IMPACT OF STRATOSPHERIC AIRCRAFT EMISSIONS ON OZONE: A TWO DIMENSIONAL MODEL STUDY

M. Natarajan[1], L. B. Callis[2], R. E. Boughner[2], and J. D. Lambeth[1]

[1] Science Applications International Corporation
Hampton, Virginia 23666, USA.

[2] Atmospheric Sciences Division, NASA Langley Research Center
Hampton, Virginia 23681, USA.

ABSTRACT

Atmospheric perturbations caused by the emission of nitrogen oxides from a projected fleet of stratospheric aircraft are studied with a two dimensional chemistry, transport model. Photochemistry of the lower stratosphere, the region where these aircraft may fly, is now known to be influenced by heterogeneous reactions involving sulfuric acid aerosols. This study examines the sensitivity of the atmospheric effects of aircraft to heterogeneous reactions. Information on background aerosols based on the SAGE II measurements have been used in the parameterization of the heterogeneous conversion rates. It is found that heterogeneous reactions make the lower stratospheric ozone less sensitive to perturbations in the odd nitrogen level. The calculated reduction in global ozone due to NO_x injection from a fleet of Mach 2.4 aircraft is 1.28% if gas phase reactions only are considered in the model, and 0.06% if heterogeneous reactions are included.

1. INTRODUCTION

Recent interest in the development of high speed civil transport aircraft has revived the concerns regarding the effects of aircraft exhaust gases on atmospheric ozone. Results from early model calculations of the perturbation due to a projected fleet of aircraft have been documented by the High Speed Research Program (NASA RP-1272, 1992). Ambient atmosphere with source gas mixing ratios projected for the year 2015 has been used in these studies as the baseline case. Aircraft fuel usage scenario as a function of latitude, and NO_x emission index in the range 5 to 45 gms of NO_x /kg of fuel have been adopted. Model results for NO_x injection at three different altitudes (based on the Mach number of the aircraft) have been reported. The calculated reductions in global ozone for different models ranged from 0.72% to 2.1%, for Mach 2.4 aircraft with an emission index of 15 for NO_x. All these models considered only gas phase reactions. The identification of the heterogeneous reactions on polar stratospheric clouds as a necessary component of the mechanism leading to the springtime ozone loss in the antarctic region, and the subsequent advances in laboratory studies of possible heterogeneous reactions on sulfuric acid droplets have revolutionized our understanding of the lower stratospheric photochemistry. Reaction probability for the reaction,

$$N_2O_5 + H_2O \rightarrow HNO_3 + HNO_3 \qquad (1)$$

involving sulfuric acid aerosols is believed to be high. Recent work by Hanson and Ravishankara (1991) suggests that the probability of the reaction,

$$ClONO_2 + H_2O \rightarrow HOCl + HNO_3 \qquad (2)$$

on sulfuric acid aerosols is a function of the acidity of the aerosol, and this reaction could be important under colder temperatures and/or larger aerosol loading. The conversion of reactive odd nitrogen into HNO_3 will also shift the partitioning of odd chlorine species, and recent measurement campaigns have reported data on ClO that are consistent with this theory. It is therefore essential that model studies of the aircraft effects should take into account the heterogeneous reactions on sulfuric acid aerosols. This paper describes one such study. Weisenstein et al. (1991) have also reported a similar study of the impact of reaction (1) on ozone response to aircraft emissions.

2. MODEL DESCRIPTION

The basic tool used in this study is a two dimensional chemistry, transport model developed on a potential temperature - latitude grid. This model extends from the ground to 2700 K surface. Main features of the model have been described by Callis et al.(1991). We have used NMC temperature data and ozone climatology in a radiative transfer code to calculate the monthly averaged diabatic heating rates. The advective fields are derived from these heating rates. Tropospheric fields have been calculated from heating rates based on FGGE data. We have adopted the horizontal mixing coefficients from Yang et al. (1990). The following chemical species are transported in the model: O_x, NO_y, HNO_3, Cl_y, CH_4, H_2O, CO, N_2O, CCl_4, CH_3Cl, CH_3CCl_3, $CFCl_3$, CF_2Cl_2, HCl, and CHF_2Cl. SAGE II H_2O data have been used in the troposphere. Photochemical and kinetic data are adopted from NASA Evaluation JPL 90-1.

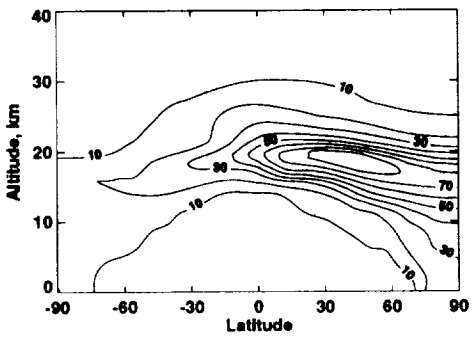

Figure 1: Percent change in total odd nitrogen in July due to NO_x injection from Mech 2.4 aircraft.

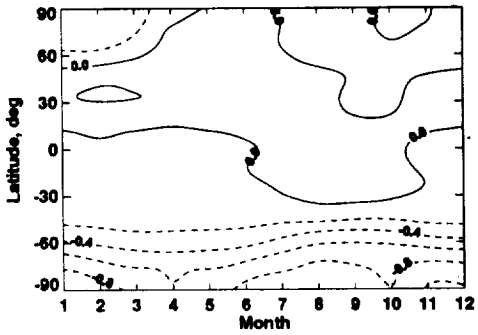

Figure 3: Same as figure 2, for Model H.

For the model with heterogeneous reactions, the distribution of the background aerosol surface area density is based on the information derived from the SAGE II measurements for 1989. γ_1, the reaction probability for reaction (1) is 0.1, and γ_2 is computed as a function of the acidity of the aerosol (Hanson and Ravishankara, 1991). Heterogeneous reations involving polar stratospheric clouds have not been included. The boundary values for the source gases are taken from NASA RP-1272, and these are the recommended data to simulate the conditions in the year 2015. The baseline atmosphere in this study includes the NO_x injection from the subsonic aircraft flying in the altitude range 6.1–12.2 km. The fuel usage for the subsonic aircraft is 170 x 10^9 kg/year and that for the supersonic fleet is 70 x 10^9 kg/year. The latitudinal distribution of the fuel use is taken from NASA RP-1272. We have considered only the effects of NO_x emission in this study.

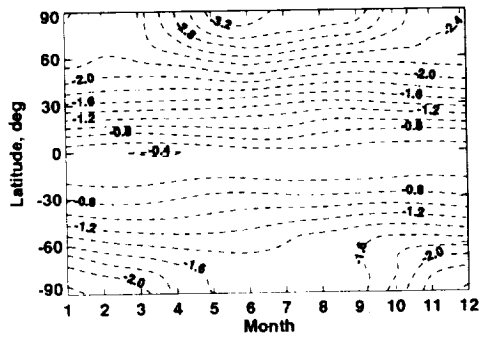

Figure 2: Percent change in total ozone for Model G (gas phase chemistry only) due to NO_x injection from Mech 2.4 aircraft.

3. RESULTS AND DISCUSSION

In the following discussion, 'Model G' denotes the case with only gas phase chemistry, and 'Model H' includes, in addition, the heterogeneous reactions on the background aerosols. NO_x injection from Mach 2.4 aircraft occurs in the altitude range 16.8–19.8 km. The percent change in the total odd nitrogen due to this injection for July is shown in Figure 1. The maximum change is about 80%, and this occurs between 30° N and 60° N. Total ozone perturbation caused by this increase in NO_y is shown in Figures 2 and 3 for the model G and H respectively. Model G yields ozone reductions at all latitude regions. Maximum reduction of 3.2% occurs in the northern high latitudes. The response of the atmospheric ozone to NO_x injection in the presence of heterogeneous reactions is dramatically different. In this case, most regions in the northern hemisphere show small increases in total ozone. In the high latitude southern hemisphere ozone reductions of 0.8% are seen. It should be noted that the aircraft fuel usage and hence the NO_x input are weighted heavily towards the northern latitudes. The vertical distribution of the ozone perturbation in July are shown in Figures 4 and 5. When only gas phase reac-

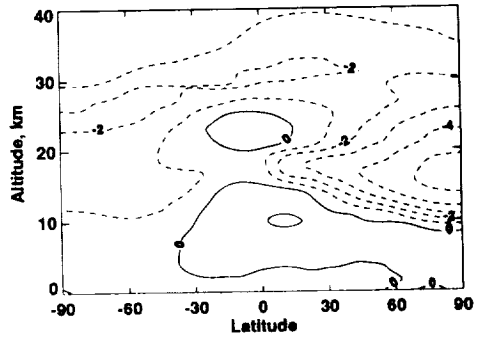

Figure 4: Latitude–Altitude distribution of percent change in ozone due to NO_x injection, Model G.

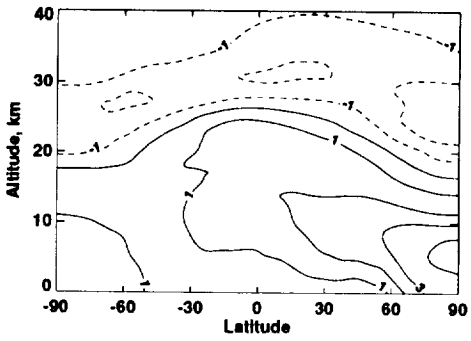

Figure 5: Same as figure 4, for Model H.

Aircraft Type	Gas Phase Chemistry	Gas Phase + Het. Chemistry
Mach 2.4 (16.8–19.8 km)	-1.28	-0.06
Mach 3.2 (21.3–24.4 km)	-4.5	-2.2

Table I: Calculated percent change in global ozone.

tions are considered (model G), ozone losses occur in the region above 10 km where the NO_x catalytic cycle is dominant. There is some increase in the tropospheric ozone which is partly due to increased production from smog reactions. For the model with heterogeneous reactions, ozone reductions are confined to the region between 22 and 35 km. Below this region ozone increases by about 1 to 3%.

The main reason for this change in the ozone response to NO_x injection is the modification of the photochemistry by the heterogeneous reactions, especially reaction (1). By converting reactive NO_x into HNO_3, this reaction effectively alters the relative importance of various catalytic cycles in destroying odd oxygen. The ratio of the odd oxygen loss rate due to NO_x cycle as a percent of the total odd oxygen loss rate is shown in Figure 6 for July. The solid lines represent results from model H, and the dashed lines represent model G. The contribution of NO_x catalytic cycle is clearly reduced to about 10% in the lower stratosphere because of the heterogeneous reactions. Correspondingly, there are increases in the odd oxygen loss

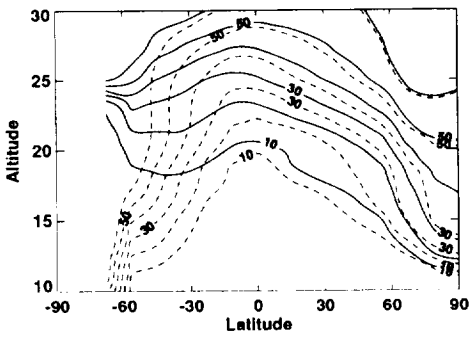

Figure 6: Ratio, in percent, of odd oxygen loss rate due to NO_x catalytic cycle to the total odd oxygen loss rate. Solid lines denote model with heterogeneous chemistry; dashed lines represent model with gas phase chemistry only.

due to Cl_x catalytic cycle. With the reduced significance of the NO_x cycle, the atmosphere with heterogeneous reaction becomes less sensitive to NO_x perturbations from stratospheric aircraft. Ozone increases in the lower altitudes nearly balance the ozone reductions in the upper regions, with the result that there is very little change in the total ozone in model H for the Mach 2.4 case. If the cruise altitude is higher, as in the case of Mach 3.2 aircraft, there will be larger ozone reductions in the mid stratosphere, and some reduction in the total ozone also. But even in this case, total ozone reduction is less for an atmosphere with heterogeneous reactions. The results from our model study are summarized in Table I.

REFERENCES

Callis, L. B., R. E. Boughner, M. Natarajan, J. D. Lambeth, D. N. Baker, and J. B. Blake,Ozone depletion in the high latitude lower stratosphere: 1979-1990,*J. Geophys. Res., 96*,2921-2938,(1991)

Hanson, D. R., and A. R. Ravishankara, The reaction probabilities of $ClONO_2$ and N_2O_5 on 40 to 75% sulfuric acid solutions, *J. Geophys. Res.,96*,17,307-17,314, (1991)

NASA Panel for Data Evaluation, Chemical kinetics and photochemical data for use in stratospheric modeling, 9, *JPL Publication 90-1*,(1990)

NASA RP-1272,The atmospheric effects of stratospheric aircraft: A first program report,(1992)

Weisenstein, D. K., M. K. W. Ko, J. M. Rodriguez, and N. D. Sze, Impact of heterogeneous chemistry on model-calculated ozone change due to high speed civil transport aircraft, *Geophys. Res. Lett, 18,* 1991-1994, (1991)

Yang, H., K. K. Tung, and E. Olaguer, Nongeostrophic theory of zonally averaged circulation. Part II: Eliassen-Palm flux divergence and isentropic mixing coefficient, *J. Atmos. Sci., 47,* 215-241, (1990)