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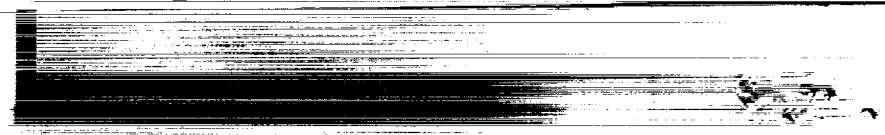

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The Atmospheric Effects of Stratospheric Aircraft: A Current Consensus

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PREFACE

The Upper Atmosphere Research Program (UARP) of the National Aeronautics and Space Administration (NASA) has recently undertaken the responsibility of directing scientific research needed to assess the atmospheric impact of supersonic transports. Supersonic aircraft operate most efficiently in the stratosphere where their emissions, primarily engine exhaust, accumulate and can perturb the chemical cycles controlling stratospheric ozone. The UARP has the duty of responding to the Congressional directive given to NASA in June 1975, and in the Clean Air Act Amendments of 1977, to "develop and carry out a comprehensive program of research, technology, and monitoring of the phenomena of the upper atmosphere so as to provide for an understanding of and to maintain the chemical and physical integrity of the Earth's upper atmosphere," particularly the ozone layer.

The Atmospheric Effects of Stratospheric Aircraft (AESA) studies were initiated by the UARP under the sponsorship of NASA's High-Speed Research Program (HSRP) with the intent of establishing a base of scientific knowledge about how aircraft emissions impact the stratosphere. The HSRP provides overall management for AESA as well as for additional studies on key environmental issues regarding high-speed civil transports. This research should provide a foundation for the national and eventually, international assessment of the environmental impacts of a commercial fleet of such aircraft.

In the late 1960s, the aircraft industry became interested in developing a fleet of supersonic transports, then denoted as SSTs. Some atmospheric scientists noted that certain exhaust products from the engines, notably NO_x ($NO+NO_2$), would increase the chemical destruction of stratospheric ozone and might lead to substantial decreases in the column abundance of ozone with corresponding increases in solar ultraviolet and damage to human health and the biosphere. The effort to conduct an environmental assessment culminated in the U.S. with the Department of Transportation's Climatic Impact Assessment Program (CIAP) in 1972-1975. That assessment must be regarded today as inconclusive in view of the great advances in stratospheric science since then. However, the predicted environmental impacts were not to be tested, the SSTs were not a commercial success, and the current operating fleet of 13 Anglo-French Concordes is too small to have a global impact on ozone.

There is now renewed commercial interest in supersonic aircraft, now denoted as high-speed civil transports (HSCTs). Compared with the Concorde, the new HSCT would be more fuel efficient, carry triple the number of passengers, and have twice the range (about 6000 nm, which is necessary for Pacific routes). The possibility of a large fleet of HSCTs has once again raised basic questions concerning the environmental acceptability of supersonic aircraft, including airport community noise, sonic boom, and stratospheric ozone. More than 15 years have elapsed since the last formal assessments of the environmental impact of stratospheric, supersonic aircraft.

The UARP and the HSRP asked a group of atmospheric scientists, under the chair of Dr. Anne Douglass, to report on our current knowledge of the potential atmospheric impact of stratospheric aircraft and on our ability to predict the environmental consequences of a fleet of HSCTs. This ad hoc

committee took Professor Harold Johnston's topical review, which is also a NASA Reference Publication, as the basis of this report. The group expanded on the topics covered in the Johnston review and forged a consensus document. This report has been reviewed by the Advisory Committee for AESA studies. We believe that it represents the current consensus of the community and wish to thank those scientists who contributed to its writing and review.

Michael J. Prather
Robert T. Watson
July 1990

SUMMARY

The Atmospheric Advisory Committee of the High-Speed Research Program (HSRP) of the National Aeronautics and Space Administration (NASA) requested a review of the history and current status of scientific studies of the effect of exhaust gases from stratospheric aircraft on global ozone. This review should serve as an introduction and benchmark for its new atmospheric research program on this subject. The possible environmental impact of supersonic transports (SST's) was previously considered in three programs: the Climatic Impact Assessment Program (CIAP) conducted by the U. S. Department of Transportation between 1972 and 1975; The Committee on Meteorological Effects of Stratospheric Aircraft (COMESA) in Great Britain, and Comite d'Etude sur les Consequences des VOls Stratospheriques (COVOS) in France. Since the scheduled termination of CIAP in 1975, there has been very little research specifically concerned with the environmental effects of stratospheric aircraft.

NASA sponsorship of stratospheric research began in 1976, and subsequent environmental work has been heavily concerned with the role of increasing chlorine abundance in the stratosphere. Great advances in understanding the stratosphere have been made since 1975, but to date little research has applied these advances to the problem of stratospheric aircraft. Although much of the work of the early 1970's is out of date, some is still relevant to this problem and should not be overlooked or unnecessarily duplicated. This report is a brief history of work in this field since 1970, and a presentation of recent model calculations of the effect of nitrogen oxides (NO_x) from the aircraft exhaust.

Critical elements of this problem are summarized in the following three statements:

1. For stratospheric aircraft flying at about 20 km, calculations made by three independent groups using two-dimensional chemical models (Johnston, Kinnison and Wuebbles, Lawrence Livermore National Laboratory, Livermore, CA (LLNL); Ko, Weisenstein, Sze, Rodriguez and Helsey, Atmospheric and Environmental Research, Inc. Boston, MA (AER); Isaksen, Stordal and Berntsen, Institute of Geophysics, Oslo, Norway (OSLO)) predict a significant reduction in total ozone (see Table 2). The model calculations indicate that for a given injection of NO_x, the calculated effect on total ozone is a strong function of altitude, and aircraft flying well below 20 kilometers give lower calculated ozone reductions. For the basic Boeing scenario for 70.6×10^9 kg fuel per year, aircraft flying at 20 km, Mach 2.4, and an emission index of 39.5 gm NO/kg fuel, the global ozone change calculated using the LLNL model is about 16 percent, a value greater than the worst case scenarios for continued emission of chlorofluorocarbons (excluding the impact of heterogeneous processes on polar stratospheric clouds that lead to the formation of the Antarctic ozone hole). Larger injections of NO (greater fuel consumption or greater emission of NO/kg fuel) would produce even larger ozone reductions.

2. NASA Engineers are optimistic that the emission index (gm of NO/kg fuel, 39.5 in the above example) can be reduced to 5 by use of advanced emission reduction technology (Niedzwiecki, 1988). Although this obviously would greatly decrease the calculated ozone reductions from those calculated for

current values of the emission index, it should be noted that an emission index as low as 5 may still produce substantial ozone reductions. Successful development of such engines is not assured, however, and safety must be maintained. Engineers at both Pratt and Whitney and at General Electric "have identified combustor concepts that they believe have the potential to achieve a cruise NO emission index of 5 to 10...it would be a high risk development program with potential barriers to success being premixing duct flashback and auto ignition..." (Boeing, 1989, pp 41-42).

3. Although current calculations are made using state-of-the-art atmospheric models, such models lack some features that are needed for modeling the atmospheric effects of high speed stratospheric aircraft. These include the following: A) heterogeneous chemical processes that occur on polar stratospheric clouds and are key to the formation of the Antarctic ozone hole; B) aircraft exhaust plume physics and chemistry; C) realistic representations of mixing in the lower stratosphere and of cross tropopause transport processes. According to present speculations, including (A) and (B) may increase the predicted ozone reduction (release of active chlorine from chlorine reservoirs by heterogeneous reactions) or it may decrease the predicted ozone reduction (removal of NO_y through fall-out of solid nitric acid). The improved representations of transport and mixing in both the upper troposphere and lower stratosphere (C) may require an increase in vertical resolution in the 5 to 25 km altitude range, as well as an increase in horizontal resolution. In addition to these model improvements, credible model calculations must account for the changes in the background atmosphere that are likely to occur over the next few decades.

To judge the effect of stratospheric aircraft on ozone properly, it is necessary to carry out some further atmospheric observations, laboratory studies, and model development specifically devoted to this problem. The following are recommendations for future research.

SPECIFIC RECOMMENDATIONS

1. Relevant work done by the Climatic Impact Assessment Program (CIAP) in the United States, the Committee on Meteorological Effects of Stratospheric Aircraft (COMESA) in Great Britain, and Comite d'etude sur les COnsequences des VOls Stratospheriques (COVOS) in France should not be unnecessarily duplicated, although much of this work from the early 1970's is out of date (Summary, vii).

2. For future evaluations, a consistent set of scenarios for emissions, including fuel consumption, emission index, altitude, latitude and season of injection, must be developed (p. 14).

3. A range of options for both NO_x emissions (annual fuel consumption times NO molecules/kg fuel) and cruise altitude of aircraft (related to the Mach number) should be considered (p. 15).

4. Models (2D and 3D) must include improved representations of stratosphere/troposphere exchange and mixing in the lower stratosphere. If increased vertical resolution is required, the latitudinal resolution must be compatible (p. 16).

5. Improvements in understanding and model representation of stratosphere/troposphere exchange and horizontal and vertical mixing processes in the 12-20 km region should be based on observational studies. Data sets currently available include carbon-14, other radioactive bomb debris, and species (e.g., O_3 and HNO_3 profiles). Future studies may require long duration flights by high altitude research aircraft (p. 16)

6. Model calculations must include the impact of water from the exhaust, and include realistic estimates of background gases and their probable future changes. The background gases include methane, carbon dioxide, nitrous oxide and chlorine species (p. 17 and 21).

7. Parameterizations of the impact of the heterogeneous processes which explain the evolution of the Antarctic ozone hole should be included in assessment models (p. 20 and 23).

8. Detailed model calculations of plume chemistry and physics as reported in CIAP Monograph 3, with current rate constants and additional reactions, should be compared with engine measurements reported there. Definitive experiments and theoretical arguments should be developed for this area, and for related heterogeneous processes (p. 17-20).

9. Aerosol effects should be considered, building on significant CIAP and post-CIAP studies that are still applicable to this problem (p. 23).

INTRODUCTION

Although the development of supersonic stratospheric aircraft was abandoned in the United States in the early 1970's for economic, political, and environmental reasons, the prospect of technological advancement has renewed interest in stratospheric aviation. The First International Conference on Hypersonic Flight in the 21st Century was held in Grand Forks, North Dakota September 20-23, 1988. A European Forum on Future Supersonic/Hypersonic Transportation was held in Strasbourg in November, 1989. The National Aeronautics and Space Administration (NASA) has initiated a preliminary study of High Speed Civil Transport (HSCT) with participation and cost sharing from Boeing Commercial Airplanes and McDonnell Douglas. NASA opened this project to the general scientific public on July 28, 1989 with the research announcement "The Atmospheric Effects of Stratospheric Aircraft: Modeling and Measurement in Support of the High-Speed Research Program" (NRA-89-OSSA-16). This report addresses the current scientific understanding of the environmental effects of a fleet of stratospheric aircraft. It is designed to evaluate the need for this research program and to provide a benchmark against which to evaluate its future progress.

Aircraft flying at higher speeds also fly at higher altitudes, and have specialized fuel requirements. For comparison, the scheduled airtime between Los Angeles and Sydney (7500 miles) is 14.5 hours for current aircraft flying at Mach 0.85 at 11 km, using Jet-A fuel. For a proposed aircraft, the airtime is 5.1 hours at Mach 2.4, 20 km, also using Jet-A fuel. Aircraft at higher Mach numbers (4, 6, 12) would fly at higher altitudes (25, 30, 35 km) with shorter airtimes (3.7, 3.0, 1.7 hours) but with specialized fuel and engine requirements.

The NASA High Speed Research Program project recognizes the challenge to resolve at least the following four potential problems in developing high speed flight:

1) the cost of development and operation must not be so large that there is no market for the service;

2) high temperature operation (associated with higher Mach numbers) requires thermally stable jet fuels or cryogenic fuels, and also special structural materials;

3) airport noise and sonic booms must not exceed acceptable levels;

4) possible environmental effects of exhaust gases, including ozone reduction by nitrogen oxides (NO_x), must be minimal.

Representatives of the U.S. aircraft manufacturers have stated that there would be no interest in constructing supersonic aircraft unless all four of the above problems can be given satisfactory solutions (Boeing, 1989). Ott (1988) states that research and development of a high speed transport system would cost between 2 and 4 billion dollars, and construction costs would be 15 billion more. Both Boeing and Douglas aircraft have analyzed future markets for high speed transportation, and the number of aircraft that would be required to pay off the business investment in this area (Boeing, 1989; Douglas, 1989). Boeing rejects aircraft at Mach numbers 4.5 and higher on

economic grounds and has tentatively selected the Mach 2.4 system, and worked out probable flight corridors. Douglas aircraft has selected a Mach 3.2 system as superior to a Mach 5.0 system. The proposed fleets of aircraft include a scenario which burns 7.1×10^{10} kg fuel per year. For comparison, some calculations made in the 1970's assumed 7.7×10^{10} kilograms of fuel per year.

Following is a discussion of factors which led to abandoning stratospheric aircraft in 1971, and a discussion of the first efforts to evaluate the impact of stratospheric exhaust on ozone. The major accomplishments of subsequent stratospheric research programs, the Climatic Impact Assessment Program (CIAP) 1971-1975, and the NASA sponsored program of stratospheric research, 1976 - present, are summarized. The factors which must be considered to assess the environmental impact of aircraft exhaust are delineated. In the final section, the results of recent model investigations are reported, and areas in which improvements must be made to reduce the uncertainty in model calculations are identified.

PROPOSED STRATOSPHERIC AIRCRAFT IN 1971

During the late 1960's and into 1971, the United States government was financing the design and construction of two prototype supersonic transports (SST). The government also proposed to share costs of fleet production, and was to be repaid by the airframe manufacturers from profits from the sale of a planned fleet of about 500 to 800 aircraft. During 1970 and early 1971, intense political debate, concerned primarily with the economics, national priorities and the sonic boom, focused on the role of government in this program. Late in the debate, another environmental issue, the impact of stratospheric pollution on ozone, was introduced. The potential impact of hydrogen radicals derived from H_2O on stratospheric ozone had already been recognized (Hampson, 1965). For example, Harrison (1970) suggested that free radicals derived from water vapor from the exhaust of 500 SST's would reduce global ozone by about three percent. At the height of debate on these issues, the Department of Commerce Advisory Board for SST Environmental Effects held a special meeting for an exchange of ideas and positions among a varied group of experts (Boulder, Colorado, March 18-19, 1971).

At this meeting, atmospheric scientists presented tutorials on stratospheric motions, trajectories, chemical interactions between water vapor and ozone, and lifetimes of radioactive debris in the stratosphere. Measurements of nitric acid vapor in the stratosphere were reported (Murcray et al., 1968) but Crutzen's (1970) proposal that NO_x played an important role in the natural ozone balance was not mentioned at this meeting. Aircraft experts presented: a) the planned size of the American fleet (500 SST's); b) the service time, cruise altitude, and amount of fuel consumed (7.7×10^{10} kg per year); c) the amounts of water vapor (1250 g H_2O per kg fuel) and nitrogen oxides (42 g NO per kg fuel) emitted in the engine exhaust; d) the assumption that near main flight corridors, local vertical columns of engine exhaust would exceed the globally average column by a factor of 10 (the "corridor effect").

Even though the importance of NO_x to the natural stratospheric ozone balance was not understood at this time, there was some mention at this meeting that nitrogen oxides from SST exhaust might affect stratospheric

ozone. In the concluding session, a formal motion that "the effect of nitrogen oxides on ozone may not be neglected" was strongly defeated.

Shortly after this meeting, Langley and McGrath (1971) reported an essentially zero rate for the key reactions between ozone and free radicals derived from water, based on their laboratory measurements. Although this work was later found to be in error, the report had a strong impact on understanding stratospheric chemistry in mid 1971.

In 1971 it was not feasible to carry out calculations of the vertical and horizontal spread of the exhaust gases and their chemical and photochemical reactions. Statistical mechanics provided an approach to studying this highly complicated system. The general idea is to apply the available knowledge as rigorously as possible, and assume equal a priori probability for conditions where needed information is incomplete.

The problem of time dependent dynamics was overwhelmingly difficult; information about the system was obtained by considering a static ensemble average. A two-dimensional (altitude and longitude at 45^{o} latitude), static, steady-state photochemical atmospheric model was set up, using known information. With the image of the Brewer circulation (tropical source and a polar sink, Brewer, 1949), the NO_x from the SSTs flying at 20 km was thought to spread up and down as well as horizontally. Based on the Boulder tutorial, uniform lateral spread was associated with a 10 km vertical spread. The stratospheric lifetime of NO_x from the exhaust gases was taken to be two years. Quantities varied in the spirit of statistical mechanics included: (1) the background stratospheric NO_x profile, which was taken to be both uniform and non-uniform, and varied from zero to values so high that astronomers would long ago have complained about spectral interference from atmospheric NO_2; (2) the quantities of aircraft-produced NO_x, which were varied over a range of 1000 for distributions of uniform background NO_x, and over a range of 30 for non-uniform background distributions; (3) the vertical and horizontal spread of the exhaust gases, which were taken to be 1, 4, 7, 10, 13, or 16 kilometers in the vertical, and globally uniform in the horizontal (Johnston, 1971(a), 1971(b)).

For the non-uniform NO_x profile that gave the best fit to the observed mid-latitude ozone profile, for NO_x exhaust rates equal to one-third of the 42 g NO per kg fuel (the estimate presented at the March 1971 meeting), and for uniform global horizontal spread of the exhaust gases, the steady state ozone reductions, calculated for each of the six vertical spreads, varied from 3 to 23% (Figure 1). It was assumed that the actual vertical spread would be close to some combination of these bands. The change of global average ozone was judged to be -13% ± 10%. Similar calculations, but including the ten-fold corridor effect, gave a maximum local column ozone change of -50%. These two results used 7.7×10^{10} kg fuel per year, and an emission index of 15 g NO/kg fuel, equivalently 1.8×10^{12} g of NO_2/year. Over the full range of recognized uncertainties, the large NO_x injections from the proposed fleet of U.S. stratospheric aircraft gave non-negligible calculated ozone reductions (Johnston, 1971(a), 1971(b)).

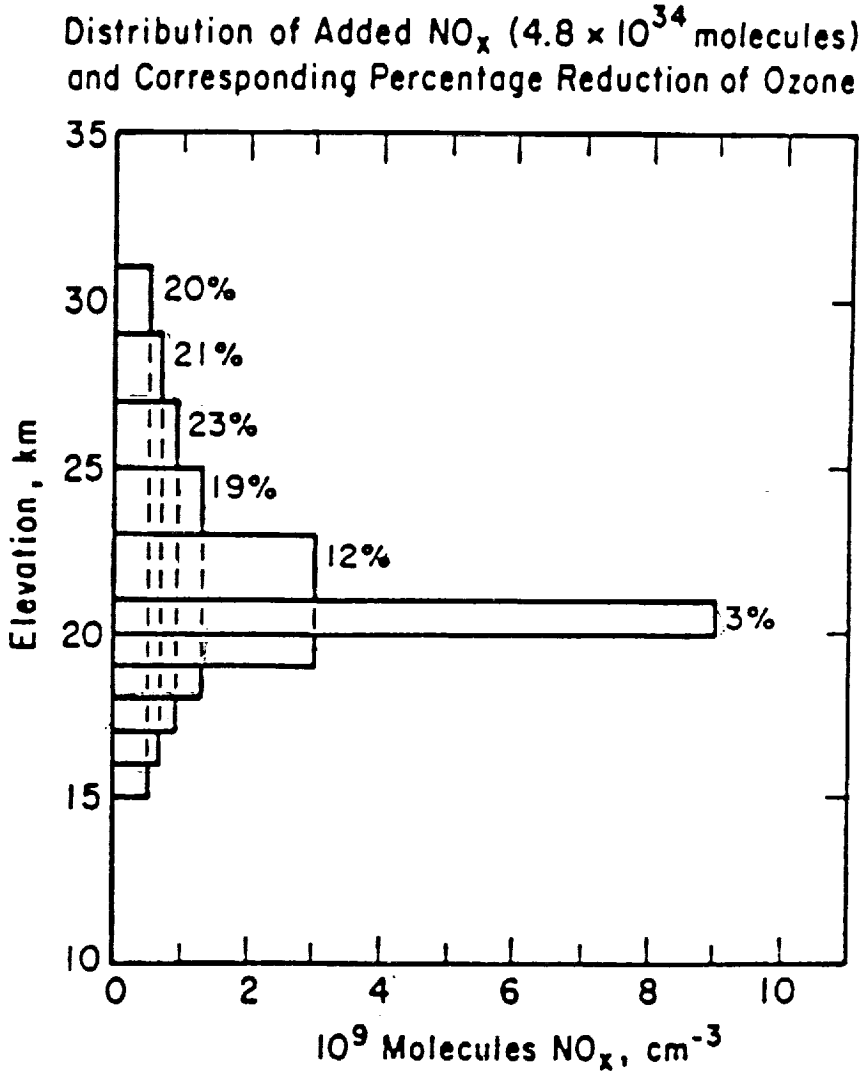

Figure 1. Steady-state calculations of the change in 24 hour average ozone columns at 45°N, spring equinox, for added artificial NO_x at 20 km with various assumed vertical spreads above and below 20 km. The natural NO_x profile was one found to give the observed vertical profile of ozone. The artifcial NO_x, added to the natural background, was based on a source of 1.8 MT (as NO_2) per year and a two-year stratospheric residence time. The percentage reduction of ozone for each assumed spread of artificial NO_x is indicated on the figure (Johnston, 1971(a), 1971(b)).

MAJOR STRATOSPHERIC RESEARCH PROGRAMS, 1972 TO PRESENT

Senate Bill S2555, "The Stratospheric Protection Act of 1971," set up a Federal program of stratospheric research that was to report to the Congress within two years (Senate Congressional Record, September 21, 1971). In the fall of 1971, Congress directed the U.S. Department of Transportation (DOT) to conduct the Climatic Impact Assessment Program (CIAP), to be completed by the end of 1974. Highlights of this productive research program follow.

(1) Observations (many for the first time) of stratospheric profiles of N_2O, NO, NO_2, HNO_3 and O confirmed Crutzen's 1970 postulate and established the magnitude of the natural rate of production of NO_x in the stratosphere. Stratospheric measurements were carried out by ground-based methods, aircraft, balloons, and NASA satellites.

(2) Through support of laboratory measurements of chemical and photochemical reaction rates, a critical data base was prepared for use by atmospheric modelers.

(3) Photochemical "smog" reactions were incorporated in stratospheric ozone considerations (Figure 2). This produces the cross over at 13 km between ozone destruction by NO_x and ozone formation via the methane-NO_x smog reactions (Johnston and Quitevis, 1975; CIAP Monograph 1, 1975a, pp 5-98 5-107).

(4) Following development of theories of motions and transport in the stratosphere, photochemical, time-dependent models of the stratosphere were formulated.

(5) The climatic impact of stratospheric particulates produced by sulfur in jet engine fuels was examined.

(6) Environmental and economic impacts of ozone reduction and of climate change were considered.

(7) In 1973 it was found that ozone is destroyed by a catalytic system involving Cl and ClO that is similar to the catalytic system involving nitrogen oxides.

(8) The possibility of heterogeneous processes on stratospheric aerosols was examined (CIAP Monograph 1, 1975(a); CIAP Monograph 3, 1975(c)). However, the importance of heterogeneous processes acting on polar stratospheric clouds in both the Antarctic and Arctic stratospheres was not considered at this time.

(9) A combined three-dimensional and two-dimensional model study showed that NO_x from stratospheric aircraft would reduce ozone (e.g., Figure 3 from Cunnold et al., 1975, 1977). This study indicated that for an emission index of 15 g NO/kg fuel, and 7.7×10^{10} kg fuel/year, nitrogen oxides injected in a narrow corridor in the northern hemisphere reduced global average column ozone by 12% with a worst case local ozone column reduction of 25% near the flight corridor.

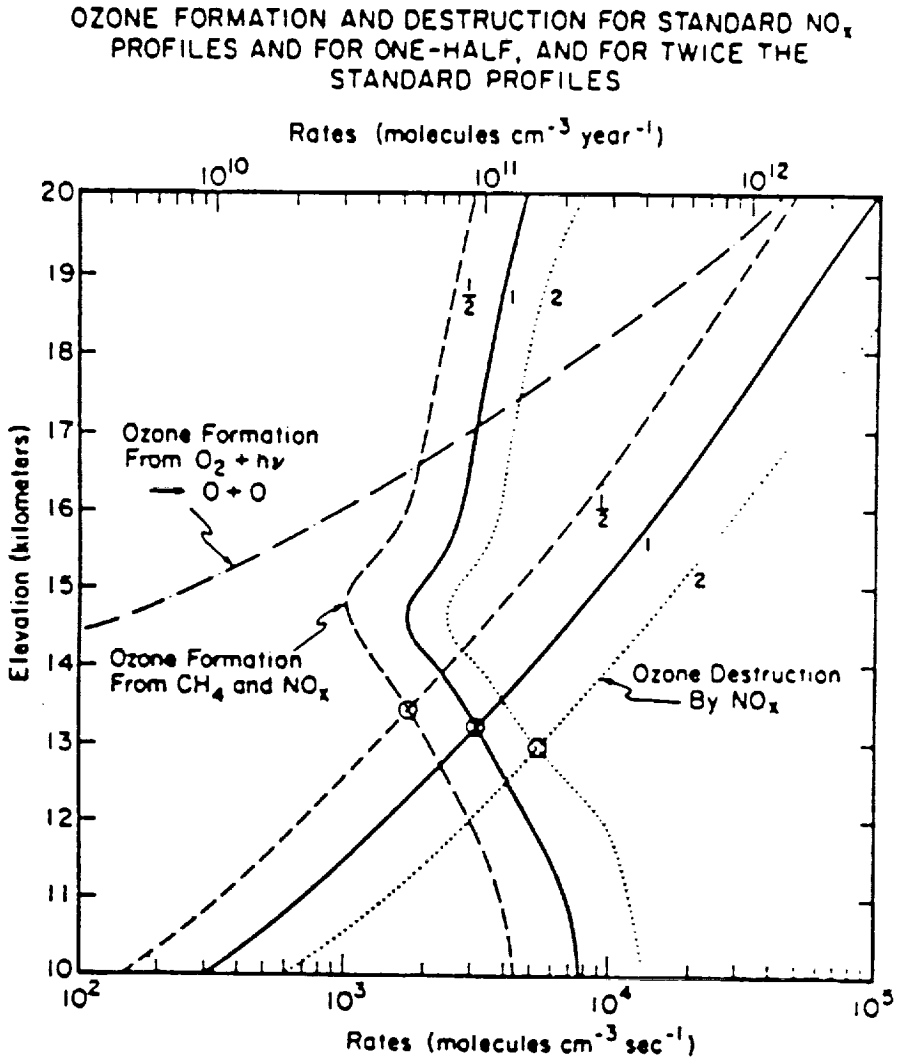

Figure 2. The rate of ozone formation from the methane-smog reactions equals the rate of destruction from the NOx catalytic cycle at about 13 km. For half and for double the assumed natural background of NOx, the cross-over point is essentially unchanged (Johnston and Quitevis, 1975).

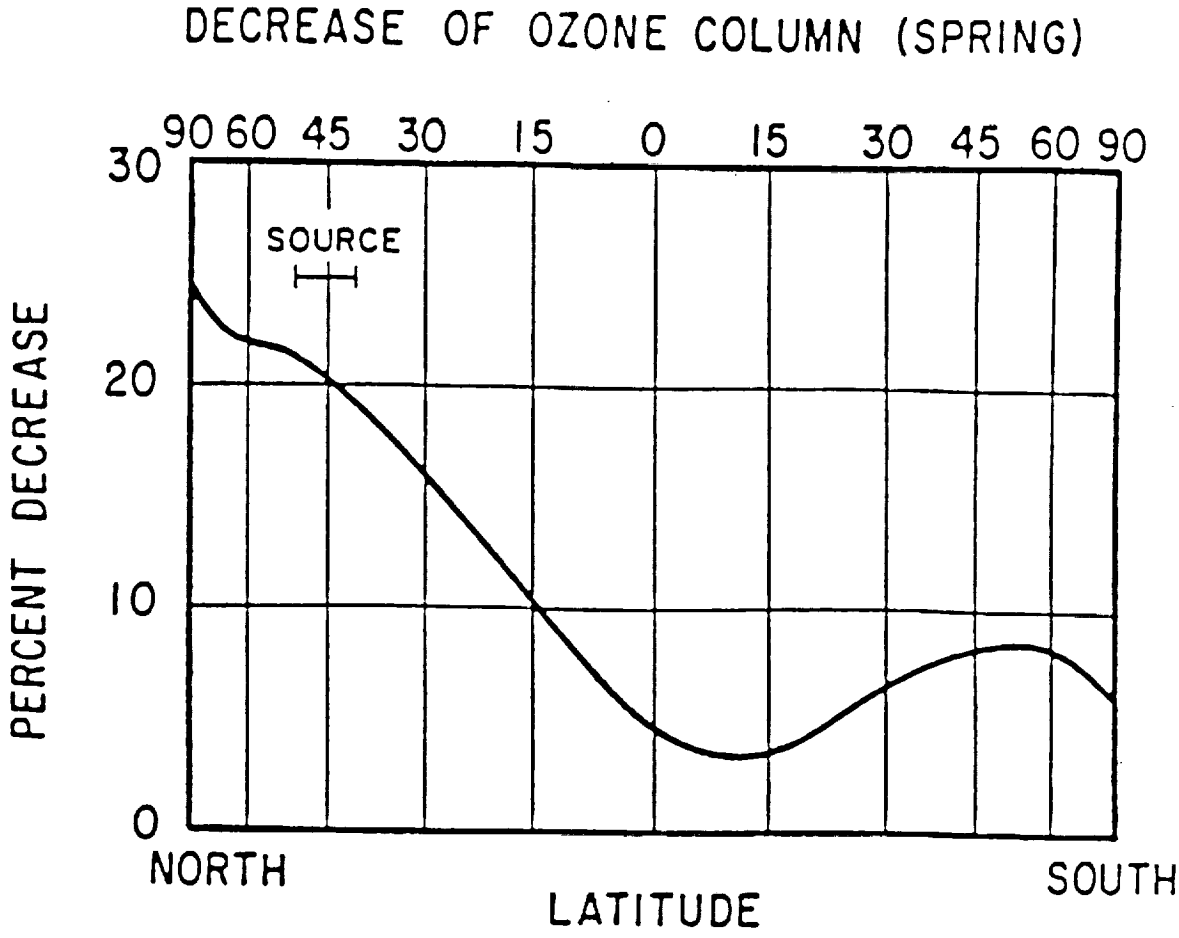

Figure 3. For 1.8 MT NO_2/yr (77 x 10^9 kg fuel/year, emission index 15), injected over a 10 degree corridor in the northern hemisphere, the local maximum decrease in ozone column is 25% near the north pole. The global average ozone decrease for this perturbation is 12%, based on a three-dimensional model of atmospheric motions and a two-dimensional model of photochemistry (redrawn from Cunnold et al., 1975, 1977).

(10) CIAP recommended the redesign of aircraft engines to achieve a 60-fold reduction in the amount of exhaust NO_x in order to protect stratospheric ozone.

The stratospheric research program was transferred to NASA in 1976; NASA brought other agencies and international organizations into the program. High quality scientific research has been carried out on a large scale since 1976, including laboratory studies, preparation of critical data bases, atmospheric measurements using aircraft, balloons, and satellites, studies of the theory of atmospheric motions and development of dynamical-radiative-photochemical models. The analysis of the effect of chlorofluorocarbons on stratospheric ozone (Rowland and Molina, 1974, 1975) led to the emphasis of the NASA program on the effects of chlorine. Selected results of the NASA stratospheric program follow.

1. The postulate of Rowland and Molina that stratospheric chlorine species would be increased by photolysis of fluorocarbons, was confirmed in the laboratory, through theoretical models and through observed vertical profiles of many chlorine containing species in the troposphere and stratosphere.

2. Approximate balance of the global ozone budget was achieved using satellite data and two-dimensional models. The vertical profiles of processes now recognized as important in destroying ozone, as calculated by the Lawrence Livermore National Laboratory (LLNL) one dimensional model, are given in Figure 4. For the region of ozone formation in the middle stratosphere and in the region of maximum ozone mixing ratio in the lower-middle stratosphere, the NO_x catalytic cycle is the most important mode of natural ozone destruction. The O_x, HO_x, Cl_x and NO_x cycles are all important in the upper stratosphere; HO_x reactions are most important in the mesosphere and troposphere.

3. An expedition in 1987 unambiguously ascribed the chemical component of the Antarctic ozone hole to stratospheric chlorine species, to heterogeneous reactions on stratospheric cloud particles, and to reactions involving the ClO dimer (Molina and Molina, 1987). The heterogeneous reactions convert NO_x into nitric acid and release active chlorine from its inactive forms, hydrogen chloride and chlorine nitrate.

Although the NASA program has not been directly concerned with the effect of stratospheric aviation on ozone, the effect of standard NO_x injections though the years of maturing of stratospheric science has been calculated using the LLNL one-dimensional model. Figure 5 shows the calculated future, steady-state ozone column changes for two separate perturbations: one, the continuous usage of chlorofluorocarbons at the 1974 rate; and two, for an assumed continuous world-wide injection of 2×10^8 NO_x molecules $cm^{-2} sec^{-1}$ (7.7×10^{10} kg fuel/year, emission index 20 g NO/kg fuel). For these constant perturbations, the calculated ozone changes have varied with the date of the calculation, which used the current best photochemical, diffusion and boundary conditions. The variations of the results between 1976 and 1981 were largely caused by new laboratory measurements of rate coefficients of the HO_x family (OH, HO_2, H) of reactions (e.g., the rate of $NO+HO_2$) and the discovery of new species to include in the models (e.g., $ClONO_2$). The ozone column was calculated to increase as a result of the NO_x injection between 1978-1980. The ozone reduction by NO_x in the middle stratosphere was counteracted by

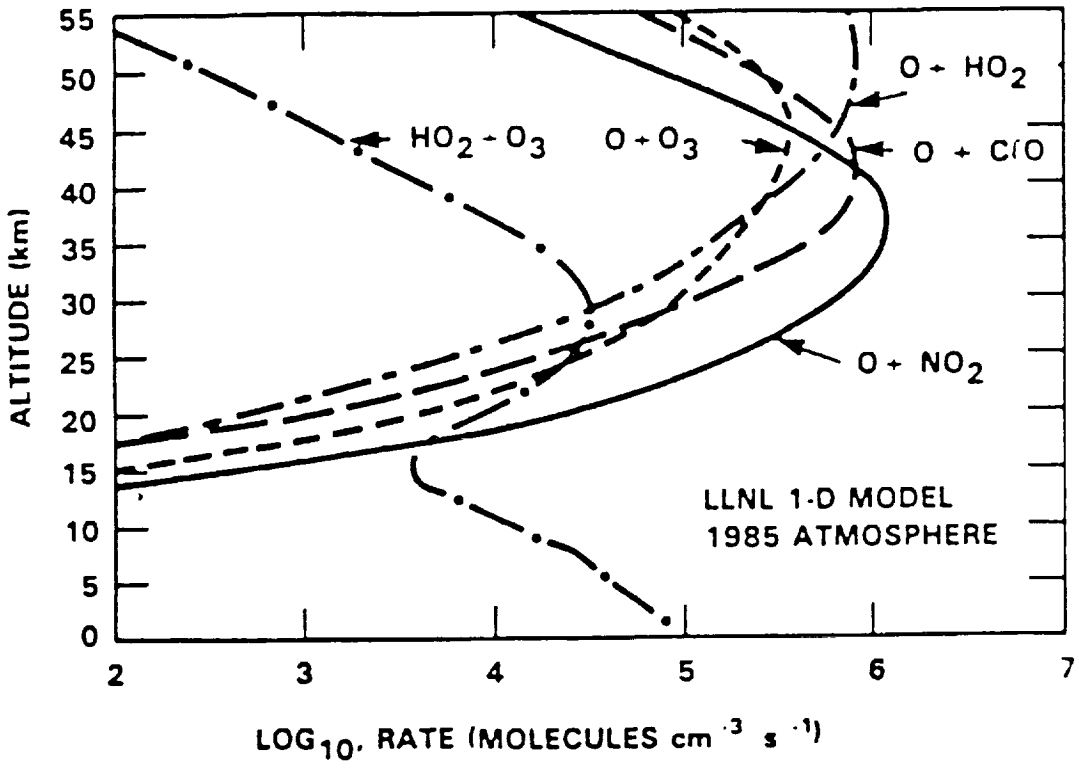

Figure 4. The ozone destruction from major catalytic cycles as a function of altitude using the LLNL 1-D model with 1985 photochemical data (D.J. Wuebbles, private communication, 1989).

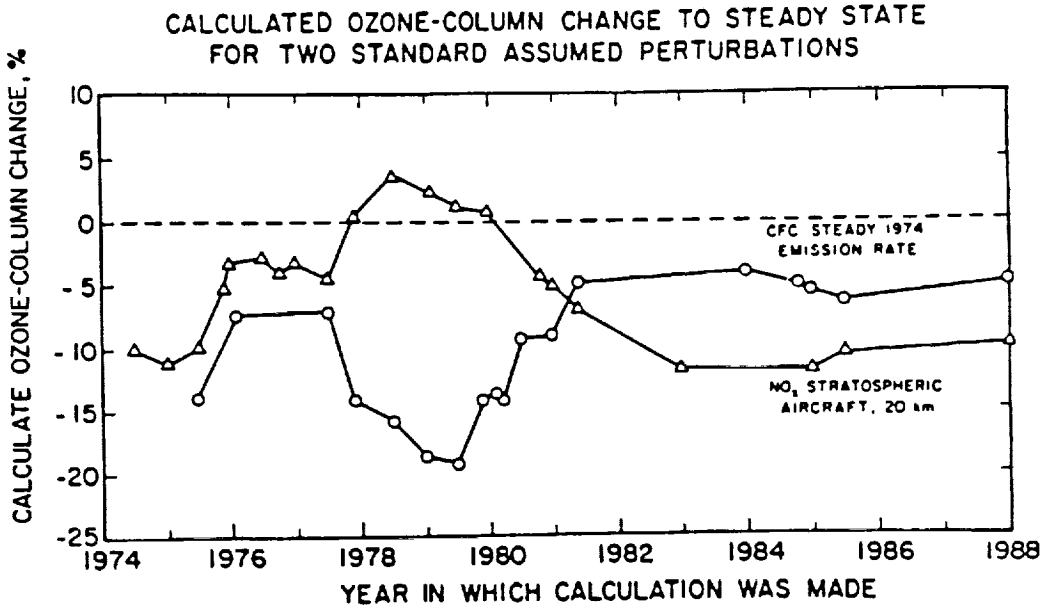

Figure 5. The calculated ozone column changes at steady state are given for two standard assumed perturbations: (a) 2000 molecules cm(-3)s(-1) over a 1 km interval centered at 20 km; (b) CFC-11 and CFC-12 emitted continuously at 1974 rate. These calculations were made using the LLNL 1-D model each year during this period using current photochemical parameters, eddy diffusion functions, and boundary conditions (WMO, 1986, p 773, updated to 1988).

ozone production via methane-NO_x smog reactions in the upper troposphere and lower stratosphere. Using the most current values for HO_x reaction rates, the crossover point of Figure 2 returns to the 12 to 14 kilometer range, and NO_x injected at 20 kilometers has been calculated to reduce the ozone column since 1980. The ozone reduction calculated for this aircraft emission is greater than that calculated for the continued use of CFC's at 1974 levels (excluding the effects of heterogeneous processes on polar stratospheric clouds). There have been no laboratory measurements of reaction rates or cross sections that would change the relative impact of aircraft emissions and the continued use of CFC's at 1974 levels since 1981.

CURRENTLY PROPOSED STRATOSPHERIC AIRCRAFT

1. Model Inputs and Sensitivities

Recent model calculations of the impact of stratospheric aircraft are discussed in the second half of this section. However, the problem of obtaining a quantitative estimate of the environmental impact of a fleet of commercial supersonic aircraft is multi-faceted. Reasonable estimates of the total amount of NO_x injected per year, the latitude and altitude of injection may be obtained. Although there are important small scale physical and chemical processes which are not resolved, it is possible to calculate the global impact of aircraft exhaust on stratospheric ozone using current models which contain parameterizations of these processes. A comprehensive study of the impact of changes in the level of NO_x emissions, the altitude and latitude of injection, and levels of background gases has been reported by Johnston et al. (1989). Their principal findings are reported in the modeling section, along with the results of more limited studies by Ko et al. (1989) and Isaksen et al. (1989). Following is a brief discussion of the above and other topics which are considered in performing model studies. Some suggestions of needed improvements are noted.

a. Number of aircraft, amount of fuel burned, plume composition, and emission index

It must be presumed that several hundred stratospheric aircraft will be in use, because the cost of developing and building a new kind of commercial aircraft is prohibitive unless it is expected that several hundred copies of the aircraft will be bought and used (Ott, 1988; Boeing, 1989). The amount of fuel burned per aircraft depends upon the size and type of aircraft and the Mach number.

The principal components of the aircraft plume are expected to be H_2O, CO_2, CO, unburnt hydrocarbons, NO, NO_2, SO_2, soot, lubricating oil, and trace metals. Emission rates similar to those described by English (1974) are assumed. The SO_2 emission arises from combustion of sulfur-containing species in the fuel; if necessary the fuel can be desulfurized to very low levels at an acceptable cost.

The emission index (EI, grams of NO_x produced per kilogram of fuel consumed) is critical to this problem. By drastic redesign of jet engines, it is theoretically possible to have high performance and low EI (Boeing, 1989, pp 83-85). NASA engineers are optimistic that the index may be reduced by a factor of 8 to 10 below the value of 40 to 50 assigned to current conventional combustors by advanced emission reduction technology. They have the goal of an emission index of 5 for an actual engine by the year 1997 (Niedzwiecki, 1988). The successful development of an engine with an emission index as low as 5 is not guaranteed. According to the Boeing 1989 report (pp 41-42), such a reduction in the EI "...will require a significant commitment to an aggressive research effort..." Furthermore, the safety of such an engine must also be considered. "GE...reported ...that it would be a high-risk development program with potential barriers to success being premixing duct flashback and auto-ignition."

In 1974, the Concorde was reported to have an EI value of 12 (CIAP, 1974, 1975), although the EI assigned to current jet engines is between 40 and 50. In 1988 some at NASA spoke of a reduction of the EI by a factor of 100 below this value as "theoretically possible" but involving very high risk (Niedzwiecki, 1988).

For model calculations, emissions may be reported as a statement of the annual rate of fuel consumption at cruise altitudes and the engine manufacturers' EI grams(NO)/kg(fuel). This specifies the number of molecules of NO emitted per year. Until actual values from real engines are demonstrated, ozone reduction calculations should be made with a range of EI values: 45, 15 and 5 which are the conventional combustor value, the standard value used during CIAP (regarded as achievable since it once existed), and the goal of theoretical models for advanced emission reduction technology. Calculated ozone changes are larger for larger emissions of NO_x as considered in the modeling section. A full range of both EI values and annual fuel consumption must be considered.

b. Cruise altitude

Because the effect of transport and because different chemical processes dominate at different altitudes, the impact of aircraft NO_x depends upon the altitude of injection. In the upper troposphere, methane NO_x smog reactions produce ozone, whereas in the lower stratosphere ozone is destroyed by the NO_x emissions. Thus the calculated change in ozone for aircraft flying just above the tropopause (12 km at middle latitudes) is much smaller than that calculated for aircraft flying at higher altitudes. In the upper troposphere, ozone is produced via methane-NO_x smog reactions; in the lower stratosphere, ozone is destroyed by the NO_x (Johnston and Quitevis, 1975; CIAP Monograph 1, 1975a pp 5-98 5-107). These processes approximately balance at 13 km (Figure 2). Although Boeing has tentatively identified the Mach 2.4 aircraft flying at 20 km as its favored model, and Douglas has selected a Mach 3.2 system, there are still proposals for hypersonic transport with Mach numbers as high as 5 (San Francisco Chronical, Cracking Japan's Aircraft Market", December 4, 1989, p c2). Depending on the actual Mach number, such craft might fly in the altitude range of 18 to 35 km (see introduction). Because the net change in ozone column calculated from models is sensitive to the altitude of injection, model calculations should explore the full range of possible cruise altitudes.

Table 4 in the modeling section presents results for the LLNL model when the altitude of injection is varied from 16.5 to 34.5 km in 3 km intervals. The global O_3 depletion for injection at 19.5 km is more than 10 times the calculated depletion for injection at 16.5 km. Such extreme sensitivity indicates the importance of transport and mixing at these altitudes. A high priority of the new NASA atmospheric research program should be to develop models with better representations of stratosphere/troposphere exchange and horizontal mixing in the lower stratosphere. In the likely event that such representations require increased vertical resolution, it will also be necessary to increase horizontal resolution to enable a consistent representations of the transport processes. The model dispersion and transport in the lower stratosphere should be calibrated with respect to the altitude of sharp transition between the stratosphere and troposphere in the 12 to 21 kilometer region against measurements of stratospheric tracers.

These include ozone and NO_x measurements, as well as measurements of radioisotopes carbon-14 and strontium-90 (Johnston, 1989).

The altitude of the aircraft is also important because the temperature at cruise altitude and separation from the tropopause affect the behavior of the plume with regard to the possibility of ice or nitric acid trihydrate formation and plume dispersion.

c. Flight corridor

Flight corridors are largely determined by population and market centers. Sensitivity of model calculation to the latitude of injection is expected to parallel the model sensitivity of the stratospheric lifetime of the pollutant. For example, injection at tropical latitudes above cloud tops, where the global circulation is upward and poleward, is expected to be more detrimental to ozone than injection at higher latitudes, where the return to the troposphere will be quicker. Such a sensitivity study is given for the LLNL model in the modeling section (Table 5).

d. Stratosphere/Troposphere exchange

The temporal and spatial distributions of stratosphere/troposphere exchange, and transport and mixing in the lower stratosphere, are particularly sensitive issues for aircraft corridors in the 12-20 km altitude range in midlatitudes. This sensitivity is closely related to the large change in tropopause height that coincides with the core of the subtropical jetstream. Below about 15-16 km isentropic surfaces lie in the troposphere at low latitudes and in the stratosphere at high latitudes. Thus, in this altitude range quasi-adiabatic motions may rapidly transfer material between the troposphere and the stratosphere. Such transfer will be strongly dependent on season since the meteorological disturbances that produce rapid meridional transport are seasonally varying. Pollutants emitted by aircraft in this region should become diluted and mixed into the troposphere rather easily.

Above 16 km, isentropic surfaces lie entirely in the stratosphere. Diabatic cooling is required for transport from such levels into the troposphere. It is therefore reasonable to assume that the exchange time will depend on the distance above 16 km and the radiative cooling rates, which in turn depend on the rate at which eddy processes drive the motion away from radiative balance. Thus, the turnover times should depend very sensitively on the cruise altitude of the proposed aircraft. During summer, when large scale eddy motions are weak or nearly absent in the region above 16 km, the time scale for mixing and transport is long compared to that in winter when large scale eddies propagate into the stratosphere.

At present, the possible role of gravity waves in contributing to the dynamical driving of the mean circulation in the lower stratosphere, as well as in directly producing mixing through turbulent breakdown, is not well understood. In addition, it is not clear whether stratosphere-troposphere exchange through sporadic tropopause folds, can be adequately modeled in terms of the mean circulation and mean species gradients as must be done in two-dimensional models. Both observational and theoretical work are needed to elucidate this question.

e. Background chlorine, methane, nitrous oxide, water, carbon dioxide

In addition to the direct perturbations to the stratospheric composition through the aircraft exhaust (NO_x, CO, CH_4, and H_2O), changes are occurring in the stratospheric background gases which may impact on these calculations. The background chlorine is expected to continue to increase for at least the next few decades, even with controls on the production of fluorocarbons CF_2Cl_2 and CF_3Cl, and these increases will lead to increased ozone destruction. There are significant measured trends in N_2O and CH_4 (WMO report no. 16, 1986: WMO report no. 18, 1990). Trends in N_2O and CH_4 tend to mitigate the increases in Cl_x through formation of chlorine resevoir species. Increases in N_2O leads to increases in odd nitrogen formation to increases in the resevoir $ClONO_2$; increases in CH_4 lead to increases in HCl. These changes in the background stratospheric composition must be considered in evaluating the future impact of aircraft emissions. Results for the LLNL model are presented in Figure 12 and Table 7 in the modeling section; results for the AER model are given in Figure 13 also in the modeling section.

f. Plume dispersion

The process by which the engine exhaust undergoes the transition from the initial high temperature, high velocity jet to a later state of thermal and translational equilibrium is a complex problem in fluid dynamics, and is a process which will differ depending on aircraft characteristics. Hoshizaki et al. (1972) defined the wake regime as that portion of the emission process, lasting about ten minutes, during which jet mixing shifts from control by aircraft effects to control by natural atmospheric effects. The wake regime was further divided into three parts, called the jet, vortex, and dispersion regimes. The jet portion lasts about 10 seconds, and represents the period during which the jet exhaust expands in diameter by several orders of magnitude, is mixed with the aircraft wake, and is incorporated into the trailing vortex pair produced by wing lift. Important admixing and cooling effects take place in this time period. The vortex regime lasts about 100 seconds, after which time the vortex pair has broken up due to hydrodynamic instability. It is during this period that the system undergoes the transition from aircraft-dominated motion to natural atmospheric motion. Within the dispersion period vortex effects are negligible, and further mixing and dispersion occur by wind shear, residual turbulence, and other mechanisms such as gravitational settling. Following the wake regime the plume undergoes zonal, meridional, and vertical mixing on time scales of days to years, in the manner characteristic of various atmospheric tracers (e.g., Bauer, 1972). During this time, the ice crystals that may have been formed in the plume will be evaporating (subliming), probably on a time scale of one or more hours. The exact amount of time required for plume evaporation does not appear to be well known, and may vary considerably depending on factors such as the meteorological conditions of flight altitude and latitude.

Hofmann and Rosen (1978) report on a chance observation of enhancement in the concentration of condensation nuclei at 23 km, approximately 18 hours after the passage on an SR-71 (Mach 3 strategic reconnaissance aircraft). Although this single observation provides only qualitative information concerning stratospheric diffusive processes, the behavior of this plume underscores the need for measurements under diverse meteorological conditions, at different seasonal/altitudes.

g. Chemistry of the Plume

1) Homogeneous Processes

Chemical modeling of the plume, including NO_x, HO_x and Cl_x chemistry, was reported in CIAP Monograph 3, Chapter 2 (Anderson and Meyer, 1974; Anderson et al., 1974). The modeling was checked against measurements of a jet engine in a test tunnel; a summarizing table from that monograph is reproduced here as Table 1. Similar calculations should be repeated with current values of rate constants, and the results should be checked against the engine measurements reported in CIAP Monograph 3 (McGregor et al., 1973). There may also be differences in the chemical behavior for an engine operating in a test tunnel compared with an engine in the real atmosphere.

According to Hoshizaki et al. (Chapter 2, CIAP Monograph 3, 1974), chemical-kinetic results from a reacting wake model, based on analytical results and observations of contrails formed by subsonic, tropospheric aircraft, show no environmentally significant chemical changes of odd nitrogen species in the wake regime. Some conversion of nitrogen oxides to nitric acid occurs in the jet region, but model predictions give a conservative upper bound of 10-20 percent conversion. Chemical changes of other pollutant species, CO, total hydrocarbons, and SO_2 are predicted to be small. Since the most significant plume chemistry takes place in the near jet flow field where the structure is dominated by aircraft characteristics, the results of the chemical kinetics are equally valid at all flight altitudes. At the end of the wake regime, the wake height is of the order of 100 m, and the wake width is of the order of 1 km. The concentration of the engine emission species is reduced by three or four orders of magnitude relative to the nozzle exit values.

For an assessment calculation, only the results of chemical changes and mixing to scales that are resolved by the model may be considered. This is illustrated by the behavior of the exhaust species NO. In the initial jet plume, near the engine nozzle, the NO concentration is of the order of a few hundred ppm, based on an emission index of 40 gms(NO)/kg fuel, and a fuel/air ratio of about 0.02. By the end of the wake regime, the concentration has been reduced through expansion and dilution to the sub-ppm range. Since air entrained into the jet plume during the wake mixing process contains ppm concentrations of ozone, the NO is subject to rapid conversion to NO_2 by reaction with O_3. With complete mixing of ambient (ozone-containing) air, the ozone concentration would be in excess of the NO concentration, and the time constant for this reaction would be comparable to the wake mixing time. However, in the absence of complete mixing with ambient air, the reaction may be somewhat diffusion controlled, and the time constant for this reaction would be longer. It is not completely certain just when the NO conversion to NO_2 will have reached equilibrium. During daylight hours, some reversal of this reaction will occur by photolysis of NO_2, as in the normal stratosphere, and the NO/NO_2 ratio should approach a photochemical steady state ratio.

Table 1. Aircraft Wake Regime Characteristics. This table is a copy of Table 2.1 of CIAP Monograph 3, Chapter 2, page 2-5.

JET REGIME	VORTEX REGIME	WAKE DISPERSION
	Fluid dynamics	
Flow time: 1 - 10 seconds	Flow time 10-100 seconds	Flow time 100-1000 seconds
Symmetrical jet growth followed by jet merging	Primary growth in vertical direction due to vortex sinking	Dispersion mechanisms: wind shear, wake and atmospheric turbulence gravitational collapse, buoyant rise
Size: 10-100 m	Size: 100-300 m	Size Km
Dilution ratio: 10^{-2}	Dilution ratio: 10^{-3}	Dilution ratio: 10^{-4}
	Chemical kinetics	
Flow temp T_0 ≈1000 K	Flow temp.: $T \approx T_\infty$	Flow temp.: $T \approx T_\infty$
Reactive species: H, O, OH, HO_2, HCO. Large non-equilibrium concentration of HO_x radicals observed.	$[HNO_3]_{JET} \approx [HNO_3]_{vortex}$	$[HNO_3]_{JET} \approx [HNO_3]_{dispersion}$
Important reactions $NO_2 + OH + M \rightarrow HNO_3$ $NO + HO_2 \rightarrow NO_2 + OH$ $NO_2 + O \rightarrow NO + O_2$ $NO_2 + H \rightarrow NO + OH$ Characteristic chemical times 10^{-2} to 10^{-1} sec	No significant chemistry	Half-time for NO_x/O_3 catalysis >> 1000 sec
	Conclusions	
$NO_x \rightarrow HNO_3$ conversion < 10 - 20 %	No significant chemistry	No significant chemistry Contrail critical temperature 190-200 K

Further insights into the behavior of plumes are gained by considering aircraft measurements of the atmospheric composition in clean air and in what is identified to be "pollution episodes." Recently, measurements of NO, NO_2, O_3, H_2O and NO_y (NO_y = total odd nitrogen) were made from the NASA DC-8 aircraft during December, 1988 and January-February, 1989, as part of the Airborne Arctic Stratospheric Expedition (Carroll et al., 1989; Hubler et al., 1989; Kelly et al., 1989). Mixing ratios of NO, NO_2, NO_y, O_3 and H_2O are shown as functions of universal time (SAM seconds after midnight) for observations obtained during pollution episodes in Figure 6(a) and (b). The ratio of NO_x (NO_x = NO + NO_2) to NO_y was found to be an excellent indicator of fresh pollution; such indications are not often observed in the behavior of O_3 unless the airmass has reached photochemical equilibrium. Mean tropospheric NO_x/NO_y was 0.26 in the Arctic and 0.36 near 40 N; in the Arctic stratosphere mean NO_x/NO_y was 0.03. During pollution episodes, defined by abrupt changes in NO_x (and particularly NO), the mean ratio was 0.61. In the troposphere (stratosphere), positive (negative) correlations are observed between NO_x and O_3, but no correlation is observed for measurements obtained during pollution events. The data shown in Figure 6(a) were obtained during a flight out of Moffett Field, California. Highly elevated mixing ratios of NO (and NO_y) were observed for an extended period without concurrent increases in NO_2 and O_3. In fact, O_3 mixing ratios remained stable at 25-30 ppbv, levels that are often indicative of clean air. This pollution episode lasted considerably longer than all the other events encountered, and it is speculated that measurements might have been made in a commercial flight corridor. The other pollution episodes were typically on the order of 10's - 100's of seconds, suggesting crossing another aircraft's or the DC-8's contrail (e.g., Figure 6(b)). No analysis has been performed to determine the likely extent of dilution for the instances where the aircraft encountered its own exhaust.

2) Heterogeneous Processes

If ice crystals or nitric acid trihydrate particles are formed in the aircraft plume, the effects of heterogeneous chemical reactions on particle surfaces must be considered. These include surface reactions involving exhaust effluent during and after the ice crystallization process and possible plume processing of ambient atmospheric species on the surface of ice crystals.

For flights over the polar regions, the plume lifetimes may be quite long, and it is possible that significant chemical transformations will take place on the plume ice crystals before evaporation. The following processes, on naturally occurring polar stratospheric clouds, are believed to be responsible for the formation of the Antarctic ozone hole:

$$HCl\ (\text{in ice}) + ClONO_2\ (\text{gas}) \rightarrow Cl_2\ (\text{gas}) + HNO_3\ (\text{in ice}) \quad (R1)$$

$$HCl\ (\text{in ice}) + N_2O_5\ (\text{gas}) \rightarrow ClONO\ (\text{gas}) + HNO_3\ (\text{in ice}) \quad (R2)$$

$$H_2O\ (\text{ice}) + ClONO_2\ (\text{gas}) \rightarrow HOCl\ (\text{gas}) + HNO_3\ (\text{in ice}) \quad (R3)$$

$$H_2O\ (\text{ice}) + N_2O_5\ (\text{gas}) \rightarrow 2\ HNO_3(\text{ice}) \quad (R4)$$

The principal effects are the conversion of chlorine reservoir species such as HCl and $ClONO_2$ into photochemically active forms and suppression of NO_x; the latter affects ozone destruction in two ways. Both the ozone loss process

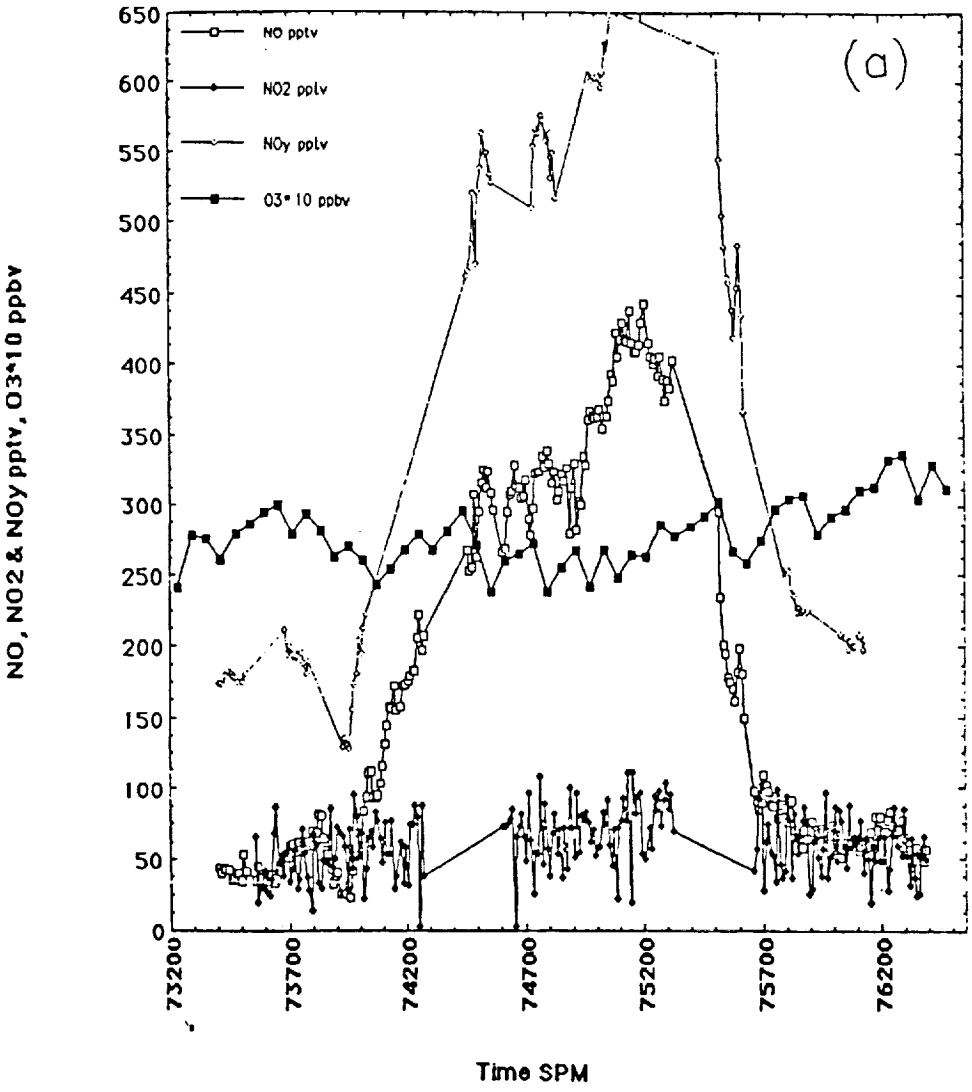

Figure 6(a) For a pollution episode encountered during a flight out of Moffett Field, California, the ratio of $(NO + NO_2)/NO_y$ jumps abruptly. The O_3 values remained relatively constant throughout the encounter. The long duration of the high values of NO and NO_y suggest that the measurements are made in a commercial flight corridor.

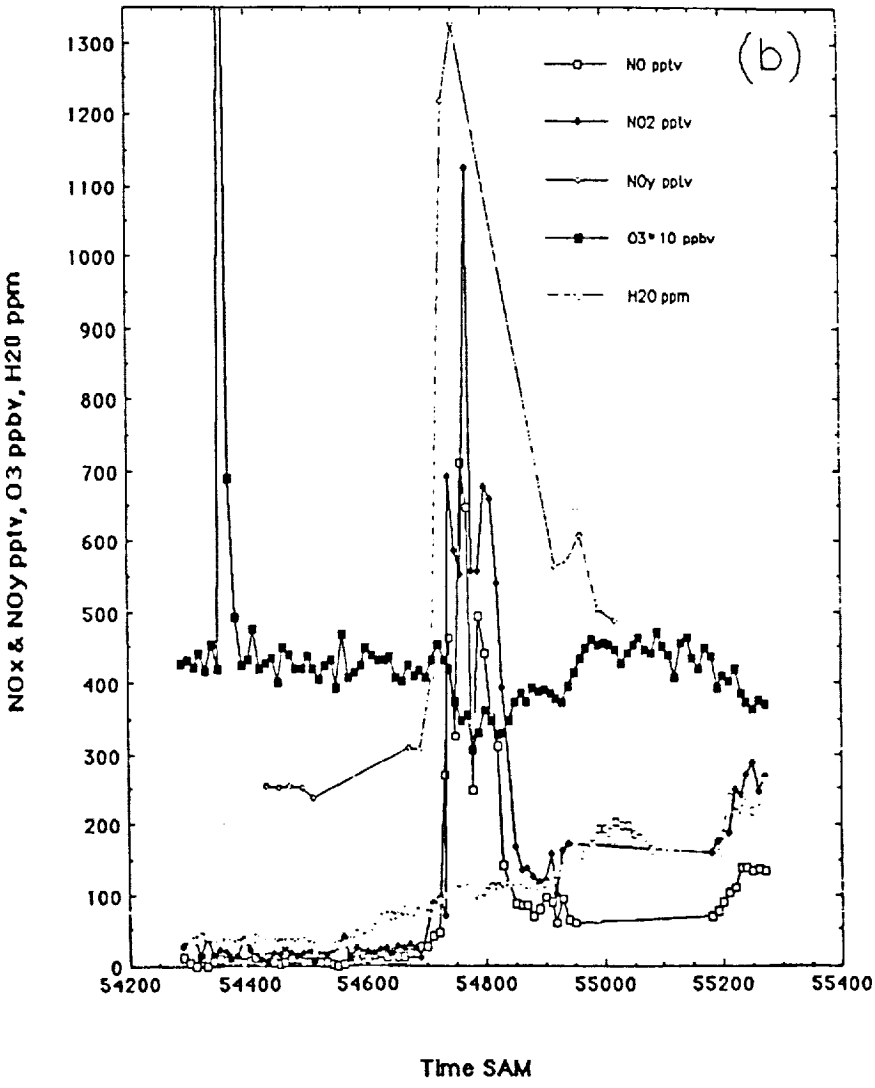

Figure 6(b); In this pollution episode, elevated levels of NO_y and NO_2 are observed for 10's to 100's of seconds, suggesting that the aircraft crossed its own or another aircraft's contrail.

involving NO_x, and NO_x interference with chlorine catalyzed ozone destruction through $ClONO_2$ formation are reduced. The HNO_3 tends to remain on the ice crystal until evaporation, whereas the products of R1, R2 and R3 (Cl_2, HOCl and ClONO) are in the gas phase and may be photolyzed. If this type of chemical process occurs in aircraft plumes, a possibly significant effect would be redistribution of chlorine from inactive to active forms. The magnitude of the effect would depend on several factors, such as the lifetime of the plume ice crystals and the collision efficiency of the surface processes. The natural HCl conversion to the active form, by reaction with OH

$$OH + HCl \rightarrow H_2O + Cl \qquad (R5)$$

requires about one month. The plume process could compete with the natural process in corridors of high flight activity, changing the balance of chlorine species, and depending upon the concentration of NO_x, possibly resulting in localized ozone reduction. On the other hand, if large heavy particles of nitric acid trihydrate are formed by the plume and fall out of the stratosphere, the nitrogen oxides from the aircraft may be removed. To date there has been no effort to include the effects of heterogeneous processes in aircraft plumes in two-dimensional models.

h. Aerosol Effects

The quantity of SO_2 emitted by a fleet of aircraft using non-desulfurized fuel, based on emission budgets such as those given by English (1974), would equal or exceed the estimated source of stratospheric SO_2 from oxidation of carbonyl sulfide (Crutzen, 1976). The total emission for one year is about one or two orders of magnitude less than the estimated SO_2 injection from the El Chichon volcano. Turco et al. (1980) have used a particle-gas model to study the effect, primarily on climate, of both SO_2 and soot from a hypothetical fleet of supersonic aircraft, and have concluded that global surface temperatures would be negligibly affected. The large particle concentration was predicted to increase by approximately 20%, however, and the possibility of enhanced surface catalysis must be considered, particularly in areas of dense aircraft traffic.

Based on their observation of the plume of an SR-71 (Mach 3 strategic reconnaissance aircraft), Hofmann and Rosen (1978) suggest that a fleet of stratospheric aircraft would produce about the same concentration of condensation nuclei as the background of 10-15 cm^{-3}. Such a large increase in the concentration ocould also have an impact on larger particles which are formed through coagulation and gas-particle interactions.

2. Recent Model Calculations

The impact of aircraft exhaust on ozone has been evaluated by three groups using one and two-dimensional models of the stratosphere. In addition, several of the sensitivities which are important to this calculation as outlined above have been considered. These include sensitivity to the altitude of injection, latitude of injection, and background concentrations of such species as CH_4, CO_2, N_2O, and Cl_x.

a. Response of O_3 to NO_x

Model studies for various NO_x emission scenarios have been carried out for the preliminary High Speed Civil Transport program of NASA. Johnston et al. (1989) have used the LLNL one-dimensional and two-dimensional models to examine a wide range of NO_x injections, and also examined the sensitivity of the calculated ozone response to altitude of injection, stratospheric chlorine background, and latitude of injection. A much more limited range of scenarios has been considered by two other groups; in general results corroborate the results of Johnston et al. (1989). Ko et al. (1989) have examined Boeing and McDonnell-Douglas scenarios for emissions of NO_x, CO, CH_4 and H_2O using the AER two-dimensional model. Isaksen et al. (1989) have examined scenarios for NOx emissions using two different two-dimensional models: a) a tropospheric model with detailed tropospheric chemistry; and b) the OSLO stratospheric model, which includes simplified tropospheric chemistry. These two-dimensional models are all described in the Two-Dimensional Intercomparison of Stratospheric Models (Jackman et al., 1989), and are identified here by the same acronyms used in that document. The two-dimensional cases examined by the various groups are summarized for comparison in Table 2.

The three models show similar responses for ozone to NO_x emissions. Figures 7, 8, and 9 show the change in ozone as a function of latitude and altitude and the change in column ozone as a function of latitude and time for the LLNL, AER and OSLO models respectively. The cases considered are starred in Table 2. For all three models, the largest ozone depletions are in the

Table 2: Scenarios examined using two-dimensional models; ozone depletions for cases marked by * are given in Figures 7, 8, and 9. For AER and OSLO calculations the NO_x is injected over a distributions of altitudes, and additional injection from the subsonic fleet in included. For LLNL NO_x is injected at one model level, and the altitude range indicates the model vertical resolution.

group		NO 10^{33} molecules/year	10^9 kg fuel/year	EI	alt of injection	global O_3 depletion
LLNL		63	77	40	22.5±1.5 km	-19.1
*		24	77	15	22.5±1.5 km 19.5±1.5 km 16.5±1.5 km	- 8.6 - 7.6 - 0.7
		8	77	5	19.5±1.5 km	- 2.8
AER	B7	6.3	62.5	5	18 - 22 km	- 0.8
	B8	12.8	70.6	9	18 - 22 km	- 1.7
	B10	6.3	62.5	5	18 - 22 km	- 0.8
*	A3	18	22.4	39.5	18 - 22 km	- 5.7
	A4	6	22.3	12.1	18 - 22 km	- 1.4
	A5	3	28.9	5.2	18 - 22 km	- 0.7
OSLO	B7	6.3	62.5	5	19 km	
*	B7	6.3	62.5	5	21 km	
	B7	6.3	62.5	5	25 km	

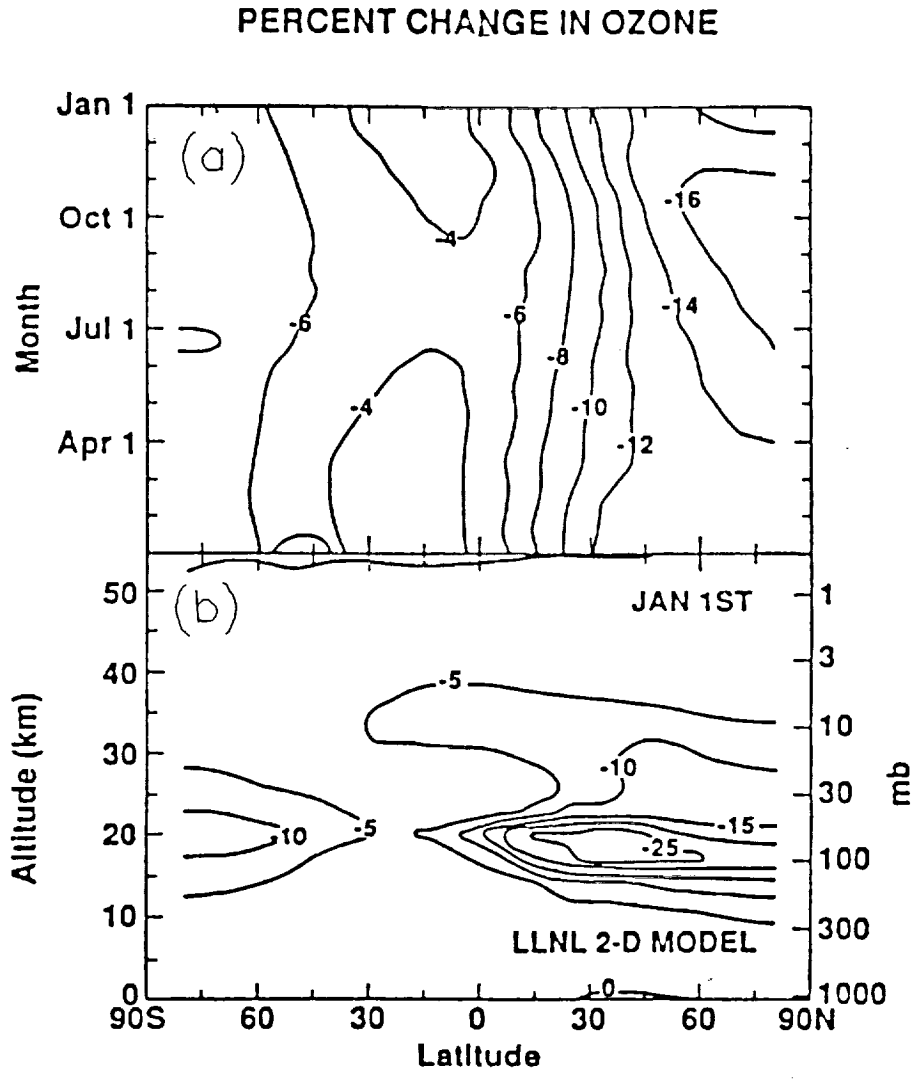

Figure 7. The O_3 change for injection of $24x10^{33}$ molecules NO/year (77 x 10^9 kg fuel/year, emission index 15) at 19 km using the LLNL model. (a) local change in O_3 as a function of latitude and height. largest changes appear in the northern hemisphere lower stratosphere. (b) total O_3 change as a function of latitude and month. The largest O_3 changes appear in the northern hemisphere winter at high latitude.

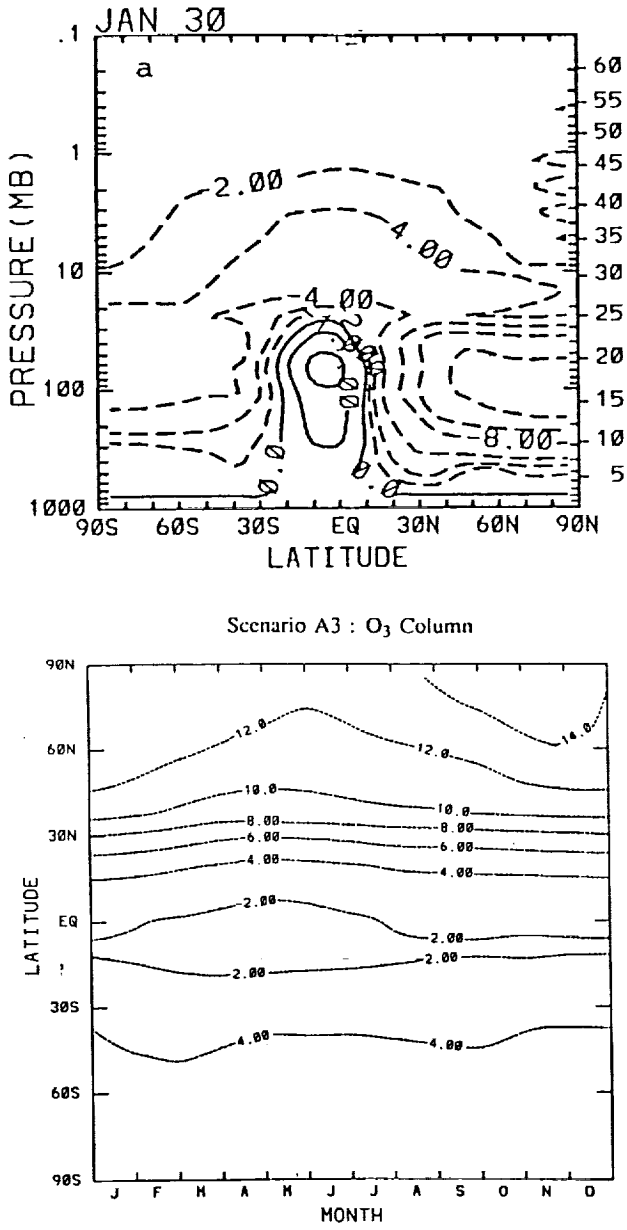

Figure 8. The O_3 reduction for 18 x 10^{33} molecules NO_x/year (22.4 x 10^9 kg fuel/year, emission index 39.5) at 22 km using the AER model. (a) local O_3 change as a function of latitude and altitude for January (contour intervals are 0, 1, 2, 4, 8, 12, 16%). The largest changes appear in the northern hemisphere lower stratosphere. (b) change in total O_3 as a function of latitude and month. The largest O_3 changes appear in the northern hemisphere winter at high latitudes.

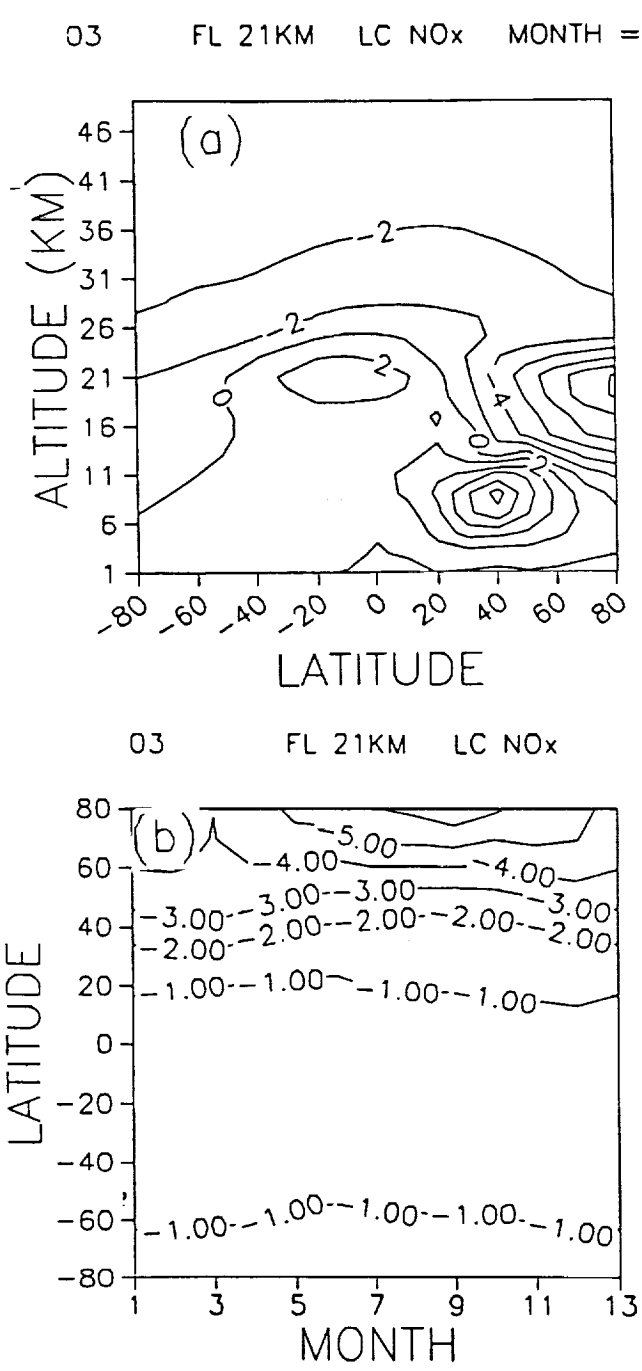

Figure 9. The O_3 reduction for 6.3 x 10^{33} molecules NO_x/year (62.5 x 10^9 kg fuel/year, emission index 15) at 21 km using the OSLO model. (a) local O_3 change as a function of latitude and latitude for June. The largest changes appear in the northern hemisphere; these are partially compensated by the O_3 production in the upper troposphere. (b) total O_3 reductions as a function of latitude and month. The largest loss of total O_3 appears in the northern hemisphere winter at high altitude.

northern hemisphere lower stratosphere, near the altitude and latitude of maximum NO_x injection. The largest differences appear in the troposphere. Both AER and OSLO show an ozone increase in the tropics. The large positive region in the northern hemisphere troposphere in the OSLO calculation results from smog production of ozone due to NO_x from a subsonic fleet. The largest total ozone depletions occur in the fall at high northern latitudes. Model transport leads to substantial local ozone changes in the southern hemisphere, but the local column changes are not nearly as large as in the northern hemisphere. Assuming that the ozone changes for these models will respond approximately linearly to changes in the level of NO_x injection, it is possible to compare the model results. The NO_x injected in the LLNL case is 1.33 times the NO_x injected in the AER case, and 3.8 times the NO_x injected in the OSLO case (note that with such a large scaling factor, there is more uncertainty in the scaled OSLO values). The scaled ozone reductions are compared to the LLNL reductions in Table 3. The three cases are in reasonable agreement. The largest calculated ozone reduction for Mach 2.4 aircraft (altitude 20 km) is considered to be the fuel usage for AER Scenario B8, with the emission index of 39.5 AER Scenario A3. This is somewhat more than double the NOx emissions of the LLNL case shown in Figure 7, and corresponds to a global ozone reduction of 16%, which is a value greater than worst case scenarios for chlorofluorocarbons. For the lowest value of the emission index considered (EI = 5) the global calculated ozone depletion is 2-3%, depending upon the cruise altitude and the actual fuel consumed.

Table 3 Comparison of calculated ozone column changes by three groups using two-dimensional models; the results are scaled to a fuel usage of 77×10^9 kg fuel/year and an emission index of 15.

	North polar	30 N	30 S
LLNL	-16%	-11%	-5%
AER x 1.33	-19%	-11%	-4%
OSLO x 3.8	-22%	- 8%	-4%

b. Sensitivity to altitude of injection

All of the studies have shown that the calculated ozone depletion is sensitive to the aircraft cruise altitude. Calculations made by Johnston et al. (1989) using the LLNL two-dimensional model are summarized in Table 4. The calculated global ozone depletion increases dramatically for injection at

Experiments using the AER model showed that emissions confined to the upper troposphere (maximum altitude of emission 14 km) had a relatively small impact on ozone, which is in general agreement with previous one-dimensional results. Ozone depletions in the middle and upper stratosphere were partially compensated by ozone increases through methane-NO_x smog reactions in the upper troposphere.

Similar results are obtained using the OSLO model. The total ozone response for the same total NO_x injection as Figure 9, but for a lower (19 km) altitude of injection, is given in Figure 10. The decrease of O3 in the lower stratosphere is smaller for the 19 km injection than for the 21 km injection, and there is also a larger increase in the upper troposphere due to increased O3 production.

Table 4 Percent change in total O3 calculated with the LLNL two-dimensional model. For all cases, the NO_x injection is 1.8MT NO_2/yr at 37^o-49^o N latitude; only the altitude of injection is varied. This corresponds to a fuel usage of 77 x 10^9 kg fuel/year and an emission index of 15.

altitude	percentage change in total O_3		
	global	Northern hemisphere	Southern hemisphere
(km)			
16.5	-0.7	-0.9	-0.4
19.5	-7.6	-10.4	-5.9
22.5	-8.6	-13.1	-3.9
25.5	-9.6	-15.6	-3.5
28.5	-10.1	-16.4	-3.5
31.5	-9.8	-16.0	-3.4
34.5	-9.2	-14.9	-3.2

c. Sensitivity to latitude of injection

Results described previously are sensitive to the various transport circulations, which largely determine the impact of emissions at various latitudes and altitudes. Emissions in the tropical regions are carried upward and poleward by the winds, dispersing globally. Emissions in the mid-and high-latitudes are carried downward towards the troposphere. NO_x which reaches the troposphere is removed rapidly by rainout, minimizing the impact below 8 km. The LLNL two-dimensional model was used to examine the sensitivity of the ozone depletion to the latitude of the NO_x injection. The global average ozone reduction was almost the same regardless of whether the injection was globally uniform or confined to the narrow latitudinal band (37^o-49^o) in the northern hemisphere. In contrast, when NO_x was injected in the equatorial region, between 0^o and 12^o N, at 22.5 km, the global average ozone reduction was substantially greater. This difference is due to tropical air rising in the equatorial stratosphere as a part of the global circulation, and spreading the injected NOx over the stratosphere of the northern and southern hemispheres. These results are summarized in Table 5.

Table 5 Percent change in total O_3 calculated with the LLNL two-dimensional model. For all cases the NO_x injection is 1.8 MT NO_2/yr at 22.5 km. The latitude of injection is varied. This corresponds to a fuel usage of 77 x 10^9 kg fuel/year and an emission index of 15.

Latitude	Percentage change in Total O_3 Global	Northern Hemisphere	Southern Hemisphere
37-49	-8.6	-13.1	-3.9
0-12	-11	-12.5	-9.5
Globally uniform	-9.8	-9.5	-10.2

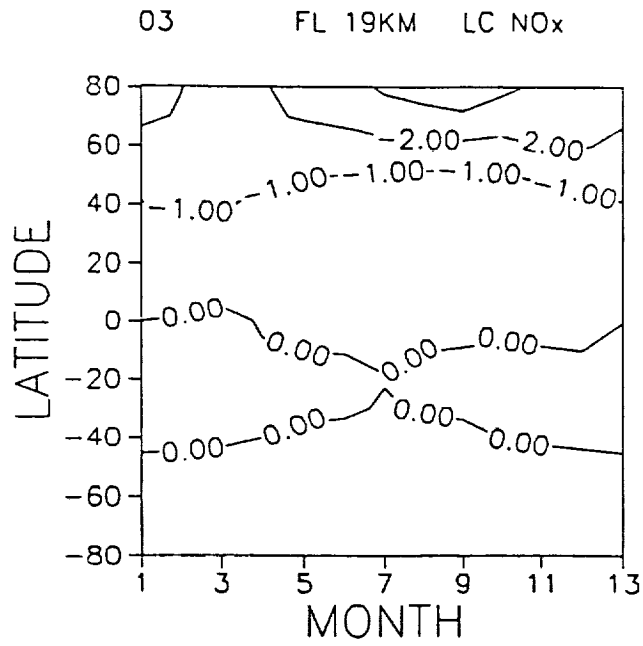

Figure 10. Same as Figure 8(b) but for a 19 km altitude of injection.

d. Sensitivity to other stratospheric gases

The calculated changes in ozone due to NOx increases must be considered within the context of two types of changes of other stratospheric constituents. The first is the impact of the direct injection of the other exhaust components in the stratosphere. The second is the changes in background gases (Cl_x, CH_4, N_2O, CO_2) that are occurring independent of stratospheric aircraft. Interference reactions among the species make it necessary to consider the net impact of all the changes.

The AER model was used to compare the impact of emissions of NO_x, CH_4, CO, and H_2O on ozone (Figure 11(a)) with the impact of NO_x alone (Figure 11(b). The O_3 response is clearly dominated by the NO_x emissions. The impact of changes in H_2O on photochemistry was considered separately, using the LLNL one-dimensional model; results are summarized in Table 6. Direct injection of H_2O at 20 km in the absence of an NO_x injection results in a small decrease in the ozone column (-0.09%). When combined with NO_x emissions, injecting H_2O reduced the calculated ozone depletion. For high levels of NO_x emission, there is very little sensitivity to injected H_2O. For various levels of NO_x emissions, the change in column ozone is decreased by 0.5 to 1% by considering the H_2O increase.

Table 6 Percent change of total 03 calculated with the LLNL one-dimensional model for various injection rates of NO at 20 km. Water injected equivalent to burning 77×10^9 kg fuel/year.

Amount of NO (10^{33} molecules/yr)	Percentage Change In O_3 H2O	NO	both	ratio
0	-0.9	0.0	- 0.9	0.
8		-3.1	- 2.66	0.88
24		-9.94	- 9.06	0.91
64		-25.02	-24.03	0.96

The impact of changing background gases must also be considered. The column ozone changes for several injection altitudes with two different backgrounds of Cl_x was calculated using the LLNL one-dimensional model, and are compared in Figure 12. The column ozone changes here are calculated with respect to reference atmosphere with 1.1 ppbv Cl_x. For low levels of NO_x injected at about 12 km, the NO_x injection decreases the ozone reduction caused by the Cl_x increase. For large NO_x injections at higher altitudes, the calculated ozone change is nearly independent of the background Cl_x.

Some calculations were also made with doubled methane using the LLNL one-dimensional model. Including the extra methane reduces the calculated ozone reduction; the relative magnitude of this effect decreases with increasing NO_x injection (Table 7).

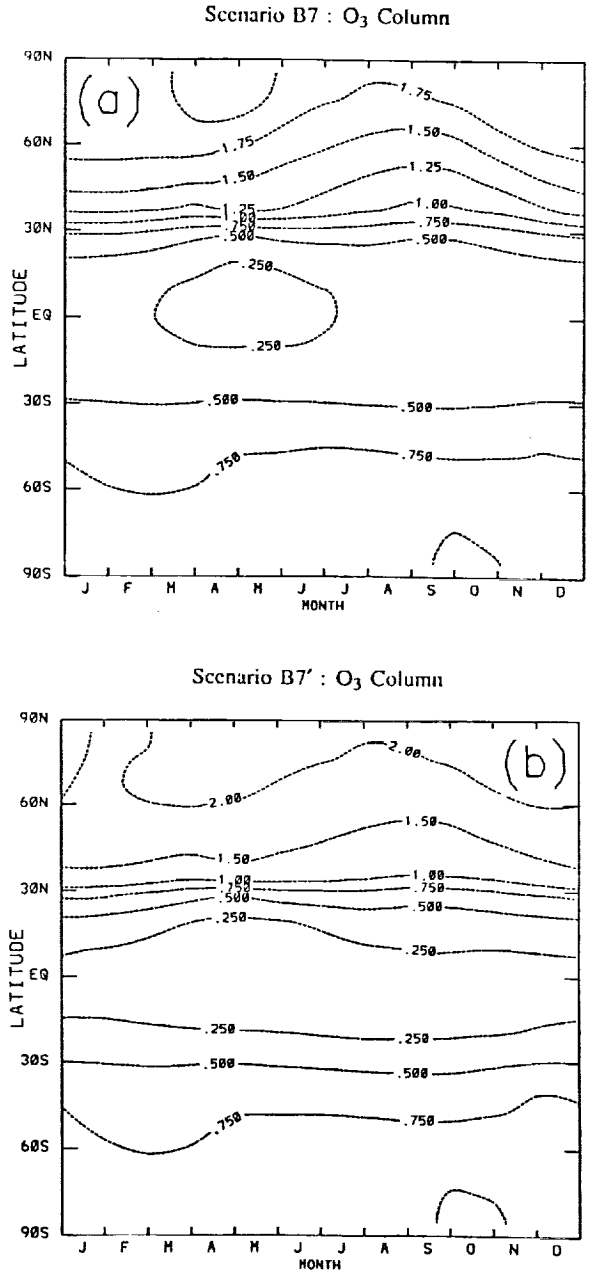

Figure 11. This scenario assumes 62 x 10^9 kg fuel/year, and an emission index of 5 gm NO/kg fuel. (a) Total O_3 change calculated by the AER model for emissions of NO_x, CH_4, CO and H_2O; (b) Total O_3 change for emissions of NO_x alone. Scenario B7 in Table 2.

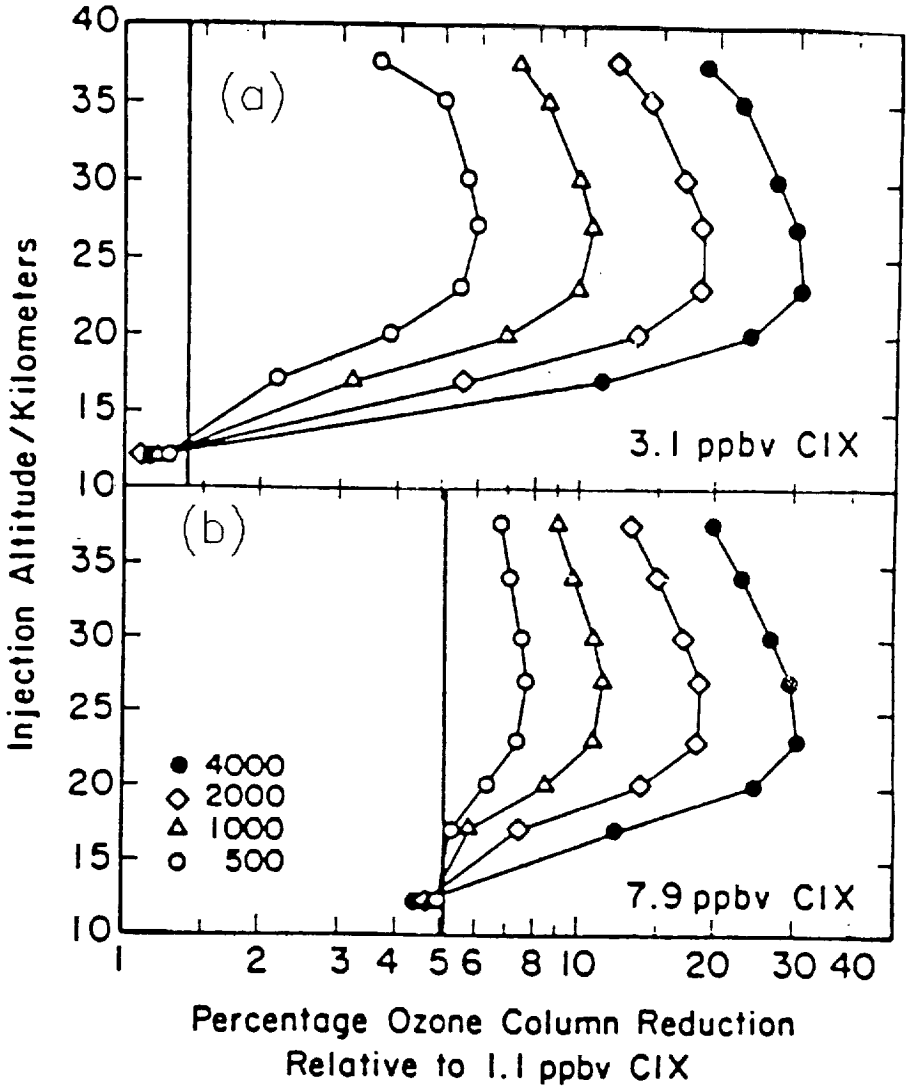

Figure 12. Percent change in the O_3 column for several values of NO_x injection and several altitudes of injection for two values of background Cl_x, calculated with respect to a base case with 1.1 ppbv Cl_x. The values of NO s^{-1} cm^{-3} over a 1 km band are given in the figure legend; for the B8 scenario of fuel usage (70.6 x 10^9 kg fuel/year), these correspond to EI values of 46, 23, 11.5 and 5.8 gm NO/kg fuel. For large values of emissions, the calculated change in O_3 is nearly independent of background Cl_x. For low altitude of injection, there is a relative increase in O_3 due to smog reactions.

Table 7 Change of vertical ozone column, 20 km injection, Cl_x 1.1ppbv, normal and double CH_4.

Amount of NO 10^{33} molecules/year	Normal CH_4	Double CH_4
0	0	+ 1.9
8	- 3.0	- 2.1
16	- 6.3	- 4.6
32	-13.1	-10.4
64	-24.7	-21.4

The AER model was used to examine a future atmosphere containing 20% more N_2O, double CH_4, 6 ppbv of odd chlorine, and perturbed temperature as a result of the CO_2 increase. The ozone response shows a complex pattern of increases at some latitudes and heights and decreases at others. The column ozone response, shown in Figure 13, shows increases of 0.5% near the equator and decreases of 2% near the polls for this future atmosphere. NO_x emissions (6.3 x 10^{33} molecules NO/year, 62.5 kg fuel/year, emission index of 5 scenario B7 Table 2) imposed on this atmosphere cause an additional increase in column ozone in the tropics and an additional decrease in mid-latitudes. However, the changes in CO_2, CH_4, N_2O and chlorine suggested for this future atmosphere are such that the impact of this aircraft scenario calculated in this future atmosphere is less than this same scenario calculated for the present atmosphere (compare figures 13(a) and 13(d)). This illustrates the importance of examining evaluations of the impact of aircraft emissions in the context of changing atmospheric composition.

e. Model improvements

Model sensitivity to the altitude and latitude of injection indicates the dependence of model results on the representation of transport in the lower stratosphere. The model representation of transport may be validated by comparison of the behavior of model tracers in this part of the stratosphere with the behavior of carbon-14 (Johnston, 1989). Improved model vertical resolution may be necessary to improve the representations of stratosphere/troposphere exchange and tracer dispersion in the lower stratosphere. The sensitivity of the calculations to the altitude and latitude of injection limits the accuracy of current models calculations of the impact of aircraft emissions on stratospheric ozone.

Heterogeneous processes, either on aerosols which may be affected by aircraft injected sulphur dioxide or on particles in the aircraft plume, may be important to partitioning nitrogen and chlorine species and therefore to the ozone behavior. These processes, which take place on scales much smaller than the model grid, should be studied further to determine if their effects are important in assessment calculations.

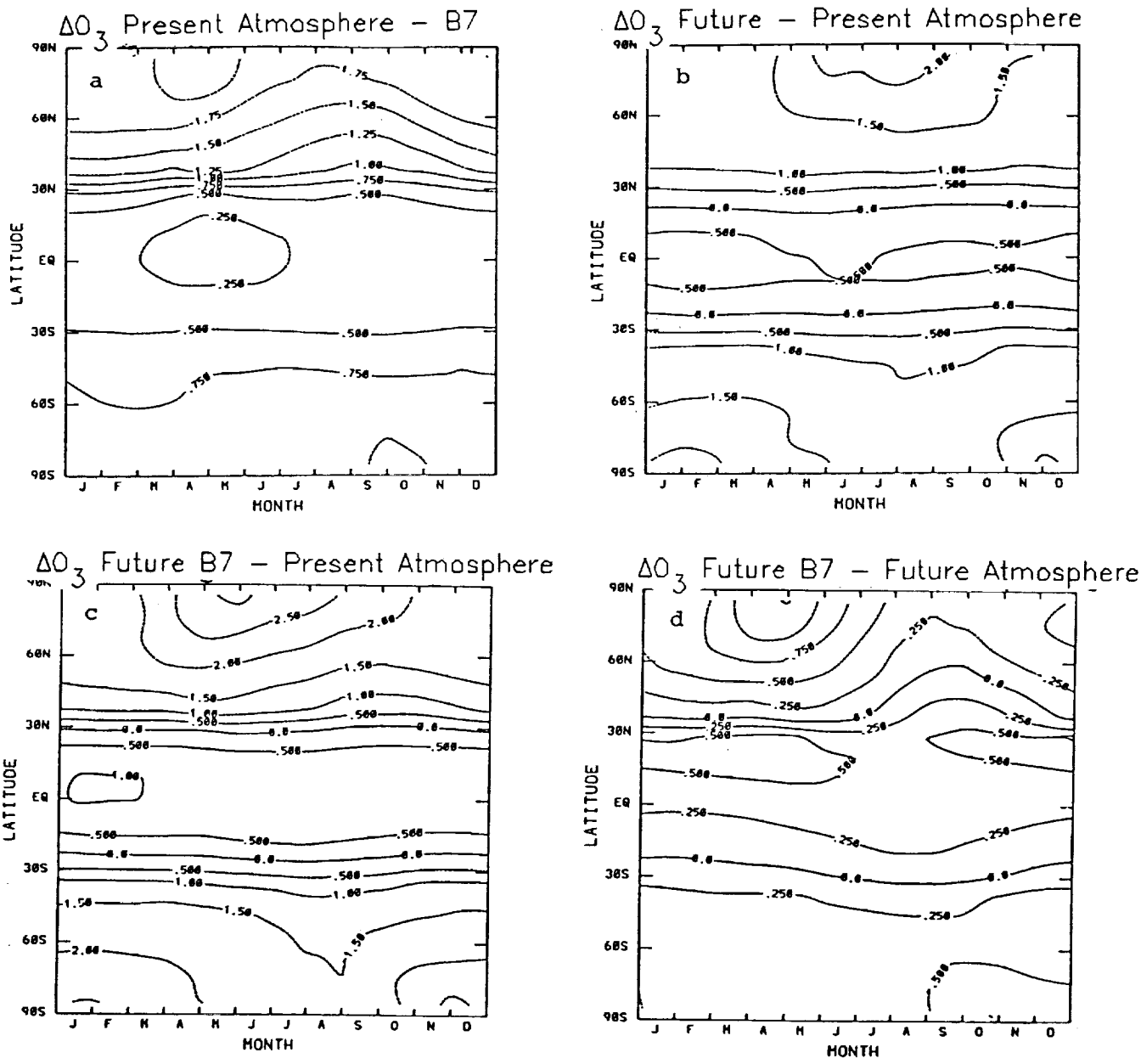

Figure 13. Total ozone changes for the B7 scenario (6.3 x 10^{33} molecules NO/year (62.5 x 10^9 kg fuel/year and EI of 5 gm NO/kg fuel) for different values of background gases. (a) Change in O_3 column as a result of B7 emissions of NO_x for the present atmosphere; (b) Change in O_3 column comparing the future atmosphere (20% more N_2O, double CH_4, increased CFC's, and double CO_2); (c) change in O_3 column comparing future atmosphere (b) with aircraft (a) to present atmosphere; (d) change in O_3 column comparing future atmosphere with aircraft to future atmosphere without aircraft.

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16. Abstract

In the early 1970s, a fleet of supersonic aircraft flying in the lower stratosphere was proposed. A large fleet was never built for economic, political, and environmental reasons. Technological improvements may make it economically feasible to develop supersonic aircraft for current markets.

Here, some key results of early scientific programs designed to assess the impact of aircraft emissions on stratospheric ozone are reviewed, and factors that must be considered to assess the environmental impact of aircraft exhaust are discussed. These include the amount of nitrogen oxides injected in the stratosphere, horizontal transport and stratosphere/troposphere exchange, and location (latitude and altitude) of flight corridors. Results using several current stratospheric assessment models are presented. Areas in which improvements in scientific understanding and model representation must be made to reduce the uncertainty in model calculations are identified.

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