NASA Reference Publication 1250

January 1991

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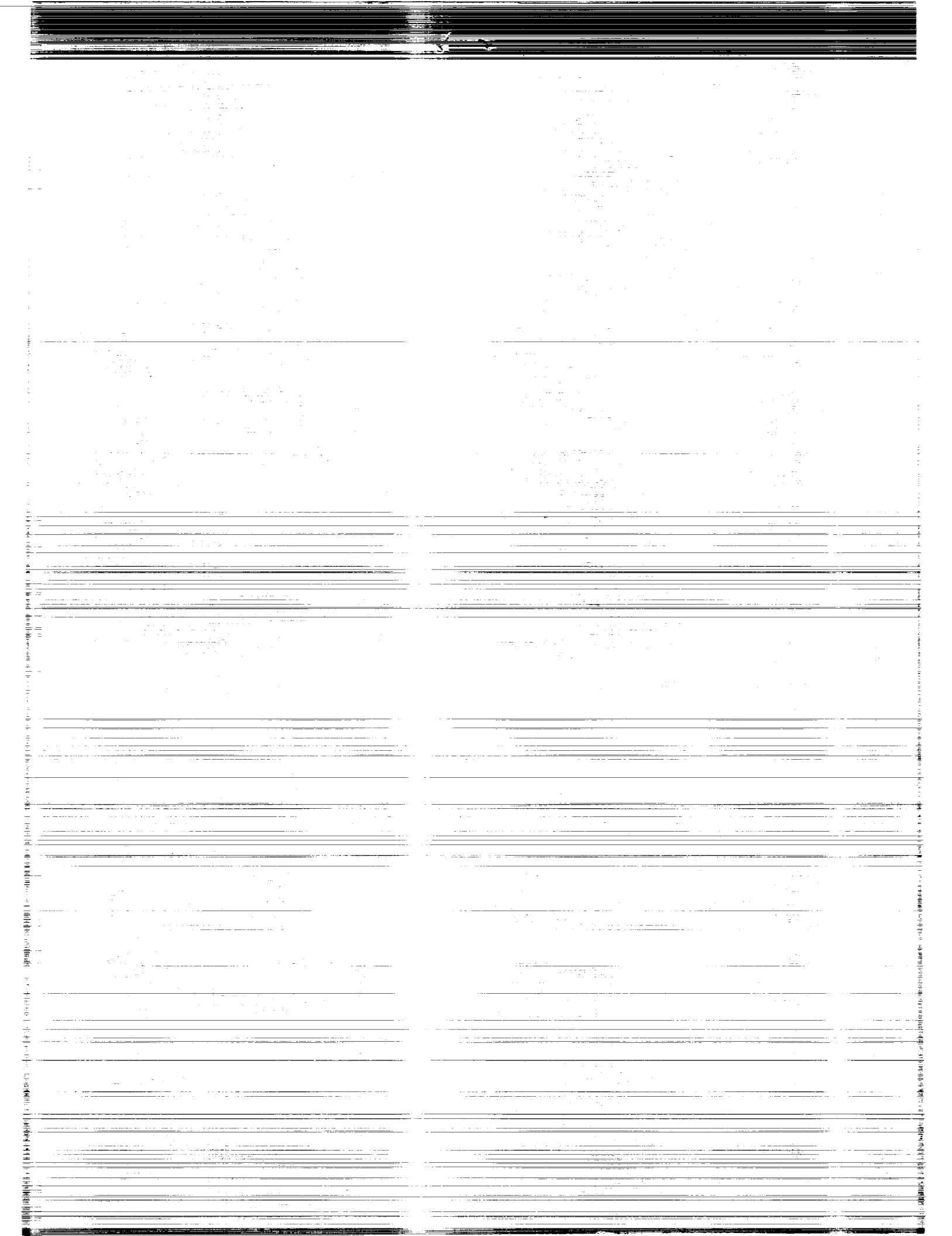

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The Atmospheric Effects of Stratospheric Aircraft: A Topical Review

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I. INTRODUCTION

Michael J. Prather and Robert T. Watson

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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 1960's, the aircraft industry became interested in developing a fleet of supersonic transports, then denoted as SST's. 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 United States with the Department of Transportation's Climatic Impact Assessment Program (CIAP) in 1974. 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 SST's 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 (HSCT's). 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 HSCT's 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 Professor Harold Johnston to review our current understanding of aircraft emissions and their impact on the stratosphere. Professor Johnston was one of the first scientists to point out the environmental danger that SST's posed to the ozone layer ("Reduction of stratospheric ozone by nitrogen oxide catalysts from supersonic transport exhaust," *Science* Vol. 173, pp. 517-522, 1971) and was a key figure in the CIAP. He and his colleagues, Drs. Doug

Kinnison and Donald Wuebbles have recently re-examined the SST problem using current models for stratospheric ozone chemistry ("Nitrogen oxides from high altitude aircraft: an update of potential effects on ozone,"*J. Geophysical Research*, Vol. 94, pp. 16,351-16,363, 1989).

This NASA Reference Publication presents Professor Johnston's topical review for the High-Speed Research Program. It provides a unique view of the current scientific issues and the lessons learned since the beginning of CIAP, and it links the current research program with the assessment process that began two decades ago. We are all indebted to Professor Johnston for this personal perspective.

II. TOPICAL REVIEW OF STRATOSPHERIC AIRCRAFT AND GLOBAL OZONE

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A. SUMMARY

The history of stratospheric aircraft and its calculated effect on ozone is reviewed for the period 1971-90, including the present problem.

(1) *The problem.* Using the most recent photochemical data, basing aircraft fuel usage on Boeing-proposed fleet size, and basing nitric oxide emissions on Pratt and Whitney emission measurements, the Lawrence Livermore National Laboratory (LLNL) two-dimensional atmospheric model calculates that nitrogen oxides from Mach 2 to 3 stratospheric aircraft cruising at 20 km would reduce global ozone by about 15 percent, a value greater than the scenarios for chlorofluorocarbons (CFC's) in which CFC emissions are assumed to remain constant from 1990 onward. [References: Boeing, 1989; Ko et al., 1989, pages 7, 27; Johnston, Kinnison, and Wuebbles, 1989; Kinnison, 1989; WMO, 1986; WMO, 1990].

(2) *Possible solutions to the problem.* The calculation in (1) above is based on aircraft cruise emission index of 39.5 grams of nitric oxide per kg of fuel consumed in the stratosphere and flying at 20 km altitude.
   (a) *Possible reduction of aircraft-produced NOx by new type of aircraft engine.* Using "advanced emission reduction technology" to design and build a new type of aircraft engine promises greatly to reduce NO_x emissions.
   (b) *The calculated effect of flight altitude.* LLNL model calculations find that for a given injection of nitrogen oxides, the calculated effect on total ozone is a strong function of altitude, and lower Mach number supersonic aircraft flying well below 20 km give much less calculated ozone reductions.
   (c) The models do not include the recently discovered heterogeneous chemistry associated with the Antarctic ozone hole, but inclusion of this feature in the models may (i) decrease or (ii) increase the calculated ozone reductions.

(3) *Some problems with these possible solutions.*
   (a) 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_x 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, pages 41-42].
   (b) Calculated ozone changes for stratospheric aircraft flying between 15 and 20 km are extremely sensitive to assigned eddy diffusion functions in some two-dimensional models, and the vertical grid height of many models is 3 km, which is much too coarse at these altitudes. Current models are not good enough to support a policy decision as to where between 14 and 19 km it is safe to emit large amounts of NO_x. There is need to carry out major model developments and calibrations, specifically devoted to the aircraft problem.

B. BACKGROUND

The Upper Atmospheric Research Program (UARP) and the High-Speed Research Program (HSRP) of the National Aeronautics and Space Administration (NASA) requested this external review of the currently understood relation between stratospheric ozone and the proposed operation of high speed passenger aircraft in the stratosphere, as an introduction and benchmark for its new atmospheric research program on this subject. Between 1972 and 1975, the U. S. Department of Transportation conducted a major research program concerning the possible environmental impacts of supersonic transports (SST's). This report gives references to eleven volumes published by this Climatic Impact Assessment Program (CIAP) and one reference to a National Academy of Sciences report that reviewed the CIAP. CIAP terminated on schedule in 1975, and very little research specifically concerned with the environmental effects of stratospheric aircraft has been done since 1975. NASA took over the field of stratospheric research in 1976, and NASA's environmental work on this program has been heavily concerned with chlorine in the stratosphere. 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 from the aircraft exhaust.

Current interest in stratospheric aviation is evidenced by:

(1) First International Conference on Hypersonic Flight in the 21st Century, September 20-23, 1988, Grand Forks, North Dakota.

(2) The NASA Langley Research Center has carried out a High Speed Civil Transport (HSCT) study project with participation and cost sharing from Boeing Commercial Airplanes and McDonnell Douglas.

(3) NASA opened the HSRP project to the general scientific public in Research Announcement NRA-89-OSSA-16, "The Atmospheric Effects of Stratospheric Aircraft (AESA): Modeling and Measurement in Support of the High-Speed Research Program, " dated July 28, 1989. Research proposals were requested up to October 31, 1989. The new atmospheric research program (AESA/HSRP) is to start by March 1990. The purpose of this report is to address the current (December 1989) scientific understanding of the possible environmental effects of a fleet of stratospheric aircraft. This review is designed to provide an evaluation of the need for this research program and a benchmark against which to evaluate its future progress.

With current aircraft flying at Mach 0.85, scheduled time between Los Angeles and Sydney (7500 miles) is 14.5 hours. If the flight Mach number is increased to include supersonic values, the trip from Los Angeles to Sydney would involve the following [Niedzwiecki, 1988]:

Mach No.	0.85	2.4	4	6		12
Trip hours	14.5	5.1	3.7	3.0		1.7
Altitude/km	11	20	25	(30	to	35?)
Fuel	Jet-A	Jet-A	T stabilized Jet	Liquid methane		Liquid hydrogen

The HCST study recognized at least the four following difficulties and challenges as it developed concepts for high speed flight:

(1) Economics.
(2) High temperatures at high Mach numbers with need for thermally stable jet fuels or cryogenic fuels and special requirements for structural materials.
(3) Airport noise and sonic booms.

(4) Ozone reduction by NO_x in engine exhaust and possible environmental effects of other components of the exhaust gases.

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. Representatives of the U.S. aircraft manufacturers state that the companies would not be interested in going ahead with constructing supersonic aircraft unless all four of the above problems can be given a satisfactory solution [Boeing, 1989]. With respect to economics, Boeing has worked out analyses of future markets for high-speed transportation and the number of aircraft that would be required to pay off the business investment in this area. On economic grounds, they reject aircraft at Mach numbers 4.5 and higher, and base further consideration largely on aircraft operating at Mach 2.4 to 3.2, cruising at altitudes of 18 to 24 km. They worked out probable flight corridors. The proposed fleets of SST's include Boeing scenario designated B8, which burns 7.1×10^{10} kg fuel per year. For comparison, some calculations made in the 1970's assumed 7.7×10^{10} kg of fuel per year (references are given in the next section).

The new NASA High-Speed Research Program is concerned with all four of the areas of concern given above, but the new research program on The Atmospheric Effects of Stratospheric Aircraft (AESA/HSRP) is concerned only with area (4). This report reviews the environmental effects of stratospheric aviation with major emphasis on the problem of ozone reduction. It includes topics in the history of this subject and recent studies that precede the initiation of the new program of atmospheric research.

C. 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), it proposed to share costs of fleet production when the time came, and the cost of these prototypes and construction 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, there was intense political debate whether the government should continue to finance the two prototypes and later to expand this program. The debate was primarily concerned with economics, national priorities, and sonic boom. Late in the debate another environmental issue was introduced; for example, Harrison [December, 1970] published a paper that said the water vapor from the exhaust of 500 SST's would reduce global ozone by about three percent, by way of chain reactions of free radicals derived from water. 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 on March 18-19, 1971).

At this meeting, atmospheric scientists presented tutorials on stratospheric motions, trajectories and lifetimes (years) of radioactivity in the stratosphere after the nuclear bomb tests of 1961-62. From the thousand-to-one ratio of horizontal to vertical spread of radioactive gases in the stratosphere, where there is a strong temperature inversion, the ratio of horizontal to vertical eddy diffusion coefficient was said to be about a million to one. Ed Danielson explained that the far northern and far southern lower stratosphere has lines of constant potential temperature that connect the stratosphere and troposphere, but above a certain altitude there are no lines of constant potential temperature that connect stratosphere and troposphere. In the first case aircraft exhaust gases can leave the stratosphere by horizontal transport, but in the second case transport out of the stratosphere requires vertical motions working against the temperature inversion. In the aircraft exhaust problem, these two regions of the stratosphere are qualitatively different. The American

SST's proposed in 1971 were to operate in the region where there are no lines of constant potential connecting stratosphere and troposphere. Other speakers presented the chemical interactions in the stratosphere, especially between water vapor and ozone. Recent measurements of nitric acid vapor in the stratosphere were reported [Murcray et al., 1968], but Crutzen's [1970] proposal that the oxides of nitrogen play an important role in the natural ozone balance was not mentioned at this meeting. Aircraft experts presented:

(1) The planned size of the American fleet (500 supersonic transports).

(2) The average service time, cruise altitude, and the amount of fuel consumed, 7.7×10^{10} kg per year.

(3) The amount of water vapor (1250 g H_2O per kg fuel) and the amount of oxides of nitrogen (42 g NO per kg fuel) emitted in the engine exhaust [a quotation from General Electric engineers].

(4) Since the stratospheric aircraft were expected to fly primarily at mid-latitudes in the Northern Hemisphere, it was regarded as reasonable and prudent to assume that near the main flight corridor local vertical columns of engine exhaust would exceed the global average value by a factor of 10, the "corridor effect."

Although there was some mention at this meeting of nitrogen oxides from SST exhaust as possibly affecting 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 laboratory measurements that indicated essentially zero rate for the key reaction between ozone and the free radicals derived from water. This work was later found to be in error, but the report itself had an impact on understanding stratospheric chemistry in mid-1971.

With the state of ignorance of the stratosphere that existed in 1971, it was not feasible to carry out calculations of the time-dependent vertical and horizontal spread of the exhaust gases and their chemical and photochemical reactions. However, statistical mechanics provides an alternate method of studying highly complicated systems. If a problem of time-dependent dynamics is overwhelmingly difficult, a static ensemble average may give meaningful conclusions. Statistical mechanics is largely concerned with fundamental problems in physics and chemistry, but its concepts are applicable in many areas. It is a systematic approach to the question: "What do you do with a problem when you don't know everything about it?" The general idea is: "Apply the knowledge that you have as rigorously as possible, and assume equal *a priori* probability for conditions where needed information is incomplete."

A global, static, steady-state photochemical atmospheric model was set up, using such information as was known. Calculations were made as a function of longitude and altitude, one latitude at a time, but primarily at 45 degrees north. Quantities varied in the spirit of statistical mechanics included:

(1) Uniform and non-uniform background stratospheric nitrogen-oxide profiles that varied from zero to values so high that astronomers would long ago have complained about spectral interference from atmospheric nitrogen dioxide.

(2) Quantities of aircraft-produced nitrogen oxides that covered a 1000-fold range. The upper limit was based on the ten-fold "corridor effect" and the G.E. emission index.

(3) With the mental image of the Brewer [1949] circulation and a polar sink, the stratospheric lifetime of NO_x from the exhaust gases emitted at 20 km was taken to be two years.

(4) In lieu of the (impossibly difficult) calculation of the vertical and horizontal spread of the exhaust gases, the vertical spread above and below the 20-km cruise height was taken to be 1, or 4, or 7, or 10, or 13, or 16 km, and the horizontal spread was assumed to be globally uniform, which is consistent with the order-of-magnitude spread of radioactive gases in the stratosphere after the nuclear bomb tests of 1961-62 (see page 4) [Johnston, 1971a 1971b].

For the non-uniform NO_x background profile that gave the best fit to the observed mid-latitude ozone profile, for NO_x exhaust rates equal to one-third the G.E. estimate, and for uniform global horizontal spread of the exhaust gases, the steady-state ozone reductions calculated with each of the six assumed vertical spreads varied from 3 to 23 percent, Figure 1. It was assumed that the actual vertical spread would be close to some combination of these bands. From this line of reasoning, the change of global average ozone was judged to be:

$$\text{Change of global } O_3 = -13 \pm 10 \text{ percent}$$

Similar calculations, but including the ten-fold corridor effect, gave:

$$\text{Maximum change of local ozone column} = -50 \text{ percent}$$

These two results used 7.7×10^{10} kg fuel per year and an emission index of 15 g NO/kg fuel, which is equivalent to 1.8 MT (10^{12} g) of NO_2 per year or to 24×10^{33} molecules of NO_x per 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, 1971a, 1971b]. Similar, less extensive results concerning aircraft were later given by Crutzen [1971].

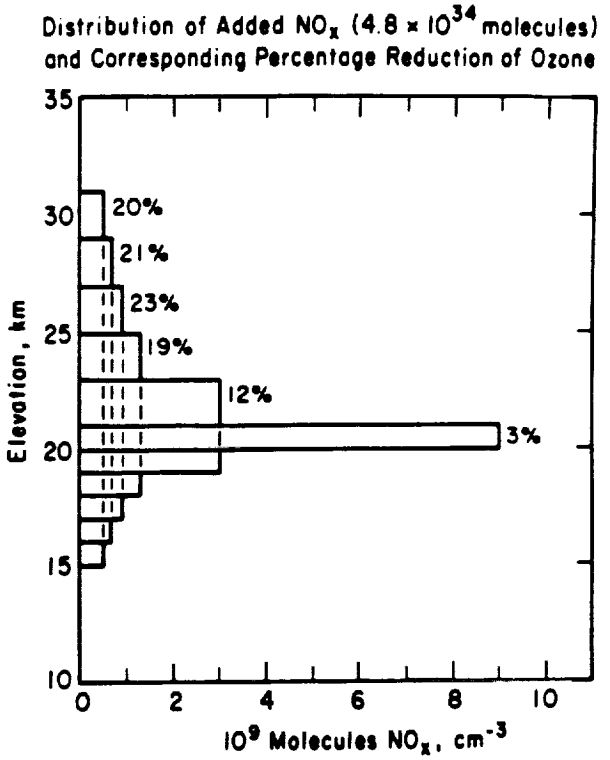


Figure 1. Steady-state calculations of 24-hour average ozone columns at 45°N, spring equinox, for non-uniform background of natural stratospheric NO_x and added artificial NO_x at 20 km with various assumed vertical spreads above and below 20 km. The natural NO_x profile was one found to give the observed vertical profile of ozone. The artificial 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. Conclusion: If actual spread of NO_x fell between these wide limits, it would significantly affect global ozone [Johnston, 1971a, 1971b]

D. MAJOR STRATOSPHERIC RESEARCH PROGRAMS, 1972 TO PRESENT

CIAP, 1972-1975

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 assigned the U.S. Department of Transportation (DOT) to conduct the Climatic Impact Assessment Program (CIAP), and to complete it by the end of 1974. This highly productive research program, among other things, carried out the following developments and discoveries:

(1) Observed (many for the first time) stratospheric profiles of NNO, NO, NO_2, HNO_3, and O, which confirmed Crutzen's postulate [1970] 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) Supported laboratory measurements of chemical and photochemical reaction rates.

(3) Prepared critical database of chemical and photochemical reaction rates for use by atmospheric modelers.

(4) Incorporated photochemical "smog" reactions in stratospheric ozone considerations, Figure 2, which gave at 13 km the cross-over between ozone destruction by NO_x and ozone formation by way of the methane-NO_x smog reactions [Johnston and Quitevis, 1975; CIAP Monograph 1, 1975a, Chapter 5, pages 98-107].

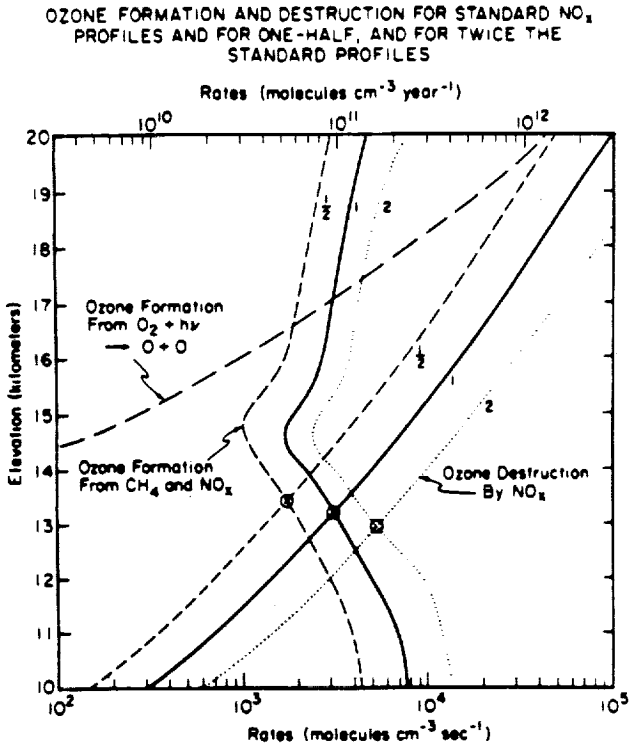


Figure 2. Vertical profiles of the rate of formation of ozone from the methane-smog reactions and of the rate of ozone destruction from NO_x catalytic cycle. There is a cross-over at about 13 km between ozone destruction and ozone formation so far as NO_x is concerned. For half and for double the assumed natural background of NO_x , the cross-over point was essentially unchanged [Johnston and Quitevis, 1975; presented at meeting on Radiation Research, Seattle, WA, July 1974, and to joint CIAP-NAS meeting, Woods Hole, MA, July 1974].

(5) Developed theories of motions and transport in the stratosphere and developed the first serious photochemical, time-dependent models of stratosphere.

(6) Studied the climatic impact of stratospheric particulates produced by sulfur in jet engine fuels.

(7) Studied environmental and economic impacts of ozone reduction and of climate change.

(8) Discovered (1973) that Cl and ClO catalytically destroy ozone in reactions that have many features in common with the nitrogen oxides system, and provided some of the concepts used by Molina and Rowland [1974] in their analysis of the chlorofluorocarbon problem. (The analogy between tropospheric nitrous oxide as a source of stratospheric NO_x and tropospheric CFC's as a source of stratospheric Cl and ClO is one of several "hand-in-glove" parallels between the NO_x and Cl_x systems.)

(9) Considered heterogeneous chemistry in the stratosphere [CIAP Monograph 1, 1975a, pages 5-55-70; CIAP Monograph 3, Chapter 6, 1975c].

(10) Projected the future of aviation to 2025.

(11) Concluded that NO_x from stratospheric aircraft reduces ozone, for example, Figure 3 [Cunnold et al., 1975, 1977]. This partly three-dimensional and otherwise two-dimensional model study, supported by CIAP, calculated that nitrogen oxides (7.7×10^{10} kg fuel per year, emission index 15 g NO per kg fuel) injected in a narrow corridor in the Northern Hemisphere reduced the global-average ozone column by 12 percent and found a worst-case local ozone-column reduction of 25 percent near the flight corridor.

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

(13) In a timely fashion (during 1975) published reports detailing its massive findings [see CIAP, all entries].

(14) Terminated on schedule after the 1974-75 fiscal year.

NASA Stratospheric Program

A major development in stratospheric science coincided with the completion of CIAP, namely, Rowland and Molina's [1974, 1975] analysis of the effect of chlorofluorocarbons on stratospheric ozone. The large stratospheric research program dropped by the Department of Transportation in 1975 was transferred to NASA in 1976, which brought other agencies and international organizations into the program. Low-profile, high quality, scientific research has been carried out on a large scale since 1976, including laboratory studies, preparation of critical data bases, atmospheric measurements (with aircraft, balloons, and satellites), studies of the theory of atmospheric motions, and dynamical-radiative-photochemical atmospheric models. The NASA program emphasized the effects of chlorine on atmospheric ozone. It dropped the CIAP supportof economic, biological, and environmental studies of the effects of ozone change. Selected results of the NASA stratospheric program are:

(1) Confirmed the postulates of Rowland and Molina.

(2) Discovered a major surprise about every couple of years.

Figure 3. Calculated decrease of the ozone column as a function of latitude for injection of 1.8 MT (as NO_2) per year over a 10-degree corridor in the Northern Hemisphere, spring season. The global average ozone decrease was 12 percent and the local maximum decrease is 25 percent near the North Pole. Based on MIT 3-D model of atmospheric motions and two-dimensional model of photochemistry [Redrawn from Cunnold et al., 1975, 1977].

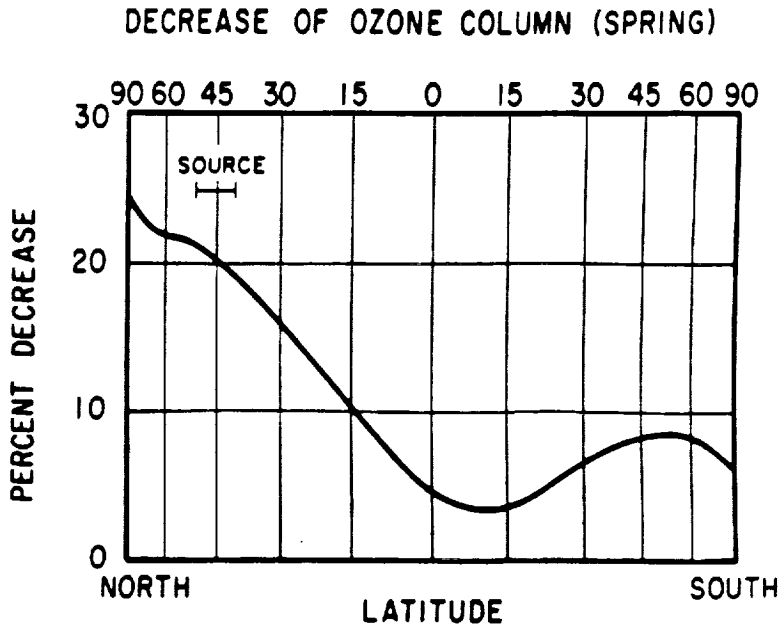
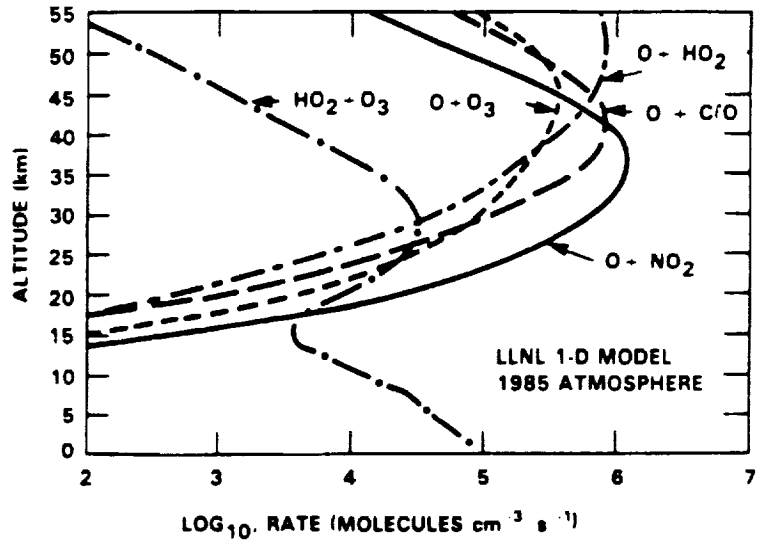


Figure 4. Calculated ozone destruction from major catalytic cycles as a function of altitude using the Lawrence Livermore National Laboratory (LLNL) one-dimensional model with 1985 photochemical data. [D.J. Wuebbles, private communication, 1989]. NO_X is the most important natural ozone destroyer in the 17- to 40-km range, and the natural rate of NO_X production in the stratosphere is known, Table I, [Crutzen and Schmailzl, 1983].



(3) Completed approximate closure of the global ozone balance using satellite data and two-dimensional models. As found by the LLNL one-dimensional model, the vertical profiles of six processes now recognized as destroying ozone are given in Figure 4. In the region of ozone formation in the middle stratosphere and in the region of maximum ozone mixing ratio in the lower-middle stratosphere, by far, the most important mode of natural ozone destruction is the nitrogen oxides catalytic cycle. The processes by O_x, HO_x, and NO_x are all about equally important in the uppermost stratosphere, and HO_x cycles are most important in the troposphere.

(4) Mounted an expedition in 1987 that determined the mechanism of the Antarctic "ozone hole." The Airborne Antarctic Ozone Experiment (AAOE) unambiguously ascribed the chemical component of the phenomenon to stratospheric chlorine species and to heterogeneous reactions of hydrogen chloride and chlorine nitrate on stratospheric cloud particles. These heterogeneous reactions lock oxides of nitrogen into the cloud particles as nitric acid, and release active chlorine from its inactive forms (hydrogen chloride and chlorine nitrate).

Although the NASA UARP program has not been explicitly concerned with the effect of stratospheric aviation on ozone, scientists at LLNL have maintained a baseline of the calculated effect of standard NO_x injections through the years of the maturing of stratospheric science, Figure 5. One curve shows the calculated future, steady-state, ozone-column change for the continuous usage of chlorofluorocarbons at the 1974 rate. The other curve shows the calculated future, steady-state, ozone-column change for an assumed continuous worldwide injection of 2×10^8 NO_x molecules $cm^{-2}s^{-1}$ (7.7×10^{10} kg fuel per year, emission index 20 g NO per kg fuel). For these constant assumed perturbations, the calculated future ozone changes have varied with the date of calculation, which used the then-current knowledge of the stratosphere. The variations of the results between the years of 1976 to 1981 were largely caused by new laboratory measurements of rate coefficients of the HO_x family of reactions and the discovery of new species to include in the models. A noteworthy feature of Figure 5 is that for about two years, 1978-1980, NO_x from stratospheric aircraft was calculated to increase the ozone column. The model gave ozone reduction by NO_x in the middle stratosphere, but it gave even larger ozone production by the methane-NO_x smog reactions in the upper troposphere and lower stratosphere (the "crossover point" of Figure 2 appeared to move up above 20 km). When all HO_x reactions were redone by modern direct methods, the "crossover point" of Figure 2 returned to the 12- to 14-km range, and NO_x injected at 20 km has been calculated to reduce the ozone column since 1980. Since 1981, the calculated ozone reduction by aircraft for this scenario is greater than that calculated for CFC's assuming fixed 1974 emission rates. (N.B. These models did not include the heterogeneous chemistry that is responsible for the current Antarctic ozone hole.). In 1988, the calculated ozone reduction by NO_x from stratospheric aircraft is about the same as it was at the conclusion of CIAP in 1974.

E. CURRENTLY PROPOSED STRATOSPHERIC AIRCRAFT

Results of Atmospheric Model Calculations Published in 1989

Using the well-developed (1973-1988) LLNL one-dimensional time-dependent model of the troposphere and stratosphere and using the newly developed LLNL zonally averaged two-dimensional chemical-radiative-transport time-dependent model, Johnston, Kinnison, and Wuebbles [1989] carried out calculations concerning stratospheric aircraft and ozone, and a part of it was included in a paper given at the 1988 Hypersonic Flight Conference cited above. The LLNL

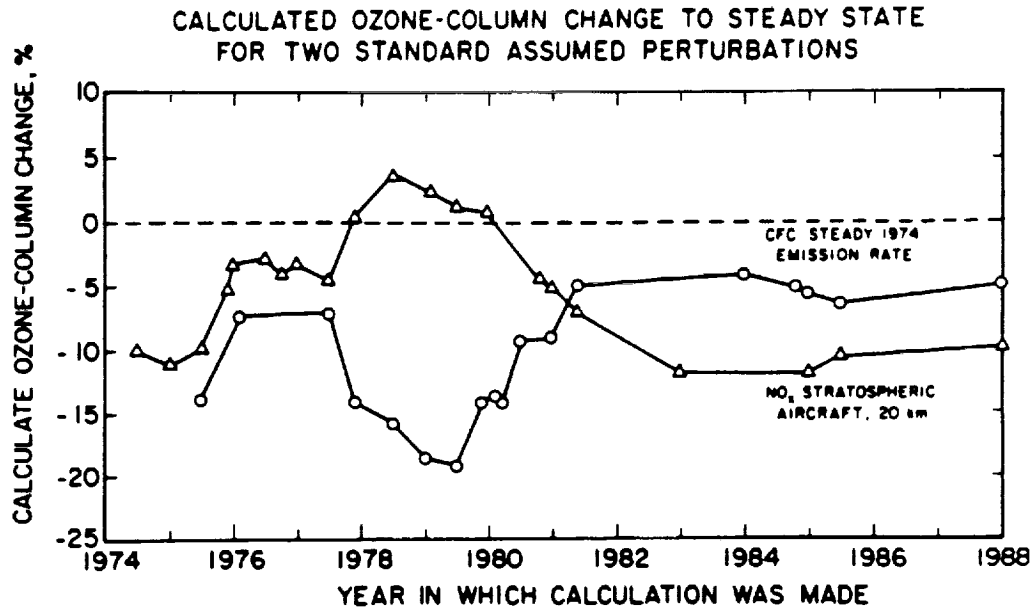


Figure 5. Calculated ozone-column change to steady-state for two standard assumed perturbations: (a) 2000 molecules $cm^{-3}s^{-1}$ over one km interval centered at 20 km; (b) CFC-11 and CFC-12 emitted continuously at 1974 rate. These calculations were made at LLNL (1-D model) over this 14-year period using then current photochemical parameters, eddy diffusion functions, and boundary conditions (Source, WMO, 1986, page 773, updated to 1988).

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calculations cover a wide range of NO_x injection rates, injection altitudes, stratospheric chlorine background, methane background, water vapor injection, and latitude patterns. In various places, the NO_x emission rates are expressed in three different units:

(1) Molecules of NO_x, divided by 10^5, injected per square centimeter per second over the globe.

(2) Metric megatons (MT), 10^9 kg per year, calculated as if NO_2, even though NO is the principal form of NO_x in engine exhaust.

(3) Emission index (EI), grams of NO per kg of fuel, and a statement of the annual usage of fuel in stratospheric flight. In this case the annual fuel used in the stratosphere is taken to be 7.7×10^{10} kg per year, a value carried over from the CIAP period. There is confusion at present whether quoted "NO_x emission index" is based on NO or NO_2 and care is needed in using written or verbal statements of this quantity. [Editors' note, July 1990: The definition of emission index (EI) for odd nitrogen has been ambiguous at times, but the AESA/HSRP working group on emissions has agreed that the program shall use the definition as grams of total NO_x (= NO + NO_2) weighed as NO_2 per kg of fuel burned.] For NASA's High Speed Civil Transport Program, Ko et al. [1989] carried out two-dimensional model studies for Boeing emission scenarios B7, B8, and B10 and for McDonnell Douglas scenarios A3, A4, and A5. Studies were made of varying background gases. Global NO_x emission rates are expressed in molecules per second.

Table I gives the range of emissions considered by Johnston, Kinnison, and Wuebbles [1989] and by Ko et al. [1989]. In each case, global NO_x inputs are presented (a) in units of NO_x molecules per year and (b) in terms of annual fuel consumption and nitric oxide emission index. For the

cases in Table I, the rate of SST-produced NO_x varies from one-eighth to 2.5 times the natural rate of NO_x production in the stratosphere [Crutzen and Schmailzl, 1983]; NO_x is the most important agent in the natural destruction of ozone, Figure 4; and this simple, model-free comparison indicates that there may be a serious ozone problem.

Table I. Fuel consumption by stratospheric aircraft, nitric oxide emission index, and molecules of nitric oxide injected into the stratosphere (i.e., above 14 km) per year, according to model calculations discussed in this report. LLNL calculations [Johnston, Kinnison, and Wuebbles, 1989]; calculations for the Boeing and McDonnell Douglas scenarios [Ko et al., 1989]

Scenario	NO_x 10^{33} Molecules/yr	Fuel 10^9 kg /yr	EI (as NO) g/kg (fuel)	EI (as NO_2)
LLNL 1-D	8 to 64	77	5 to 40	8 to 61
LLNL 2-D	63	77	40	61
	24	77	15	23
	8	77	5	8
Boeing B7	6.3	66.1	5	7.3
Boeing B8	12.8	75.3	9	13
Boeing B10	7.7	91.3	4	6.4
Douglas A3	18.1	25.3	36	55
Douglas A4	5.7	25.3	11	17
Douglas A5	3.1	25.3	6	9
Natural source [Crutzen and Schmailzl, 1983]	24			

One-Dimensional Model Calculations

The calculated ozone reduction is, obviously, a strong function of the rate of injection of nitrogen oxides into the stratosphere. Figure 6 presents the calculated (LLNL one-dimensional model) ozone changes as a function of NO_x injection rate at 20 km from zero to 4000×10^5 molecules $cm^{-2}s^{-1}$ (0 to 64×10^{33} molecules per year, globally) and for 1.1 and 7.9 ppb of stratospheric Cl_y. The value of 8 ppb of Cl_y is the calculated steady-state value about a century from now, if CFC's are used throughout the future at the present rate. The value of 1.1 ppb of Cl_y corresponds to the 1960 stratosphere, and the value in 1989 is taken to be 3 ppb. For large NO_x injection rates (2000 to 4000) such as would occur with engines using current technology, the ozone-column reduction is large, 12 percent to 22 percent; and the degree of ozone reduction is almost independent of the Cl_y background. For the relatively small NO_x injection rate (500), which approximately corresponds to proposed future high-technology engines, the addition of NO_x causes additional ozone reduction: 3.0 percent for 1.1 ppbv Cl_y, 2.4 percent for 3.1 ppbv Cl_y, and 1.2 percent for 7.9 ppbv Cl_y.

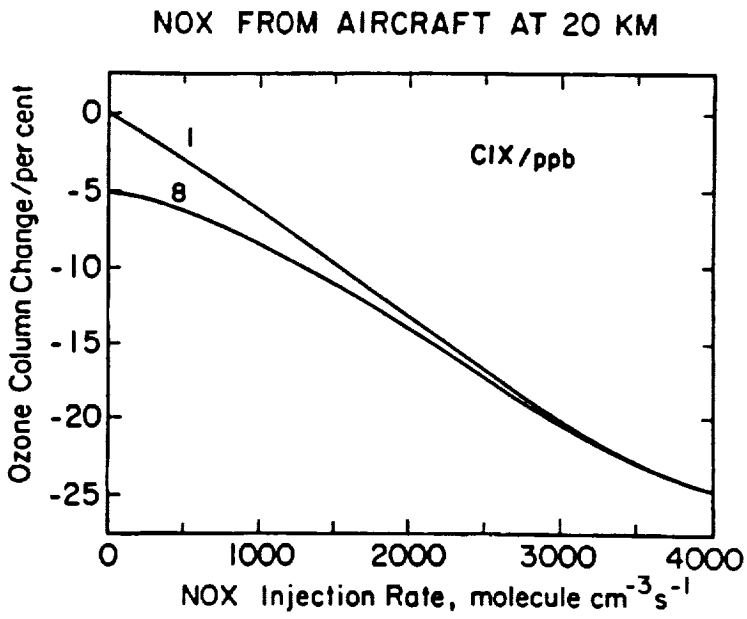


Figure 6. Percentage change of the ozone vertical column as calculated by the LLNL one-dimensional model, where the reference conditions include upper boundary value of stratospheric Cl_y at 1.1 ppbv, and the lower boundary value of nitrous oxide is 300 ppbv. For each value of Cl_y at 1.1 and 7.9 (nominally 1 and 8) ppbv, nitric oxide injection rates of 0, 500, 1000, 2000, and 4000 molecules $cm^{-3}s^{-1}$ over a one-km thick band at 20 km are considered.

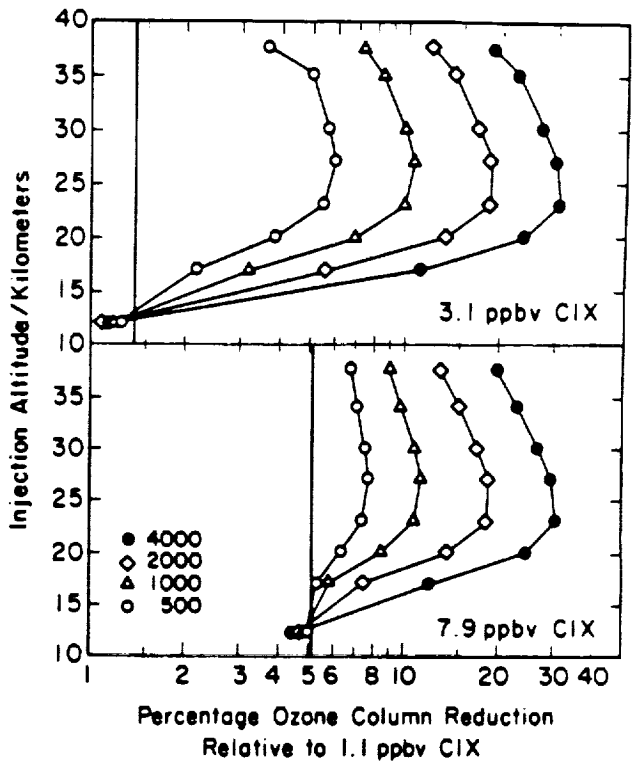


Figure 7. Calculated percentage reduction of the ozone vertical column as a function of altitude and magnitude of NO injections, according to LLNL one-dimensional model. The ozone reduction is calculated relative to the reference atmosphere with 1.1 ppbv of Cl_y. The background of Cl_y is 3.1 ppbv in the upper panel, and the vertical line on the left-hand side is the ozone reduction caused by 3.1 ppbv of Cl_y. The background of Cl_y is 7.9 ppbv in the lower panel, and the vertical line in the center of the panel is the ozone reduction brought about by 7.9 ppbv of Cl_y. For a given Cl_y background and a given NO injection rate, the injection altitude that produces the maximum ozone reduction is 25 ± 2 km. For injections at 12 km, added nitric oxide increases ozone thus reducing the ozone reduction caused by Cl_y.

Another important variable in the calculated ozone reduction by SSTs is the altitude of NO_x injection. Figure 7 presents the calculated (LLNL one-dimensional model) ozone reduction as a function of altitude of injection for five rates of injection (units as in Figure 6) and for two values

of stratospheric chlorine. The upper panel corresponds to the present chlorine background, and it should be applicable for the next decade or two. The lower panel corresponds to the chlorine background expected about a century from now, unless chlorofluorocarbon use is held constant. In general, the injection altitude that gives the maximum reduction of ozone is about 25 km. NO_x injected at about 34 to 37 km has about the same effect on the ozone column as that injected at 20 km. The crossover altitude between ozone production by methane-NO_x smog reactions and ozone destruction by added NO_x is 12 km; compare Figure 2.

The vertical grid height of the one-dimensional model is one km. For 3.1 ppbv Cl_y and 500 NO_x molecules $cm^{-3}s^{-1}$ injected over the one-km band, the ozone column change for NO_x injection at 17 km is -0.7 percent, but the ozone column change for NO_x injection at 20 km is -2.4 percent. To model accurately the effect of aircraft in the 12 to 21-km flight range, a necessary (but not sufficient) condition is that the model have a vertical grid height no greater than one km.

Some calculations were made with doubled methane, and some with and without considering water vapor from the exhaust, Table II. With no added NO_x, doubling methane produced a 1.9 percent increase of the ozone column. Doubling methane and injecting $8x10^{33}$ molecules NO_x per year at 20 km produce a 2.1 percent ozone reduction relative to normal methane. In general, added NO_x at 20 km causes less calculated ozone column reduction with doubled methane than with the reference value for methane, Table II-B.

Table II. (A) Change of the vertical ozone column (percent) as a result of various injection rates of nitric oxide (in units of 10^{33} molecules per year) from stratospheric aircraft at 20 km, where the Cl_y background is 1.1 ppbv. 1-D LLNL model.

NO(10^{33}/yr)	Normal CH_4	2xCH_4
0	0.0	+1.9
8	-3.0	-2.1
16	-6.3	-4.6
32	-13.1	-10.4
64	-24.7	-21.4

(B) Effect of injecting water vapor in an amount equivalent to burning $77x10^9$ kg of fuel (CH_2 empirical formula) per year. Stratospheric Cl_y is 3.1 ppbv.

	Percent Change of the Ozone Column			
NO(10^{33}/yr)	H_2O only	NO only	H_2O & NO	Ratio Both/NO
0	-0.09			
8		-3.0	-2.66	0.88
24		-9.94	-9.06	0.91
64		-25.02	-24.03	0.96

Water vapor is a necessary exhaust component from hydrocarbon fuel combustion, and for the standard assumed fuel consumption rate of Table II-B the rate of water vapor emission is 2.0×10^5 molecules cm^{-3} over a one km vertical band. With no injected nitric oxide, this large water vapor injection at 20 km is calculated to reduce the ozone column by only 0.09 percent. When both NO and water are emitted together, the calculated ozone reduction is smaller (by factors that vary from 0.88 to 0.96 in Table II-B) than for calculations made ignoring the addition of water.

Two-Dimensional Model Calculations

The calculations made by Johnston, Kinnison, and Wuebbles [1989] with the LLNL two-dimensional model are given in Table III. The model has a 3-km vertical grid height. Calculations were made for aircraft flights at average altitudes of 16.5, 19.5, 22.5, 25.5, 28.5, 31.5, and 34.5 km. Other things being equal, maximum ozone reductions were found at about 28.5 km. Most calculations were made with 1.8 MT NO_2 yr^{-1} which corresponds to an emission index of 15 g NO per kg fuel or to 24×10^{33} molecules NO_x per year. Global average ozone reduction for NO_x injected at 22.5 km was about 9 percent. For an NO emission index of 40 (engines with current technology), the global average ozone reduction was 19 percent. For an NO emission index of 5 (proposed engines for about the year 2000), the global average ozone reduction was 2.8 percent . When all flights are in the Northern Hemisphere, ozone reduction in the Southern Hemisphere is about one-third that calculated for the Northern Hemisphere. The ozone reduction calculated for aircraft at 16.5 km is one-tenth that calculated at 19.5 km, which is only one grid height away, illustrating the necessity for better vertical resolution for the model if it is to be used to locate safe altitudes for stratospheric aircraft.

A comparison between two different models is given by Figures 8 and 9, for the LLNL and the AER two-dimensional models.

For 1.8 MT NO_2 per year injected at 18 to 21 km altitude and between 37 and 49 degrees north latitude (LLNL), Figure 8 presents contour plots of percentage ozone-column change as a function of season and latitude in the upper panel, and in the lower panel it gives percentage change of the January ozone concentration as a function of latitude and altitude. This case is the second entry in Table III, which is the third entry in Table I (NO emission index of 15). The average reduction of the ozone column is 7.6 percent for the globe, 5.9 percent in the Southern Hemisphere, and 10.4 percent in the Northern Hemisphere. The maximum local ozone-column reduction, 16 percent, is seen in the polar region.

Using the AER model, Ko et al. [1989] made calculations for all the Boeing and McDonnell-Douglas scenarios shown in Table I. The closest comparison to be made between calculations by AER and by LLNL is Ko's McDonnell-Douglas A3 scenario (Figure 9) relative to the second entry in Table III (Figure 8). The LLNL case injects 1.33 more NO_x into the stratosphere than scenario A3. Figure 9 was prepared by Ko et al. for the A3 scenario, and it shows: maximum column reduction, 14 percent in north polar winter; 3 percent at 45 degrees south; and 8 percent at 30 degrees north. In Table IV, these ozone reductions are multiplied by 1.33 and compared with similar numbers from the LLNL calculation.

This comparison shows that the calculated ozone changes by Ko et al. and by Johnston, Kinnison, and Wuebbles are in reasonably good agreement when applied to the same NO_x input.

Using 1989 jet engine technology, the calculated ozone reduction for Mach 2.4 aircraft (altitude 20 km) implicit in Boeing B8 fuel usage [Ko et al., 1989, page 27] and McDonnell-Douglas A3 emission index [Ko et al., page 7] is found by interpolation among the LLNL two-dimensional calculations given in Table III. This scenario gives a calculated global ozone reduction of 16

Table III. Calculated changes in global and hemispherical ozone vertical columns as a result of NO_x injections by stratospheric aircraft. LLNL two-dimensional model. The boundary values include 300 ppbv of nitrous oxide and enough chlorofluorocarbons to give 2.8 ppbv of stratospheric chlorine at 50 km. Photochemical and chemical constants based on JPL 1987 [DeMore et al., 1987].

NO_x Injection as if NO_2			Percent Ozone Change		
km	Lat.N	MT/yr	Global	N.H.	S.H.
16.5 ± 1.5	37-49	1.8	-0.7	-0.9	-0.4
19.5 ± 1.5	37-49	1.8	-7.6	-10.4	-5.9
22.5 ± 1.5	37-49	4.8	-19.1	-27.9	-6.7
22.5 ± 1.5	37-49	1.8	-8.6	-13.1	-3.9
22.5 ± 1.5	37-39	0.6	-2.8	-4.3	-1.2
22.5 ± 1.5	0-12	1.8	-11.0	-12.5	-9.5
25.5 ± 1.5	37-49	1.8	-9.6	-15.6	-3.5
28.5 ± 1.5	37-49	1.8	-10.1	-16.4	-3.5
31.5 ± 1.5	37-49	1.8	-9.8	-16.0	-3.4
34.5 ± 1.5	37-49	1.8	-9.2	-14.9	-3.2
34.5 ± 1.5	37-49	0.9	-5.1	-8.4	-1.6
34.5 ± 1.5	37-49	0.45	-2.6	-4.4	-0.8
34.5 ± 1.5	N.Hemi.	1.8	-9.1	-13.6	-4.5
34.5 ± 1.5	Global	1.8	-9.8	-9.5	-10.2
22.5 ± 1.5	Global	1.8	-9.4	-9.4	-9.3
19.5 ± 1.5	Global	1.8	-7.2	-7.2	-7.1

percent, which is a value much greater than scenarios for chlorofluorocarbons in which emissions were frozen at 1974 levels, and which is comparable or larger than predicted ozone losses with high growth in CFC production.

In the case of CFC's it must be remembered that model predictions of ozone depletion did not include the Antarctic ozone hole. However, these predictions of moderate ozone loss by the year 2060 (4-12 percent at high latitudes in early spring), coupled with observations of the ozone hole, has led to international agreements (Montreal Protocol, 1987) whereby governments shall reduce substantially and eventually phase out CFC production.

Although Boeing has tentatively converged on Mach 2.4 and Douglas on Mach 3.2 as favored models, there are still proposals for the Hypersonic Transport with Mach number 5 [San Francisco Chronicle, "Cracking Japan's Aircraft Market," December 4, 1989, page C2]. Depending on actual Mach number, such craft might fly in the altitude range of 25 to 35 km. Figure 10 presents the results of LLNL two-dimensional model for injecting nitric oxides between 33 and 36 km (entry 10 of Table III). The rate of injection is the same as that for Figure 8. The maximum reduction of the ozone column is 34 percent in the Arctic spring, the column reduction between 30 and 45 N is 12 to 20 percent, and the ozone column reduction is 2 to 6 percent from the Equator over the Southern Hemisphere. According to CIAP Monograph 3, Chapter 2, page 75, "The high combustion temperature in the HST (Hypersonic Transport) scramjet results in a nitric oxide emission index about an order of magnitude greater than that of current jets." According to this statement, Figure 10 underestimates ozone reduction by HST's.

Figure 8. Calculated percentage change in ozone according to LLNL two-dimensional model for the injection of 1.8 MT NO_2 per year between 18 and 21 km altitude and between 37 and 49 degrees north: (upper panel), change in ozone vertical column as a function of latitude and season; and (lower panel), change in local ozone during January as a function of latitude and altitude. [Johnston, Kinnison, and Wuebbles, 1989].

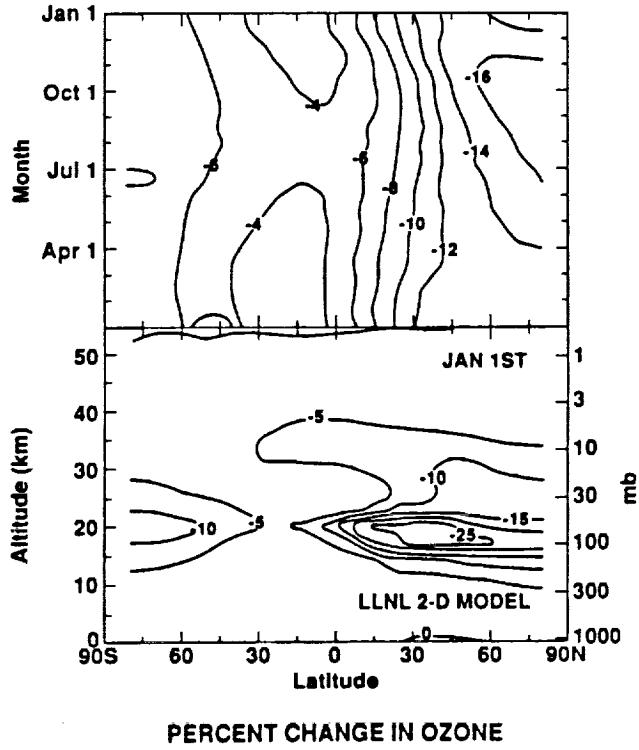
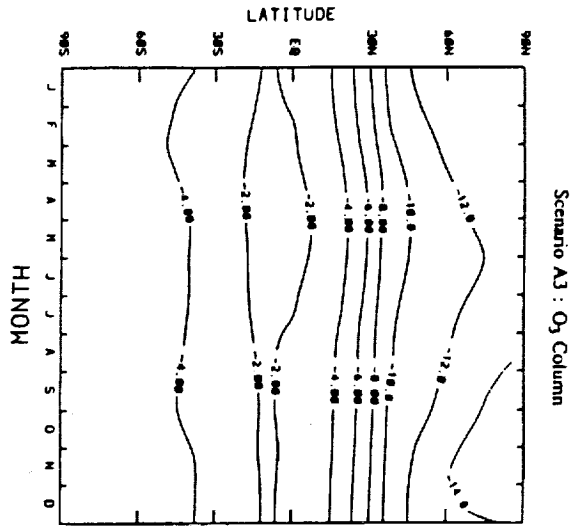


Figure 9. Calculated percentage change of ozone vertical column as a function of latitude and time of year, according to AER two-dimensional model. McDonnell-Douglas scenario A3, which gives 3/4 the nitrogen oxide injection as that for the LLNL model in Figure 8. The latitude of injection is mostly in the Northern Hemisphere between 30 and 60 degrees, but there is also significant injection in equatorial regions and in the Southern Hemisphere. [Ko et al., 1989, pages 29, 35, and 54].



Sensitivity of Two-Dimensional Model to Assigned Eddy Diffusion Coefficients

With the LLNL two-dimensional model, a test was made of its aircraft-ozone sensitivity to small variation of the assigned eddy diffusion coefficients [Kinnison, 1989]. Although the two-dimensional models include a substantial amount of atmospheric physics, there is need for some

Table IV. Comparison of calculated ozone column changes by two studies using two-dimensional models.

	North Polar	30N	30S
LLNL (Figure 8)	-16 %	-11 %	-5 %
AER x 1.33 (A3 scenario and Figure 9)	-19 %	-11 %	-4 %

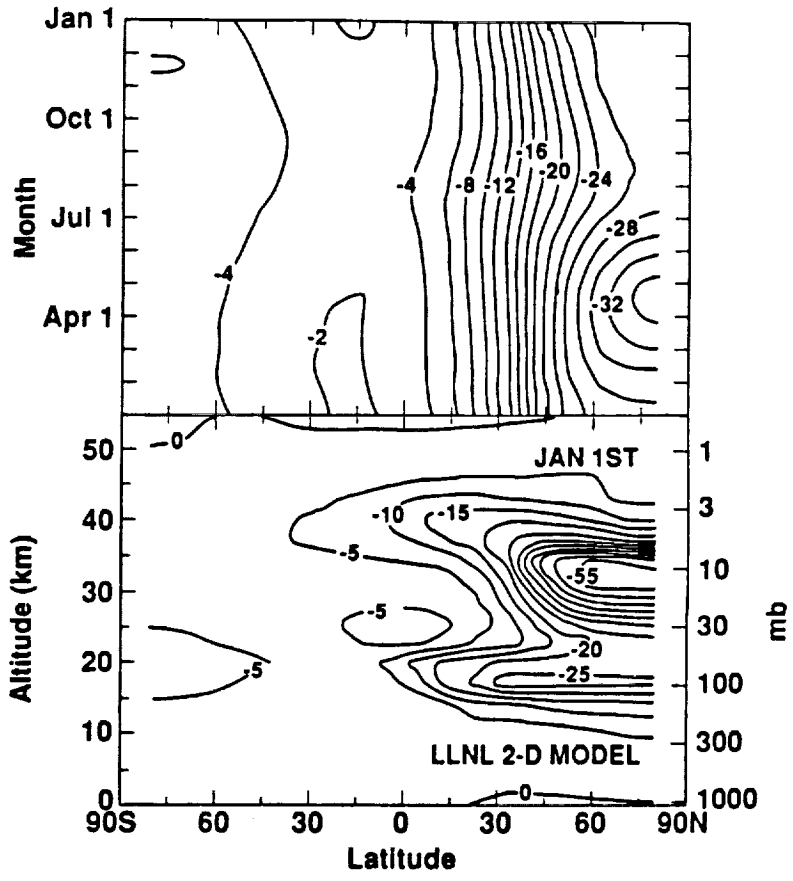


Figure 10. Calculated percentage change in ozone according to LLNL two-dimensional model for the injection of 1.8 MT NO_2 per year between 33 and 36 km altitude and between 37 and 49 degrees north: (upper panel), change in ozone vertical column as a function of latitude and season; and (lower panel), change in local ozone during January as a function of latitude and altitude. [Johnston, Kinnison, and Wuebbles, 1989].

residual empirical parameterization of atmospheric motions. D. J. Wuebbles, author of the LLNL two-dimensional model, proposed two equally plausible sets of such parameters, as given in Table V.

These two sets of parameters were used to calculate ozone changes from aircraft-injected NO_x (1.8×10^{12} grams "NO_2" per year) and to calculate the spread of carbon-14 during and after the

nuclear bomb tests of 1961-62 [Kinnison, Johnston, and Wuebbles, manuscript in preparation, 1990]. The parameters of Case B gave a distinctly better (but, even so, not very close) fit to the

Table V. Two cases of assigned K_{yy} and K_{zz} for study of aircraft-ozone perturbations in LLNL two-dimensional model [Kinnison, 1989, page 173].

Description	Case A	Case B
K_{yy}, troposphere	$1x10^{11}$ cm^2 s^{-1}	$5x10^{10}$ cm^2 s^{-1}
K_{yy}, stratosphere	$2x10^9$	$2x10^9$
K_{zz}, troposphere	$1x10^5$	$5x10^4$
K_{zz}, low-mid stratosphere	$1x10^3$	$1x10^3$
K_{yy} & K_{zz} transition near tropopause	Gradual	Relatively sharp
K_{yy} & K_{zz} variation with latitude and season	None	None

carbon-14 data, and Case B was used in the calculations reported by Johnston, Kinnison, and Wuebbles [1989]. For these two cases, a comparison between the calculated effect of aircraft NO_x on ozone for several altitudes of injection is given in Table VI. It can be seen that a small change of assigned eddy diffusion coefficients makes a 70-fold change in the calculated ozone reduction for injection of NO_x at 15-18 km and more than a two-fold change for injections at 18-21 km. This extremely high sensitivity of calculated ozone change to eddy diffusion coefficients in the critical 12 to 21 km range strongly calls for extreme effort in improving the physics in the models in this region and in calibration of the models against available, significant observables, especially radioactive tracers, chemical species, temperature, etc. Use of the radioactive tracers as observed from 1960 to 1970 is especially attractive in that they involve no chemistry, which complicates the use of reactive species as tracers of atmospheric motions.

NASA carried out a "Two-Dimensional Intercomparison of Stratospheric Models" in a workshop held in Virginia Beach, Virginia, September 1988 [Jackman et al., 1989]. These models have been developed primarily to calculate the effects of CFC's on ozone, and the region between 12 and 21 km was relatively unimportant. Very little of these intercomparisons contributed to calibrating the models for those features needed for the stratospheric aircraft problem. A quotation from page 33 illustrates the situation: the current typical (residual mean circulation) "RMC models appear to require a source of odd nitrogen in the upper troposphere, perhaps attributable to lightning. The alternative is to use much larger eddy-diffusion coefficients in the lower stratosphere (15-25 km) than currently believed reasonable, [i.e. $K_{yy} > 10^{10}$ vs. an average value of $3x10^9$]." This statement, including the rest of the workshop report, shows that two-dimensional models of 1988 are primitive relative to what is needed for the AESA/HSRP assessment. Soon atmospheric scientists may be asked to testify as to what altitude between 15 and 19 km it is "safe" (a policy maker's stated percentage ozone reduction) to operate a fleet of aircraft, even though this fleet is predicted to give an "unacceptable" (again, a policy maker's number) degree of ozone reduction if it operates at or above 20 km. At the present state of atmospheric modeling, atmospheric science is not ready to make such a statement. An aggressive program to improve our knowledge of

atmospheric dynamics and sufficiently to calibrate model results against atmospheric observables would probably make it possible for atmospheric scientists to give a useful answer to that question -- in, perhaps, 2 to 4 years.

Table VI. Calculated changes in global ozone vertical columns as a result of NO_x injections by stratospheric aircraft (24×10^{33} molecules NO_x per year). LLNL two-dimensional model. The boundary values include 300 ppbv of nitrous oxide and enough chlorofluorocarbons to give 2.8 ppbv of stratospheric chlorine at 50 km. Photochemical and chemical constants based on JPL 1987. Two different sets of eddy diffusion coefficients as given by Table V.

NO_x Injection Altitude/km	Latitude N	% Ozone Change Case A	% Ozone Change Case B
15-18	37-49	-0.01	-0.7
18-21	37-49	-3.3	-7.6
30-33	37-49	-8.0	-9.8
33-36	37-49	-7.2	-9.2
33-36	N Hemisphere	-7.1	-9.1
33-36	Global	-8.0	-9.8

F. DISCUSSION

The model calculations briefly presented above contribute to four out of seven components that will need to be addressed in the AESA/HSRP studies which are listed here:

(1) Number of aircraft and amount of fuel burned
(2) Emission index
(3) Cruise altitude
(4) Flight corridors
(5) Background chlorine, methane, nitrous oxide, carbon dioxide
(6) Heterogeneous chemistry as identified in Antarctic ozone hole
(7) Physics and chemistry of aircraft wake in stratosphere

These components are discussed here in light of the model calculations and of future options for high-speed stratospheric aircraft.

Number of aircraft and amount of fuel burned. Considering the large cost of developing and building a new kind of commercial aircraft [Ott, 1988; Boeing, 1989], it is unlikely that airframe builders would go ahead with a decision to construct unless prospects were good for several hundred (300 to 800, perhaps) copies eventually to be bought and used. The amount of fuel

Emission index. The greatest potential for reducing the expected ozone reduction by stratospheric aircraft [Johnston, Kinnison, and Wuebbles, 1989] is to reduce the NO_x emission index . As aircraft engines were modified for higher performance and greater fuel efficiency between 1971 and 1988, the "NO_x" emission index increased from 20-35 to 35-55 [Niedzwiecki, 1988]. The "*future goal of advanced emission reduction technology*" is to reduce the NO_x emission index to its "*lowest theoretical*" value of 5±2 by the year 1997 [Niedzwiecki, 1988]. McDonnell-Douglas [quoted by Ko et al., 1989] gives 39.5 as current (1988) NO_x emission index and 5.2 as future goal of NO_x emission index. By drastic redesign of jet engines, it is theoretically possible to have high performance and low nitric oxide emission [Boeing, 1989, pages 83-85]. However, Boeing [1989] has quotations from aircraft-engine engineers, who are pessimistic about the operability and the possibility of achieving the NO_x emission index of 5 grams per kg of fuel: "To reduce the NO_x emissions further, both P&W and GE have identified combustor concepts that they believe have the potential to achieve a cruise NO_x emissions index level of 5 or 10 rather than the 32 predicted for conventional combustor design .... Development and validation of the innovative combustor concepts will require a significant commitment to an aggressive research effort. Figure 3-13 [Pratt and Whitney, not shown] presents a lean premixed and prevaporized (LPP) combustor that has the potential for an 84 percent reduction in cruise NO_x emission index from 32.1 to 5.0 .... The GE innovative combustor was a LPP concept, and had a potential for a 61 percent reduction in cruise NO_x emission index from a baseline of 31 to 9. GE ... reported ... that it would be a high-risk development program with potential barriers to success being premixing duct flashback and auto-ignition; these potential problems are applicable to the P&W LPP concept as well" [Boeing, 1989, pages 41-42].

But what is the meaning of "*grams of NO_x per kg of fuel*" in the quotations above? Ferri [1972], in discussing nitrogen oxides from SST exhaust, said: "This amount varies from 15 to 70 grams of oxides per 1000 grams of fuel..." In most of the article, Ferri was explicit in referring to nitric oxide, NO, but this quotation is ambiguous as to whether it is nitric oxide, nitrogen dioxide, or both. In 1974, the Concorde was reported to have a nitric oxide emission index of 12 [CIAP, 1974, 1975] or a "NO_2 " emission index of 18, which is greater than that for nitric oxide by 46/30 = 1.53, the ratio of molar masses. Emission index as "equivalent NO" or "equivalent NO_2 " gives the same information, and from either statement one can calculate the number of molecules of NO_x = NO + NO_2 , the quantity needed for model calculations. However, over the last year and a half, statements made on the definition of "NO_x emission index" have been indefinite and conflicting. In measuring the nitrogen oxides emission index, NO and NO_2 are measured individually. As an example, assume that it is found that one kg of fuel produces x grams of NO and y grams of NO_2. There are three different usages of the expression "NO_x emission index" :

(1) Equivalent NO emission index = $x + (30/46)y$
(2) "NO_x" emission index = $x + y$
(3) Equivalent NO_2 emission index = $(46/30)x + y$

Expression (3) is about 1.5 times greater than expression (1), but either one is satisfactory if it is clearly and accurately defined. Expression (2) is chemically ambiguous. Although this matter remains unresolved, it appears that some of the aircraft industry use expression (2), some use expression (3), and it is a matter of priority to clarify the situation. If no attention is given to reducing nitric oxide from engine exhaust, the emission index of proposed future stratospheric aircraft would probably be about 40 grams of nitric oxide, NO, per kg of fuel. For proper perspective on this problem, it is recommended that ozone reduction calculations be made for at least the three *nitric oxide* emission indices: 40, 15, and 5, which are, respectively, the value that would probably exist if no attention is given toward reducing NO_x from engine exhaust, a standard value used during CIAP and regarded as achievable since it once existed, and the goal of future "advanced emission reduction technology." The effect of all three values of *nitric oxide* Emission

Index, 40, 15, and 5, should be evaluated and kept in mind until actual values from real engines are demonstrated.

[Editors' note, July 1990: The AESA/HSRP has adopted definition (3) above, equivalent NO_2 emissions, and has set EI = 5 as the programmatic goal for NO_x reductions. Further, AESA/HSRP has established a special subcommittee on emission scenarios that met on May 1 and July 12, 1990, and is preparing a report on aircraft emissions.]

Flight corridor. Although flight corridors are largely determined by population and market centers, certain aspects of their choice is an option in the ozone reduction problem. Very limited studies of this variable were made by the LLNL study, but some statements can be made based on various paired comparisons. So far as global average ozone reduction is concerned, there is little difference between midlatitude injection and uniform global injection. Injection at tropical latitudes causes somewhat greater global ozone reduction than similar injections at midlatitudes, and the explanation for this appears to be features in the two-dimensional LLNL model that are similar to the Brewer circulation, lifting and spreading the tropical injection of nitrogen oxides.

Background chlorine, methane, nitrous oxide, water, carbon dioxide. Ozone reduction by small injections of nitrogen oxides in the stratosphere are strongly affected by the background value of chlorine species (see Johnston, Kinnison, and Wuebbles, 1989, for a detailed study of this problem), but the ozone reductions caused by large injections of NO_x are almost unaffected by large changes of chlorine in the atmosphere (Figure 6). Large injections of water alone into the stratosphere have very little affect on stratospheric ozone, but the water and nitrogen oxides emitted together produce less ozone reduction by factors of 0.85 to 0.95 than that calculated for NO_x alone. Doubling methane reduces the calculated ozone reduction by aircraft injected NO_x by factors of 0.7 to 0.9, Table II. In making further model calculations, it is important to include water from the exhaust and to make realistic estimates of all of these background gases, including their probable future changes.

Heterogeneous chemistry as identified in Antarctic ozone hole. The model calculations from LLNL and AER are based on homogeneous atmospheric photochemistry and the 1987 data base for these quantities. As soon as two-dimensional models are able to include heterogeneous reactions and quantitatively to explain the evolution of the Antarctic ozone hole, this branch of chemistry should be included at all latitudes in models exploring the effect of stratospheric aircraft on ozone. At this time there are no firm grounds for predicting even the sign of the effect of this heterogeneous chemistry on the aircraft problem. The added NO_y may form stratospheric clouds and convert inactive HCl and chlorine nitrate into active ozone-destroying free radicals, and this process may increase the ozone reduction calculated by current models. On the other hand, consideration of the nitric-acid cloud-forming process may reveal that the oxides of nitrogen are removed from the stratosphere by fall-out of nitric acid ice more rapidly than the rate considered by current models, and the currently calculated ozone reductions would be reduced.

CIAP, Monograph 3, made a special study of the formation of contrails by stratospheric aircraft, as a function of altitude, latitude, and temperature. They were found to occur only rarely and preferentially near the tropical tropopause and in the polar winter lower stratosphere.

Chemistry of the plume. The composition of the aircraft plume is expected to be hot air, H_2O, CO_2, CO, unburnt hydrocarbons, NO, NO_2, HNO_3, SO_2, soot, lubricating oil, and trace metals. Detailed chemical modeling of the plume, including NO_x, HO_x, and Cl_y chemistry, was reported in CIAP Monograph 3, Chapter 2, pages 2-5, Table VII. The modeling was checked against measurements of a jet engine in a test tunnel, and photographs were taken of sub-sonic aircraft in the high troposphere and a Mach 2.4 craft in the stratosphere at about 19 km. Although the detailed model calculations reported in CIAP Monograph 3 [Anderson and Meyer, 1974; Anderson

et al., 1974] should be repeated using modern chemical coefficients, CIAP Monograph 3 has much insight pertinent to these problems in the 1990's.

G. CONCLUSIONS AND SCIENTIFIC RECOMMENDATIONS

The salient fact in this area is that state-of-the-art 1990 atmospheric models predict that a commercially viable fleet of Mach 2.4 aircraft cruising at about 20 km and using current (1990) technology jet engines would emit so much nitrogen oxides into the stratosphere that global ozone would be reduced about 15 percent . This calculated ozone reduction is far greater than that calculated for chlorofluorocarbons if the emissions are continued at present rates. It should be noted for the case of CFC's that model predictions of ozone depletion did not include the Antarctic ozone hole. However, these predictions of moderate ozone loss by the year 2060 (4-12 percent at high latitudes in early spring [WMO, 1990]) coupled with observations of the ozone hole has led to international agreements (Montreal Protocol, 1987) to reduce and eventually to phase out CFC production. This comparison is not a worst-case scenario for supersonic aircraft, but rather it is the motivation for the necessary research programs like AESA/HSRP.

Representatives of NASA and the aircraft industry agree that one objective of the High-Speed Research Program is to "establish technologies and operational procedures that insure no significant ozone depletion." The promising options for this purpose are: (A) using "advanced emission reduction technology" to design and build a new type of aircraft engine that will greatly reduce NO_x emissions and (B) to fly the SST's at low Mach number and low stratospheric altitudes where ozone is relatively unaffected by added nitrogen oxides. There may be other options such as the unknown effects of heterogeneous chemistry.

(1) *NO_x emission index.* As demonstrated earlier in this paper, there is confusion as to the meaning of the term "NO_x emission index." The literature on this subject appears not to be in refereed abstracted journals but in difficultly accessed company, agency, ASME, etc. reports, and we need to obtain and make available the reports that give the experimental methods, computations, and calibrations used to determine NO, NO_2, and possibly N_2O and HNO_3 in engine exhaust. Modelers should be assured that their model input of emission index can be traced back to its measured origin.

(2) *Two-dimensional stratospheric models with emphasis on the 12- to 21- km range.* All atmospheric models in 1990 that include methane chemistry probably will agree that NO_x injected at about 12-km altitude will not decrease the ozone column (Figures 2 and 7) and that large injections of nitrogen oxides at 21-km altitude will strongly reduce the ozone column (see Tables III and IV). The 9-km interval between 12 and 21 km spans the great complexities discussed by Danielson in 1971. This 9-km range spans only three steps of vertical resolution in many (not all) 1990 two-dimensional models. For example, the LLNL two-dimensional model calculates 7.6 percent global average ozone reduction for a given NO_x injection between 18 and 21 km, but only a 0.7 percent global average ozone reduction for the same injection between 15 and 18 km (Table III). This 11-fold difference between adjacent vertical cells clearly shows that 3-km vertical resolution is not good enough for this altitude range. Furthermore, the eddy diffusion function for the LLNL model was modestly adjusted to improve agreement with carbon-14 data, and there was a 70-fold change in its prediction of the ozone reduction caused by aircraft flying between 15 and 18 km. The first priority should be to improve the vertical resolution, atmospheric dynamics, and calibration of the two-dimensional models, especially in the 12- to 21-km range. Although it may be difficult to model this region from first principles of atmospheric physics, that should be

done as far as possible, and, supplementing this, there are a number of atmospheric properties in this region for testing model parameterizations, such as:

(a) Excess carbon-14 between 1961 and 1970 [Johnston, 1989; Kinnison, 1989] and other nuclear bomb test tracers.

(b) *In situ* vertical profiles of ozone and appropriate satellite ozone data for the lower stratosphere. (The Umkehr method and Solar Backscattered Ultraviolet satellite data do not have adequate vertical resolution).

(c) Observed vertical profiles at many latitudes and seasons for temperature, nitric acid, hydrochloric acid, hydrogen fluoride, aerosols of the Junge layer, water, nitrous oxide, chlorofluorocarbons, methane, etc.

In 1990, I have some confidence in the ozone changes that the models will predict for NO_x injection below 12 and above 21 km, but I believe no model is good enough to support a policy decision as to where between 14 and 19 km it may be safe to emit large amounts of NO_x.

Within 2 to 4 years, atmospheric scientists and modelers, if given strong support, could probably develop models, spanning 0 to 50 km, that are adequate for this job. We need "advanced stratospheric modeling technology" as much as we need "advanced emission reduction technology."

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NASA
National Aeronautics and Space Administration

Report Documentation Page

1. Report No.	2. Government Accession No.	3. Recipient's Catalog No.
NASA RP-1250		

4. Title and Subtitle
The Atmospheric Effects of Stratospheric Aircraft: A Topical Review

5. Report Date
January 1991

6. Performing Organization Code
SEU

7. Author(s)
H. S. Johnston,* M. J. Prather, and R. T. Watson

8. Performing Organization Report No.

10. Work Unit No.

9. Performing Organization Name and Address
NASA Office of Space Science and Applications
Earth Science and Applications Division

11. Contract or Grant No.

13. Type of Report and Period Covered
Reference Publication

12. Sponsoring Agency Name and Address
National Aeronautics and Space Administration
Washington, DC 20546

14. Sponsoring Agency Code

15. Supplementary Notes
*Harold S. Johnston, University of California, Berkeley, Calif.

16. Abstract

In the late 1960s the aircraft industry became interested in developing a fleet of supersonic transports (SSTs). Between 1972 and 1975, the Climatic Impact Assessment Program (CIAP) studied the possible environmental impact of SSTs. For environmental and economic reasons, the fleet of SSTs was not developed.

The Upper Atmosphere Research Program (UARP) has recently undertaken the responsibility of directing scientific research needed to assess the atmospheric impact of supersonic transports. The UARP and the High-Speed Research Program asked Harold Johnston to review the current understanding of aircraft emissions and their effect on the stratosphere. Johnston and his colleagues have recently re-examined the SST problem using current models for stratospheric ozone chemistry. This report provides a unique view of the current scientific issues and the lessons learned since the beginning of CIAP, and it links the current research program with the assessment process that began two years ago.

17. Key Words (Suggested by Author(s))	18. Distribution Statement
stratospheric chemistry, ozone, upper atmosphere research, stratospheric aircraft, high-speed civil transport	Unclassified - Unlimited Subject Category 45

19. Security Classif. (of this report)	20. Security Classif. (of this page)	21. No. of pages	22. Price
Unclassified	Unclassified	36	A03

NASA FORM 1626 OCT 86

NASA-Langley, 1991

For sale by the National Technical Information Service, Springfield, VA 22161-2171