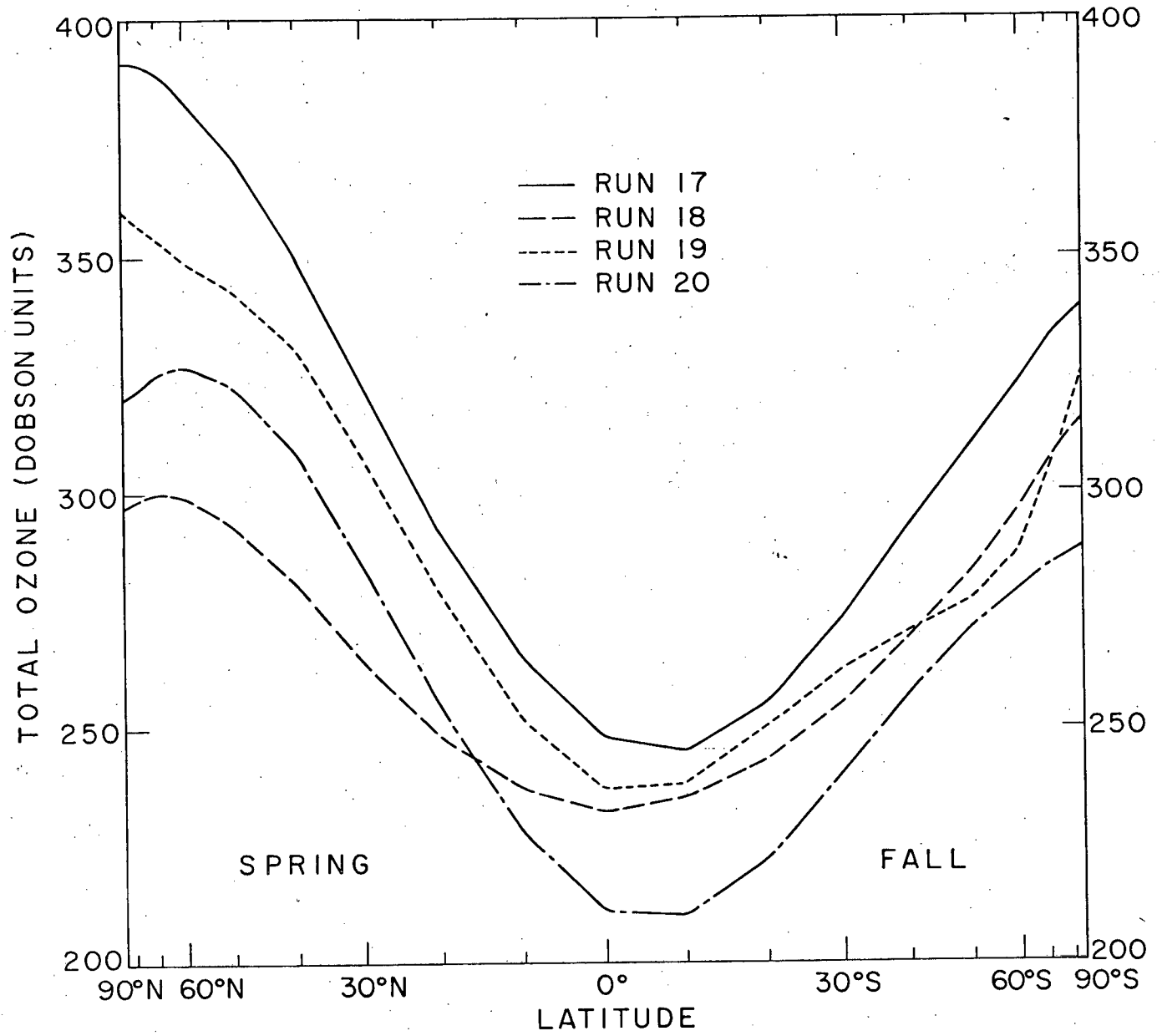


Figure 1, (a): Spring - Fall

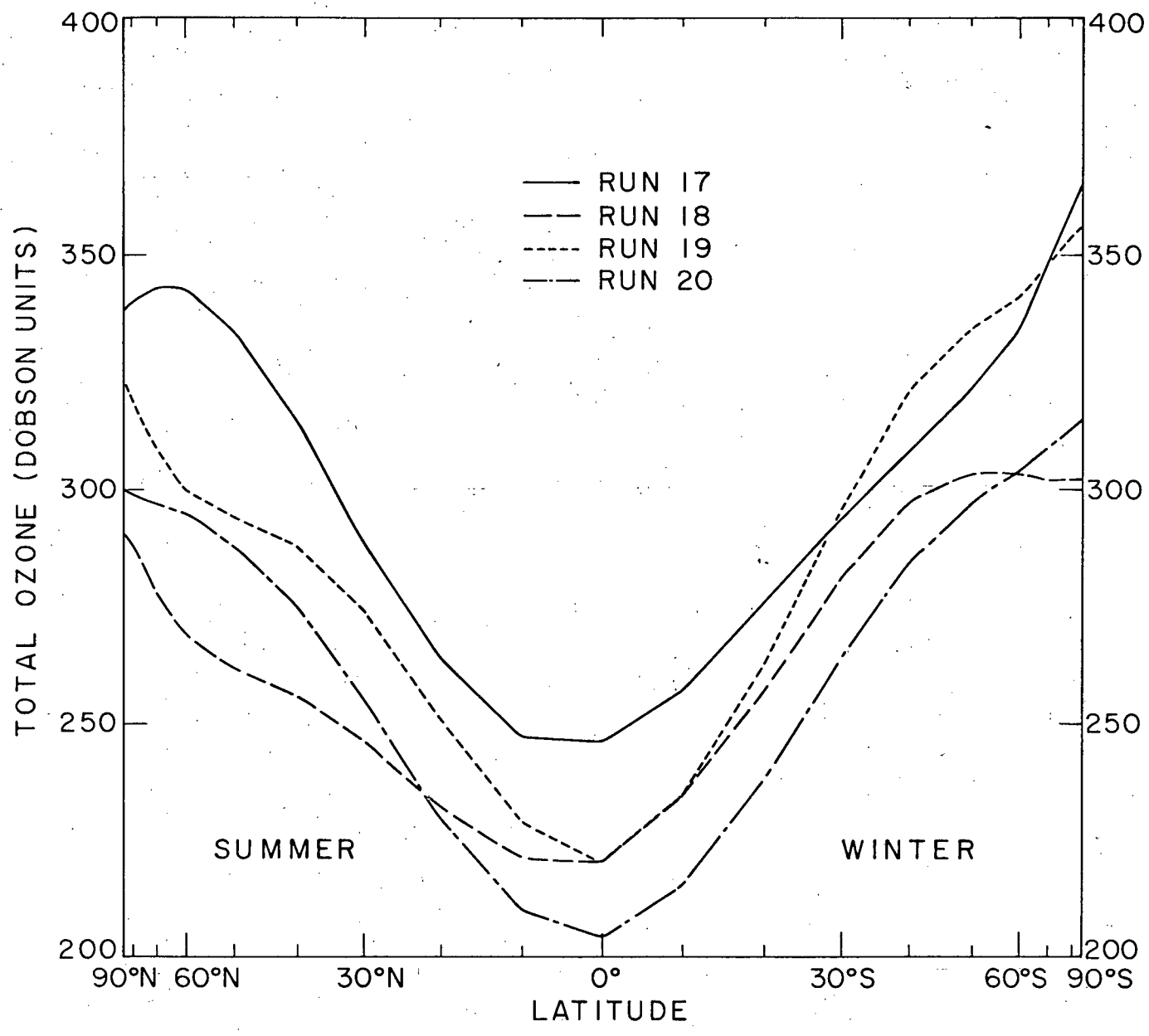


Figure 1, (b): Summer - Winter

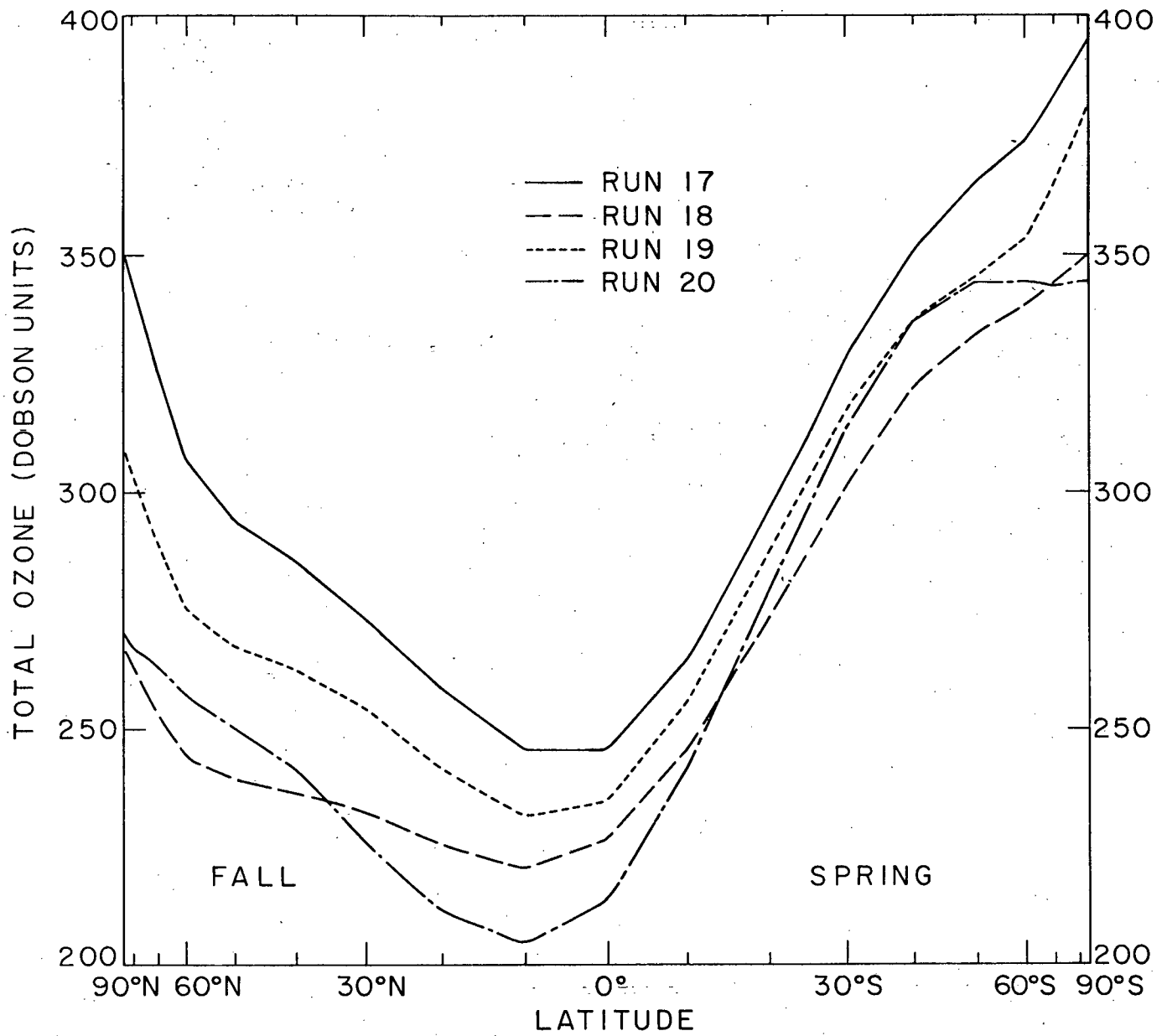


Figure 1, (c): Fall - Spring

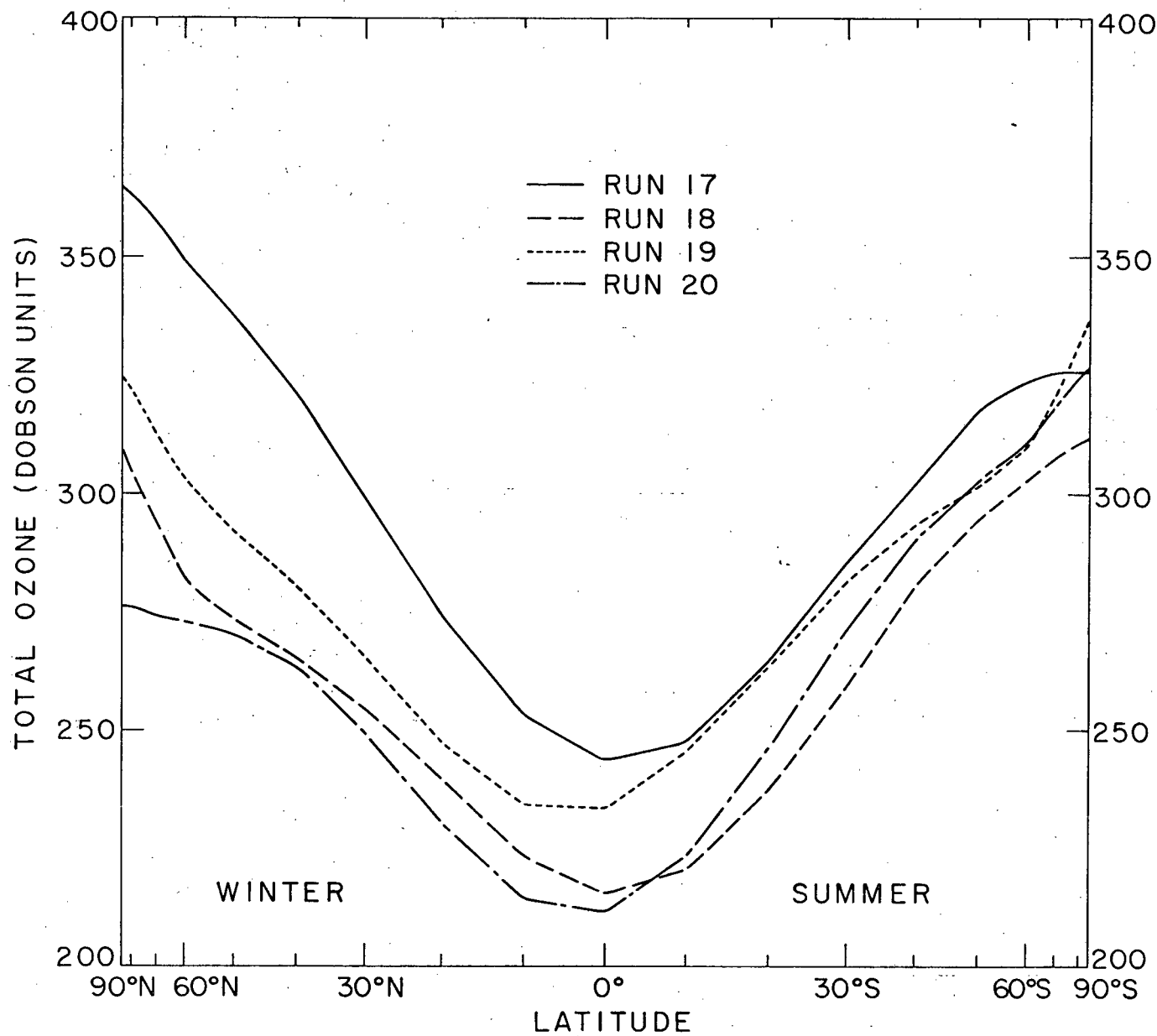


Figure 1, (d): Winter - Summer

Reports prepared under complete or partial support from contract AT(11-1)-2249

1. "A general circulation model of stratospheric ozone," by D. Cunnold, F. Alyea, N. Phillips, and R. Prinn, AIAA/AMS International Conference on the Environmental Impact of Aerospace Operations in the High Atmosphere, June 11-13, 1973, Denver, AIAA paper no. 73-529.
2. "Heating of the stratosphere by SST aerosols," by H. Malchow, C.S. Draper Laboratory, 75 Cambridge Parkway, Cambridge, Massachusetts, 1974.
3. "Diagnostic studies of the general circulation of the stratosphere," by R. Newell, G. Herman, J. Fullmer, W. Tahnk, and M. Tanaka, Report no. COO-2195-11, Department of Meteorology, M.I.T., Cambridge, Massachusetts 02139. Presented at IAMAP Conference, Melbourne, Australia, January, 1974.
4. "A general circulation model of stratospheric ozone," by D. Cunnold, F. Alyea, N. Phillips, and R. Prinn, Proceedings of the International Conference on structure, composition and general circulation of the upper and lower atmospheres and possible anthropogenic perturbation, Jan. 14-25, 1974, Melbourne, Australia, pp 932-970.
5. "Ozone kinematics and transports in unstable waves," by W. Klein, PhD thesis, Department of Meteorology, M.I.T., Cambridge, Massachusetts 02139, January 1974.
6. "Preliminary results of the M.I.T. photochemical-dynamical ozone model," by D. Cunnold, F. Alyea, N. Phillips, and R. Prinn, Proceedings of the Third conference on the Climatic Impact Assessment Program, Feb 26 - Mar 1, 1974, Cambridge, Massachusetts, pp 403-421.

7. "First results of a general circulation model applied to the SST-NO_x problem," by D. Cunnold, F. Alyea, N. Phillips and R. Prinn, Proceedings of the 2nd International Conference on the Environmental Impact of Aerospace Operations in the High Atmosphere, July 8-10, 1974, San Diego, published by American Meteorological Society, pp 187-193.
8. "The distributions of odd nitrogen and odd hydrogen in the natural and perturbed stratosphere," by R. Prinn, F. Alyea, D. Cunnold, and A. Katz, Proceedings of the 2nd International Conference on the Environmental Impact of Aerospace Operations in the High Atmosphere, July 8-10, 1974, San Diego, published by American Meteorological Society, pp 180-185.
9. "A three-dimensional dynamical-chemical model of atmospheric ozone," by D. Cunnold, F. Alyea, N. Phillips, and R. Prinn, J. Atmos. Sci., 32, 170-194, 1975.
10. "Stratospheric ozone destruction by aircraft-induced nitrogen oxides," by F. Alyea, D. Cunnold, and R. Prinn, Science, 188, 117-121, 1975.
11. "Stratospheric distributions of odd nitrogen and odd hydrogen in a two dimensional model," by R. Prinn, F. Alyea, and D. Cunnold, J. Geophys. Res., In Press, 1975.

Appendix

Papers which have appeared in the reviewed literature:

1. "A three-dimensional dynamical-chemical model of atmospheric ozone,"
by D. Cunnold, F. Alyea, N. Phillips, and R. Prinn, J. Atmos. Sci.,
32, 170-194, 1975. *Removed*
2. "Stratospheric ozone destruction by aircraft-induced nitrogen oxides,"
by F. Alyea, D. Cunnold, and R. Prinn, Science, 188, 117-121, 1975. *Removed*
3. "Stratospheric distribution of odd nitrogen and odd hydrogen in a
two dimensional model," by R. Prinn, F. Alyea, and D. Cunnold, J. Geophys.
Res., In Press, 1975. *Removed*