Conf-9110217--3

UCRL-JC-108035
PREPRINT

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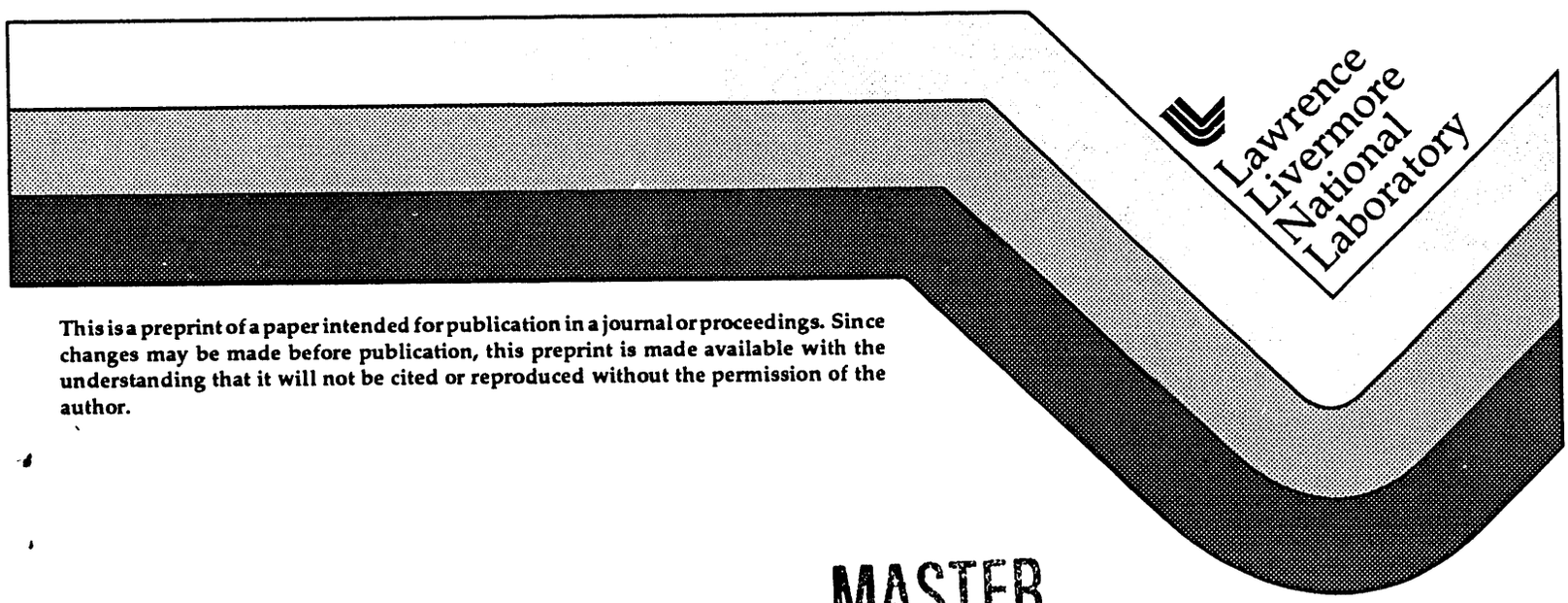
This paper was prepared for submittal to the 42nd Congress
of the International Astronautical Federation
October 5–11, 1991, Montreal, Canada

October 1, 1991



UCRL-JC--108035

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FUTURE AIRCRAFT AND POTENTIAL EFFECTS ON STRATOSPHERIC OZONE AND CLIMATE

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Abstract

The purpose of this study is to extend the recent research examining the global environmental effects from potential fleets of subsonic and supersonic commercial aircraft.[1,2] Initial studies with LLNL models of global atmospheric chemical, radiative, and transport processes have indicated that substantial decreases in stratospheric ozone concentrations could result from emissions of NO_x from aircraft flying in the stratosphere, depending on fleet size and magnitude of the engine emissions. These studies used homogeneous chemical reaction rates (e.g., gas-phase chemistry). Recent evidence indicates that reactions on particles in the stratosphere may be important. Heterogeneous chemical reactions, for instance, N_2O_5 and $ClONO_2$ on background sulfuric acid aerosols, convert NO_x (NO and NO_2) molecules to HNO_3. This decreases the odd oxygen loss from the NO_x catalytic cycle and increases the odd oxygen loss from the Cl_x catalytic cycle. By including these heterogeneous reactions in the LLNL model, the relative partitioning of odd oxygen loss between these two families changes, with the result that emissions of NO_x from proposed aircraft fleets flying in the stratosphere now increase ozone. Having these heterogeneous processes present also increases ozone concentration in the troposphere relative to gas-phase only chemistry calculations for emissions of NO_x from subsonic aircraft.

Introduction

Concern for the impact that large fleets of aircraft might have on the distribution of global ozone and climate was initiated in the 1970s. These studies suggested that the emissions of trace gases from fleets of aircraft flying in the upper troposphere and lower stratosphere could cause a significant reduction in ozone with accompanying increases in ultraviolet radiation reaching the earths surface.3,4 Emissions of nitrogen oxides (NO_x), water vapor (H_2O), carbon monoxide (CO), hydrocarbons (HC), sulfur dioxide (SO_2), and aerosols were investigated in detail. In the U.S., research was conducted by the Department of Transportation's Climatic Impact Assessment Program (CIAP) and the National Academy of Sciences (NAS) [5,6,7,8] Results from this work concluded that emissions of trace gases, primarily NO_x, could affect the global distribution of ozone.

Recently, using the LLNL one- and two-dimensional chemical-radiative-transport models of the global atmosphere, with a complete set of currently recommended gas-phase chemical reaction rates, a detailed sensitivity analysis was completed to reinvestigate the effects of aircraft trace gas emissions on ozone.[1] Results of this study reaffirmed the findings in the 1970s that NO_x produced thermally in the aircraft exhaust can reduce stratospheric ozone. This study found that the altitude at which NO_x emissions caused the largest reduction in column ozone is about 28 km. In addition, for a given altitude and magnitude of NO_x emissions, the LLNL two-dimensional model indicates that the reduction of global ozone depends on the latitude of the injections, with the maximum ozone reduction for a tropical injection. For a given injection, the largest ozone column reductions occur in the polar regions. Water vapor was also investigated and found to cause a small ozone reduction when injected by itself. When coupled with NO_x emissions, H_2O vapor reduced the ozone reduction caused by NO_x by a factor between 0.85 and 0.97, depending on the altitude and magnitude of the emissions.

Realistic emission scenarios were developed to more accurately account for the spread in emissions with latitude and altitude for an assumed fleet of supersonic aircraft.[2] A matrix of supersonic scenarios evaluated over a wide range of mean fight altitudes and magnitudes of NO_x emissions confirmed previous analysis showing that ozone destruction becomes larger as the emissions of NO_x increases and as the altitude of injection increases. From this analysis, the effect on global ozone of a given supersonic prototype can be obtained.

The purpose of this study is to build on previous analyses of potential aircraft emission effects on ozone in order to better define the sensitivity of ozone to such emissions. With specific attention on the effects that heterogeneous chemical processes on background sulfuric acid aerosols may have on changes in global

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and local ozone from emissions of trace gases from propose subsonic and supersonic aircraft fleets.

The LLNL Two-Dimensional Model

The LLNL zonally averaged two-dimensional chemical-radiative-transport model currently determines the atmospheric distributions of 57 chemically active trace constituents in the troposphere and stratosphere.[2,9,10,11] The model domain extends from pole to pole, and from the ground to 56 km. The sine of latitude is used as the horizontal coordinate with intervals of about 10°. The vertical coordinate corresponds to the natural logarithm of pressure, $z^* = -H_0 \ln(p/p_0)$, where H_0 is the assumed scale height of 7.2 km and p_0 is the surface pressure (1013 mbar). The vertical resolution in (p/p_0) is 0.417 km or about 3 km.

Over 170 chemical and photochemical reactions are included in the model. Reaction rates, solar flux data, absorption cross sections, and quantum yields are based on the latest NASA panel recommendations.[12] Photodissociation rates, including the effects of multiple scattering, are computed as a function of time at each zone, with optical depths consistent with calculated species distributions.

The diabatic circulation for the ambient atmosphere is determined using net heating rates calculated in an internally consistent way with the derived species distributions. The technique for deriving the diabatic circulation is similar to that used by Solomon et al..[13] The vertical and horizontal velocities are determined from the zonally averaged residual Eulerian thermodynamic equation.

The net diabatic heating rates are determined using the best available solar and infrared radiative models. The solar model includes absorption and scattering effects for O_3, O_2, and NO_2 at ultraviolet and visible wavelengths, and for H_2O, CO_2, and O_2 in the near infrared. The solar model for visible and ultraviolet wavelengths uses a two-stream model to calculate reflection and transmission operators for scattering of diffuse incident radiation by a single layer.[14] Scattering from the solar beam is calculated for each layer using the delta-Eddington technique.[15] Merging of individual layers, including multiple-scattering , is accomplished via a flux formulation of the adding technique.[16] The longwave emission and absorption by O_3, CO_2, and H_2O are included in the infrared submodel.

Temperatures for the ambient atmosphere vary continuously, over the annual cycle, based on the reference model of Barnett and Corney.[17] The derived diabatic circulation depends strongly on the temperature distribution; by using observed temperatures for the ambient atmosphere, a more accurate representation of the diabatic circulation can be derived.

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

The chemical continuity equation for each individual species is solved using a variable time step, variable order, implicit technique for solving stiff numerical systems with strict error control. Advection terms are treated accurately using the two-dimensional transport algorithm of Smolarkiewicz.[18]

The diurnal-average concentrations for each species at each zone are calculated at each time step. Diurnal calculations are used to derive time-varying factors for each chemical and photochemical reaction included in the diurnal-averaged version of the model.

Turbulent eddy transport is parameterized through horizontal (K_{yy}) and vertical (K_{zz}) eddy diffusion coefficients.

Emission Scenarios for Supersonic and Subsonic Aircraft

Emission scenarios for this study are taken from a recent investigation conducted by NASA's High Speed Research Program.[19] In these scenarios both subsonic and supersonic aircraft fleets are represented for the year 2015. The trace gas emissions for the 2015 subsonic scenario are based on the Boeing B6 scenario.[20] The subsonic emission scenario was divided into two regions, flights under 400 miles or short range, and flights greater than 400 miles or long range. The total amount of fuel consumed is 20×10^9 kg/year and 150×10^9 kg/year for short and long range flights respectively. The altitude of injection for short range flight is between 0 and 9.1 km and between 9.1 and 12.2 km for long range flights. In all scenarios conducted in this study the subsonic scenario is held constant. The supersonic scenarios are divided into three different airframe prototypes; Mach 1.6, Mach 2.4, and Mach 3.2. Each airframe prototype has a characteristic cruise altitude (Mach 1.6, 14.3-17.4 km; Mach 2.4, 16.8-19.8 km; Mach 3.2, 21.3-24.4 km). The total amount of fuel consumed for the three prototypes is kept constant at 70×10^9 kg/year, which represents approximately 500 aircraft. For the above prototypes, fuel use during take off, climb, and descent is ignored.

The latitudinal distribution of fuel use in percent and the emission indices (EI) for NO_x, H_2O, CO, and hydrocarbons (HC) given in gm/kg fuel consumed are shown in Table 1. Currently, the sum of all hydrocarbon emissions are treated as CH_4. This is not correct, but was modeled in this manner due to the limited information about the actual chemical breakdown for the hydrocarbon emission values. In both the subsonic and supersonic emission scenarios, the NO_x emitted is 90% NO and 10% NO_2 on a molecular basis. As mentioned above, calculations using multi-dimensional models have shown that emissions of NO_x reduce ozone in the regions where the proposed supersonic fleets will fly. Therefore, in order to represent the uncertainties in future engine emission values, supersonic emission indices for NO_x are varied between 5 and 45 gm/kg of fuel consumed.

Table 1 Parameters for both subsonic and supersonic aircraft scenarios.

Latitudinal Distribution of Fuel Use (percent)		
Latitude (degrees)	Subsonic	Supersonic
80-90 N	0	0.6
70-80 N	0.4	0.7
60-70 N	2.9	0.7
50-60 N	15.7	12.3
40-50 N	25.2	28.4
30-40 N	31.6	18.4
20-30 N	11.0	8.4
10-20 N	3.7	6.7
Eq-10 N	2.4	6.3
Eq-10 S	1.7	4.9
10-20 S	1.6	4.2
20-30 S	1.6	4.0
30-40 S	2.0	3.1
40-50 S	0.2	1.3
50-90 S	0	0

Emission Indices (gm/kg fuel)		
Species	Subsonic	Supersonic
NO_x	20.7	as specified
H_2O	1230	1230
CO	1.1	1.5
HC (as CH_4)	0.2	0.2

Results of Proposed Aircraft Fleet Scenarios

In Figure 1, the percent change in NO_x ($NO+NO_2$) relative to a ambient 2015 atmosphere that does not include emissions of either subsonic or supersonic aircraft are shown for the Mach 1.6, 2.4, and 3.2 proposed aircraft fleets (including subsonic emissions). In this case, the EI of NO_x is 15 gm/kg of fuel consumed for each of the supersonic fleets represented in Figure 1. The increase of NO_x from the subsonic fleet peaks between 10-12 km at over 160% of the ambient value. There is a secondary peak attributed to the supersonic contribution near the cruise altitude for each prototype. The increase in NO_x is between 80% and 100%, depending on the supersonic prototype. It is interesting to note that even though over 90% of the NO_x from these proposed fleets are emitted in the Northern Hemisphere, increases of NO_x between 20% and 30% are calculated in the Southern Hemisphere. This indicates the important role that dynamics plays in the distributions of trace gases in the atmosphere, and also implies that uncertainties in the model treatment of these processes need to be carefully evaluated.

In this study, we have evaluated a total of fifteen proposed 2015 aircraft fleet scenarios. Table 2 describes each of these scenarios. For each of the 15 scenarios, Table 2 shows the calculated global averaged change in column ozone relative to a 2015 ambient atmosphere with and without trace gas emissions from proposed subsonic aircraft. For the results listed in Table 2, all model calculations used the normal gas-phase (or homogeneous) chemistry reactions. In all cases, column ozone decreases when NO_x is emitted by supersonic aircraft. There is an increased sensitivity to ozone depletion as the cruise altitude increases. The emissions of NO_x from the subsonic scenario increases column ozone by approximately 0.7% globally. This increase is due to the well known CH_4-NO_x-smog mechanism that occur in the troposphere. In Figures 2a-2e, both the local (altitude vs latitude, July) and column (latitude vs month) percent change in ozone relative to an ambient atmosphere without trace gas emissions from aircraft are shown for a proposed Mach 1.6, 2.4 and 3.2 fleet. The supersonic EI's for NO_x are identical for each prototype represented in Figure 2. The percent change in local ozone at 10 km increases as the cruise altitude decreases, showing the competition between odd oxygen loss from catalytic reactions occurring in the stratosphere and production from the CH_4-NO_x-smog reactions in the troposphere (see Figure's 2b, 2d, and 2e). For the Mach 1.6 scenario (Figure 2a), the subsonic contribution to percent change in column ozone is greater than the loss from the supersonic emissions of NO_x. This is not calculated to occur for the Mach 2.4 and Mach 3.2 cases.

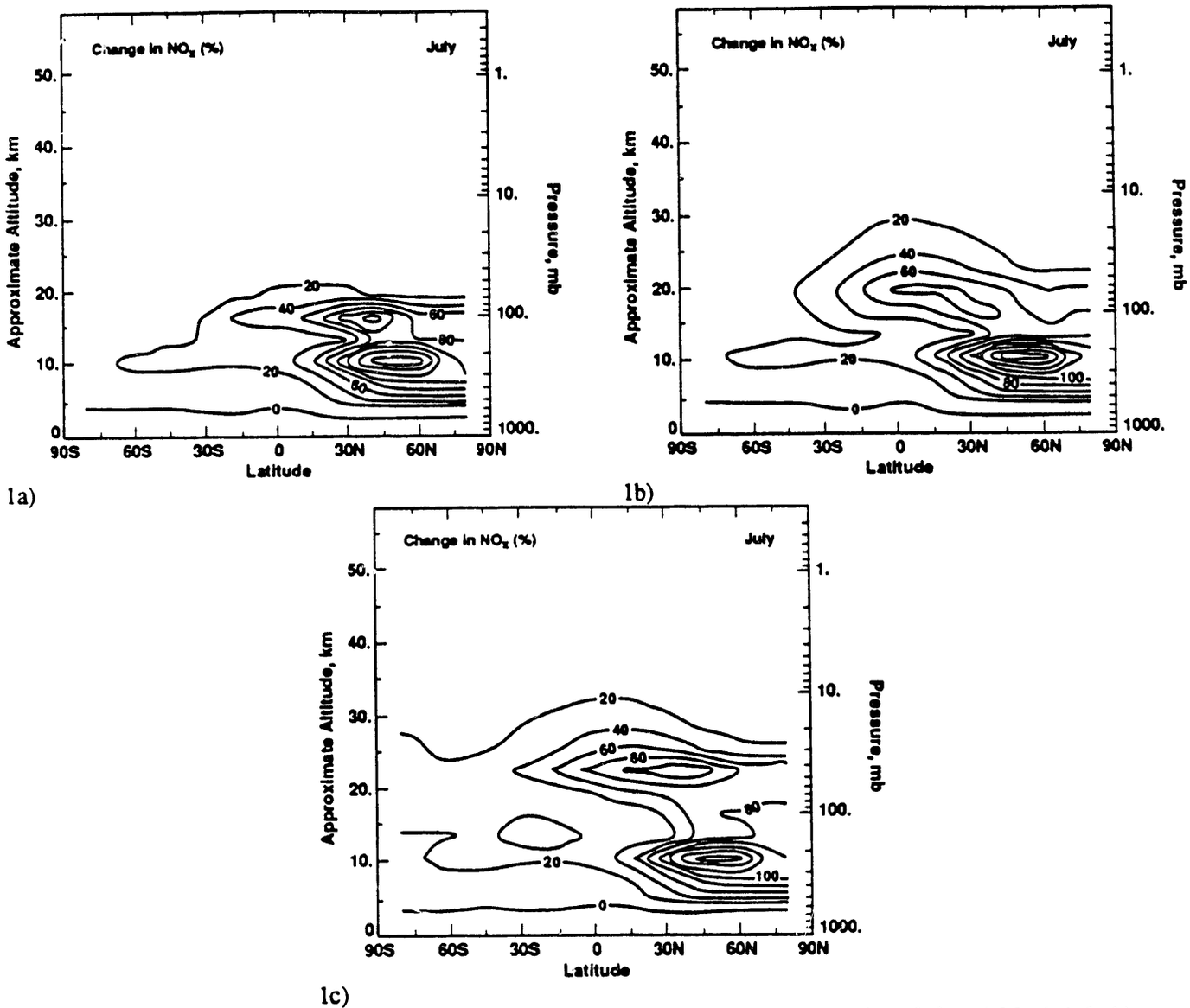


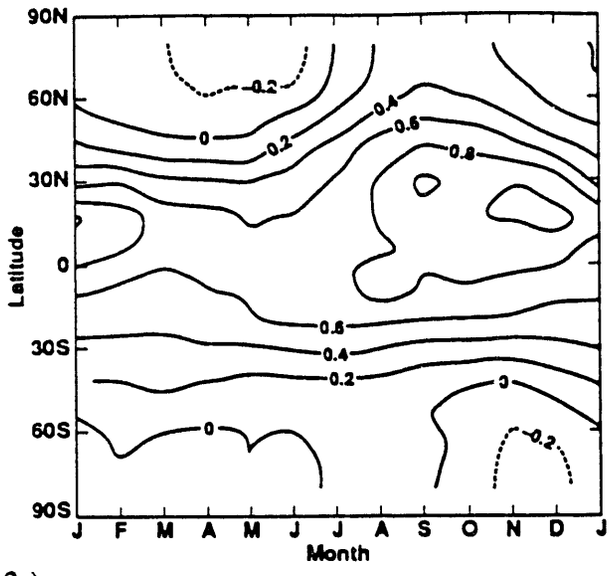
Fig. 1 Percent change in NO_x ($NO + NO_2$) relative to a 2015 atmosphere with no aircraft for: a) Mach 1.6 ; b) Mach 2.4; c) Mach 3.2 proposed aircraft fleets. Subsonic aircraft fleets are included in each scenario and are identical.

The stratospheric water vapor emissions due to aircraft emissions from hydrocarbon fuel combustion are much larger than the NO_x emissions. In Figure 3, the percent change in H_2O vapor for a Mach 3.2 fleet is shown relative to a 2015 ambient atmosphere without aircraft. Maximum increases of 28% in H_2O vapor occur at mid-latitudes in the Northern Hemisphere at approximately 21 km. Previous studies have shown that the effect of H_2O vapor emissions by itself is small, but it has a larger impact when included in combination with the NO_x emissions (see Table 2).[1] 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 to the interactions between NO_x and HO_x chemistry. It is also interesting to point out that there is an increase in H_2O vapor between 4% and 8% at high latitudes in the Southern Hemisphere. The impact that this increase may have on Polar Stratospheric Cloud formation probabilities, with resulting effects on the Antarctic ozone "hole" is unknown at this time.

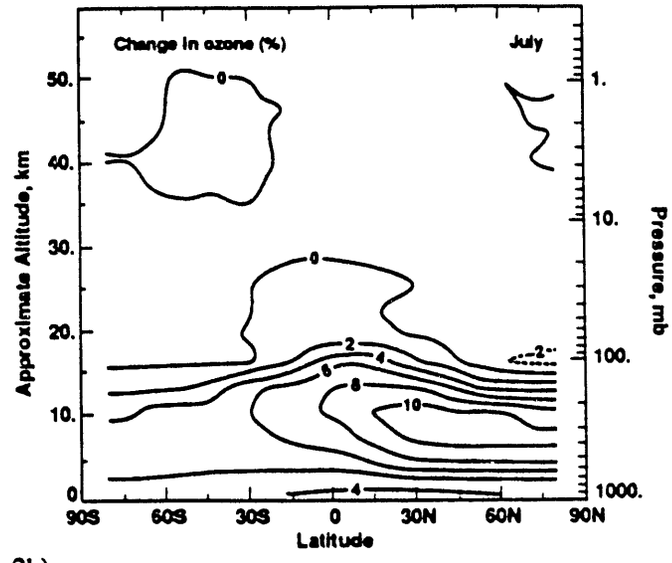
Table 2: Results of proposed 2015 aircraft scenarios using gas phase chemical reactions rates only.

Mach Number	HSCT EI (g NO_2) /kg of fuel	HSCT NO_x (molec/yr) x 10^{34}	NO_x Emissions only	NO_x,H_2O Emissions	NO_x,H_2O CO, CH_4 Emissions	Percent Change in Annually and Globally Averaged Ozone	
						+	++
1.6	5	0.46	YES	-	-	0.61	-0.10
1.6	5	0.46	-	-	YES	0.61	-0.10
1.6	15	1.37	YES	-	-	0.40	-0.31
1.6	15	1.37			YES	0.40	-0.31
2.4	5	0.46	YES	-	-	0.18	-0.53
2.4	5	0.46	-	-	YES	0.20	-0.51
2.4	15	1.37	YES	-	-	-1.06	-1.76
2.4	15	1.37	-	-	YES	-1.04	-1.74
2.4	45	4.12	YES	-	-	-5.42	-6.06
3.2	5	0.46	YES	-	-	-0.47	-1.17
3.2	5	0.46	-	YES	-	-0.22	-0.92
3.2	5	0.46	-	-	YES	-0.23	-0.93
3.2	15	1.37	YES	-	-	-3.08	-3.77
3.2	15	1.37	-	YES	-	-2.70	-3.38
3.2	15	1.37	-	-	YES	-2.71	-3.39

+ Effect of both subsonic and supersonic emissions on ozone
++ Effect of supersonic emissions only on ozone



2a)



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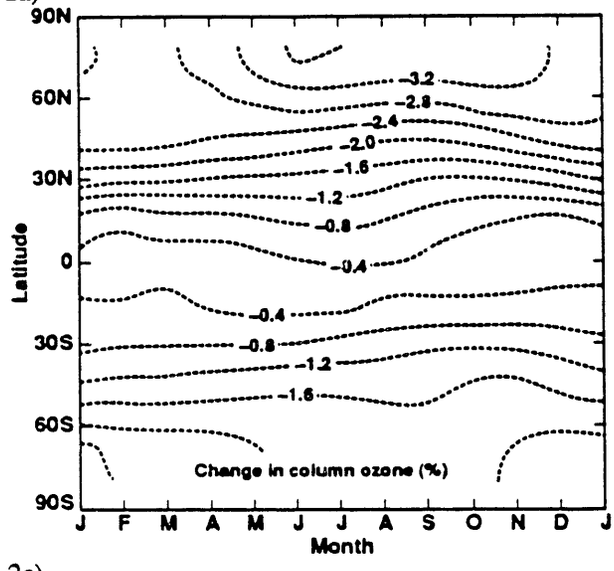

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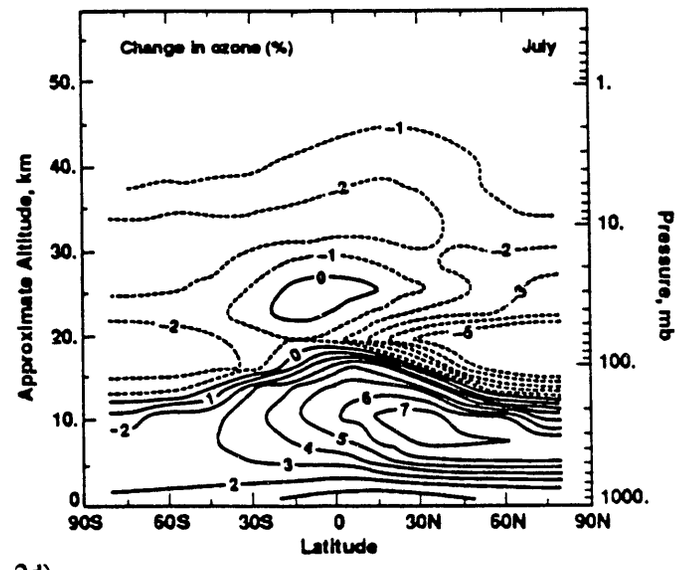

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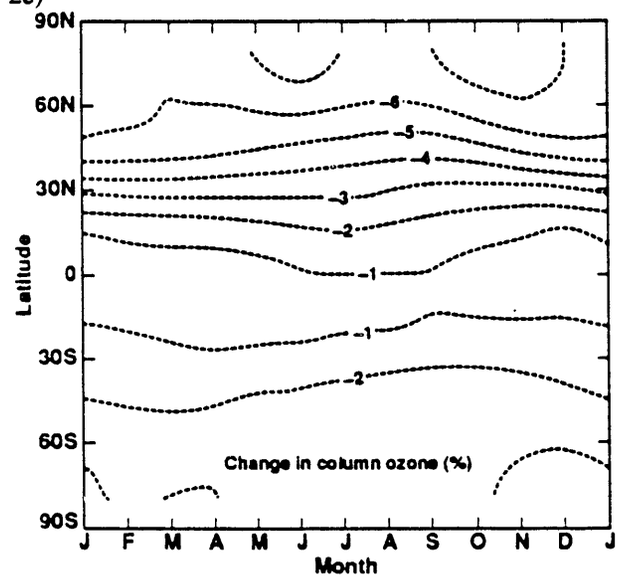

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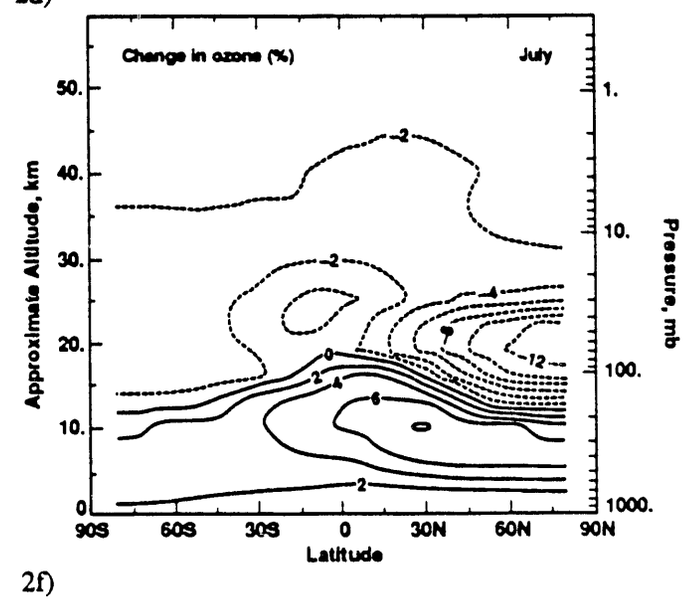

2f)

Fig. 2 Percent change in ozone relative to an ambient atmosphere without aircraft emissions for : a) column ozone, Mach 1.6; b) local ozone July, Mach 1.6; c) column ozone, Mach 2.4; d) local ozone for July, Mach 2.4; e) column ozone, Mach 3.2; f) local ozone for July, Mach 3.2.

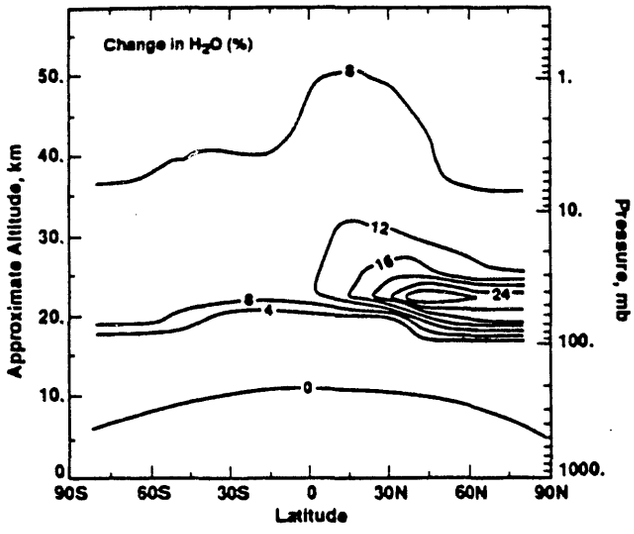

Fig. 3 Percent change in H_2O vapor for a Mach 3.2 aircraft fleet relative to an ambient atmosphere without aircraft emissions.

Heterogeneous Chemistry on Background Sulfuric Acid Aerosols

In the previous section, the chemical reactions included in the model were all gas-phase (or homogeneous) reactions. Recently, laboratory studies suggest that heterogeneous chemical processes may play an important role in lower stratospheric chemistry. Studies indicate that heterogeneous chemistry is important in explaining the Antarctic ozone "hole".[21,22] In addition, ozone depletion from heterogeneous chemical processes may be important globally. One very recently theoretical study suggested that including heterogeneous reactions that convert N_2O_5 and $ClONO_2$ to HNO_3 on the background sulfuric acid aerosol layer will change the partitioning of odd oxygen loss processes between the total odd nitrogen (NO_y), total odd chlorine (Cl_y), and total odd hydrogen (HO_y) families, in such a manner, that additional increases in NO_x (i.e., from aircraft) would produce ozone.[23] Where odd oxygen loss is defined as the process of converting either oxygen atoms or ozone molecules into another species (i.e., O_2). In this study, we also investigated these issues by adding the following reactions to the LLNL 2-D model chemistry package:

$$N_2O_5\ (g)\ [+H_2O \text{ in aerosol}] \rightarrow 2\ HNO_3\ (g) \qquad (1)$$

$$ClONO_2\ (g)\ [+H_2O \text{ in aerosol}] \rightarrow HNO_3\ (g) + HOCl\ (g) \qquad (2)$$

Since a complete treatment of these reactions would require a sophisticated aerosol microphysical model, which currently has both theoretical and practical limitation, we treated these reactions using the following relationship to calculate the two rate constants for the above cases.

$$K = (V)(\gamma)(\text{Surface Area})$$

V= Effective collision velocity = 5200 cm/s.

Surface Area = Based on analysis of Sage II data by Poole, Thomason, and Yue (see chapter's 3 and 8 in the UNEP/WMO international assessment on ozone, 1991). The surface area distribution has altitude (12-32 km), latitude (90N-90S), and temporal resolution. Because of the uncertainty in calculating an aerosol surface area distribution, two different approaches were used. The two approaches varied in magnitude from each other by a factor of four. We designated the two surface area distributions in this study as "large" and "small".

γ = Reaction probability per collision. In this study, the reaction probability was varied for reaction 1 (see Table 3). Laboratory measurements suggest that the reaction probability for N_2O_5 on sulfuric acid aerosols is around 0.1 and does not have a temperature dependence (see UNEP/WMO,1991)[24] In this study, we reduced the reaction probability by a factor of five in order to estimate the sensitivity of this reaction on the chemical composition in the lower stratosphere. The reaction probability for $ClONO_2$ on sulfuric acid aerosols does have a temperature dependent expression:[24]

$$\gamma_2=0.006\exp(-0.15(T-200)).$$

The sensitivity of reaction 2 was not investigated in this study.

When including these two heterogeneous reaction in the LLNL 2-D model, we also calculated a repartitioning of odd oxygen loss between the NO_y, Cl_y, and HO_y families. In fact, the column ozone distribution decreased in our 2015 ambient atmosphere when these heterogeneous reactions were included. This is due to the replacement of odd nitrogen in the lower stratosphere from its active catalytic form to a species like nitric acid, HNO_3, which has a long photochemical lifetime. With reduced concentrations of odd nitrogen in this region, the interactions between the NO_y and the Cl_y chemical families are decreased, releasing active forms of odd chlorine that are more effective in

destroying ozone than the odd nitrogen species they replaced (e.g, the total integrated odd oxygen loss in this region of the stratosphere increased).

In Figures 4a-d, the effects of converting NO_x to HNO_3 is calculated by the LLNL 2-D model. For example, in Figure 4a, the heterogeneous reactions described above are not included and the maximum mixing ratio values for the high latitude, Northern Hemispherical distribution of HNO_3 is calculated to be less than 6 ppbv. When comparing this distribution to data observed on the Nimbus 7 satellite, using the LIMS instrument, one observes values in the range of 12 ppbv.[25] After incorporating the heterogeneous reactions described above, the HNO_3 distribution increases at high latitudes, when the magnitude of the reaction probability for N_2O_5 on background sulfuric acid aerosols is increased. In this study, the observed distribution for HNO_3 compares best with Figure 4c. This does not mean that having a reaction probability of 0.1 for N_2O_5 on sulfuric acid aerosol is necessarily the correct magnitude. In order to completely understand these processes, a complete distribution of all the important trace gases that are involved in odd oxygen loss in this region of the stratosphere would need to be measured and compared to model calculations. Currently this is not possible, although data from the Upper Atmospheric Research Satellite (UARS) should help this over the coming year.

In Table 3 and Figures 5a-f, the effect on ozone from using heterogeneous reaction rates for the conversion of $ClONO_2$ and N_2O_5 on background sulfuric acid aerosols is shown for the Mach 2.4 aircraft fleet scenario. As the reaction probability for N_2O_5 on background sulfuric acid aerosols is increased the percent decrease in column ozone relative to a 2015 atmosphere without aircraft becomes less negative. In fact, as the magnitude of the reaction probability is changed from 0.02 to 0.1 or by a factor of five, the percent change in column ozone increases. Even when the reaction probability is 0.02, the percent change in globally and annually averaged ozone is only -0.36% (Table 3), instead of -1.76% when gas-phase only chemistry is used (Table 2). In addition, the percent change in tropospheric ozone from emissions of NO_x from the proposed subsonic fleet increases as the reaction probability magnitude increases (see Figures 5b,d,f). This region of the atmosphere, as Lacis et al., points out, is the most sensitive to an increase in radiative forcing from a given changes in ozone.[26] Therefore, increased ozone in this region, from subsonic aircraft emissions, could potentially have a significant effect on radiative forcing of climate contribution to global warming.

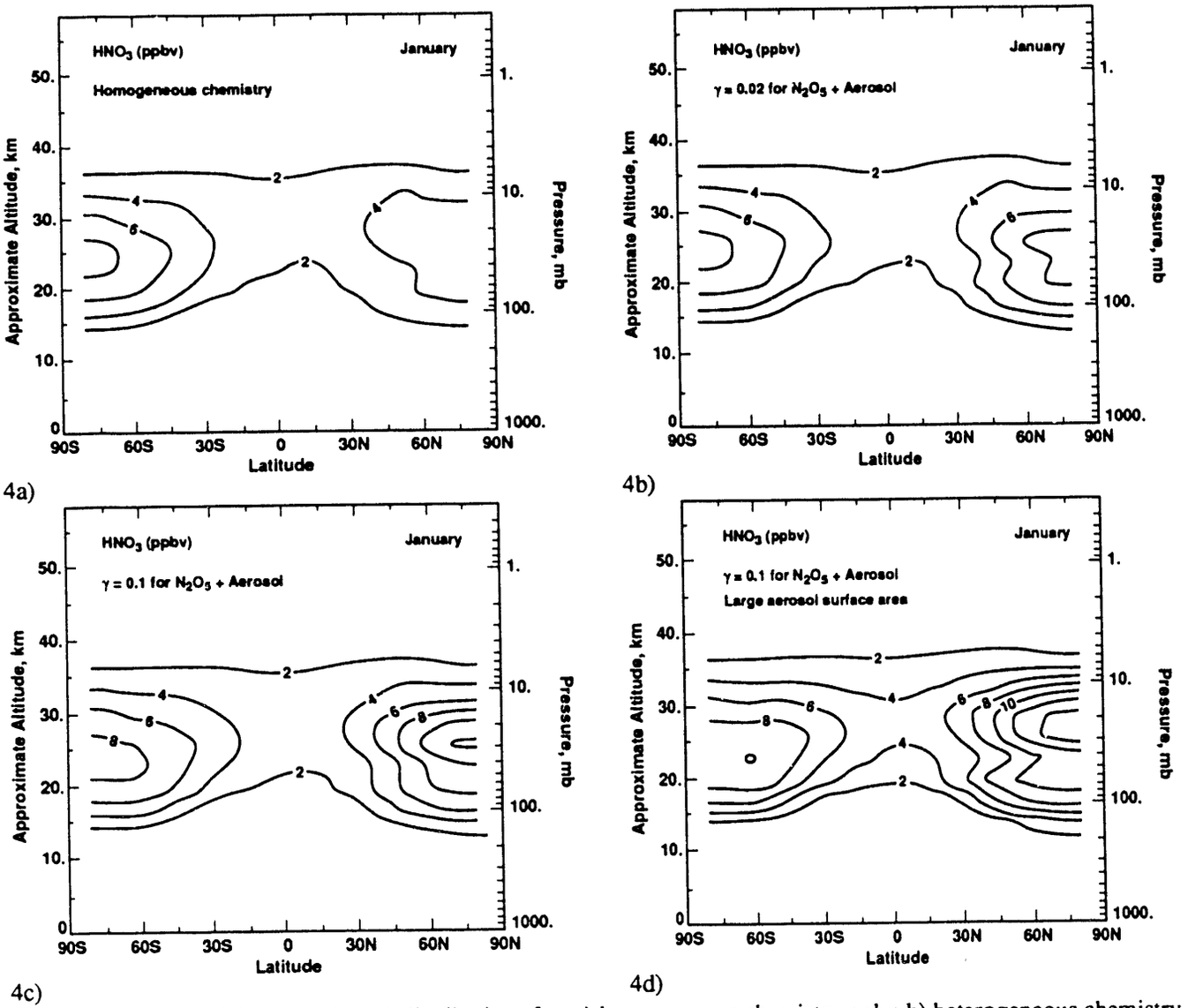

Fig.4 Nitric acid mixing ratio (ppbv) distributions for: a) homogeneous chemistry only; b) heterogeneous chemistry set with γ =0.02 for N_2O_5 + aerosol, small aerosol surface area; c) γ =0.1; small aerosol surface area; d) γ =0.1, large aerosol surface area.

Table 3 Results of proposed aircraft scenario Mach 2.4, EI NO_x 15, NO_x emissions only, using heterogeneous reaction rates for N_2O_5 and $ClONO_2$ on background sulfuric acid aerosols. Relative to a 2015 ambient atmosphere with no aircraft.

Factor	Relative Aerosol Surface Area	N_2O_5 Reaction Probability	Percent change in column ozone (%) Global	Northern Hemisphere	Southern Hemisphere
0.2	small	0.02	-0.36	-0.48	-0.24
1.0	small	0.1	0.28	0.33	0.24
4.0	large	0.1	0.82	0.98	0.66

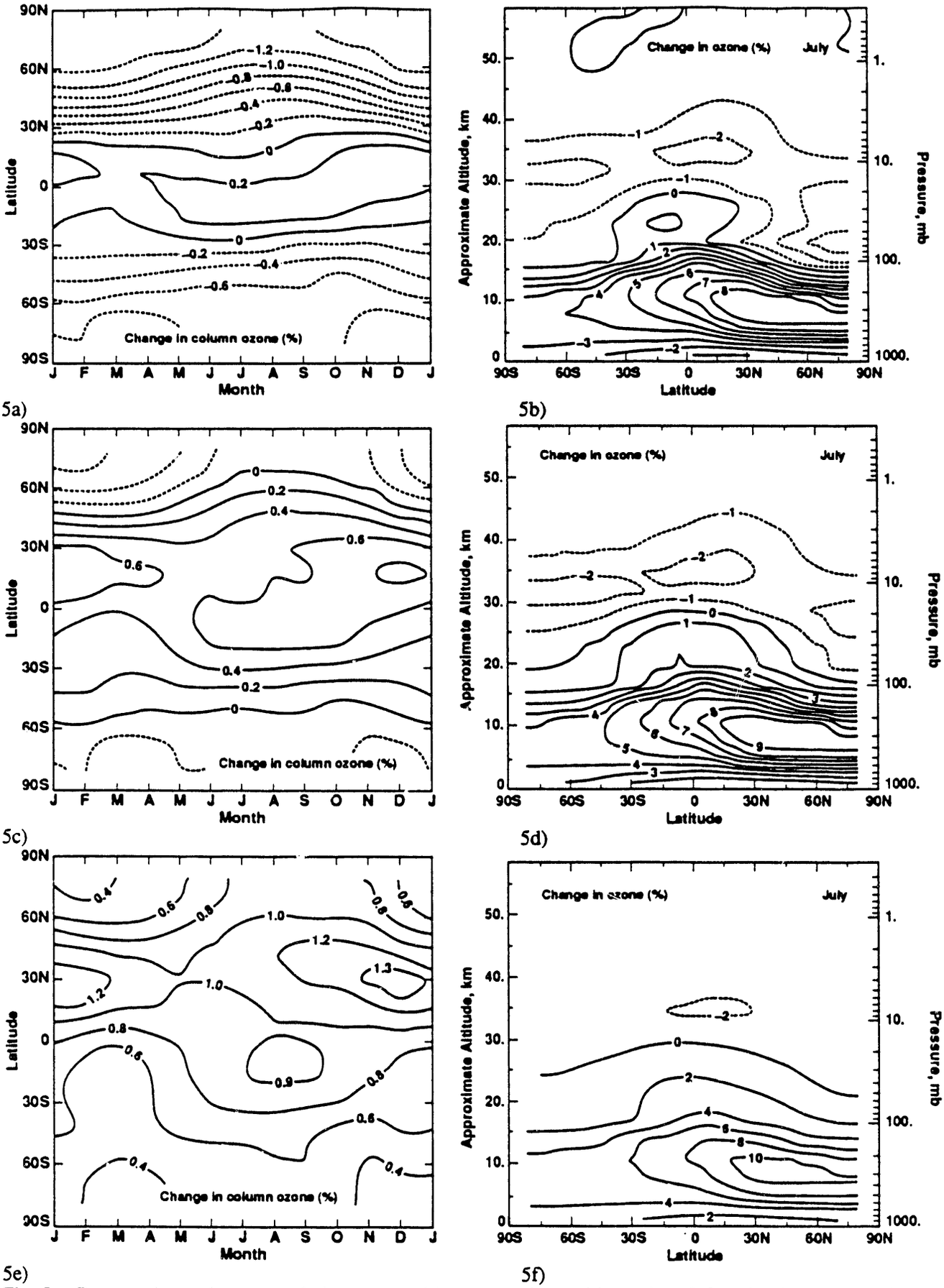

Fig. 5 Percent change in ozone relative to an ambient atmosphere without aircraft for: a) column ozone, heterogeneous chemistry set with γ =0.02 for N_2O_5 + aerosol, small aerosol surface area; b) local ozone, with γ=0.02, small aerosol surface area; c) column ozone, γ =0.1, small aerosol surface area; d) local ozone, with γ =0.1; small aerosol surface area; e) column ozone, γ =0.1, large aerosol surface area, with γ =0.1; f) local ozone, with γ=0.1, large aerosol surface area.

Conclusions

In this study heterogeneous reactions that convert $ClONO_2$ and N_2O_5 on background sulfuric acid aerosols to HNO_3 were found to change the partitioning of odd oxygen loss between the NO_y, Cl_y, and HO_y families. Decreasing the odd oxygen loss from the NOy family and increasing the odd oxygen loss for both the Cl_y and HO_y families. The overall integrated odd oxygen loss was greater when the heterogeneous reactions were present.

Incorporating these heterogeneous reaction, has major implications on the amount of ozone depletion from a given aircraft emission of NO_x. For the emission of NO_x from supersonic aircraft, depending on the choice for the reaction probability of N_2O_5 on sulfuric acid aerosols, what was once a decrease in ozone from a given injection of NO_x (using gas-phase chemistry only), now shows an increase. In fact, having these reactions present actually increases the concentration of ozone in the middle to upper troposphere from the subsonic aircraft fleet by a larger amount than is observed for the gas-phase chemistry only case.

However, one should keep in mind that these results are preliminary, and that more research needs to be completed in the laboratory to measure the magnitude of the reaction probabilities of N_2O_5 and $ClONO_2$ on sulfuric acid aerosol under conditions that are representative of stratospheric composition, temperatures, and pressures. In addition, the sulfuric acid aerosol surface area needs to be determined more accurately, with the ultimate goal of modeling the growth and destruction of these aerosols from microphysical processes. This will allow us to estimate the environmental impact of natural and anthropogenic emissions (i.e., volcanic eruptions or aircraft emissions) of sulfur containing species on the surface area available for heterogeneous reactions.

Using more accurate values for the reaction probabilities and the sulfuric acid aerosol surface area, coupled with a more complete observed species data base to compare model derived species distributions with, we will have a better understanding of the effects that emissions of trace gases from both subsonic and supersonic aircraft will have on ozone and other chemical and radiatively important trace gases.

Acknowledgments

Work at Lawrence Livermore National Laboratory was performed under the auspices of the U.S. Department of Energy under Contract W-7405-ENG-48 and was supported by the NASA's High Speed Research Program.

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DATE FILMED

12/10/91