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SENSITIVITY STUDY OF GLOBAL OZONE TO NOX EMISSIONS FROM AIRCRAFT

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There recently has been new interest in the development of faster and more efficient aircraft for intercontinental passenger flights [Ott, 1988]. Such aircraft would probably spend a large fraction of their flight time in the stratosphere, perhaps as high as 35 km. As a natural progression from studies that were done in the early 1970's [Johnston, 1971; CIAP, 1975; WMO, pages 727, 740, 773, 1985], this study investigates the sensitivity of stratospheric ozone to NO_X emissions in conjunction with current understanding of atmospheric chemical and physical processes.

The LLNL one-dimensional and new two-dimensional chemical-radiative-transport models of the troposphere and stratosphere are used in this investigation. The two-dimensional model gives latitudinal, altitudinal, and seasonal information; and the computationally faster one-dimensional model is used for sensitivity studies. Because of uncertainties in possible future emissions, it is necessary to examine the model sensitivity to a wide range in magnitude, altitude, and latitude of assumed NO_X emissions. As an initial study, the models are used in simulation of a typical scenario from the CIAP era, and results are compared with those of a

published two-dimensional model, which includes some three-dimensional features [Alyea et al., 1975; Cunnold et al., 1977]. This investigation lays the ground work for future studies including different background amounts of ClX and for other aircraft emission scenarios.

ATMOSPHERIC MODELS

1. Description of LLNL Two-Dimensional Model. The LLNL zonally-averaged two-dimensional chemical-radiative-transport model currently determines the atmospheric distributions of 31 chemically active atmospheric trace constituents in the troposphere and stratosphere. The model domain extends from pole to pole, and from the ground to 0.56 mb (approximately 0 to 54 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 mb). The vertical resolution is $\ln(p/p_0) = 0.417$ or about 3 km. Ninety-five 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 [DeMore et al., 1987], except that the old value is used for the rate of the reaction, $O + NO_2 = O_2 + NO$. Photodissociation rates, including the effects of multiple scattering, are computed as a function of time at each zone, with optical   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. [1986]: the vertical velocity is determined from the zonally averaged residual Eulerian thermodynamic equation, while the horizontal velocity is determined using the equation for mass continuity. The net heating rates are determined using accurate solar and infrared radiative models.

Temperatures for the ambient atmosphere vary continuously, over the annual cycle, based on the reference model of Barnett and Corney [1985]. 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.

Turbulent eddy transport is parameterized through diffusion coefficients K_{yy} and K_{zz}. In the current version of the model, a value of K_{yy} of 2×10^9 cm^2 s^{-1} is assumed at all stratospheric altitudes and latitudes; and values of are assumed in the troposphere, with a transition region at opause. Values of

K_{zz} are 1×10^3 cm^2 s^{-1} in the lower stratosphere, increasing slowly with altitude based on gravity wave modeling studies.

The 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 (1984). The diurnal-averaged concentrations for each species at each zone are calculated at each time step. Accurate diurnal calculations are used to derive time-varying factors for each chemical and photochemical reaction included in the diurnal-averaged version of the model.

2. One Dimensional Model. The LLNL one-dimensional eddy-diffusion model of the troposphere and stratosphere was also used in this study. The details of this model are discussed in Luther et al. [1979] and Wuebbles et al. [1983], with photochemical rate coefficients from DeMore et al. [1985, 1987]. Briefly the 1-D model contains 37 species. There are 119 chemical and 46 photochemical reactions included in the model. The 1-D model atmosphere is divided up into 44 vertical layers which extend from the surface up to 56 km. Transport of trace species is controlled by an empirical eddy diffusion function (K_z) which was determined by long-lived atmospheric tracers (N_2O, CH_4, CFC-11, CFC-12, and CH_3Cl).

The boundary values of the reference atmosphere were selected to reproduce the composition of 1988. The mixing ratios and altitudes of various families of species are: 3.1 ppbv of ClY at 55 km, 13 pptv of BrY at 55 km, and 19:4 ppbv of NOY at 37.5 km.

RESULTS

Hemispherical and global average ozone changes from 2-D model calculations are given in Table 1-A; and results of one-dimensional model calculations at similar altitudes are given in Table 1-B. The standard injection is taken to be 1.8 million metric tons (10^9 kg) of NO_x (as NO_2) per year [CIAP, 1975; Alyea et al., 1975; Cunnold et al., 1977]. For this standard NO_2 injection, spread uniformly at 19.5 ± 1.5 km and between 37 and 49 degrees north, the calculated percentage changes of the ozone vertical column are presented as a function of latitude and season in the top panel of Figure 1, and the local ozone percentage changes are given as a function of latitude and altitude in the lower panel of Figure 1. For the same NO_2 injection at 31.5 ± 1.5 km, the column and local percentage changes of ozone are given in Figure 2.

With the less expensive 1-D model, a wide range of calculations was made, and these results are summarized as a contour plot of percentage ozone-column change as a function of injection rate and altitude of injection, Figure 3. In the 1-D model the injection rate of 1500 molecules $cm^{-3}s^{-1}$ over a kilometer vertical band corresponds to a global injection of 1.8 MT yr^{-1} of NO_2, so that Figure 3 spans injection rates from zero to twice the standard rate. In terms of aircraft, these altitudes roughly correspond to current commercial aircraft (12 km), to the French-British Concorde (17 km), to the 1971 American-planned supersonic transport (20 km), and to possible future hypersonic aircraft (30 to 34 km).

DISCUSSION OF RESULTS

For the standard NOX injection at low altitude (19.5±1.5 km) in the 2-D model, the maximum ozone column change of -9 % occurs in the polar region of the northern hemisphere; the maximum local concentration change is -16 % near the injection corridor (Figure 1); the global average ozone change is -3.3%; and the ratio of total ozone change in the northern to southern hemisphere is 3.1 (Table 1). For injections at high altitude (31.5±1.5 km), the 2-D model gives a maximum ozone column change of -32 % in the winter polar regions; the maximum local ozone change is -55 % (Figure 2); the global average ozone change is -8.0 %; and the hemispherical ratio for ozone change is 6.9 (Table 1). In comparison, the older three/two dimensional calculations for a 20 km injection altitude gave a global average ozone change of -12 % and a hemispherical ratio of 2/1 [Cunnold et al., 1977]. There have been many changes in photochemical parameters and methods of treating atmospheric motions since 1977, and these comparisons are primarily of historical interest.

The 1-D results for ozone vertical column may be compared with 2-D results for total global ozone (Table 1). For the standard injection centered near 20 km, the calculated global-average ozone change is -3.3 % for the 2-D model and -8.7 % for the 1-D model, using the same photochemistry. For the same injection near 30 km, the ozone changes are -8.0 % for the 2-D model and -12.5 % for the 1-D model. For an NO_2 injection of 1.8 MT yr^{-1} at 34.5±1.5 km, the 2-D global-average ozone change is -7.2 % when the injection is

made between 37 and 49 degrees north, -7.1 % when spread uniformly
over the northern hemisphere, and -8.0 % when spread uniformly over
the globe; the corresponding ozone change calculated by the 1-D
model is -10.5 %. It is interesting that the 2-D model gives
approximately the same global-average ozone change regardless of
latitude spread of the injection band. When the injection is all in
the northern hemisphere, the average ozone reduction in the northern
hemisphere is four to seven times that in the southern hemisphere.

For the standard NOX injection at 34.5 km, one calculation was
made omitting temperature feedback, and the global average ozone
change is -7.2 % with temperature feedback and -7.7 % without
temperature feedback, a small difference. For NOX injections of
0.45, 0.9, and 1.8 MT yr^{-1}, the global-average ozone changes are
-2.1 %, -4.0 %, and -7.2 %, respectively, which is slightly slower
change than linear.

The 1-D model gives larger ozone reductions than the 2-D model:
by a factor of 1.5 when injections occur at 30 km and by a factor
of 2.6 for injections at 20 km. Reasons for these differences are
being investigated, and it appears to be related to differences in
transport rates in the two models. The lifetime of excess NOX
injected into the stratosphere was calculated from the relation

$$\text{lifetime} = t(NOY) = \frac{\text{global excess NOY at steady state}}{\text{global NOX injection rate}}$$

for both 1-D and 2-D models, and these values are included the
last column in Table 1. For NOX injection at 30 to 35 km. 1-D
lifetime of excess NOY is twice that for the 2-D model;
injection altitude the 1-D lifetime is almost five ti. s

that for the 2-D model. As can be seen from Figures 1 and 2, the 2-D model sweeps NOY out of the northern hemisphere faster than it transports NOY to the southern hemisphere. Studies are underway testing these models against atmospheric carbon-14 data to see which model gives the more nearly correct stratospheric lifetime to an inert tracer.

The calculated ozone reduction is slightly greater (10 to 20 %) when we use the new [DeMore et al., 1987] temperature dependent value for the rate coefficients for the reaction $O + NO_2$, relative to that for the constant value, 9.3×10^{-12} cm^3 s^{-1}, Table 1-B.

The ozone-column changes calculated with the 1-D model for altitudes between 12 and 34 km and for injection rates between 0 and 3000 molecules $cm^{-3}s^{-1}$ over a one km band (0 to 3.6 MT yr^{-1} of NO_2) are shown as a contour plot in Figure 3. These calculations were made with the 1985 photochemical coefficients [DeMore et al. 1985] before the 1987 values were published, and stratospheric ClX is taken to be 3.1 ppbv. According to Figure 3, injected NOX below 13 km in any amount increases the ozone column. Above 13 km, injected NOX decreases the ozone column, increasing with rate of injection and with altitude. Above 30 km, the reduction of the ozone column decreases with altitude for a given injection rate; this feature is also given by the 2-D model. There are not enough calculated points for this contour plot to be valid in fine detail; it should be used only to get a broad picture of the joint effects of injection altitude and injection magnitude. In particular, this contour plot does not accurately give the injection altitude for maximum ozone-

column reduction. Further study of this problem is in progress, including large changes in ClX.

TITLES TO FIGURES

Figure 1. Percent change in ozone for injection of 1.8 MT yr^{-1} of NO_2, where the latitude of injection is between 37 and 49° N and the altitude of injection is between 18 and 21 km. The upper panel shows the change of the ozone vertical column as a function of latitude and month of the year. The lower panel shows the change of local ozone (as of Jan. 1) as a function of latitude and altitude. Photochemical coefficients are from DeMore et al. [1987], except for use of older value [DeMore et al.; 1985] for reaction $O + NO_2$.

Figure 2. Same as Figure 1, except the injection altitude is between 30 and 33 km, nominally 31.5 km.

Figure 3. Percentage change of the ozone vertical column as calculated by the one-dimensional model as a function of altitude of NOX injection and magnitude of NOX injection in units of molecules cm^{-3} s^{-1} over a one kilometer band centered at the stated altitude. The stratospheric $\therefore$ is 3.1 ppbv, which is approximately the 1988

value. An injection of 1500 in these units corresponds of 1.8×10^{12} g (as NO_2) yr^{-1}. Photochemical coefficients are from DeMore et al. [1985].

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Table 1.   Calculated changes in global
and hemispherical ozone vertical columns as a
result of NOX injections by stratospheric
aircraft.

A. LLNL two-dimensional model. The boundary
values include 300 ppbv of nitrous oxide and
enough chlorofluorocarbons to give 2.8 ppbv of
stratospheric chlorine at 50 km. JPL 1987,##.

NOX Injection as NO_2			% Ozone Change			t(NOY)*
km	Lat.N	MT/yr	Global	N.H.	S.H.	yr
16.5	37-49	1.8	+0.01	-0.02	+0.04	0.3
19.5	37-49	1.8	-3.3	-5.0	-1.6	1.2
31.5	37-49	1.8	-8.0	-13.8	-2.2	1.8
34.5	37-49	1.8	-7.2	-12.4	-1.8	1.8
34.5	37-49	1.8 #	-7.7	-13.1	-2.3	
34.5	N.Hemi.	1.8	-7.1	-11.3	-2.9	1.8
34.5	Global	1.8	-8.0	-7.5	-9.6	1.9
34.5	37-49	0.9	-4.0	-6.9	-1.0	
34.5	37-49	0.45	-2.1	-3.6	-0.5	

B. One-dimensional model results. 3.1 ppbv
ClX, #.

		JPL [DeMore et al.]			t(NOY)*
km	MT yr^{-1}	1985##	1987##	1987	yr
12	1.8	+0.2			
17	1.8	-2.9	-2.9	-3.5	1.5
20	1.8	-8.6	-8.7	-10.0	2.7
30	1.8	-12.1	-12.5	-13.4	3.9
34	1.8	-10.1	-10.5	-11.4	3.6

* Stratospheric residence time of excess NOY
calculated as ratio of steady-state excess
inventory of NOY over injection rate of NOX.
\# Temperature feedback is not included in
this case.
\#\# Old, slow rate for reaction O + NO_2.
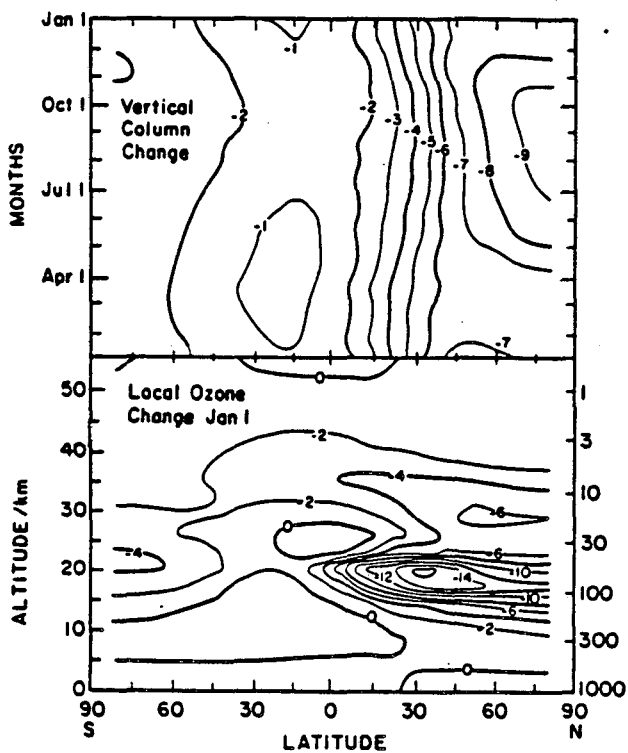
PERCENT CHANGE IN OZONE INJECTION AT 19.5 KM

Fig. 1

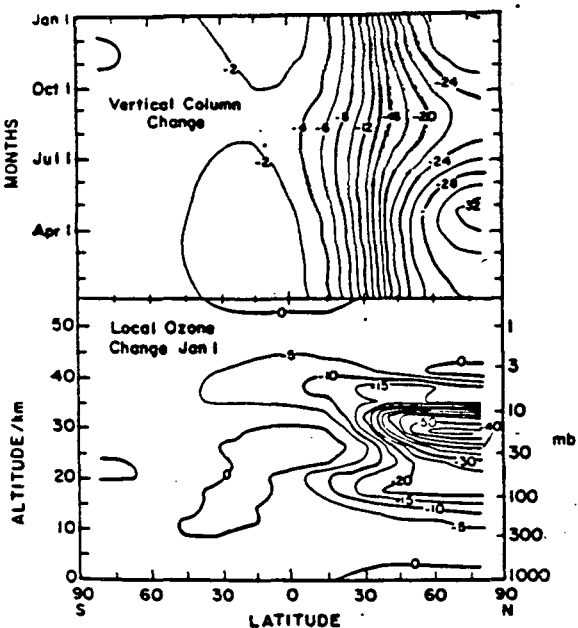PERCENT CHANGE IN OZONE INJECTION AT 31.5 KM

Fig. 2

PERCENTAGE CHANGE OF OZONE VERTICAL COLUMN
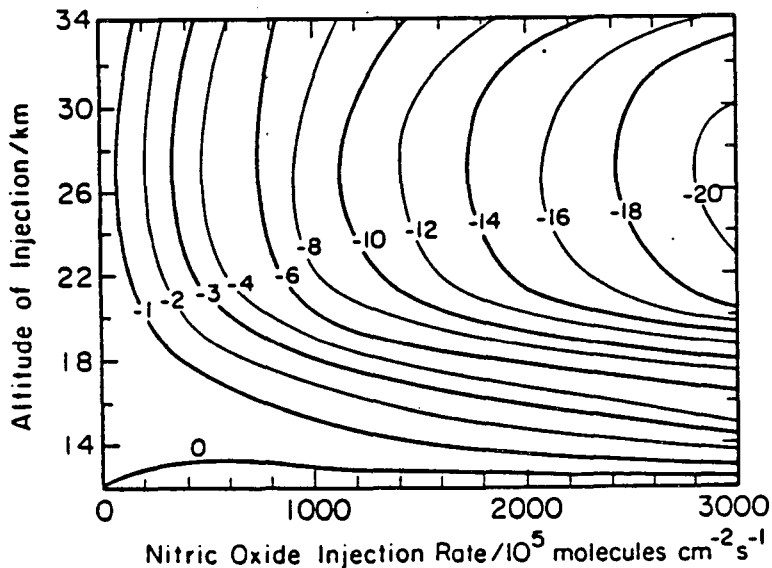STRATOSPHERIC CIX, = 3.1 ppbv

Fig. 3