CHAPTER 9

Predicted Aircraft Effects on Stratospheric Ozone

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Chapter 9

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SCIENTIFIC SUMMARY

Engine emissions from subsonic and supersonic aircraft include oxides of nitrogen (NO_x), water vapor, unburned hydrocarbons, carbon monoxide, carbon dioxide, and sulfur dioxide. Addition of NO_x to the atmosphere is expected to decrease ozone in the stratosphere and increase ozone in the troposphere. Resulting changes in ozone, water vapor, and aerosol loading in the altitudes around the tropopause may have a climatic impact since the response of radiative forcing to changes in concentrations is most sensitive here.

The first step in assessing these effects is to determine quantitatively the changes in background concentrations associated with emitted gases and aerosols. Most of the emissions from the projected supersonic fleet as well as one-quarter to one-half of the emissions from the subsonic fleet are deposited directly into the lower stratosphere, to be redistributed by large-scale transport. Current models cannot accurately simulate the accumulation of emitted materials in the stratosphere and their eventual removal, as much of the injection is close to the tropopause. Model predictions in this chapter are based on simulations from two-dimensional models that assume that the emitted material is zonally mixed. The impact of flight corridor effects on the predictions has not been assessed.

The current fleet of subsonic aircraft may be sufficiently large to have increased background concentrations of NO_x, ozone and the sulfate layer. Gas phase modeling studies predict that the current subsonic aircraft fleet (injection of 2 $Tg(NO_2)$/year) contributes to 5–10 percent of the total amount of ozone in the troposphere around 40°N. These models also predict a decrease in ozone in the lower stratosphere of less than 1 percent. Projected increases in the subsonic fleet could lead to further increases of tropospheric ozone with associated changes in OH and additional (although still small) reduction of ozone in the stratosphere. These estimates are subject to considerable uncertainties due to uncertainties in the magnitude and distribution of emissions and to differences among models. The role of heterogeneous chemical processes in the troposphere needs to be assessed.

The additional impact on ozone from projected fleets of supersonic aircraft [high-speed civil transports (HSCT)] operating in the year 2015 (cruise altitudes around 15, 19, and 22 km) has been examined by a model intercomparison exercise using gas phase models. With the operation of aircraft assumed to be concentrated in the Northern Hemisphere, the largest decrease in local ozone is found poleward of 30°N around the cruise altitude. There is very little increase in upper tropospheric ozone due to HSCT alone because of the reduced ozone flux from the stratosphere. For cruise altitudes below 28 km, the calculated decrease in column abundance of ozone is larger for higher cruise altitudes and larger NO_2 emission indices. The calculated decrease in the Southern Hemisphere is typically a factor of 2 to 3 smaller than that in the Northern Hemisphere. In one of the cases (a fleet of aircraft flying Mach 3.2 between 21 and 24 km with NO_2 emission index of 15 and annual fuel use of 70 Tg per year), the calculated decrease in ozone column abundance at northern mid-latitudes ranges from 7 percent to 12 percent. This spread in model results increases with decreasing cruise altitude and reflects differences in transport and photochemical balance in the various models. In the Mach 2.4 case (cruise altitude of 17-20 km), the range of model-calculated decreases in column abundance in the same region is 2 to 6 percent.

Heterogeneous reactions occurring on polar stratospheric clouds (PSCs) or the global sulfate aerosol layer have a large impact on the ozone chemistry in the lower stratosphere, and hence, the predicted response to supersonic aircraft. If N_2O_5 is converted to HNO_3 via known heterogeneous reactions on the global sulfate layer, the column changes for the Mach 2.4 case from two of the models are calculated to be small (–0.5 to +0.5 percent). At the same time, there is an increase in upper tropospheric ozone of about 5 percent due to HSCT alone. Nevertheless, there could be large and unpredictable changes in ozone if emissions from the aircraft cause enhanced formation of PSCs or increases in the sulfate aerosol loading, leading to repartitioning of the chlorine species and higher concentrations of ClO.

9.1. INTRODUCTION

The possibility that the current fleet of subsonic aircraft may already have caused detectable changes in both the troposphere and stratosphere has raised concerns about the impact of such operation on stratospheric ozone and climate. Recent interest in the operation of supersonic aircraft in the lower stratosphere has heightened such concerns. Previous assessments of impacts from proposed supersonic aircraft have been based mostly on one-dimensional model results although a limited number of multidimensional models were used (CIAP, 1974; COMESA, 1975; COVOS, 1976; Sundararaman, 1984). In the past 15 years, our understanding of the processes that control the atmospheric concentrations of trace gases has changed dramatically (WMO, 1986; WMO, 1990). This better understanding has been achieved through accumulation of kinetic data and field observations as well as development of new models. It would be beneficial to start examining the impact of subsonic aircraft to identify opportunities to study and validate the mechanisms that have been proposed to explain the ozone responses.

The two major concerns are the potential for a decrease in the column abundance of ozone leading to an increase in ultraviolet radiation at the ground, and redistribution of ozone in the lower stratosphere and upper troposphere leading to changes in the Earth's climate. Two-dimensional models (as discussed in Chapters 6 and 8) have been used extensively for ozone assessment studies, with a focus on responses to chlorine perturbations. There are problems specific to the aircraft issues that are not adequately addressed by the current models. This chapter reviews the current status of the research on aircraft impact on ozone with emphasis on immediate model improvements necessary for extending our understanding. The discussion will be limited to current and projected commercial aircraft that are equipped with air-breathing engines using conventional jet fuel. The impacts are discussed in terms of the anticipated fuel use at cruise altitude.

9.2 IMPACT OF AIRCRAFT OPERATION ON ATMOSPHERIC TRACE GASES

Various materials are introduced into the atmosphere associated with the operation of aircraft. Such operation can perturb the ambient concentrations of atmospheric trace gases either by direct injection of specific trace gases (such as H_2O and oxides of nitrogen) or through photochemical reactions of the injected material (such as impact on ozone from injected oxides of nitrogen and nonmethane hydrocarbons). Current analysis indicates that engine effluents have the largest relative impacts while contributions from leakage of fluids (hydraulic and sanitation), fuel dump, chipped paint, and erosion of other components are apparently trivial (see Chapter 2, HSRP [1992]).

9.2.1 Emission Indices

Emissions from engine types are specified in terms of the emission index (EI) for each material, defined as grams emitted per kilogram of fuel burned. Typical EI values for various materials are summarized in Table 9-1. The amount of H_2O and CO_2 emitted associated with fuel combustion are determined by stoichiochemistry with minor variations depending on the saturation of the fuel. Emissions associated with incomplete combustion (CO, unburned hydrocarbons) depend on engine design and operating conditions. Additional emissions such as sulfur and trace metals may come from impurities in the fuel and are conserved during the combustion process. The emitted material that has the largest impact on ozone is oxides of nitrogen (NO_x = NO + NO_2), which are produced predominantly from the reaction of N_2 and O_2 at high temperature in the combustion chamber. NO_x in the engine effluent is typically 85 percent NO and 15 percent NO_2. There has been some confusion on the definition of EI(NO_x) as tailpipe emission for NO has been reported sometimes as mass of NO_2 equivalent without explicitly stating so. The EI for NO_x is defined here in terms of grams of NO_2 equivalent emitted, *i.e.*, 46 gm times the total number of moles of NO plus NO_2 emitted per kilogram of fuel burned. The large range of values quoted for CO, HC, and NO_x in Table 9-1 is mainly caused by different power settings during take off, climb, cruise, and descent. It is interesting to note that the evolution of engine design (more efficient, cleaner burning, high-temperature combustion) in the past decades has decreased EIs for CO and hydrocarbons and increased EI(NO_x).

Table 9-1 Emission index (grams per kilograms of fuel used) of various materials for subsonic and supersonic aircraft for cruise condition. Values in parenthesis are ranges for different engines and operating conditions.

Species [gm molecular weight]	Subsonic Aircraft* Short-Range	Subsonic Aircraft* Long-Range	Supersonic Aircraft†
CO_2 [44]	3160	3160	3160
H_2O [18]	1230	1230	1230
CO [28]	5.9 (0.2-14)	3.3 (0.2–14)	1.5 (1.2–3.0)
HC as methane [16]	0.9 (0.12-4.6)	0.56 (.12–4.6)	0.2 (0.02–0.5)
SO_2 [64]	1.1	1.1	1.0
NO_x as NO_2 [46]	9.3 (6-19)	14.4 (6–19)	Depends on design (5–45)

*Mean (fuel-consumption weighted) emission indices for 1987 based on Boeing (1990). The values were calculated from a data base containing emission indices and fuel consumptions by aircraft types. The difference between short-range (cruise altitude around 8 km) and long-range (cruise altitude between 10 and 11 km) reflects different mixes of aircraft used for different flights.

†Based on Boeing (1990) and Douglas (1990).

9.2.2 Estimates of Perturbations to Background Concentrations

The extent to which the background concentration of a particular trace gas is perturbed by direct injection from aircraft can be obtained by comparing the injection rate with the local budget of the trace gas (*i.e.*, *in situ* photochemical production rate and transport divergence) in the region. The expected increase in concentration is related to the injection rate by the local residence time of the species. For a relatively inert gas injected into the lower stratosphere near the tropopause, the residence time in the stratosphere is about 1 year. If the material is deposited farther away from the tropopause, the residence time is closer to 2 years. Residence time in the upper troposphere is expected to be shorter (1–3 weeks) because of more efficient vertical overturning. Strat-trop exchange also plays an important role in this case because material injected in the tropical upper troposphere could be transported to the stratosphere where the residence time is much longer.

Although estimates exist for the annual mass exchange between the troposphere and the stratosphere, there are large uncertainties concerning the processes responsible for the exchange process (See review in Chapter 5, WMO [1986]). It is particularly important to understand those processes for aircraft operations because specific flight corridors could be adjusted systematically according to local meteorological conditions. For instance, the transverse circulation thought to be associated with jet streams (Krishnamurti, 1961; Mahlman, 1973) would imply downward motion on the cyclonic (poleward) side of the jet core. This would imply that material deposited poleward of the jet core would have shorter residence time than material injected equatorward of the jet core.

The residence time in the lower stratosphere (estimated to be about 1 year) is long enough for the emitted NO_x to be repartitioned to other forms of total odd nitrogen (NO_y) species as dictated by the local conditions. Thus, the NO_x emitted in the stratosphere could be treated as NO_y in model simulations. In contrast, NO_x deposited in the troposphere will remain as NO_x during the 1–3 weeks it stays in the atmosphere. Given the ambient concentrations of various trace gases in the atmosphere, values from Table 9-1 imply that the perturbation on a percentage basis is largest for NO_y and H_2O in the stratosphere. Emitted NO_x is also expected to have a large impact in the upper troposphere. Although any unburned HC may form a large local source of a specific hydrocarbon, its effect on atmospheric chemical cycles is small compared to CH_4.

Recently, Hofmann (1990) suggested that the stratospheric sulfate burden may have been increasing by 5 percent per year in the last decade.

Hofmann (1991) pointed out that fuel use from subsonic aircraft has also increased at the same rate during that period, suggestive that sulfur emission from aircraft engines could be a cause. Estimates based on EI for sulfur are not inconsistent with such a suggestion. However, tests of such ideas must await more careful studies incorporating microphysics of aerosol growth that can translate sulfur emissions to changes in sulfate loading. At present, the budget of the stratospheric sulfate layer is not well understood. Various theoretical estimates suggested that 8×10^7 kg of sulphur per year is needed to sustain the sulfate layer under background (nonvolcanic) conditions. The EI for SO_2 given in Table 9-1 would imply that fuel use in the stratosphere over 10^{11} kg/year would be important (>50 percent) if all the sulfur were deposited in the stratosphere. However, it should be noted that the value cited in Table 9-1 is an upper limit based on fuel standards for engines. Actual sulfur content in the fuel is expected to be smaller since industry prefers low-sulfur fuel, which is less corrosive to the engines.

Other products from engine combustion include carbon soot particles and condensation nuclei (CN). The number of CN has been estimated to be 2–4 x 10^8 particles per kilogram of fuel used (Douglas, 1989). Whether this may lead to additional formation of aerosol particles or polar stratospheric clouds is unclear since such processes are not well understood. It is believed that addition of carbon soot (light-absorbing) particles may also change the optical properties of the atmosphere.

9.3 MODEL STUDIES OF OZONE RESPONSE

As pointed out by Johnston (1971) and Crutzen (1971), increases in the stratospheric concentration of NO_x from engine effluent will lead to an increase in the NO_x-catalyzed ozone removal by the reactions:

$$\begin{aligned} &NO + O_3 \rightarrow NO_2 + O_2 \\ &\underline{NO_2 + O \rightarrow NO + O_2.} \\ &O + O_3 \rightarrow 2O_2, \text{ catalyzed by } NO_x \end{aligned} \qquad (9\text{-}1)$$

However, increases in NO_x may also affect other chemical cycles, leading sometimes to compensating effects. In the troposphere, the increases in NO_x will enhance the following reactions:

$$\begin{aligned} &NO + RO_2 \rightarrow NO_2 + RO \\ &NO_2 + h\nu \rightarrow NO + O \\ &\underline{O + O_2 \rightarrow O_3} \\ &RO_2 + O_2 \rightarrow RO + O_3, \text{ catalyzed by } NO_x \text{ and by } HO_x \text{ and by photons} \end{aligned} \qquad (9\text{-}2)$$

leading to net ozone production. (See Chapter 5 for a more detailed discussion.)

The exact response of ozone depends on the detailed balance of the competing chemical cycles and transport in different regions of the atmosphere. This is one reason why a multidimensional model is necessary for a realistic assessment of the ozone impact. Most of the results presented in this chapter are based on results from two-dimensional models discussed in Chapters 6 and 8 of this report. In interpreting these results, one must be aware of uncertainties normally associated with these models as well as additional points that are specific to the aircraft problem (see reviews by Douglass *et al.*, 1991; Johnston *et al.*, 1991). These are :

- Since the injection is very close to the tropopause, the results are particularly sensitive to how strat-trop exchange is treated in the models. This has not been a major focus of previous efforts in two-dimensional model development.
- Once the engine effluent is deposited along the flight path, it is subject to mesoscale transport before it becomes zonally mixed. In addition, with emissions concentrated at heavily-traveled flight corridors, the emitted material may retain zonally asymmetric features. Questions can be raised whether current two-dimensional models can adequately simulate these effects.
- The response of ozone to emitted NO_x will change if heterogeneous chemistry is occurring. Furthermore, H_2O and NO_x emissions may promote formation of polar stratospheric clouds that activate chlorine-catalyzed ozone depletion. Sulfur emission may lead to enhancement of the sulfate layer. These changes may cause repartitioning of the existing chemical species in the atmosphere and result in a change in ozone unrelated to other injected chemicals.

9.4 EFFECT OF SUBSONIC AIRCRAFT ON OZONE

We will restrict the review to the 10–12 km region near the extratropical tropopause where the emissions from cruising subsonic fleet are expected to have the largest impact. The effect in the lower tro-

posphere is more difficult to predict because of the much larger variabilities in both local conditions and emissions from the aircraft during ascent and descent.

9.4.1 Latitudinal Distribution of Flight Operations and Fuel Use

The current flight scenario was estimated in the Boeing report (Boeing, 1990) based on the 1987 Official Airline Guide (OAG) (see also Chapter 4 in HSRP [1992]). The OAG contains published commercial jetliner flights with the exception of the domestic former Soviet Union, Eastern Europe, and Chinese flights. Short-range travel (< 400 miles) was assumed to occur around altitudes of ≈26,600 ft (8 km). All long-range flight traffic was assumed to occur around 33,000 to 37,000 ft (10 to 11.2 km). The latitudinal distributions of fuel burned are shown in Figure 9-1 for latitudinal bands of 10°. Note that 93 percent of the long-range cruise occurs in the Northern Hemisphere.

Based on the 1987 OAG, the Boeing report estimated the figures for fuel burned for the 1987 fleet to be 1.14 x 10^{10} kg/year for the short-range cruise and 7.24 x 10^{10} kg/year for the long-range cruise. The world consumption of aviation fuel in 1987 was 15.3 x 10^{10} kg (International Energy Annual, 1988). Nuesser and Schmitt (1990) reported a figure of 15.0 x 10^{10} kg for 1988 based on ICAO statistics and information supplied by Lufthansa. Therefore, the Boeing report accounts only for 53 percent of all aviation fuel burned in 1987. HSRP (1992) has estimated that aviation in the former Soviet Union and Eastern Europe could account for 12 percent of the world total, China for 2 percent, and U.S. military and private jet use for 7 percent each. The remaining 19 percent of the world jet fuel usage could be assigned to the sum of charter, cargo, and turboprop aircraft. For comparison, 11 percent of the German Lufthansa fleet fuel consumption was used for cargo flights in 1988 (Reichow, 1990).

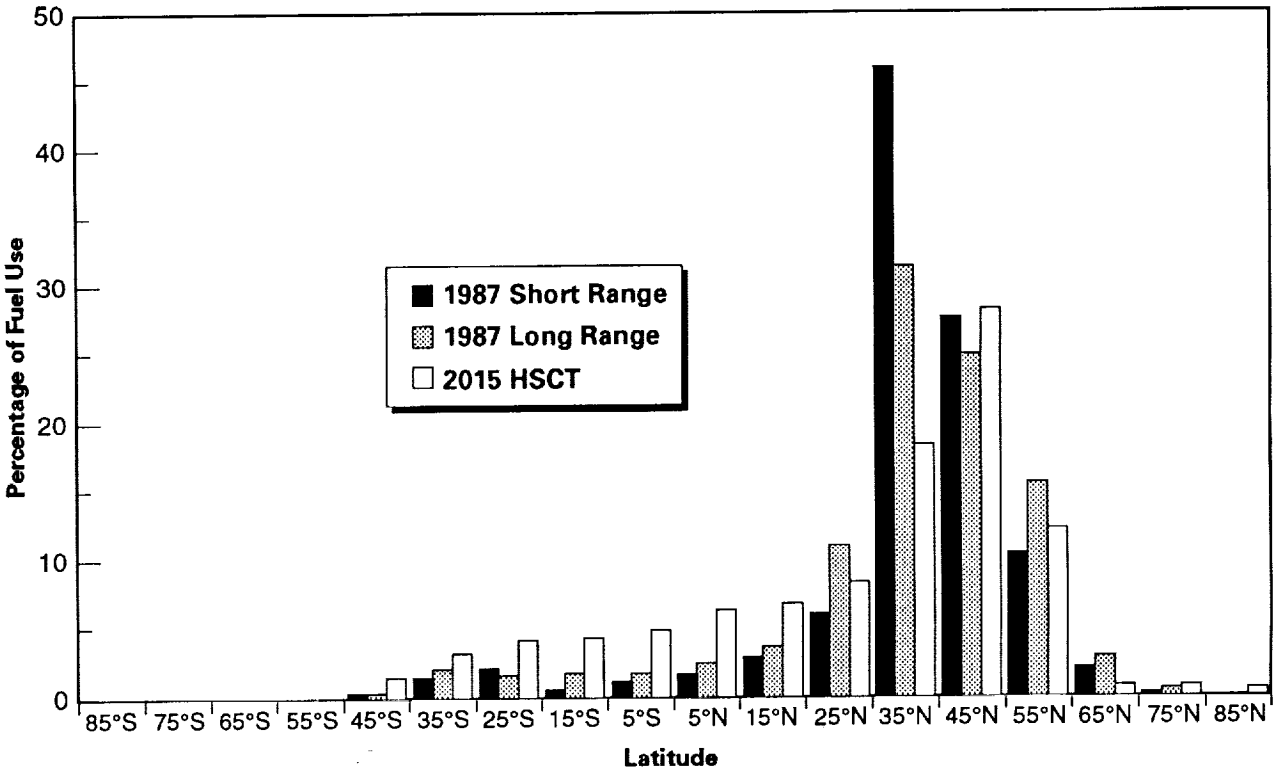

Figure 9-1 Fractional distribution of fuel use as a function of latitude band for 1987 commercial jet air traffic and projected 2015 HSCT aircraft. The fractional distributions sum to 100 percent for each of the three cases.

Table 9-2 Estimates of percentage of fuel burn in the stratosphere for the subsonic fleet.

Month	Cruise Altitude = 10 km		Cruise Altitude = 11 km	
	Latitude of tropopause *λ_T at 10 km (°N)	Portion of Northern Hemisphere fuel burn that occurs in the stratosphere (percent)	Latitude of tropopause *λ_T at 11 km (°N)	Portion of Northern Hemisphere fuel burn that occurs in the stratosphere (percent)
January	38	56	33	72
February	42	41	33	72
March	43	40	34	72
April	46	27	35	72
May	47	26	40	56
June	55	16	45	40
July	65	2	55	16
August	65	2	55	16
September	64	2	50	26
October	58	8	45	40
November	52	16	45	40
December	49	27	40	56
Annual†		22		48

*Based on atmospheric cross sections for the year 1980 (Danielsen *et al.*, 1983).
†Assume same fuel burn for each month

9.4.2 Allocation of Fuel-Related Emission to Troposphere and Stratosphere

The cruise altitude, 10–11 km, is within the stratosphere during certain times of the year. Table 9-2 gives the values for the latitude (λ_T) in the Northern Hemisphere where the tropopause is below and above the cruising altitude to the north and south of λ_T, respectively. Also given is the percent of fuel burned in the Northern Hemisphere that occurs in the stratosphere based on the fuel use distribution given in Figure 9-1. Table 9-2 reveals that, over the course of a full year, 22 and 48 percent of the 1987 Northern Hemisphere fuel burn occurs in the stratosphere for a cruise altitude of 10 and 11 km, respectively. This corresponds to an injection of about 0.2 to 0.5 x 10^9 kg(NO_2) in the Northern Hemisphere stratosphere. The result illustrates that the estimates critically depend on the assumed cruise altitude. A recent case study reported by Schumann and Reinhardt (1991) utilized actual flight paths and tropopause heights along the flight paths as determined by a numerical weather prediction model to calculate the fraction of the time spent in the stratosphere by the flights between Frankfurt and New York in December 1990 and June 1991. The results show that the time fraction depends on actual flight routing designed to avoid the jet stream. Future analysis should take into account these routing practices.

9.4.3 Estimates of Impact on Ozone

9.4.3.1 Impact on Stratosphere

Since part of the flight operation actually occurs within the stratosphere, stratospheric NO_x can be perturbed by materials directly deposited in the stratosphere as well as transport of engine effluents deposited in the tropical troposphere. The amount of NO_x transported from the troposphere is estimated to be small and confined to tropical and subtropical latitudes. Previous work of Ko *et al.* (1986) used a two-dimensional zonal mean model to estimate the bulk transport of lightning-generated NO_x to the lower stratosphere. Results from the same model indicate that the current subsonic fleet may have increased the NO_y concentrations in the lower stratosphere in the Northern Hemisphere by 0.5 ppbv. It is unclear whether measurements can detect an interhemispheric NO_y difference of 0.5 ppbv in the lower stratosphere and ascribe it to specific causes. One may expect a hemispherical asymmetry (with more NO_y

and H_2O in the Northern Hemisphere) to result from the export of denitrified and dehydrated air from the Antarctic ozone hole. Results from the Ko *et al.*, model and from Wuebbles and Kinnison (1990) show that the effect on ozone is small with a decrease in local ozone of less than 1 percent. Clearly, these results can only be considered as rough estimates, as two-dimensional models have limited utility in the description of the small scale features of strat-trop exchange as evident from the fact that they are not capable of accurately modeling the observed water vapor concentration in the lower tropical and subtropical stratosphere.

9.4.3.2 Impact on the Troposphere

The impact of aircraft emissions on the troposphere is to increase the NO_x content of the upper troposphere and to change the ozone concentration. Since ozone production in the current atmosphere is believed to be NO_x-limited, the effect of NO_x emissions is to enhance the ozone concentration.

Kley *et al.* (1981) measured the altitude distribution of NO_x in the upper troposphere and showed that input of NO_x from the stratosphere plus aircraft emissions are large enough to produce the observed increase with altitude of NO_x. Ehhalt *et al.* (1992) have compared measured profiles of NO with NO_x profiles that were calculated based on emission from ground-based and aircraft sources plus natural sources using a simple two-dimensional model. Figure 9-2 shows their measured NO data that were obtained during quasi-meridional flights close to the east coast of North America and to the west coast of Europe. Based on EI(NO_x) = 10, they concluded that 30–40 percent of the present-day tropospheric NO_x between 6 and 10 km at northern mid-latitudes results from aircraft emissions. Their figure would increase to 45–60 percent if the values for EIs from Table 9-1 are used.

The effect of aircraft on tropospheric ozone was addressed by Hidalgo and Crutzen (1977), Liu *et al.* (1980), Isaksen (1980), and Derwent (1982). More recent studies include work by Crutzen and Bruehl (1990), Wuebbles and Kinnison (1990), Beck *et al.* (1992), and Johnson and Henshaw (1991). Although the amount of NO_x emissions from aircraft is small compared to surface NO_x emission, the effect on tropospheric ozone and global warming is significant because the amount of ozone produced per unit emission is some 20 times larger for aircraft emission than surface emission, and the change in ozone occurs at altitudes of maximum radiative response (Johnson *et al.*, 1992).

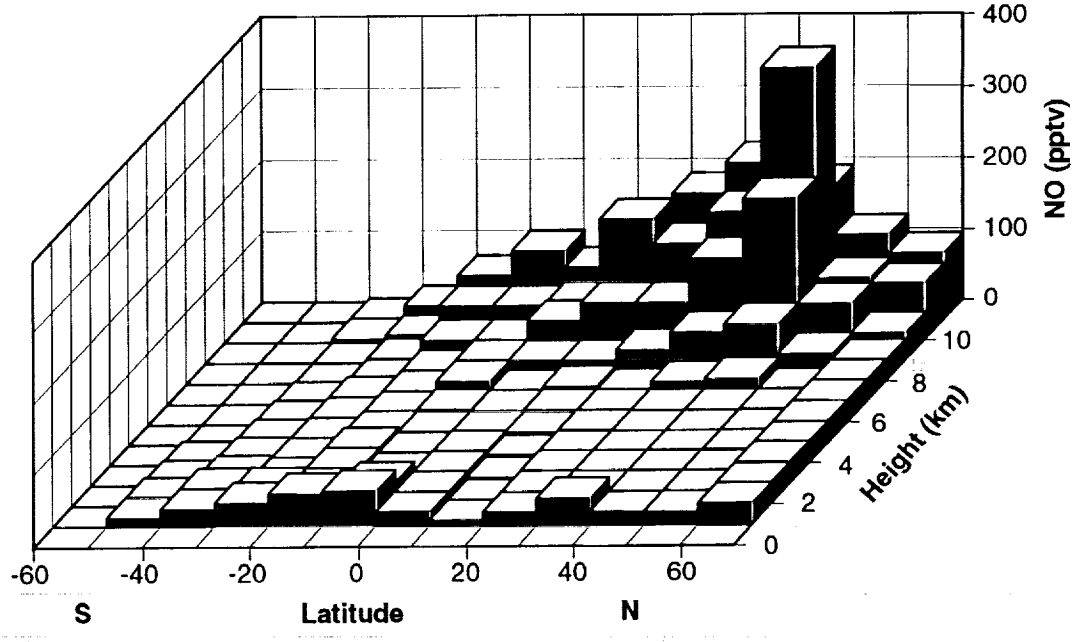

Figure 9-2 Observed distribution of NO from quasi-meridional flights close to the east coast of North America and to the west coast of Europe. The results are reported in Ehhalt *et al.* (1992).

Typical model results indicate that local ozone increases of 3–12 percent between 8 and 12 km for NO_x injections of 1.5–2.0 x 10^9 kg of NO_2 per year. There are associated changes to OH and other species. There has been no concerted effort to understand the differences among the models and there are considerable differences in the distribution of the emissions with height and latitude. Current research effort is being directed to improve the quality of the emissions data base. Finally, the role of heterogeneous reactions in the troposphere has not been explored.

9.5 EFFECT OF SUPERSONIC AIRCRAFT ON OZONE

There are several recent studies of the effects of high-speed aircraft (Johnston *et al.*, 1989; Ko *et al.*, 1991). It is difficult to compare the results since the emission scenarios are different in each study. The High Speed Research Program (HSRP) from National Aeronautics and Space Administration (NASA) organized a model intercomparison workshop in which the modelers were asked to use the same input to perform the simulations. The discussion in this section is based on the results of that workshop. The reader is referred to the workshop report (HSRP, 1992) for additional information.

As an assessment, the result presented here is necessarily interim. These model calculations should be viewed more as sensitivity studies, primarily designed to serve the following purposes:

- Identify parameters that are needed to characterize the ozone response,
- Allow for intercomparison of model predictions,
- Focus on the range of fleet operations and engine specifications giving minimal environmental impact, and
- Provide the basis for future assessment studies.

The basic scenarios were chosen to be as realistic as possible using available information on anticipated technology. They are not to be interpreted as a commitment or goal for environmental acceptability.

It should be emphasized that the calculations reported in HSRP (1992) are performed using gas-phase chemistry only. Heterogeneous chemistry occurring on the global sulfate layer (Weisenstein *et al.*, 1991) and the PSCs is expected to modify the results in a significant way. These effects could be further enhanced if there is an increase in aerosol loading or occurrence of PSCs due to the operation of the HSCT.

9.5.1 Parameters That Affect the Calculated Ozone Response

9.5.1.1 Emission Scenarios

For two-dimensional models that simulate the zonal mean (averaged over longitude) distributions of the trace gases, one must specify the distributions of the emitted materials as functions of latitude, height, and season. The calculations were performed for supersonic fleets with cruise fuel use of 70 x 10^9 kg/year. This corresponds to a realistic fleet of approximately 500 or more aircraft that represents an economically feasible size for the HSCT fleet. Fuel use during takeoff, climb, and descent was ignored in these calculations. The adopted latitudinal distribution of fuel use is given in Figure 9-1. Detailed distribution for any specific fleet will of course depend on flight routes, anticipated demands between city pairs, and routing to avoid sonic booms over land. The chosen distribution is based on two independent studies that take into account each of these concerns (Boeing, 1989, 1990; Douglas, 1989, 1990).

The fuel use is distributed according to projected flight paths and the emitted materials are assumed to be deposited along the flight paths. No adjustment is made to account for the vertical and meridional transport of the plume that may occur in the first few weeks before the emitted material becomes zonally mixed. Aircraft with particular cruise speeds (Mach numbers) operate most efficiently at specific altitudes. The adopted pressure-altitude ranges for each Mach number are 14–17 km for Mach 1.6 cruise, 17–20 km for Mach 2.4 cruise and 21–24 km for Mach 3.2 cruise. The assigned spread in altitude is in accord with possible traffic control and the natural climb of cruise altitudes towards the end of a trip as the fuel is being used up. Fuel use and emissions are assumed to be uniform throughout the year.

The emission indices for various engine effluents were discussed in Table 9-1. Recent modeling results (Johnston *et al.*, 1989; Ko *et al.*, 1991; Wuebbles and

Kinnison, 1990) showed that the main impact on ozone is due to the total amount of NO_x emitted and the altitude of injection. For fixed fuel use, the amount of NO_x emitted is related to the EI(NO_x). The Mach number (*i.e.* altitude of injection) and the EI for NO_x are used as the only two independent parameters in the HSRP scenarios, which represent cases with cruise speeds of Mach 3.2, 2.4, and 1.6, and selected cases with EI(NO_x) ranging from 5 to 45.

9.5.1.2 Background Atmosphere

The predicted HSCT fleet could be fully operational only by about the year 2015 when atmospheric concentrations of several trace gases are expected to be different from what they are today. It was decided that the calculations should be performed relative to a background atmosphere for the year 2015. (See Chapter 8 for a description of the typical 2015 atmosphere.) It is assumed that there will be no reduction in the subsonic fleet with the introduction of the supersonic fleet. Thus, the impact of the supersonic fleet will be compared to the baseline atmosphere that includes a projected subsonic fleet (which is twice the present fleet size) operating in the 2015 background atmosphere.

9.5.2 Model Results Using Gas Phase Chemistry

The modeling groups that participated in the intercomparison are:

AER:	Atmospheric and Environmental Research Inc., M. Ko and D. Weisenstein
GSFC:	NASA Goddard Space Flight Center, C. Jackman, A. Douglass, and K. Brueske
LLNL:	Lawrence Livermore National Laboratory, D. Wuebbles and D. Kinnison
NCAR:	National Center for Atmospheric Research, G. Brasseur
CAMED-P:	University of Cambridge, University of Edinburgh, J. Pyle, R. Harwood, and A. Jones
Oslo:	University of Oslo, I. Isaksen, F. Stordal.

The procedure for this intercomparison assessment made use of the infrastructure set up for previous model intercomparison workshops (see Jackman *et al.*, 1989). Model results were collected in digital format at the Upper Atmosphere Data Program (UADP) at NASA Langley to facilitate comparisons.

Except for the CAMED-P model, the transport circulation and temperatures in the models are fixed so that the effects of dynamical feedbacks are ignored. Changes in O_3 are responses to modifications in the chemical removal rates resulting from aircraft emissions. This, to first order, should be related to the amount of injected NO_y retained in the atmosphere at steady state. Typical residence time of the injected materials depends on the altitude of injection and ranges from 1.7 to 2.6 years for Mach 3.2 injection, 1.1 to 1.5 years for Mach 2.4 injections, and 0.5 to 0.8 years for Mach 1.6 injection. Note that a large residence time implies that the emitted NO_y is retained in the stratosphere for a longer period of time so that more NO_y will be added to the stratosphere at steady state for a particular emission rate. As a result, more O_3 will be removed for the same emission. For instance, the calculated decrease in ozone at northern mid-latitudes for EI (NO_x) = 15 ranges from 7–12 percent for the Mach 3.2 case, as compared with 2–6 percent for the Mach 2.4 case. The residence time could also depend on the time of the year the emissions occur and the actual three-dimensional nature of the flight path. Neither of these is considered in the results discussed here.

The calculated changes in column ozone and in local ozone are shown in Figures 9-3 and 9-4 for the case with Mach 2.4 and EI(NO_x) = 15. The calculated perturbations in global ozone content are summarized in Table 9-3. The following observations can be made about the results:

- The calculated impacts on O_3 are greater for larger EI and higher cruise altitudes. This result applies to all models.
- The magnitudes of the calculated O_3 changes can differ from model to model by significant amounts. The calculated changes in global content differ by a factor of 1.5 for Mach 3.2 cruise, factor of 4 for Mach 2.4 cruise. For the Mach 1.6 case, the calculated ozone responses range from no impact to a decrease of 0.7 percent for EI = 15.
- All models showed large decreases in the region north of 30°N between 10 and 25 km where most of the NO_x emissions are deposited.
- The extent to which the lower stratosphere in the Southern Hemisphere is affected in each

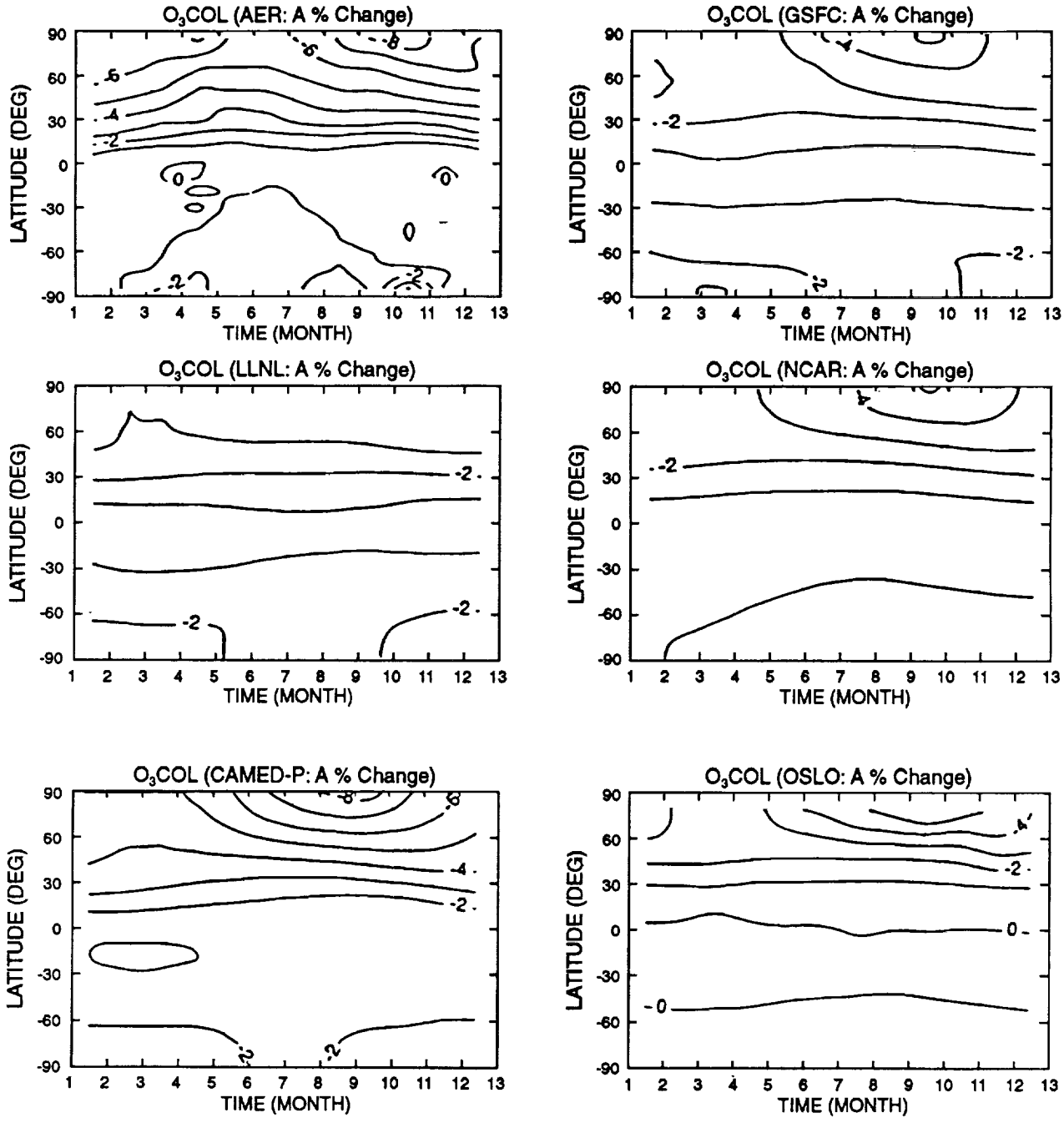

Figure 9-3 Calculated percent changes in the column abundances of O_3 as functions of latitude and season for a fleet of Mach 2.4 supersonic aircraft with $EI(NO_x) = 15$. The annual fuel use is 70×10^9 kg/year. The latitudinal distribution of fuel use is given in Figure 9-1. The percent change is calculated relative to a 2015 atmosphere with a subsonic fleet. The contour interval is one percent.

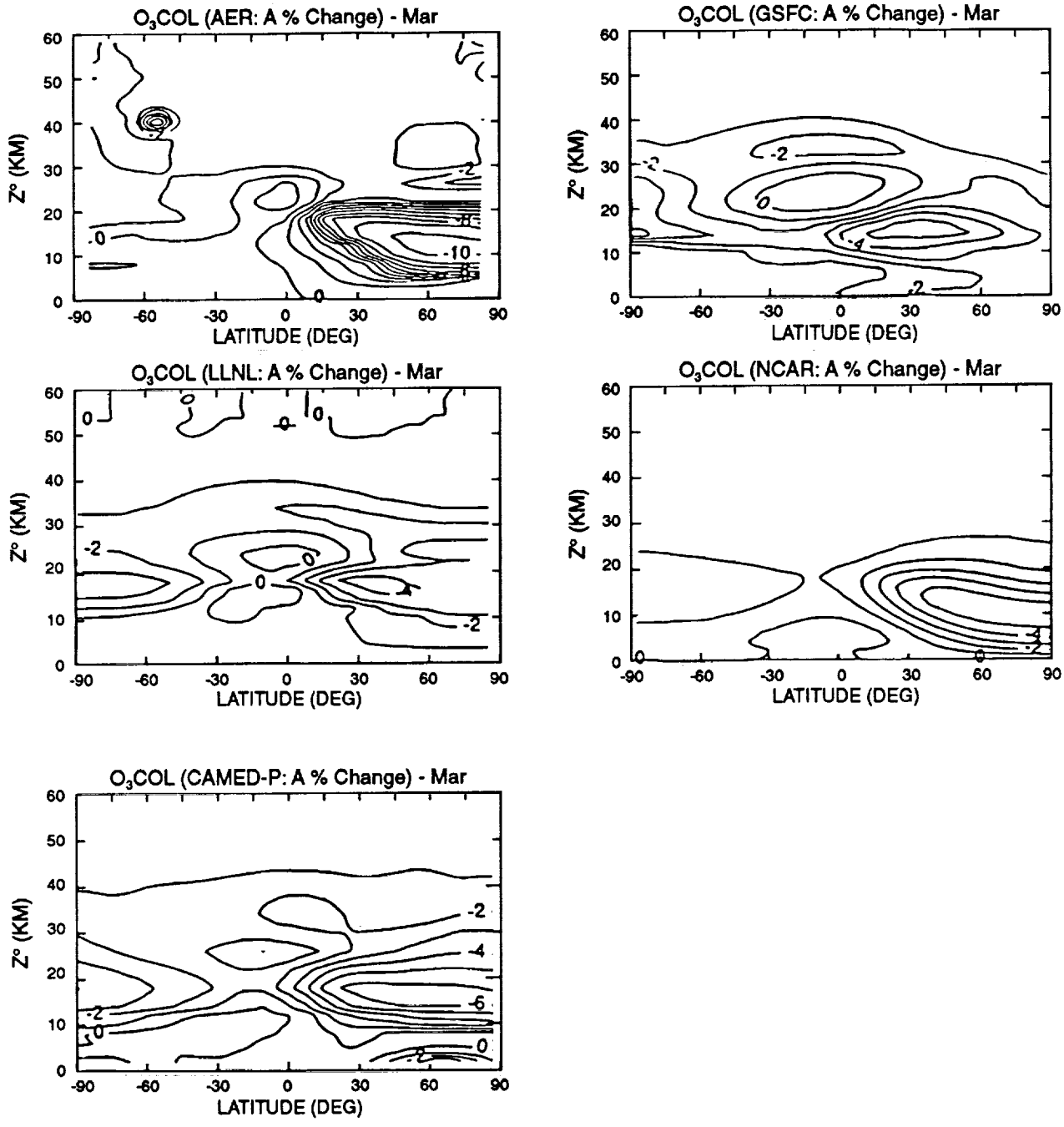

Figure 9-4 Calculated percent changes in the local concentration of O_3 as functions of latitude and height for a fleet of Mach 2.4 supersonic aircraft with $EI(NO_x) = 15$. The annual fuel use is 70 x 10^9 kg/year. The latitudinal distribution of fuel use is given in Figure 9-1. The percent change is calculated relative to a 2015 atmosphere with a subsonic fleet. The contour interval is one percent.

Table 9-3 Calculated percent decrease in annual average of global O_3 content.

	EI = 5			EI = 15			EI = 45
Mach Number	3.2	2.4	1.6	3.2	2.4	1.6	2.4
AER	1.2	0.61	0.11	4.3	2.1	0.40	7.5
CAMED-P				5.3	2.5	0.72	
GSFC	1.2	0.50	0.20	4.1	1.7	0.67	5.9
LLNL	0.9	0.50	0.085	3.4	1.7	0.29	6.1
NCAR	0.78	0.31	0.14	3.5	1.4	0.62	4.7
Oslo	1.2	0.15	0.01	4.1	0.72	.002	3.5

model is related to how efficiently NO_y is transported to the Southern Hemisphere.

- The sensitivity of the ozone response is related to the residence time of injected NO_x.
- The sensitivity of the ozone response to changes in NO_x in the lower stratosphere is different in different models as evident from the fact that the ozone changes normalized by changes in NO_x are different.
- Although changes of ozone in the upper troposphere contribute little to changes in column abundance, such a redistribution could have important implications for the radiative balance in the stratosphere.

9.5.3 Heterogeneous Chemistry in the Atmosphere

The O_3 responses to increases in NO_x shown in the previous section were calculated assuming gas phase reactions only. Heterogeneous reactions occurring on PSCs and on the global sulfate aerosol layer (see Chapters 3 and 4) are likely to change these responses in a significant way. In contrast to the case with chlorine perturbation, where heterogeneous reactions will enhance the ozone depletion, including heterogeneous reactions in the HSCT model calculations will actually give smaller ozone decreases due to NO_x increases. Figure 9-5 shows the change in ozone from the AER

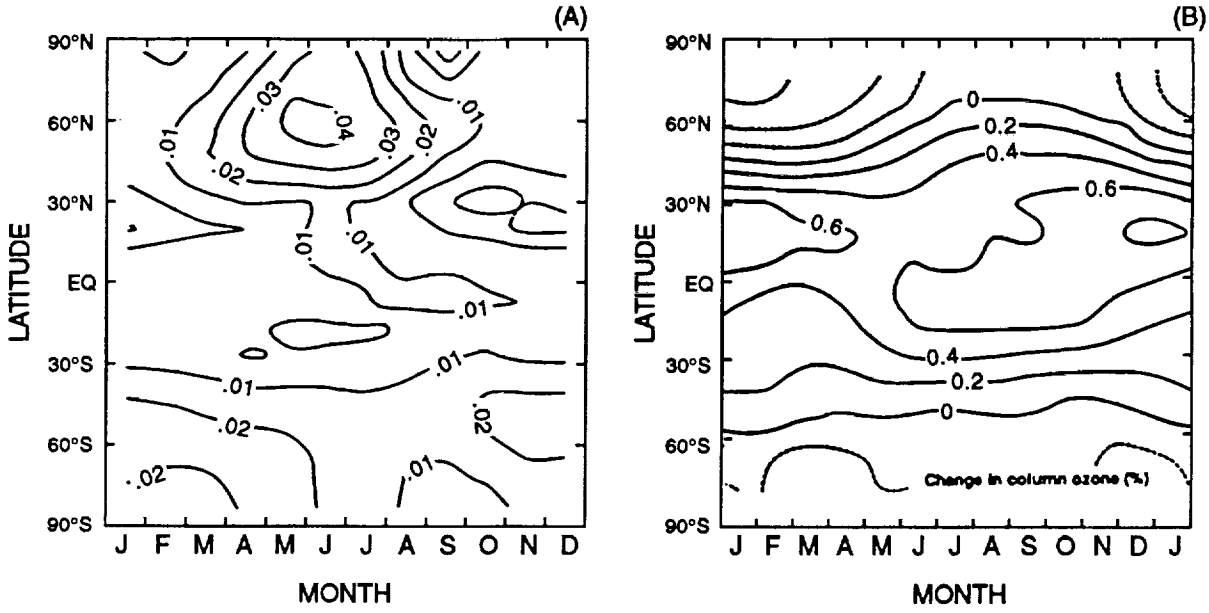

Figure 9-5 Percent changes in column abundance of O_3 calculated with the reaction $N_2O_5 + H_2O$ (aerosol) $\rightarrow$ $2HNO_3$ included. The emission scenario is exactly the same as those described in Figure 9-3. Panel A-result from AER model (Weisenstein *et al.*, 1991) Panel B-result from LLNL model using the lower limit parameterization (see Chapter 8). Note that the parameterization of aerosol collision frequency is not the same in the two models.

model (Weisenstein *et al.*, 1991) and LLNL model, (Wuebbles, private communication) calculated using the same scenario as in Figure 9-3 except that the reaction $N_2O_5 + H_2O \rightarrow 2HNO_3$ is assumed to occur on the global sulfate layer. Note that the calculated ozone change is greatly reduced compared to the results shown in Figure 9-3.

The altered response of ozone to NO_x injection with heterogeneous chemistry included can be explained by changes in the relative contributions of the catalytic cycles to ozone removal (Weisenstein *et al.*, 1991). These cycles involve NO_x, Cl_x, HO_x, O_x, and Br_x radical species. The NO_x cycle accounts for more than half of the ozone loss in both winter and summer with gas phase chemistry only. The effect of the heterogeneous reaction $N_2O_5 + H_2O \rightarrow 2HNO_3$ is to repartition the odd nitrogen family, resulting in NO_x concentrations which are reduced by 80 percent in winter and 40 percent in summer between 10 and 20 km at mid and high latitudes as compared with gas phase-only calculations. Thus the ozone loss due to the NO_x cycle becomes a much smaller fraction of the total ozone loss. Increases in OH and ClO due to inclusion of the heterogeneous reaction cause the contributions from the HO_x and Cl_x cycles to nearly double.

Addition of nitrogen oxides from HSCT emissions results in a 20–30 percent increase in the NO_x cycle with or without the $N_2O_5 + H_2O$ reaction. However, the ozone response is much less sensitive to the NO_x increase with heterogeneous chemistry included because of the reduced role of NO_x in regulating total ozone loss. Furthermore, an increase in the NO_x concentration will enhance the concentration of $ClONO_2$, HNO_3, and HO_2NO_2 and thus reduce the Cl_x and HO_x catalytic destruction of ozone. Depending on the background levels of Cl_x and HO_x and the overall rate of the $N_2O_5 + H_2O$ reaction, HSCT emissions can even result in small ozone increases.

The rates for many heterogeneous reactions have been measured in the laboratory (see Chapter 3). There is no reason to exclude these reactions in model simulations. However, whether they actually occur in the atmosphere and whether there are other significant heterogeneous reactions occurring on aerosol particles are unresolved issues. A concerted campaign of atmospheric measurements of radical species should help to elucidate the role of heterogeneous reactions occurring on aerosol particles and on PSCs in the lower stratosphere where HSCT aircraft will fly.

9.5.4 Future Research

The analysis reported in the HSRP (1992, Chapter 5) suggests that the difference among the model-predicted ozone responses can be understood in terms of the residence time for the injected NO_x and O_3 response sensitivity factor peculiar to each model. One must look for observations in the present atmosphere that can help define these quantities and predict how they may change. Future research should emphasize model developments and acquisition of kinetic data to better define model input and field data for model validation. Specific emphases should be given to the following issues (see also review by Johnston *et al.*, 1991 Douglass *et al.*, 1991).

9.5.4.1 Stratospheric-Tropospheric Exchange

The issue of stratospheric-tropospheric exchange is important for determining the residence time for the injected material and how they are redistributed. Changes in ozone concentration near the tropopause are particularly sensitive to the changes in NO_x in the same region. The ^{14}C data from atmospheric nuclear tests seem ideally suited for deriving stratospheric residence times for comparison with model results (Johnston *et al.*, 1976; Johnston, 1989). Analyses using two-dimensional models has been reported by Shia *et al.* (1989), Jackman *et al.* (1991) and Kinnison *et al.* (1991). Other data such as ^{238}Pu and ^{90}Sr may also be useful (see *e.g.*, Telegadas and List, 1969). Analysis of data for H_2O, O_3, and NO_y near the tropopause may provide clues for the actual mechanisms responsible for the troposphere-stratosphere exchange rate.

9.5.4.2 Ozone Budget in the Lower Stratosphere

To get a handle on the O_3 sensitivity, one can make use of measurement programs that are designed to provide simultaneous observations of many species [such as Atmospheric Trace Molecule Spectroscopy Experiment (ATMOS), balloon measurement campaigns, and aircraft campaigns] to provide directly measured or derived concentrations for the radical

species to define the local chemical removal rates for ozone for the present-day atmosphere. Getting a handle on the removal rate by transport is much more difficult. Application of the data assimilation technique to derive transport wind fields from observations may serve as a starting point for deriving transport fluxes of O_3 in the lower stratosphere.

9.5.4.3 Plume Dispersion and Plume Chemistry

The source function for the emitted materials used in the calculations is assumed to have the same latitude-height distribution as the flight paths, and the chemical composition is assumed to be identical to that of the emission at the tail pipe. Plume subsidence and subsequent dispersion in the first few weeks before the emitted materials become zonally mixed could provide an effective distribution of sources that differs from the flight paths. Chemical transformation, occurring homogeneously and heterogeneously, may alter the composition of the materials.

The possibility that the emitted material may not be well mixed zonally raised the question of the importance of the proper treatment of nonlinear chemistry (Tuck, 1979) and chemical eddies (Pyle and Rogers, 1980). Recent results from Zhadin and Bromberg (1991) showed their treatment of chemical eddies resulted in a decrease in calculated ozone loss due to more efficient removal of NO_x in the lower stratosphere. Resolution of this issue may have to await results from three-dimensional model simulations.

9.5.4.4 Effects of Sulfur and Particulates

One must also consider the possibility that operation of the HSCT may increase the loading and size distribution in the sulfate layer through emissions of sulfur and particulates. The possible climate effects from the change in radiative properties must be considered in conjunction with the effects from ozone redistribution.

9.5.4.5 Effects of PSCs and Coupling to Chlorine Chemistry

The saturation temperature for the formation of PSCs is a direct function of the local concentration of HNO_3 and water vapor (see discussion in Chapter 3). Injection of NO_y and H_2O by HSCT could promote formation of PSCs. A recent study by Peter *et al.* (1991) estimated that there could be a significant increase in the occurrence of type-I and type-II PSCs in the polar region. However, our ability to model PSCs to provide quantitative predictions of future effects remains extremely limited. How this may enhance the ozone removal by the chlorine cycle needs to be investigated.

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