

CONF-890692-4-Rev.1

UCRL- 99926, Rev. 1
PREPRINT

Received by OSTI

JUN 22 1989

**Preventing Depletion of Stratospheric Ozone—
Implications on Future Aircraft Emissions**

Douglas E. Kinnison and Donald J. Wuebbles

This paper was prepared for submission to
the 82nd APCA Annual Meeting and Exhibition,
Anaheim, CA
June 25-30, 1989

May 1989

Lawrence
Livermore
National
Laboratory

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

**Preventing Depletion of Stratospheric Ozone—
Implications on Future Aircraft Emissions**

Douglas E. Kinnison and Donald J. Wuebbles

Lawrence Livermore National Laboratory
P.O. Box 808, L-262
Livermore, California 94550

MASTER



DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

ABSTRACT

There is much recent renewed interest in the development of faster aircraft for intercontinental passenger flights. Such aircraft would likely spend a large fraction of their flight time in the stratosphere, at altitudes as high as 35 km. It is important, in order to prevent the problems with the proposed supersonic-transport that occurred in the early 1970's, that the aircraft industry work together with the atmospheric science community to insure that future aircraft emissions will not deplete stratospheric ozone. In this study, we have used our two-dimensional model of the troposphere and stratosphere to examine the sensitivity of stratospheric ozone to such emissions. Initial results indicate, for commercial fleets as large as proposed for the original SST and depending on the odd-nitrogen emissions per engine, that substantial decreases in stratospheric ozone could result. The decrease in ozone is sensitive to the altitude and latitude in the emissions. Effects on ozone, for the same emission rate, tend to be larger as altitude increases, until a maximum effect is reached near 30 km.

I. INTRODUCTION

Interest is increasing in the development of faster aircraft for intercontinental passenger flights (e.g., First International Conference on Hypersonic Flight in the 21st Century, University of North Dakota, Grand Forks, ND, September 1988). It is important, in order to prevent the problems discovered in the early 1970's with the development of the proposed supersonic transport¹ that the aircraft industry work together with the atmospheric science community to insure that future aircraft emissions will not significantly deplete stratospheric ozone or cause other significant effects on the environment. The purpose of this study is to build on our previous analyses^{2,3,4} of potential aircraft emissions on ozone to determine useful limits on NO_x emissions. The LLNL two-dimensional model of the troposphere and stratosphere has been used to examine the sensitivity of stratospheric ozone to such emissions. The two-dimensional model provides latitudinal, altitudinal, and seasonal resolution, which is an advantage over previous studies that primarily used one-dimensional models, to calculate the effect of NO_x emissions on ozone.⁵

II. DESCRIPTION OF LLNL TWO-DIMENSIONAL MODEL

The LLNL zonally-averaged two-dimensional chemical-radiative-transport model currently determines the atmospheric distributions of 31 chemically active atmospheric trace constituents in the troposphere and stratosphere. The model domain extends from pole to pole, and from the ground to 0.56 mb (approximately 0 to 54 km). The sine of latitude is used as the horizontal coordinate with intervals of about 10°. The vertical coordinate corresponds to the natural logarithm of pressure ($z^* = -H_0 \ln(p/p_0)$, where H_0 is the assumed scale height of 7.2 km, and p_0 is the surface pressure, 1013 mb). The vertical resolution is $\ln(p/p_0) = 0.417$ or about 3 km. Ninety-five chemical and photochemical reactions are included in the model. Reaction rates, solar flux data, absorption cross-sections, and quantum yields are based on the latest NASA panel recommendations.⁶ Photodissociation rates, including the effects of multiple scattering, are computed as a function of time at each zone, with optical depths consistent with calculated species distributions.

The diabatic circulation for the ambient atmosphere is determined using net heating rates calculated in an internally consistent way with the derived species distributions. The

technique for deriving the diabatic circulation is similar to that used by Solomon et al. [1986]⁷: the vertical velocity is determined from the zonally averaged residual Eulerian thermodynamic equation, while the horizontal velocity is determined using the equation for mass continuity. The net heating rates are determined using accurate solar and infrared radiative models.

Temperatures for the ambient atmosphere vary continuously, over the annual cycle, based on observations.⁸ The derived diabatic circulation depends strongly on the temperature distribution; by using observed temperatures for the ambient atmosphere, a more accurate representation of the diabatic circulation can be derived.

For the perturbed atmosphere, a perturbation form of the thermodynamic equation is solved for the changes in stratospheric temperatures resulting from changes in the distribution of ozone and other radiatively active constituents. Using this approach, the diabatic circulation is assumed to be unchanged in the perturbed atmosphere from that calculated for the ambient.

Turbulent eddy transport is parameterized through diffusion coefficients K_{yy} and K_{zz} . In the current version of the model, a value of K_{yy} of $2 \times 10^9 \text{ cm}^2\text{s}^{-1}$ is assumed at all stratospheric altitudes and latitudes; and values of 5×10^{10} are assumed in the troposphere. Values of K_{zz} are $1 \times 10^3 \text{ cm}^2\text{s}^{-1}$ in the lower stratosphere, increasing slowly with altitude based on gravity wave modeling studies.

The continuity equation for each individual species is solved using a variable time-step, variable order, implicit technique for solving stiff numerical systems with strict error control. Advection terms are treated using an accurate two-dimensional transport algorithm.⁹ The diurnal-averaged concentrations for each species at each zone are calculated at each time step. Diurnal calculations are used to derive time-varying factors for each chemical and photochemical reaction included in the diurnal-averaged version of the model.

The species distributions calculated by the LLNL two-dimensional model shows good agreement compared to observed data obtained from ground based, balloon, aircraft, rocket and satellite measurements of present day atmospheres. For example, the distribution of column ozone calculated by the LLNL two-dimensional model along with observed column ozone measured by the global Dobson network is shown in Figure 1. The model simulates the ozone maximum in both hemispheres during spring. However, the model does not calculate the Southern Hemisphere spring maximum, which is observed to form off the pole. This effect is due to dynamic processes that are currently not treated in such models. Also the model results here did not include the effects of the Antarctic "ozone hole."

III. RESULTS AND DISCUSSION

Recent one and two-dimensional sensitivity calculations^{2,3,4} confirm results from earlier studies^{5,10} that ozone is sensitive to the altitude and latitude of NO_x emissions. We will first describe some results from the recent papers. Using the one-dimensional model, injections of NO_x below 13 km increased the column ozone amount (Figure 2). For emissions at altitudes above 13 km, injected NO_x decreases the ozone column, with the effect on ozone increasing as a function of the rate of injection and with altitude of emission. For emissions above 30 km, the reduction of the ozone column decreases with altitude for a given injection rate. This is due to the importance of odd-nitrogen sink processes in the upper

stratosphere. For the standard Climatic Impact Assessment Program (CIAP)¹ injection (1.8 MT yr^{-1} as NO_2 , at 19.5 km), which corresponds to about 500 of the then proposed American SST's flying 8 hours per day, the ozone column reduction was calculated in the one-dimensional model to be about 10 percent. This emission corresponds to about 2.3×10^{34} molecules NO_2 per year. The two-dimensional model, for the same magnitude and altitude of injection and with the emissions assumed to be spread globally calculates a global average reduction of 7.2 percent (Figure 3). The two-dimensional model calculates larger reductions at high latitudes (-11 percent) compared to the equatorial region (-5 percent). The lifetime of total reactive odd-nitrogen,

$$\text{NO}_y = \text{NO} + \text{NO}_2 + \text{NO}_3 + \text{HNO}_3 + \text{HNO}_4 + 2 \times \text{N}_2\text{O}_5 + \text{ClNO}_2 + \text{ClONO}_2 + \text{HONO}$$

is larger for the one-dimensional model (2.7 years) than the two-dimensional model (1.9 years). Here lifetime is defined as the global excess of NO_y at steady state divided by the global NO_x injection rate. The differences between the globally averaged two-dimensional model result and that given by the one-dimensional model appear to be related to different assumption in treating transport processes.

In Figure 4, using the one-dimensional model, the percentage change of the ozone vertical column as a function of stratospheric odd chlorine, $\text{Cl}_x = \text{Cl} + \text{ClO} + \text{HCl} + \text{HOCl} + \text{ClONO}_2 + \text{ClNO}_2$, and NO_x injection rates for emissions at 20 km is shown. With no injection of NO_x , the effect on ozone of increasing Cl_x (i.e., from chlorofluorocarbon emissions) for the present atmosphere (approximately 3 ppbv) relative to a "1960" reference atmosphere is -1.4 percent. For "future" atmospheres, 8 ppbv of Cl_x is readily attainable at present chlorofluorocarbon emission rates. With 8 ppbv of Cl_x , ozone is reduced by 5.1 percent relative to a "1960" reference atmosphere. With injections of NO_x , Figure 4 can be divided into two regions, region A, where increasing the rate of NO_x injection decreases the ozone column; and region B, increasing the rate NO_x injection increases the ozone column. For all Cl_x reference atmospheres, the addition of NO_x "buffers" the effect of Cl_x reduction on ozone. At high reference Cl_x , greater than 12 ppbv (region B), column-ozone increases for an injection between zero and 1000 molecules $\text{cm}^{-2} \text{ s}^{-1}$. This increase is small in comparison to the already large reduction of column ozone due to chlorine.

In this study, two additional scenarios are developed. These scenarios attempt to spread the emissions over a wider range of altitudes than we assumed in the previous analyses we published (which put all of the NO_x emissions into a single altitude and latitude). These scenarios were chosen to approximate some of those done during the CIAP studies in the 1970's. However, we also examine the effect of varying the aircraft cruise altitudes. The first scenario, case A, is assumed to have a maximum cruise altitude of 22.5 km, while in the second scenario, case B, we assumed a maximum cruise altitude of 25.5 km. The case A fleet cruises at a lower altitude, and injects 39 percent more NO_x per year than the case B fleet. Total global emissions in case A and B are 7.2×10^{33} and 5.1×10^{33} molecules of NO_2 per year, respectively. The maximum NO_x injection for both scenarios is assumed to be at 42°N ($\sim 1.8 \times 10^{33}$ molecules of NO_x per year at 22.5 km in case A and 1.15×10^{33} molecules NO_x per year in case B). The case A scenario has larger assumed emissions than case B in order to give a comparable change in global

ozone; this simplifies some of our upcoming discussion of the model results. The intention of these scenarios is to provide a more reasonable estimate of potential emissions as a function of latitude and season than the previous sensitivity studies.^{2,3,4} However, we need to emphasize that the magnitude of emission, as well as the variations with latitude, are entirely arbitrary, and are not intended to match any particular aircraft designs.

In Figure 5, for the case A scenario, the maximum ozone reduction calculated in the LLNL two-dimensional model occurs at high northern latitudes (-4.5 percent), during fall, with decreasing effects on ozone at lower latitudes (-1.0 percent at equator). In the Southern Hemisphere, the maximum ozone reduction is constant across season, showing a maximum decrease in column ozone of 2 percent. Figure 6 shows results for the case B scenario fleet; the column ozone reduction is similar to case A in seasonal shape and magnitude. The importance of the ozone reduction efficiency to NO_x emissions versus altitude is obvious when comparing Figures 5 and 6. The case A fleet injects 39 percent more NO_x than the case B fleet, but due to the altitude of injection does not decrease column ozone proportionally. In previous studies, for injection typical of these scenarios and ambient odd chlorine concentrations, the maximum ozone reduction efficiency to injected NO_x calculated by the LLNL one-dimensional model peaked at 25 ± 2 km.^{2,3}

In Figures 7 and 8, the local percent change in ozone is shown for the two scenarios. The maximum local percent ozone change occurs at about 19.5 km in both cases; maximum ozone depletion is about 6% and 4% at mid-latitudes in the Northern Hemisphere for the case A and case B assumed scenarios, respectively. The maximum effect on ozone actually occurs at a lower altitude than the cruise altitude because of the effects of atmospheric dynamical processes in the model. In the Southern Hemisphere, percent change in local ozone is about half the reduction in the Northern Hemisphere. Both proposed scenarios exhibit similar latitudinal and altitudinal local ozone reduction response to NO_x injections, even though the altitude of injection are different.

It is impossible for us to say what amount of ozone depletion would be acceptable to society. We will assume, for the sake of argument, that a 0.5 percent change in total ozone is acceptable (we are not saying it is) at any given latitude; with normal variability in ozone and existing measurement uncertainties such a change would likely be difficult to be accurately determined. Assuming a near linear relationship between emissions and the calculated change in ozone (at least for small O_3 perturbation), emissions of NO_x from both the case A and case B scenarios would have to be reduced by a factor greater than eight.

IV. CONCLUSIONS

The effects of two scenarios for future commercial aircraft fleets were examined in the LLNL two-dimensional model. These scenarios were extensions of those examined in prior analyses. They attempt to account for variations in emissions with latitude and altitude not considered in the other recent studies. We calculated the effect that NO_x emissions from these scenarios would have on both the local and column ozone distributions. These scenarios were chosen because they provide a crude, but not unrealistic estimate of possible variations in aircraft emissions as a function of latitude. However, polar flights between North America and Europe, although potentially important, were not addressed in these scenarios. Different assumed cruise altitudes of emissions were also examined; however,

the flight altitudes used were chosen arbitrarily and are not designed to match any given aircraft.

Major findings of this study are:

1. For the scenarios we examined (which assumed emissions comparable to fleets examined in CIAP studies), it was possible for significant decreases in ozone to occur in both the Northern and Southern Hemispheres. Emissions for the specific scenarios examined would need to be reduced substantially to limit the derived ozone change.
2. The change in ozone from any assumed scenarios depends critically on the magnitude, altitude, and latitude of the emissions.
3. The atmospheric sciences community will need to work closely with the aircraft industry to estimate the potential effects of emissions from future commercial aircraft flying in the stratosphere, especially to indicate emission levels that would affect levels of global atmosphere ozone.

V. ACKNOWLEDGEMENTS

This work was sponsored by the U.S. Department of Energy Carbon Dioxide Research Program and performed by the Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48.

VI. REFERENCES

1. CIAP Monograph 2, "Propulsion effluent in the stratosphere; Monograph 3, The stratosphere perturbed by propulsion effluent," U.S. Department of Transportation, Washington, D.C., (1975).
2. D. Kinnison, H. Johnston, and D.J. Wuebbles, "Sensitivity study of global ozone to NO_x emission from aircraft," Proceedings of the 1988 International Quadrennial Ozone Symposium, Gottingen, Fed. Rep. of Germany, Aug. 8-13, 1988.
3. D. Kinnison, D.J. Wuebbles, and H.S. Johnston, "A study of the sensitivity of stratospheric ozone to hypersonic aircraft emissions," Proceedings of the First International Conference on Hypersonic Flight in the 21st Century, Grand Forks, N.D., Sept. 20-23, 1988.
4. H. Johnston, D. Kinnison and D. Wuebbles, "Nitrogen oxides from high altitude aircraft: An update of potential effects on ozone," submitted to *J. Geophys. Res.*, (1988).
5. WMO (World Meteorological Organization), Atmospheric ozone: Assessment of our understanding of the processes controlling its present distribution and change," Global ozone research and monitoring project, Report No. 16, (1985).
6. W. DeMore, J.J. Margitan, M.J. Molina, R.T. Watson, D.M. Golden, R.F. Hampson, M.J. Kurylo, C.J. Howard, and A.R. Ravishankara, "Chemical kinetics and photochemical data for use in stratospheric modeling," Jet Propulsion Laboratory, Pasadena, CA: JPL Publication 85-37, (1985); JPL 87-41, (1987).
7. S. Solomon, J.T. Kiehl, R.R. Garcia, and W. Grose, "Tracer transport by the diabatic circulation deduced from satellite observations," *J. Atmos. Sci.*, **43**, 1604-1617, (1986).
8. J.J. Barnett and M. Corney, "A middle atmosphere temperature reference model from satellite measurements," *Adv. Space Res.*, **5**, 125-134, (1984).
9. P.K. Smolarkiewicz, "A fully multidimensional positive definitive advection transport algorithm with small implicit diffusion," *J. Comp. Phys.*, **54**, 325-362, (1984).
10. D.J. Wuebbles, "A theoretical analysis of the past variations in global atmospheric composition and temperature," Lawrence Livermore National Laboratory, Report UCRL-53423, (1983).

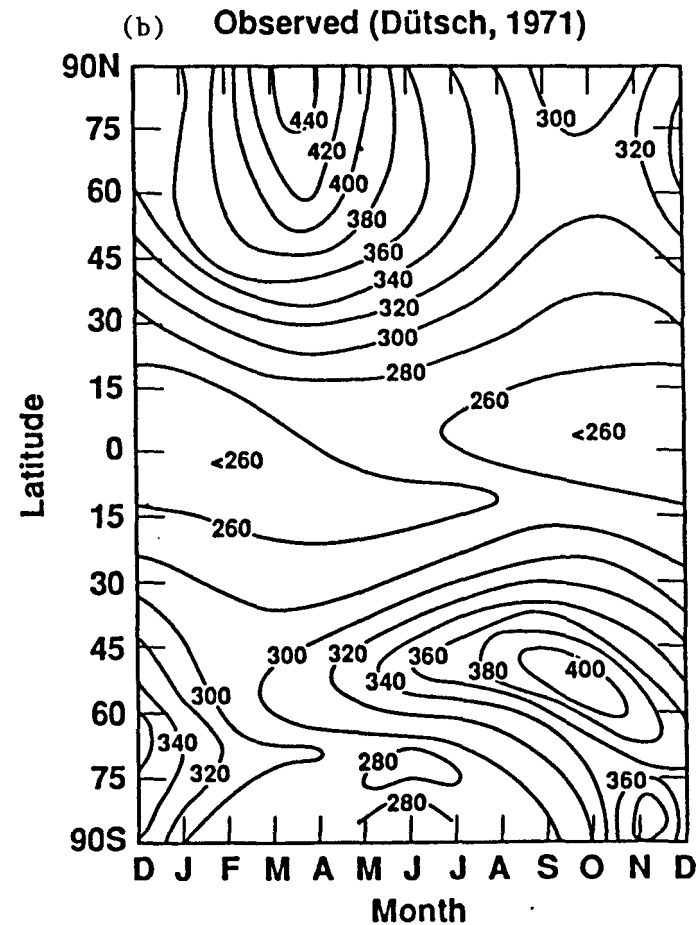
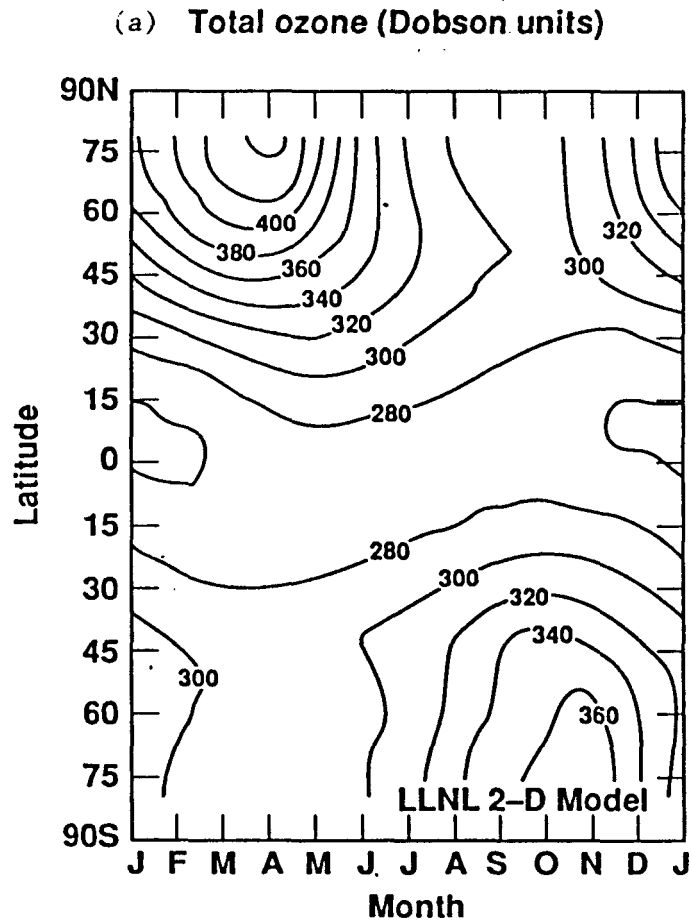


Figure 1. Column-ozone as a function of latitude and time of the year (Dobson units). (a) Calculated by the LLNL two-dimensional model. (b) Taken from Dütsch [1971], using data from the global Dobson network.

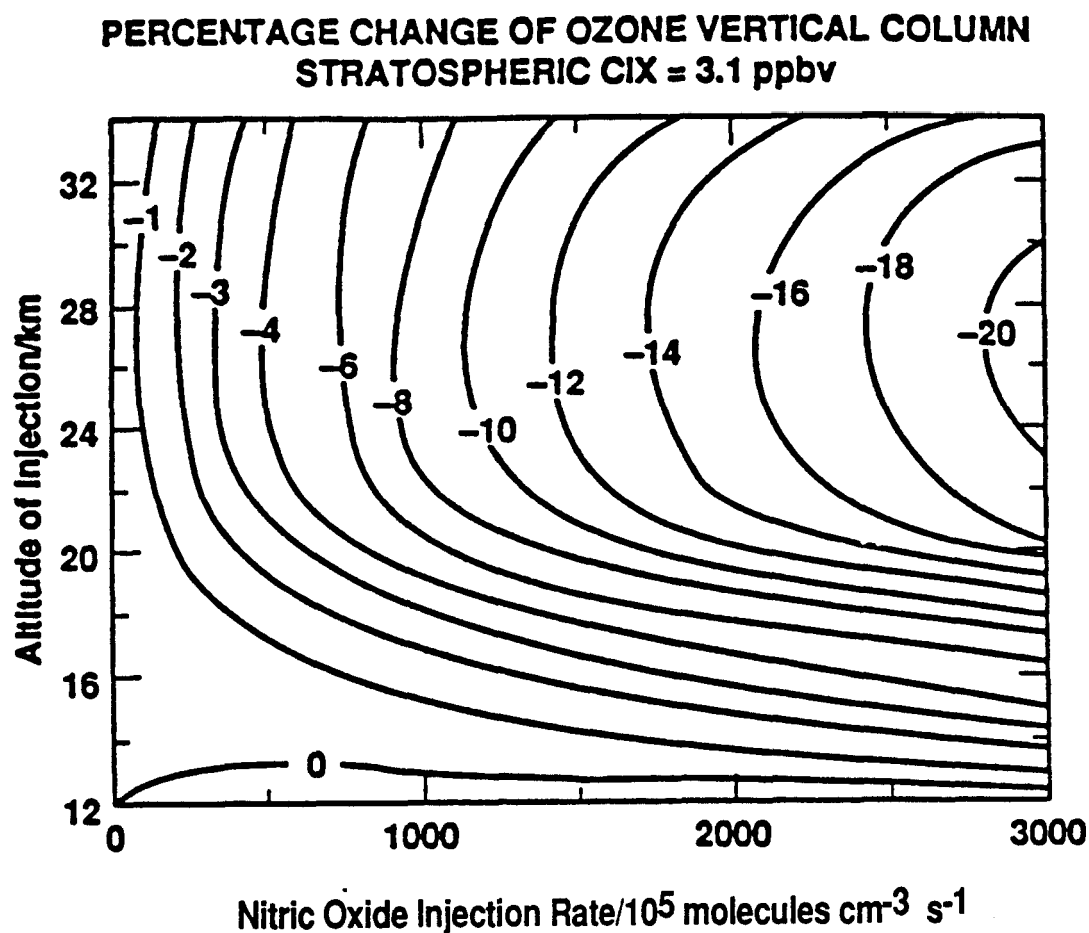


Figure 2. Percentage change of the ozone vertical column as calculated by the LLNL one-dimensional model as a function of altitude on NO_x injection and magnitude of NO_x injection in units of molecules cm⁻³ s⁻¹ over a one kilometer band centered at the stated altitude. The percentage change in ozone is relative to a reference atmosphere that contains 3.1 ppbv Cl_x. An injection of 1500 in these units corresponds to a global injection of 1.8×10^{12} g yr⁻¹ (as NO₂). Photochemical coefficients are from DeMore et al. (1985).⁶

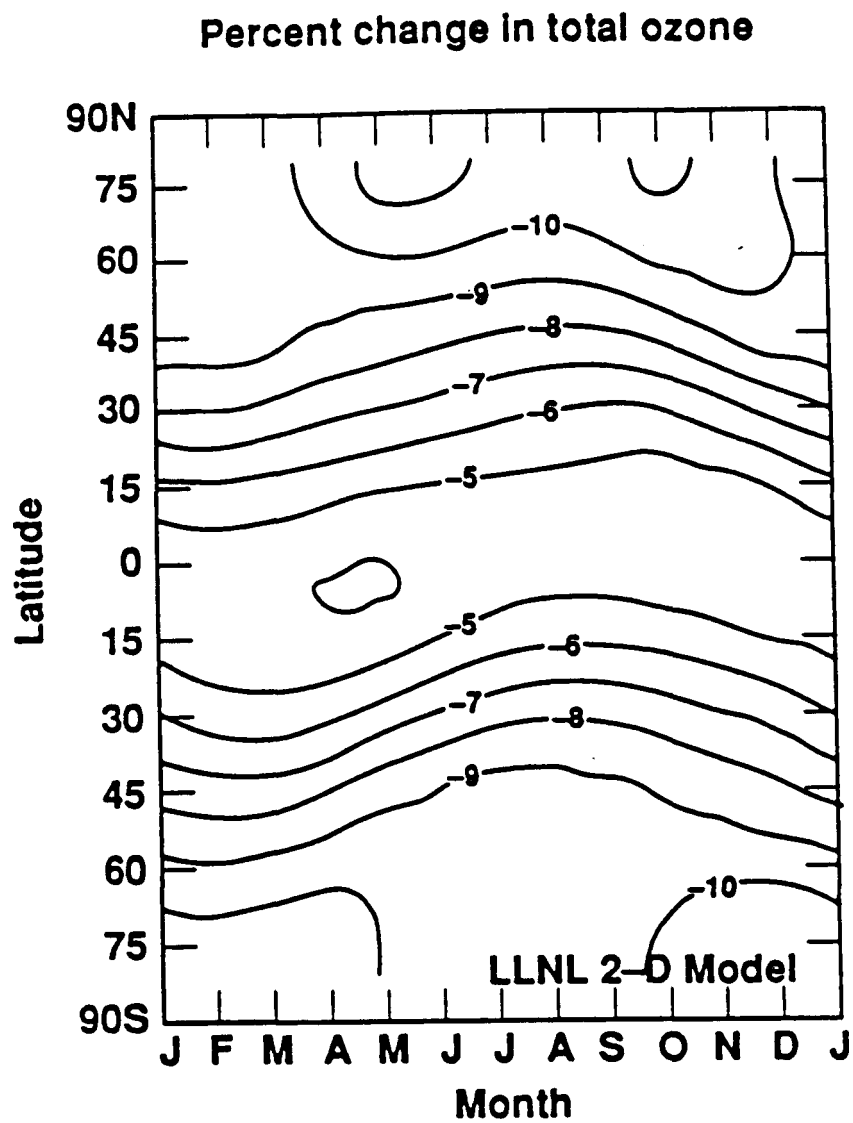


Figure 3. Percentage change in ozone-column for an injection of $1.8 \times 10^{12} \text{ g yr}^{-1}$ (as NO_2) at 19.5 km relative to a reference atmosphere with 2.8 ppbv Cl_x .

PERCENTAGE CHANGE OF OZONE VERTICAL COLUMN
ALTITUDE OF NITRIC OXIDE INJECTION, 20/KM

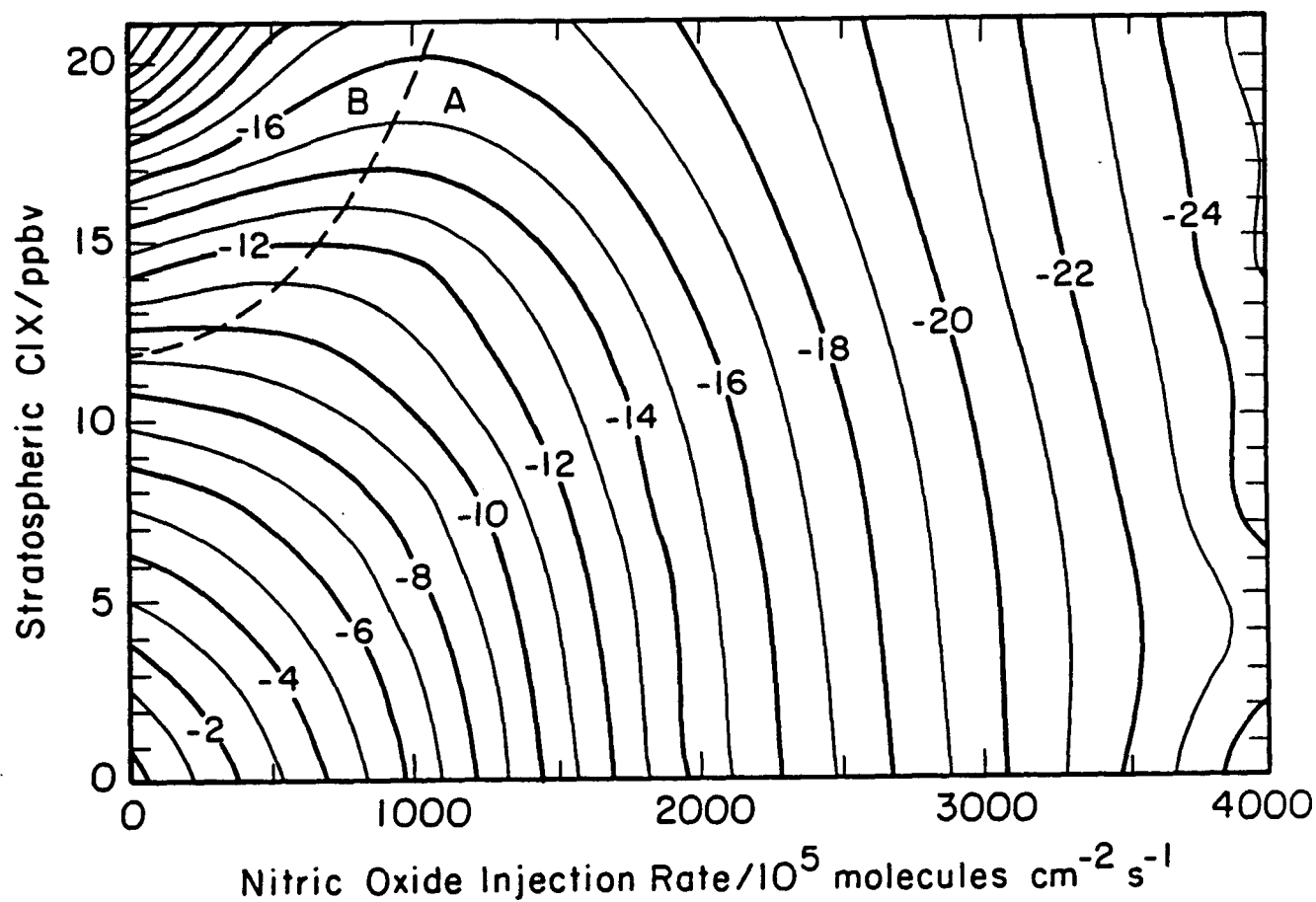


Figure 4. Contour of percentage change of the ozone vertical column as a joint function of NO_x injection rate at 20 km and stratospheric Cl_x mixing ratio. To the left of the dashed line, an increase in NO_x injection rate (e.g. from stratospheric aircraft) decreases the ozone reduction caused by Cl_x , giving a relative increase of ozone.

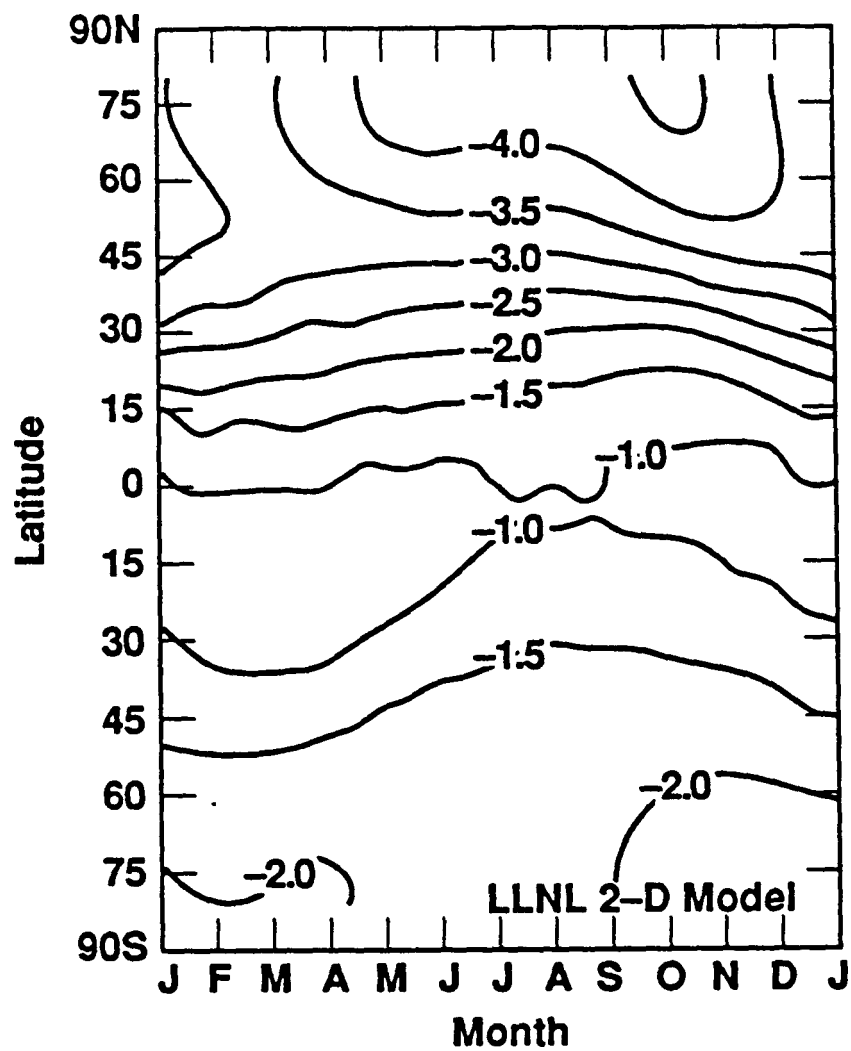


Figure 5. Percentage change in ozone column for the assumed case A scenario. The reference atmosphere has 2.8 ppbv Cl_x .

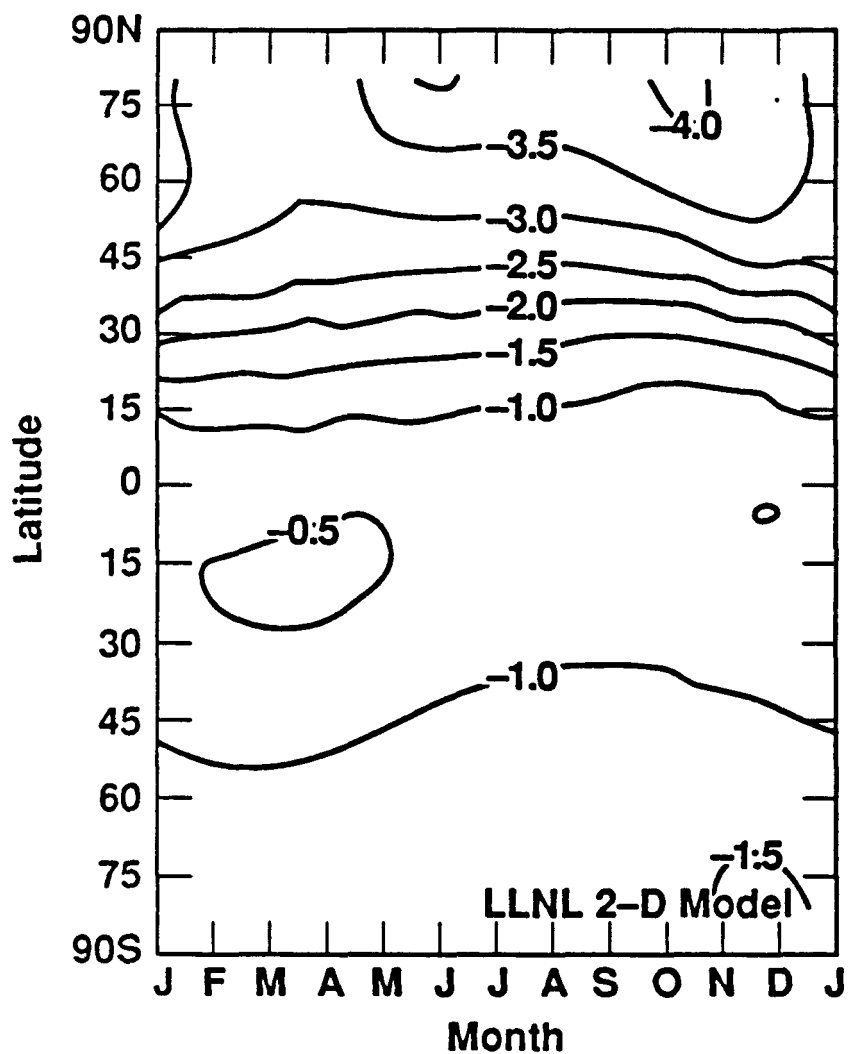


Figure 6. Percentage change in ozone column for the assumed case B scenario. The reference atmosphere has 2.8 ppbv Cl_x .

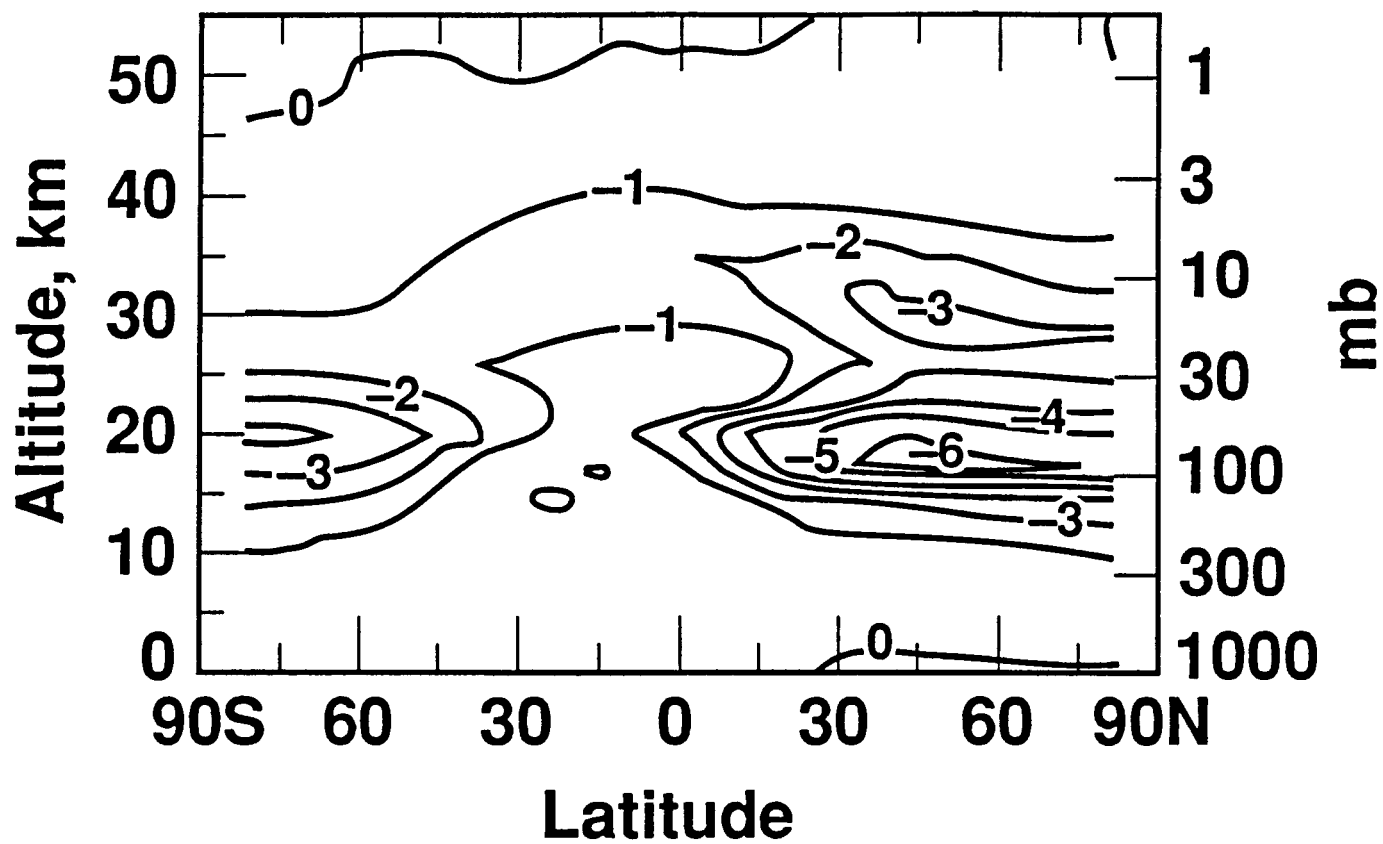


Figure 7. Percentage change in local ozone for the assumed case A scenario. The reference atmosphere has 2.8 ppbv Cl_x .

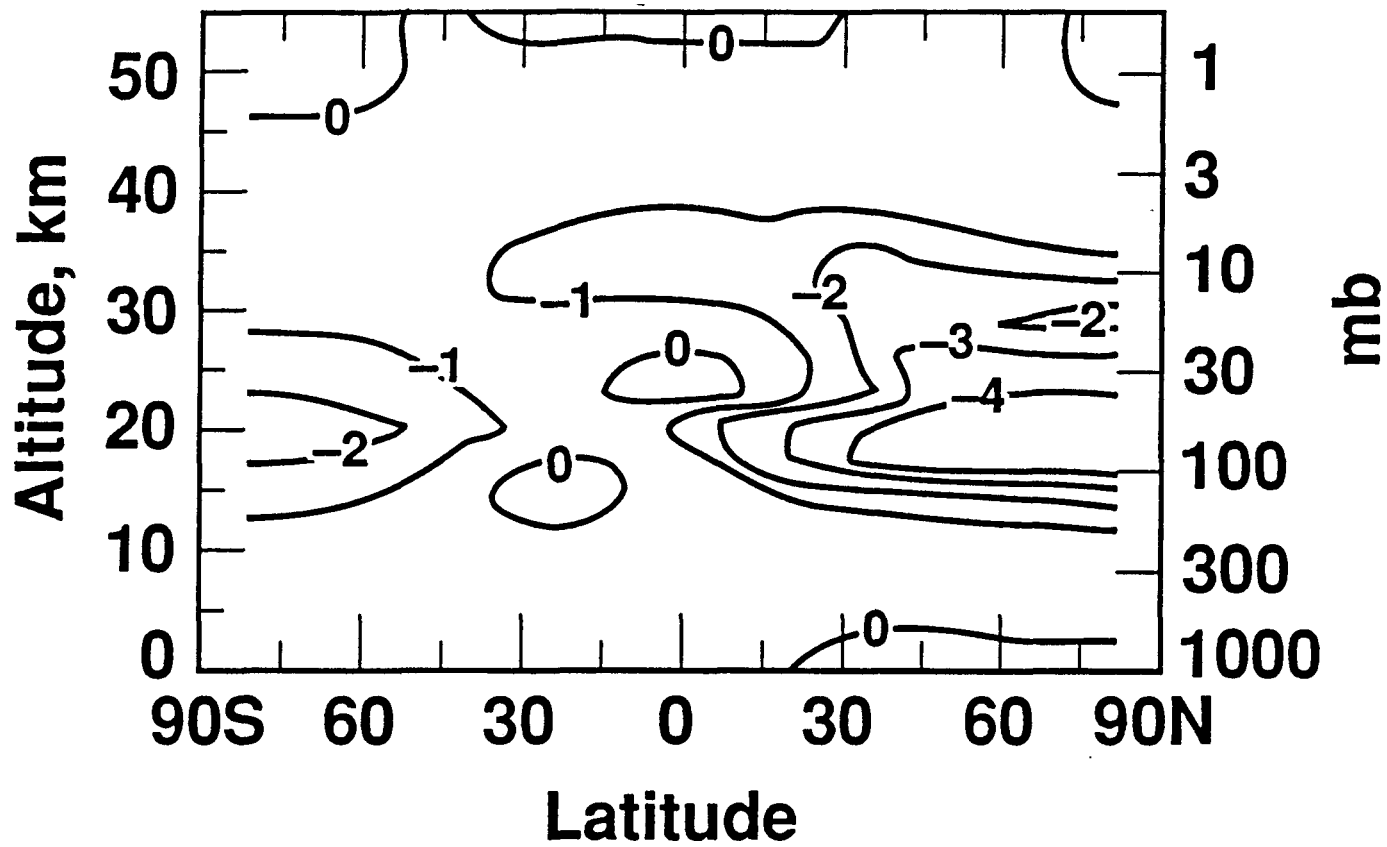


Figure 8. Percentage change in local ozone for the assumed case B scenario. The reference atmosphere has 2.8 ppbv Cl_x .