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INFLUENCE OF PRESENT AND POSSIBLE FUTURE AIRCRAFT EMISSIONS ON THE GLOBAL OZONE DISTRIBUTION

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1. INTRODUCTION

The first research studies of the potential environmental impacts from commercial fleets of aircraft flying in the stratosphere were done in the early 1970s (Harrison,1970; Johnston, 1971). These studies suggested that the emissions from fleets of such aircraft could cause a significant reduction in the concentrations of ozone in the stratosphere, with accompanying increases in the amount of ultraviolet radiation reaching the Earth's surface. The primary concern, as pointed out by Johnston, was the chemical destruction of ozone from emissions of nitrogen oxides (NO_x) produced thermally in the aircraft engine combustion process. As a consequence of these studies, various governments instigated research programs to clarify the potential environmental effects of aircraft operations in the stratosphere. In the U.S., concurrent, but independent, studies were conducted by the Department of Transportation's Climatic Impact Assessment Program (CIAP, 1974, 1975 a, b) and by the National Academy of Sciences (NAS, 1975). Several independent assessments were also carried out in Europe (COMESA, 1976; COVOS, 1976). A subsequent study to CIAP, the High Altitude Pollution Program (HAPP) sponsored by the U.S. Department of Transportation, continued studies on potential aircraft effects at a lower research level until 1980.

The first detailed sensitivity analysis in almost a decade of the effects from aircraft emissions on ozone was published by Johnston et al., in 1989 (part of this work also appeared in Kinnison et al., 1988). Both the LLNL one-dimensional and two-dimensional (2-D) models of the global atmosphere were used in this study. The one-dimensional model provides a historical perspective along with having computational advantages for multiple sensitivity calculations, while the two-dimensional model provides information on seasonal and latitude effects as well as having a stronger theoretical basis for its representation of atmospheric transport processes. Major findings from this study were:

1. Nitrogen oxides from the exhaust gases of a fleet of HSCTs can reduce stratospheric ozone on a global basis.

2. The altitude at which NO_x emissions cause the largest reduction in the ozone column is about 25 km in the one-dimensional model and about 28 km in the two-dimensional model.

3. For a given altitude and magnitude of NO_x emissions, the two-dimensional model indicates that the reduction of global ozone depends on the latitude of the injections, with the maximum ozone reduction for tropical injection. For a given injection, the largest ozone column reductions occur in the polar regions.

4. For extremely large stratospheric chlorine (Cl_x) mixing ratios (~8 ppbv compared to about 3 ppbv in the current atmosphere), NO_x emissions can increase the ozone column, partially counteracting the ozone reduction caused by the chlorine. This effect is only found in a highly Cl_x perturbed stratosphere where ozone is already greatly depleted.

5. Water vapor (without NO_x) emissions cause a small ozone reduction; water vapor and NO_x together give an ozone reduction less than that calculated for NO_x emissions alone by a factor between 0.85 and 0.97, depending on the altitude and magnitude of the emissions.

6. Calculated global reductions of ozone due to NO_x emissions are smaller in the two-dimensional model than in the one-dimensional model, by factors that range from 0.66 to 0.86 for injection altitudes between 20 to 34 km. [More recent calculations show a sensitivity in the two-dimensional model to uncertainties in the treatment of eddy transport processes (Kinnison, 1989).]

7. On the basis of an uncertain estimate of fuel consumption by future stratospheric aircraft, two-dimensional model calculations for a fleet of 500 aircraft operating at 22 km with NO_x emission properties of 1988 subsonic commercial aircraft engines give a global ozone reduction of 19% (assuming emissions occur between 37°–49°N). The effect of reducing the emission index for the amount of NO_x produced per kg of fuel burned is shown in Table 1.

The purpose of this study is to build on the previous analyses of potential aircraft emission effects on ozone in order to better define the sensitivity of ozone to such emissions. A preliminary analysis is made of the sensitivity of the ozone distribution to the current fleet of commercial aircraft. The future emissions scenarios evaluated in this study attempt to more accurately account for the spread in emissions with latitude and altitude for an assumed fleet of HSCTs. The time it would take for the atmosphere to recover from the effects on ozone after emissions are ceased is also examined. In addition to emissions of NO_x, the effects of potential emissions of carbon monoxide and water vapor are also evaluated. These analyses are all based on results from the LLNL two-dimensional chemical-radiative-transport model of the troposphere and stratosphere.

Table 1. Calculated change in global-averaged total ozone from the LLNL two-dimensional model assuming an annual fuel consumption of 7.7×10^{10} kg, with all emissions between 37°–49°N in a 3 km altitude range centered about 22.5 km altitude (relative to an atmosphere with no aircraft emissions), based on Johnston, et al. (1989).

Emission Scenario	Emission Index g of NO Kg^{-1} Fuel	Injection Rate MT of NO$_2$ Yr^{-1}	Injection Rate Molecules/ 10^5cm^{-2}s^{-1}	Change in Total Ozone
Current Subsonic Rate	40*	4.8	4000	-19.0%
Standard CIAP Rate	15	1.8	1500	-8.6%
Future Goal	5	0.6	500	-2.8%

* This emission index assumes current commercial subsonic aircraft technology being used in the stratosphere. The actual emission index for current aircraft at their flight altitudes in the upper troposphere are a factor of 2-3 smaller than this.

2. THE LLNL TWO-DIMENSIONAL MODEL

The two-dimensional model of the global atmosphere provides a zonally-averaged representation of the chemical and physical processes determining the composition and distributions of trace constituents in the troposphere and stratosphere. The concentrations of 35 chemically active trace constituents, based on more than 100 chemical and photochemical reactions, are calculated as a function of altitude, latitude, and season within the model. Essentially the model determines the atmospheric distribution of ozone and other important constituents based on the interactions of chemical, radiative, and dynamical transport processes thought to be operating in the atmosphere. For the version of the model used here, the diabatically-driven circulation is determined for the current (background) atmosphere using observed temperatures along with model-derived radiative transfer calculations of the net solar and longwave heating, determined in an internally consistent way with the derived species distributions. For the perturbed atmosphere, a perturbation form of the thermodynamic equation is solved for the changes in stratospheric temperatures resulting from the changes in ozone and other radiatively important trace constituents (note that the diabatic circulation is assumed to be unchanged from the current atmosphere for these calculations).

3. CURRENT AIRCRAFT EMISSIONS

Earlier calculations with the one-dimensional model (Wuebbles, 1983) suggested that emissions from the existing, primarily subsonic, fleet of commercial aircraft could affect upper tropospheric ozone, resulting in increased concentrations of ozone in the region just below the tropopause. However, the one-dimensional model is of limited use in evaluating changes in tropospheric chemistry. Even the two-dimensional model is somewhat limited in treating tropospheric chemistry, particularly in the lower troposphere, because of the significant impact of emissions from highly reactive species primarily produced over land; only a three-dimensional model can be used to fully evaluate the effects on the global atmosphere from such emissions. Nonetheless, the two-dimensional model provides a useful test of how sensitive the troposphere may be to aircraft emissions primarily occurring in the upper troposphere.

One of the difficulties in evaluating the effects of existing aircraft emissions is that there isn't an available evaluation of the trends in such emissions over the last several decades. The most complete estimate of current emissions comes from the analysis of commercial aircraft flights for 1987 in a recent analysis by Boeing Corporation (Boeing, 1989). Slightly more than 29,000 city-pairs were accounted for in their analysis, along with the appropriate aircraft types and flight frequencies. The analysis determined emissions of nitrogen oxides, carbon monoxide, and hydrocarbons at 26,000 feet (about 8 km) and 37,000 feet (about 11 km). Figure 1a shows a contour plot of the NO$_x$ emissions from the Boeing analysis. Bauer (1978) published an estimate of the 1975 subsonic aircraft emissions that had about a factor of 3.4 less NO$_x$ emitted than the assumed emissions for 1987. If this earlier emissions estimate is correct, there was a sizable increase in aircraft emissions during the 12 years between 1975 and 1987.

The two-dimensional model is used to evaluate the effects on ozone concentrations resulting from the 1987 fleet emissions compared to an ambient atmosphere with no aircraft emissions. Emissions of nitrogen oxides, carbon monoxide, and hydrocarbons were included in the calculation based on the Boeing analysis; the hydrocarbon emissions were assumed to have the same reactivity as methane, as there was no attempt in this preliminary study to represent the complexity of non-methane hydrocarbon chemistry in this version of the model.

The calculated response of the ozone distribution to the emissions for the 1987 fleet are shown in Figures 1b and 1c. The largest increase in ozone occurs near 9 km at midlatitudes in the Northern Hemisphere, corresponding to the altitude and latitude of the maximum emissions. Approximately a 5% maximum increase in ozone is determined in July, primarily due to the effect of the NO$_x$ (see Table 2) emissions to produce ozone in the troposphere through smog-formation reactions (e.g., see WMO, 1985). The global-averaged change in total ozone is 0.45%, with the maximum change of 0.6% occurring at high latitudes in the Northern Hemisphere.

Table 2. Sensitivity analyses to determine the relative importance of NO$_x$, CO, and hydrocarbon (represented as CH$_4$ emissions) on the calculated change in total ozone for the current aircraft emissions scenario.

	Change in total ozone (%) Global Average	Change in total ozone (%) Northern Hemisphere	Change in total ozone (%) Southern Hemisphere
NO$_x$ + CO + CH$_4$	0.45	0.53	0.37
NO$_x$ only	0.44	0.52	0.36
CO only	0.018	-0.0058	0.043
CH4 only	0.024	0.0031	0.045

A trend analysis of ozonesonde measurements from nine stations at latitudes from 32°N to 75°N has been recently made for the years from 1965 through 1986 (WMO, 1989). Significant increases in ozone concentrations were determined throughout the troposphere, with the largest increase (+0.83% per year for the period from 1970 to 1986) occurring in the bottom 3 km of the atmosphere. A secondary peak in the ozone increase (+0.38% per year or 6% over the 16 year period of the trend estimate) was found at altitudes between 8 and 10 km, just below the tropopause. Although it is premature to attribute causality, it is interesting that the secondary peak occurs at the same altitude and magnitude as the calculated effect from subsonic aircraft emissions. Aircraft emissions may explain a substantial fraction of the observed ozone increase in the upper troposphere. However, one should be extremely cautious in interpreting the global implications of such a limited data set.

4. A MATRIX OF FUTURE EMISSIONS

The amount of nitrogen oxides and other gases emitted into the atmosphere by a fleet of commercial aircraft depend on a number of factors, including the fleet size, engine fuel efficiency, the aircraft range, the cruise altitude, flight operational procedures, and the emission index for amount of NO_x produced per kg of fuel burned. Possible designs for HSCT aircraft and their engines are still at an early stage; therefore, most of the factors determining emissions of an actual fleet are still unknown. Examining a wide range of possible flight altitudes and levels of emissions for the effects on ozone will help establish the design criteria needed to prevent an unacceptable environmental problem.
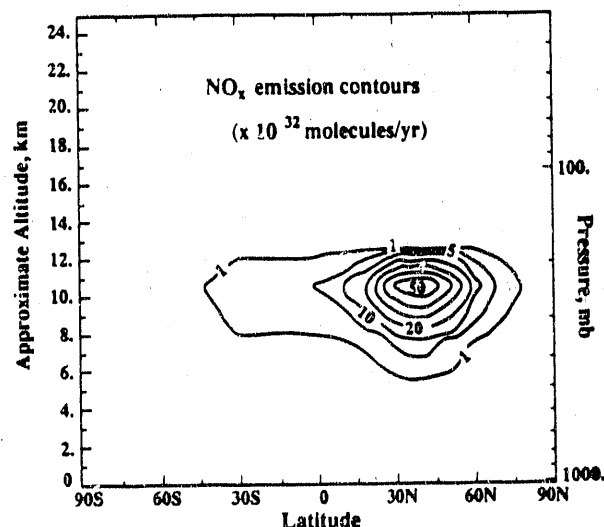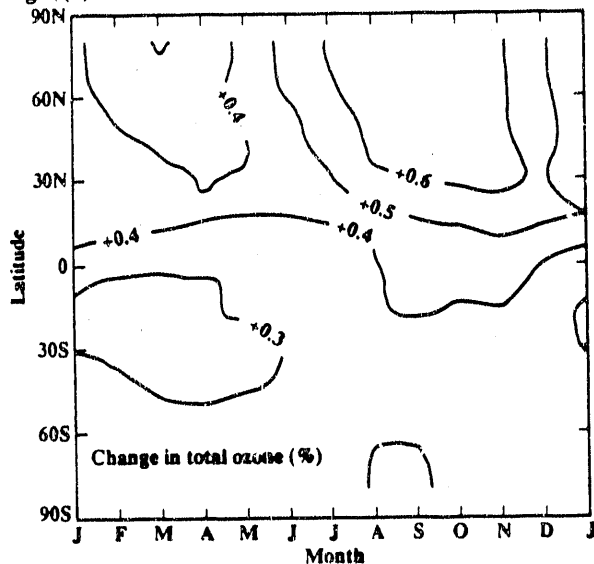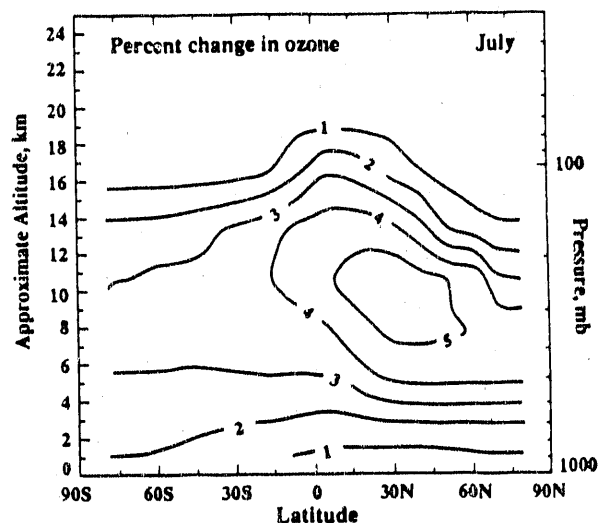

Fig. 1(a)

Fig. 1(b)

Fig. 1(c)

Fig. 1. (a) Contour of the NO_x emissions as a function of altitude and latitude determined by Boeing (1989) in their evaluation of the commercial aircraft emissions for the year 1987; (b) Calculated percentage change in total ozone at steady-state plotted as a function of latitude and time of the year as determined in the LLNL two-dimensional model for the 1987 subsonic aircraft fleet compared to an atmosphere without aircraft emissions; (c) Calculated percentage change in ozone at steady-state plotted as a function of altitude and latitude as determined for the 1987 subsonic aircraft fleet during the month of July.

In this study, we have evaluated a total of twelve HSCT fleet scenarios assuming three different cruise altitudes and varying levels of NO_x emissions. These hypothetical scenarios were developed by the HSCT group at McDonnell Douglas Corporation in a cooperative effort to establish a matrix of scenarios with realistic assumptions for flight altitudes versus magnitude of emissions. In no manner are these scenarios based on actual aircraft designs; they are intended to cover a broad range of aircraft altitudes and emissions These scenarios attempt to consider how the emissions should vary with latitude and altitude appropriate to a worldwide fleet of HSCTs. No subsonic aircraft emissions were considered in these scenarios. Three different aircraft, flying at different mean cruise altitudes, were assumed in the scenarios; the mean cruise altitudes were 15.8, 18.3, 22.9 km, although changes in flight altitudes throughout the flight patterns assumed in the scenarios are accounted for. Ten different city-pairs were chosen by McDonnell Douglas to represent the appropriate regions for intercontinental flights; these flight patterns combined with the assumed aircraft type provide the criteria for the altitude and latitude of the aircraft emissions. A wide range of NO_x emissions were assumed, based on uncertainties in fleet size, fuel burned, and NO_x emission index. All scenarios assume that a kerosene based fuel will be used in the HSCT in contrast to a more exotic fuel such as liquid methane or hydrogen.

Table 3 indicates the mean cruise altitude and the total global NO_x emissions for the 12 scenarios. Figure 2 shows the NO_x emissions assumed in scenario c1, corresponding to a mean flight altitude of 22.3 km, a total NO_x emission of 0.28 Mt per year (as NO_2) or 3.64×10^{33} molecules per year. The NO_x emissions for scenarios with the other mean cruise altitudes look very similar to Fig. 2, except maximum emissions occur at lower altitudes.

For each of the 12 scenarios, Table 3 shows the calculated change in total ozone column for each hemisphere, along with the global averaged change in ozone. These calculations are based on a background stratospheric chlorine level of 2.9 ppbv, corresponding to current concentrations.

Table 3. Annually-averaged change in ozone for the twelve emissions scenarios evaluated with the LLNL two-dimensional model.

Scenarios	Mean Cruise Altitude (km)	Total NOx Emissions (x 10^33 Molec./Yr)	ΔO3/% N.H.	S.H.	Global
a0	15.8	0.67	- 0.20	- 0.18	- 0.19
a1	15.8	4.18	- 0.43	- 0.33	- 0.38
a2	15.8	17.89	- 1.53	- 1.00	- 1.27
a3	15.8	58.31	- 5.50	- 3.35	- 4.44
b0	18.3	0.63	- 0.25	- 0.22	- 0.23
b1	18.3	3.29	- 0.71	- 0.52	- 0.61
b2	18.3	16.58	- 3.47	- 2.24	- 2.86
b3	18.3	54.02	- 12.18	- 7.63	- 9.93
c0	22.9	0.58	- 0.28	- 0.23	- 0.25
c1	22.9	3.64	- 1.00	- 0.64	- 0.82
c2	22.9	15.32	- 4.22	- 2.42	- 3.33
c3	22.9	49.98	- 14.45	- 8.17	- 11.34

Although more realistic scenarios for the assumed emissions are used here than in the prior Johnston, et al. sensitivity analyses, the main conclusion is similar: the largest effects on total column ozone occur with the largest NO_x emissions and the highest assumed flight altitude. The Northern Hemisphere has the largest decreases in total ozone. Figure 3 shows the change in globally and annually averaged total ozone as a function of the altitude and magnitude of the emissions. This figure indicates that, for these scenarios, the assumed HSCT fleet would need to keep NO_x emissions below 0.5-1×10^{34} molecules per year, depending on the aircraft flight altitude, in order to affect globally-averaged total ozone by one percent or less. Using a one percent criteria in this discussion is entirely arbitrary, since humanity has yet to determine what is an acceptable level of ozone destruction from such emissions.
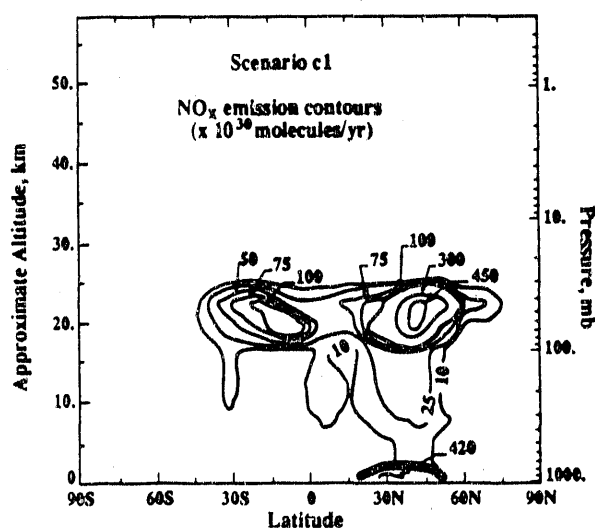
Fig. 2. Emissions of NO_x as a function of altitude and latitude assumed in scenario c1, corresponding to an aircraft flying at a mean cruise altitude of 22.3 km and a total emissions of 3.64×10^{33} molecules of NO_x per year.

Figure 4a gives the change in total ozone versus time of year for scenario c1, while Figure 4b shows the change in ozone versus altitude and latitude for the same scenario. The maximum change in total ozone occurs at high Northern Hemisphere latitudes and peaks in late summer and into fall. The minimum decrease in total ozone occurs in the tropics. Figure 4b shows that the maximum ozone destruction for this scenario occurs near 20 km altitude in Northern Hemisphere polar region.

Part of the intention in undertaking this analysis is to develop the appropriate criteria needed by the aircraft industry to ensure that the HSCT developed is not environmentally harmful. Because this matrix of scenarios extends over such a wide range of emissions we want to be careful that the results for these scenarios are not over-interpreted. We do not want to imply that the large emission cases in any way correspond to any fleet under actual consideration.

5.   OZONE RECOVERY TIME

Of interest to policymakers is the question of how long it takes for the ozone concentrations to recover if the emissions are greatly reduced. For scenarios a2, b2, and c2, Figure 5 shows that it takes about nine years after emissions begin for the maximum change in total ozone to be reached, and about an additional decade for the globally-averaged ozone column to recover to unperturbed conditions after emissions cease. The altitude of the emissions appears to make little difference in the recovery time.
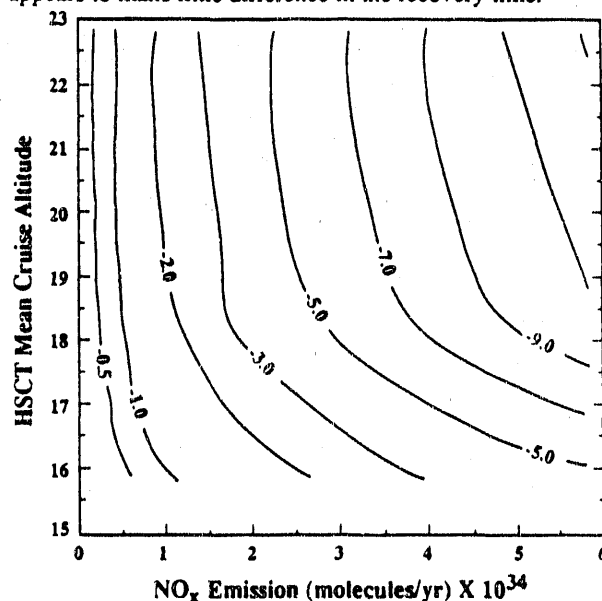
Fig. 3. Change in globally and annually averaged total ozone at steady-state plotted as a function of mean cruise altitude and amount of NO_x emissions assumed in the twelve scenario matrix.

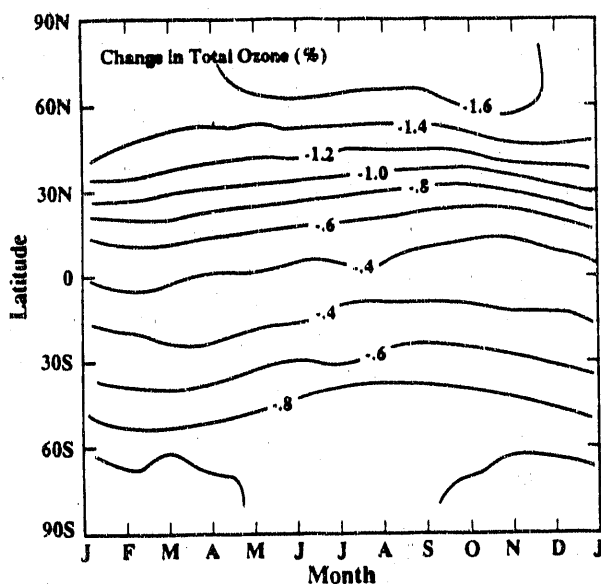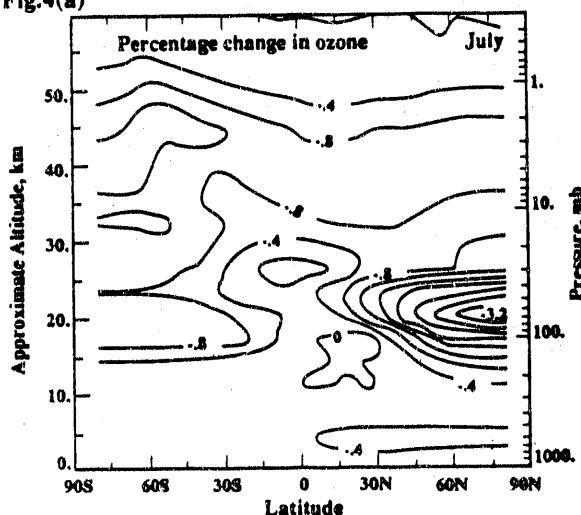Fig.4(a)

Fig. 4(b)

Fig. 4. (a) Calculated percentage change in total ozone at steady-state plotted as a function of latitude and time of the year as determined for scenario c1; (b) Calculated percentage change in ozone at steady-state plotted as a function of altitude and latitude as determined for scenario c1 during the month of July .

6. FURTHER SENSITIVITY STUDIES

This evaluation of the future HSCT fleet scenarios has, until this section, considered the effects on ozone resulting from NO_x emissions only. Several sensitivity calculations are evaluated here based on assumed emissions of water vapor and carbon monoxide. These estimated emissions are based on current engine technology. An additional sensitivity calculation will consider the effect on the calculated ozone changes of a future atmosphere with higher background stratospheric chlorine along with increased concentrations of CO_2, CH_4, and N_2O.

The stratospheric water vapor emissions due to aircraft emissions for hydrocarbon fuel combustion are much larger than the NO_x emissions. For scenario c1, this corresponds to about a factor of 370 larger number of molecules of water emitted into the stratosphere than NO_x

emitted. The effect of the water vapor emissions by itself is small, but it has a larger impact when included in combination with the NO_x emissions. The effect on ozone with the coupled NO_x and H_2O emissions case is a factor of 0.9 less than the NO_x emissions only case. This effect is due the interactions between NO_x and HO_x chemistry.

The effects of carbon monoxide emissions were also evaluated. For scenario c1, the total CO emissions are 6.24×10^{33} molecules per year, but over half of these emissions occur in the bottom 1.5 km of the atmosphere. We found that the CO emissions had a negligible effect on global ozone.

In the wake of the aircraft effluent, highly reactive species exist (e.g. HO_2) that could convert NO_2 to nitric acid (CIAP,1975b). For scenario b2, the extreme case was considered where all the NO_x ($NO + NO_2$) is converted to HNO_3. The annual averaged ratio for the calculated change in ozone with HNO_3 injected relative to NO_x being injected is 0.93 Globally; 0.91 in the Northern Hemisphere; 0.97 in the Southern Hemisphere. This suggests that the effect would be moderately important only if a large portion of the NO_x is converted to HNO_3.
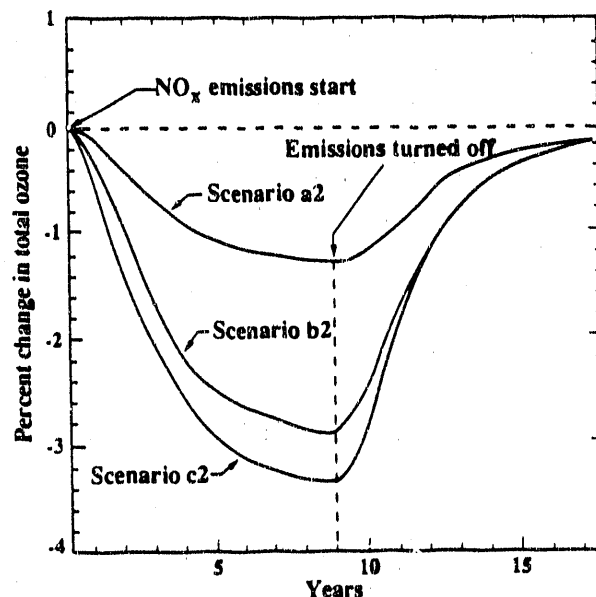

Fig. 5. Change in globally and annually averaged total ozone as a function of time after initiating the NO_x emissions for scenarios a2, b2, and c2, followed by cessation of all emissions once steady-state has been reached.

As a test of the sensitivity of the aircraft scenario effects to the background atmosphere, we evaluated the effect of the NO_x emissions in scenario c1 for an atmosphere containing 5.2 ppbv of stratospheric reactive chlorine, 395 ppmv of carbon dioxide, 2.25 ppmv of methane, and 0.334 ppmv of nitrous oxide. This corresponds to the atmosphere in 2020 if one assumes constant emissions of CFCs at 1986 levels, and continuing trends at current rates for CO_2, CH_4 and N_2O. In actuality, this level of chlorine will likely not be reached in the next few decades as a result of international efforts to reduce CFC production. However, this chlorine level is useful in evaluating the sensitivity of aircraft emissions to background chlorine levels. There appears to be a large sensitivity to the background atmosphere; the calculation of scenario c1 with the 2020 background atmosphere determines a global total ozone decrease of 0.58% as compared to the ozone decrease of 0.82% calculated using the current atmosphere as the background. The chemical

interactions of NO_x with stratospheric chlorine are likely responsible for the sensitivity, but the role of the increased background CO_2, N_2O, and CH_4 require further evaluation.

CONCLUSIONS

This study has used the LLNL two-dimensional model of the global atmosphere in an evaluation of the effects on global ozone concentrations from current subsonic aircraft emissions and from the emissions of possible future high speed civil transports. We have attempted to include more realistic representations of emissions as a function of altitude and latitude in these scenarios than were included in previous sensitivity analyses. Major findings from this study are:

1. Current aircraft emissions may be having an impact on upper tropospheric ozone, leading to increasing concentrations of ozone in the upper troposphere. However, the analysis here was very preliminary. The potential significance of the effects on ozone warrant much further study.

2. A matrix of HSCT scenarios evaluated over a wide range of mean flight altitudes and magnitudes of NO_x emissions confirmed previous analyses showing that ozone destruction becomes larger as the emissions of NO_x increase and as the altitude of injection increases.

3. Model calculations indicate that a major reduction in emissions would allow the stratosphere to recover to unperturbed conditions in about a decade.

4. Sensitivity studies indicate that water vapor emissions have a moderate effect on the change in total ozone, while carbon monoxide emissions had a negligible effect. Water vapor emissions should be considered in all future evaluations of HSCT fleets.

5. Injection of NO_x as HNO_3 had a moderate effect on the change in total ozone.

6. The calculated change in ozone for the HSCT scenarios was very sensitive to the background atmosphere, particularly to the levels of stratospheric chlorine and concentrations of carbon dioxide, methane, and nitrous oxide. This requires further evaluation, but suggests that assessment analyses of future HSCT fleets should carefully consider the appropriate background atmosphere for the time period in which the aircraft will fly. These analyses should also consider projections of subsonic emissions.

ACKNOWLEDGMENTS

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