

ENVIRONMENTAL EFFECTS OF AIRCRAFT AT CRUISE :

AN UPDATE

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SUMMARY

New laboratory determinations of chemical reaction rates and modeling refinements have shown that the effect of cruise-altitude emissions on stratospheric ozone has changed from one of ozone decrease to one of slight increase. The situation, however, is not yet fully resolved, since the uncertainties in the model predictions have not been adequately quantified. The status of the calculations of ozone change due to high altitude aircraft is critically reviewed and important areas of uncertainty identified.

HISTORICAL BACKGROUND

During 1971-1975, the U.S. Department of Transportation (DOT) completed the first comprehensive assessment of the effects of stratospheric pollution by aircraft (1). This assessment, known as the Climatic Impact Assessment Program (CIAP), was the first systematic, multidisciplinary Federal study on the stratosphere. Concurrently with CIAP, the National Academy of Sciences/National Academy of Engineering (hereinafter referred to as NAS) in the U.S., and the British and the French governments had conducted independent studies (2, 3, 4) on the same problem; the British study was conducted by the Committee on Meteorological Effects of Stratospheric Aircraft (COMESA) and the French, by the Comite d'Etudes sur les Consequences des Vols Stratospheriques (COVOS).

Studies prior to CIAP on the pollution effects of supersonic transports (see 5 for example) had suggested that the water vapor in the exhaust could cause two effects: (1) stratospheric ozone could be depleted by a catalytic chemical reaction set involving the water-related radicals; and (2) by altering the occurrence of high cirrus and by forming persistent contrails, the injected water vapor could alter the radiation balance of the earth's surface and hence its climate. In 1971, during Congressional hearings on supersonic transport (SST) development in the United States, possible adverse health effects were for the first time linked to large scale commercial SST operations (6). The health hazard envisaged was possible increase in the incidence of skin cancer in fair-skinned humans owing to the fact that less stratospheric ozone would allow more of the biologically harmful solar ultra-violet (UV) radiation to penetrate to the ground which could induce and/or promote skin carcinomas.

The CIAP and other parallel studies had concluded that the nitrogen oxides (collectively known as NO_x and comprised of nitric oxide, NO , and nitrogen dioxide, NO_2) in the exhaust had far greater potential to deplete ozone than water vapor, despite the larger water vapor emissions (as may be seen from Table 1, adapted from (7), which lists the emission indices of various exhaust species in gm per kg of fuel burned). Thus, large scale commercial SST operations could indeed harm the environment with undesirable consequences. The CIAP Report of Findings (1) in its Executive Summary stated, in 1974:

"Develop, within the next year, a plan for a proper program for international regulation of aircraft emissions and fuel characteristics for whatever stratospheric flight operations may evolve in the future."

"Accelerate combustion research and engine development programs needed to make stratospheric flight possible with specified nitrogen oxide emission standards."

As may be seen from Figure 1, taken from Oliver, 1979 (8), higher Mach numbers imply higher cruising altitudes in a region of general quiescence but increasing ozone mass density (i.e., the stratosphere). The Arctic tropopause is located much lower (ca. 8 km.) than the tropical tropopause (ca. 16 km.); thus, even the current and near-future subsonics would be flying in the lower stratosphere for operations in the mid-to-high latitudes. The severity of the problem was linked to the pollutant injection altitude—the higher the altitude, the worse the effects—and not to whether the aircraft were subsonic or supersonic, with the corollary that even the subsonic fleets could pose a threat.

CIAP AND OTHER ESTIMATES

In CIAP studies, a 0.5 percent reduction in total ozone (total ozone is all the ozone contained in a column of air extending from the ground up) in the Northern Hemisphere was nominally chosen to be a "minimum-detectable" level of change (1). This change would be brought about, it was calculated, by a fleet of 120 "Concorde-like" aircraft, cruising 4.4 hours a day every day of the year (365 days) at 16.5 km using 19,100 kg of fuel per hour at cruise and emitting 18g of NO_2 per kg of fuel burned. This number is shown in Table 2 along with the numbers adapted from the NAS and other studies, for the same ozone change. Both the CIAP and the NAS calculations were predicated upon the same data base. A spread of a factor of at least 5 in these numbers is evident from Table 2.

UNCERTAINTIES IN THE CIAP AND OTHER ESTIMATES

One-dimensional (1-D) numerical models of the stratosphere were the assessment tools used by CIAP and other studies. These models, which attempt to simulate the known physics and chemistry of the stratosphere, have a severe limitation in that they assume horizontal homogeneity of stratospheric properties, especially of motion. But only 1-D models have incorporated to the satisfactory degree the treatment of stratospheric chemistry. The elements of these models include: (1) the chemistry and photochemistry of trace species of importance (e.g., ozone, nitric oxide, atomic oxygen) and (2) highly parameterized mechanisms which transport the trace species in the vertical direction.

Based upon the admittedly incomplete treatment of transport and possible inaccuracies in the laboratory chemical rate constant determination, uncertainty estimates in the model calculations were made by the CIAP and NAS studies. These estimates were subjective and are shown in Table 2. As stated earlier, even the subsonic aircraft had the potential to deplete ozone under CIAP modeling assumptions and the calculations for subsonic fleets alone are shown in Table 3.

The uncertainty ranges and the spread in the calculations were such that the Federal Aviation Administration instituted its High Altitude Pollution Program (HAPP) following CIAP to reduce them in order to formulate viable regulatory options.

CHANGES IN MODEL CALCULATIONS SINCE CIAP

In the modeling of the chemistry of the trace species, the following three changes have occurred since CIAP:

- (1) Automobile exhaust containing NO_x is known to generate ozone in urban smog situations in the presence of methane and methane has been measured up to stratospheric altitudes. This "methane-oxidation" chemistry, which was not included in the CIAP model, has now been incorporated to a limited extent.
- (2) Trace species other than NO_x and the water-related radicals (HO_x) have been discovered to have ozone depleting effects. The most important among these are chlorine-related ClO_x (Cl and ClO) which are anthropogenic in origin. Hence the CIAP chemistry which included only the odd oxygen (O_x), odd nitrogen (NO_x) and water-related odd hydrogen (HO_x) species has been expanded to include the odd chlorine (ClO_x) species. Thus, the treatment of chemistry has been much improved.

- (3) The species in the various families such as NO_x , HO_x , and ClO_x are highly interactive; thus, changes in the rate constants of certain reactions have far-reaching consequences. Three such changes have taken place since CIAP in the following reactions:

$\text{OH} + \text{HO}_2$ About 4 times slower than the CIAP value

$\text{HO}_2 + \text{NO}$ About 40 times faster than the CIAP value

$\text{HO}_2 + \text{O}_3$ Temperature dependence different from the CIAP value.

These changes, especially the second, $\text{HO}_2 + \text{NO}$, have had profound influence on model calculations. The otherwise-very-active NO_x catalytic chain



is interrupted by the fast $\text{HO}_2 + \text{NO}$ reaction. The response of the model calculations to chronological changes in chemical input data is shown in Figure 2 taken from Luther et al., (9). The NO_x injection considered is equivalent to approximately 2000* "Concorde-like" CIAP aircraft, at two different altitudes (17 and 20 km). The 1975 chemistry included the revised $\text{OH} + \text{HO}_2$ rate (10), the 1977 chemistry the revised $\text{HO}_2 + \text{NO}$ rate (11), the 1979a chemistry the revised $\text{HO}_2 + \text{O}_3$ temperature dependence (12), and the 1979b chemistry the currently-accepted (13) ClONO_2 chemistry. (ClONO_2 is an example of the interactive chemistry between the NO_x and the ClO_x families. It is formed by $\text{ClO} + \text{NO}_2 \Rightarrow \text{ClONO}_2$.) In any case, without worrying about the details of the chemistry involved, one can see the profound effect of the $\text{HO}_2 + \text{NO}$ rate constant change. The unexpected feature of this figure, however, is the reversal of the trend between 1979a and 1979b.

The overall result of the three changes listed above has been a dramatic shift in the calculated aircraft effects from one of depletion to one of small increase in total ozone. However, as can be seen from Figure 3 (8), NO_x injections still deplete ozone above about 22 km while increasing it below; the sum of

* The NO_x injection considered was 1.2×10^9 kg per year as NO_2 . With the "Concorde-like" CIAP aircraft as defined earlier, this corresponds to 2,245 such aircraft.

the decrease above and the increase below leads to a small net increase in total ozone. It is important to keep in mind with regard to this figure that the cross-over point from increase-below-to-decrease-above is model-dependent. Changes in the transport parameterization and in the tropospheric removal processes such as rain-out or wash-out, for example, would alter the position of this point. The calculated net effect, needless to add, will depend upon its location. The numbers (500, 1000, 1500) on the three curves in Figure 3 roughly correspond to fleets of 500, 1000, and 1500 "Concorde-like" CIAP aircraft defined earlier.

UNCERTAINTIES IN THE CURRENT CALCULATIONS

There are, despite recent improvements, still uncertainties in almost every aspect of the assessment models: those associated with atmospheric chemistry and transport, and projected fleet emissions. The High Altitude Pollution Program has identified the following specific uncertainty factors to confirm, modify or clarify the present understanding:

(1) Discrepancies in the Measured NO_x Content in the Engine Exhaust

Figure 4, from Few et al., 1977 (14), is an illustration of the measurement of NO concentration as a function of fuel-to-air (F/A) ratio obtained by different techniques. The in-situ absorption method yields values about a factor of 6 higher than the others. In a joint effort with NASA, U.S. Air Force and U.S. Navy, the FAA has undertaken to resolve the uncertainty. Preliminary data indicate that the in-situ absorption method may be in error.

(2) Two-Dimensional Models and the Distribution of Natural Ozone

It is well known that the natural (i.e., unperturbed) ozone distribution is highly seasonal and latitude dependent. The reason for this is the meridional transport in the upper troposphere and the lower stratosphere. This atmospheric region is also, coincidentally, the region of current and near-future aircraft flights. In addition, the aircraft injection is also latitude-variant (witness the transAtlantic corridor). Thus 2-D models are more appropriate for the aircraft problem. However, they suffer from lack of adequate chemistry.

The "little-change" conclusion drawn in the previous section (see Figure 4) may well be misleading in that ozone depletions may actually be occurring in certain latitude bands with increases in others. A 1-D model is inherently

incapable of addressing this issue. The consequences of the latitudinal variation in the predicted effects may be quite different in the sense that steady depletions in certain regions and steady increases in others (especially if they were to occur close to the ground) may both be harmful with no cancellation of the effects, either on climate or the biosphere.

Hence, HAPP has undertaken to refine the existing 2-D models.

An example of the shortcomings of the 2-D models in their treatment of chemistry is to be seen in Figures 5a, 5b, 6, 7 and 8 taken from Widhopf and Glatt, 1979 (15). Figure 5b is the observed seasonal and latitude variation of total ozone. Figure 5a illustrates the calculated values using CIAP (no chlorine) chemistry. Figures 6, 7, and 8 show calculated distributions with (a) $\text{HO}_2 + \text{NO}$ reaction rate revised but with no chlorine, (b) $\text{HO}_2 + \text{NO}$ and $\text{HO}_2 + \text{O}_3$ reaction rates revised and no chlorine and (c) $\text{HO}_2 + \text{NO}$ and $\text{HO}_2 + \text{O}_3$ reaction rates revised and chlorine chemistry included, respectively. As may be seen, the agreement between theory and observations was good (Figures 5a and 5b), became poor (Figures 5b and 6), improved again (Figures 5b and 7) and deteriorated again (Figures 5b and 8) chronologically. This problem is being given further attention.

(3) Chemical Uncertainties

The pressure (i.e., altitude) dependence of certain reactions involving HO_x family, the temperature dependence of $\text{NO} + \text{O}_3$ reaction, and the uncertainties in methane oxidation chemistry, which are either unknown or poorly known, have the potential to alter the present understanding of the aircraft effects.

(4) Measurements of Background NO_x

The background concentration levels of NO , NO_2 and other members of the NO_x family in the atmosphere are not yet well known. There is a need to measure these species simultaneously, i.e., within the same air sample in order to verify the theoretical understanding of the partitioning among them. As may be seen from Figure 9 (15), which is a comparison of a 2-D model calculation against a set of simultaneous observations of NO , NO_2 and HNO_3 , the situation requires further resolution. There are difficulties in the comparisons of ratios of trace species such as HNO_3 to NO_2 also (9).

CONCLUDING REMARKS

The uncertainties in the present understanding of the effects of high altitude aircraft are such as to warrant continued studies. Specific uncertainty factors have been identified and their resolution should not prove difficult.

The views expressed in this paper are solely those of the author and are not intended to reflect any policy of the Federal Aviation Administration or the U.S. Department of Transportation.

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Table 1
Typical Jet Engine Emission Indices at
Cruise Altitudes. (English et al, 1975)

| Exhaust Species | Emission Index, gm/kg |
|---------------------------------------|--------------------------|
| CO ₂ | 3220 |
| H ₂ O | 1250 |
| NO _x (as NO ₂) | 6-30 |
| CO | 4 |
| SO _x (as SO ₂) | 1.0 |
| Hydrocarbon (as CH ₂) | 0.1-0.2 |
| Soot (as carbon) | 0.1 |
| Lubricating Oil | 0.1 |
| Trace Elements | 0.01 |

Table 2
Number of Concorde-like Aircraft Which
Would Lead to a Northern Hemispheric
Ozone Reduction of 0.5%, as of 1975.

| Name of Study | Number of Aircraft | Uncertainty Range (Estimate) |
|---------------|--------------------|---------------------------------|
| CIAP | 120 | (0.33-1.50) |
| NAS | 79 | (0.33-3.00) |
| COMESA | 435 | None given |
| COVOS | 326 | None given |

Table 3
Calculated Northern Hemispheric Ozone
Reduction by Subsonic Aircraft, as of 1975.

| Name of Study (Reference) | Northern Hemispheric Ozone Reduction Percent | Uncertainty Range (Estimate) |
|---|--|---------------------------------|
| 1. "1974" CIAP Fleet | | |
| CIAP | 0.09 | 0.1-2 |
| NAS | 0.10 | 0.1-10 |
| COMESA | None given | None given |
| COVOS | None given | None given |
| 2. Projected Fleet of 100 B-747 SP-like Aircraft | | |
| CIAP | 0.08 | 0.2-2 |
| NAS | 0.16 | 0.1-10 |
| COMESA | None given | None given |
| COVOS | None given | None given |

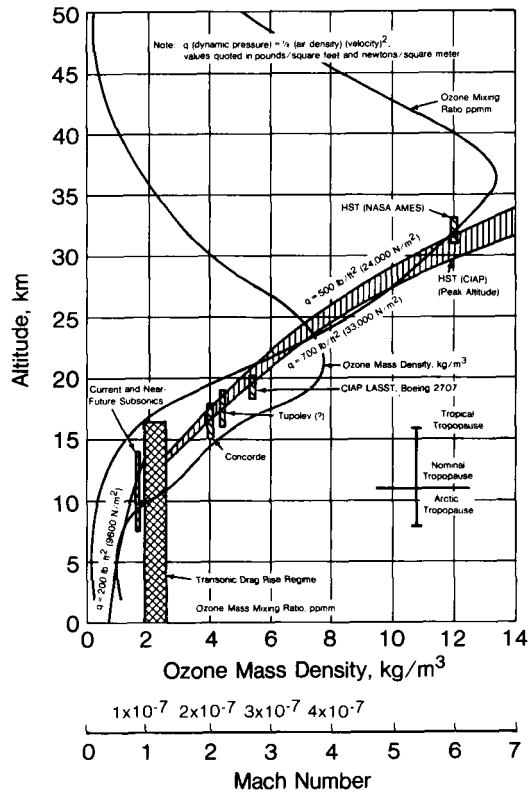


Figure 1.- Aircraft cruise altitudes and ozone data. (Oliver, 1979.)

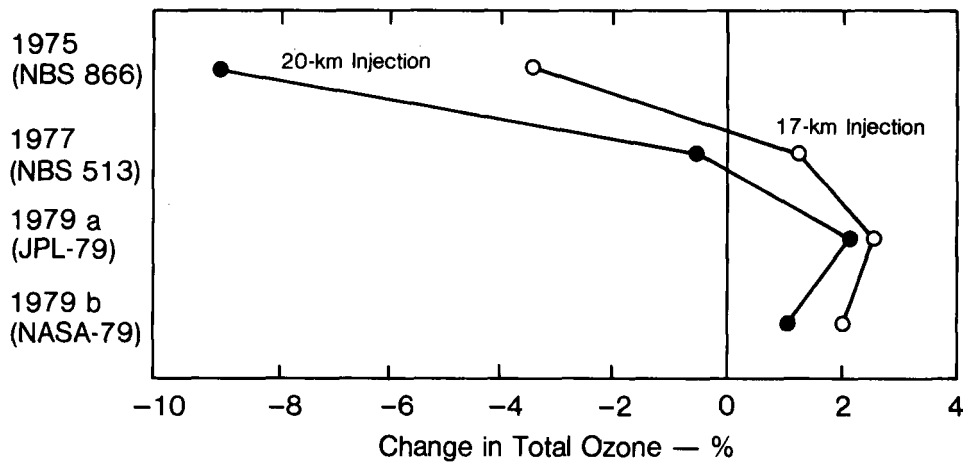
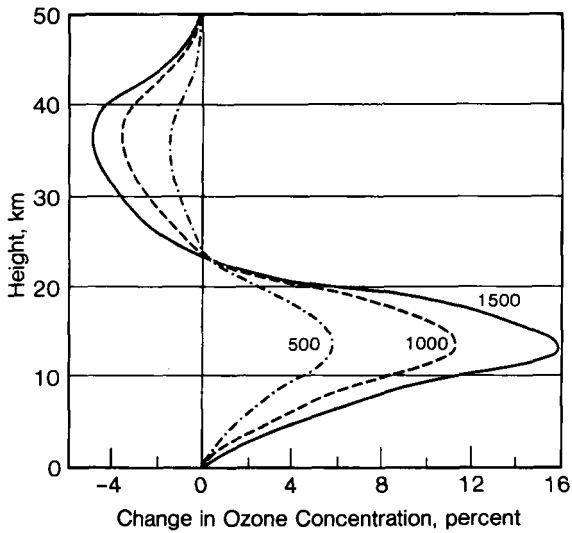


Figure 2.- Influence of changes in rate coefficients on model-predicted changes in total ozone due to NO_x injections. (Luther et al., 1979.)



Source: Lawrence Livermore Laboratory (1978).

Figure 3.- Percentage changes in ozone versus altitude for three injection rates of NO at 20 km. Rates shown are in molecules/cm³ sec over a 1-km band, and correspond to global injection rates of 0.62, 1.23, and 1.85×10^9 kg NO_x (as NO₂) per year. (Oliver, 1979.)

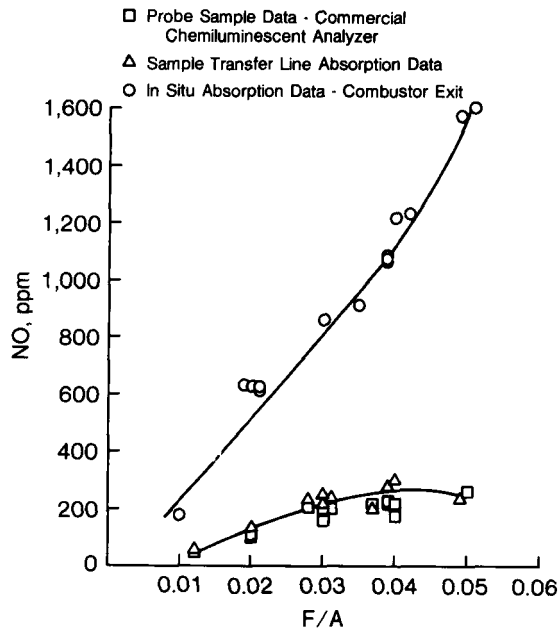
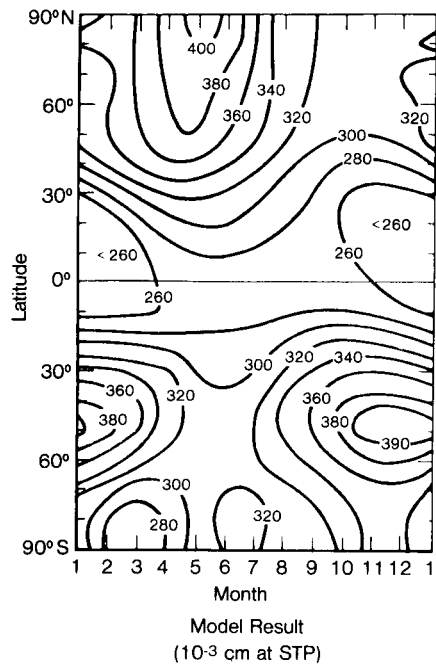
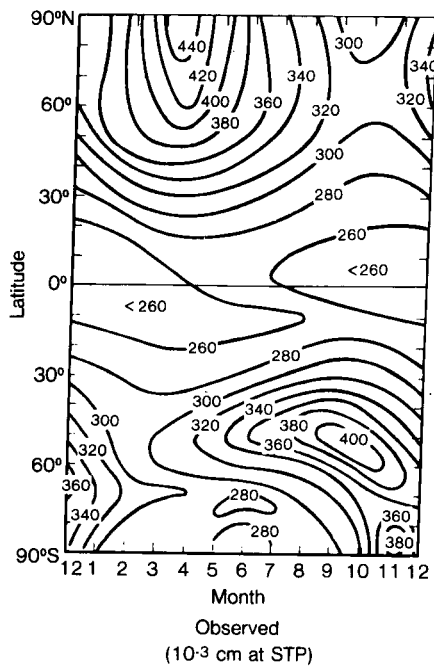


Figure 4.- NO concentration as a function of the fuel-to-air ratio for turbine engine combustor exhaust obtained by various means. (Few et al., 1977.)



(a) Calculated, without chlorine chemistry.



(b) Observed.

Figure 5.- Monthly variation of the total ozone column as a function of latitude (10^{-3} cm at STP). (After Widhopf and Glatt, 1979.)

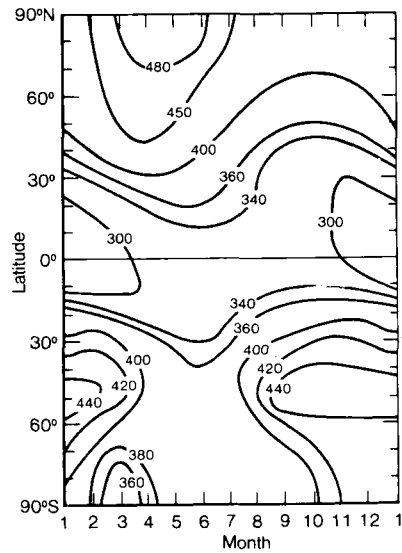


Figure 6.- Calculated monthly variation of total ozone column as a function of latitude without chlorine chemistry but with new rate coefficient for $\text{HO}_2 + \text{NO}$ (10^{-3} cm at STP). (Widhopf and Glatt, 1979.)

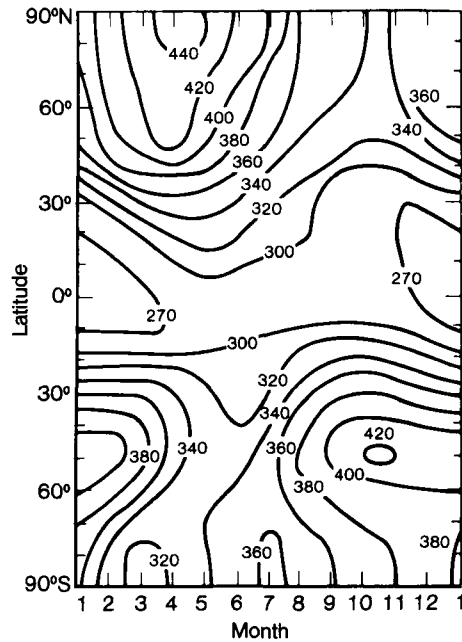


Figure 7.- Calculated monthly variation of total ozone column as a function of latitude (10^{-3} cm at STP) without chlorine chemistry but with new rate coefficients for $\text{HO}_2 + \text{NO}$ and $\text{HO}_2 + \text{O}_3$. (Widhopf and Glatt, 1979.)

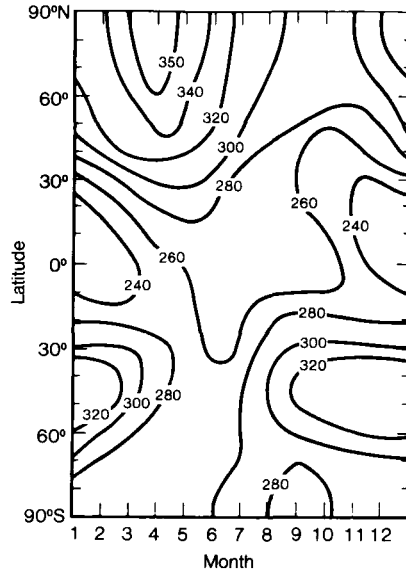


Figure 8.- Calculated monthly variation of total ozone column as a function of latitude (10^{-3} cm at STP) with chlorine chemistry and with new rate coefficients for $\text{HO}_2 + \text{NO}$ and $\text{HO}_2 + \text{O}_3$ (2 ppbv ClO_x). (Widhopf and Glatt, 1979.)

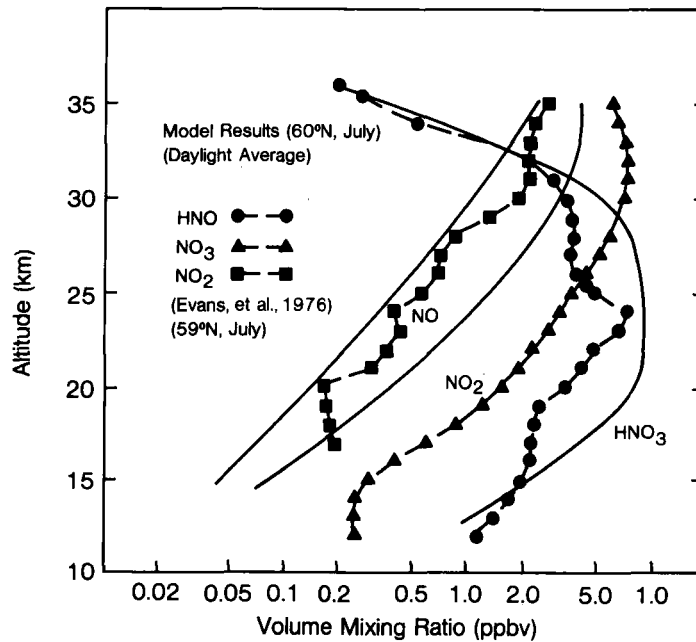


Figure 9.- Comparison of calculated and measured profiles of NO , NO_2 and HNO_3 . Calculations made with the aerospace model. (Widhopf and Glatt, 1979.)