A 2-D model simulation of downward transport of NO_x into the stratosphere: Effects on the 1994 austral spring O_3 and NO_x


Abstract. Simulations using SAMPEX and HALOE data suggest that NO_x produced by thermospheric processes and by relativistic electron precipitation in the mesosphere and lower thermosphere have been important to stratospheric NO_x and O_3 during the austral spring in 1994. The relative importance of the two NO_x sources is discussed. The results are supported by an analysis of HALOE NO_x and CH_4 data during October 1994 and are in agreement with ATMOS NO_x observations made in November 1994.

Introduction

It has been suggested that there is a possible coupling between the thermosphere, mesosphere, and stratosphere by means of the downward transport of odd nitrogen (NO_x), at high latitudes, during periods of advective descent [Solomon et al., 1982; Garcia et al., 1984; Russell et al., 1986; Callis et al., 1986, 1991a, b]. Callis et al. [1986, this issue] have provided evidence of the effects of precipitating electrons on mesospheric and thermospheric NO_x. With 2-D simulations, this paper examines the contributions to stratospheric NO_x made by the downward transport of NO_x, formed by thermospheric processes and by electron precipitation. The simulations use satellite observations of both thermospheric NO from HALOE and NO source profiles derived from electron fluxes observed by the Solar Anomalous Magnetospheric Particle Explorer (SAMPEX).

2-D Model

Simulations in this study used a modified version of the model described in Callis et al. [1991a, b] extended to ≈10^4 km. Diabatic transport fields are derived from heating rates that are calculated using SBUV ozone and an NMC temperature climatology for the stratosphere. MSIS is used for mesospheric temperatures.

Non-LTE effects are included in the calculation of the CO2 cooling rates according to Zhu et al. [1992]. Heating efficiencies given by Mlynczak and Solomon [1993] have been adopted in the evaluation of solar heating and chemical heating due to exothermic reactions. Stratospheric horizontal mixing coefficients are based on Yang et al. [1990], and in the mesosphere these are set equal to the upper stratospheric values. Vertical diffusion is negligible in the stratosphere, but values of ≈10^6 cm^2/sec are used in the mesosphere.

We include reactions involving species from the O_x, NO_x, HO_x, Cl_x, and Br_x families. The upper boundary condition for O_x is derived from the MSIS reference atmosphere. Averaging factors are used to account for diurnal variations. Heterogeneous chemical effects on sulfate aerosols are included for a background aerosol distribution. The kinetic rates and cross-sections used are taken from DeMore [1992]. We include a parameterized ozone hole [Callis et al., 1991b] and denitrification. Denitrification occurs with a 30 day time constant from Julian day 155 to 288 and from 14 to 24.5 km. The NO_x to which the parameterization relaxes was taken to be 0.2 ppbv. The three simulations which we use in this analysis are described in Table 1.

Data

The SAMPEX mission is described by Baker et al. [1993], and a description of the PFTI (Proton Electron Telescope) instrument is provided by Cook et al. [1993]. It provides electron measurements from 0.4 to 15 MeV with coverage between ±82° and all longitudes. The satellite pointing strategy and PET angular response is such that the instrument reflects both precipitating and trapped electron populations as well Long-term at-

Table 1. Summary of Simulations

<table>
<thead>
<tr>
<th>Simulation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>SS^1, NO_x from N_2O, 1990 trace gases, zero NO_x flux at upper b.c.</td>
</tr>
<tr>
<td>2</td>
<td>SS, same as 1 except HALOE NO for upper b.c.</td>
</tr>
<tr>
<td>3</td>
<td>TD^1, same as 2 including electron source of NO_x</td>
</tr>
</tbody>
</table>

^1 SS = steady state, TD = time dependent
Atmospheric energy inputs are derived from PET as follows. Assume a two-part exponential spectrum with e-folding energies $E_{0,L}$ and $E_{0,H}$ for $0.4 < E < 1.0$ MeV and $1.0 < E < 15$ MeV, respectively. Using PET electron fluxes, $P_1$, $ELO$, and $EHL$, derive daily averaged values of $E_{0,L}$ and $E_{0,H}$ over the South Atlantic Anomaly (SAA) and over $3.0 < L < 6.0$ ($L$ is the McIlwain parameter, see McIlwain [1961]). With the daily average of the integral electron flux, derive the multiplying constant for the exponential spectra. Calculate the global magnetospheric energy (GME) for the SAA and outer trapping region of the magnetosphere. Declines in GME from one day to the next, are assumed to be energy lost to the atmosphere. The average of all such declines observed in the long-term record is then conservatively assumed to be occurring during those days when there is an increase in GME. Energy input is apportioned between hemispheres according to the areas of the SAA and the region for $3.0 < L < 6.0$. Energy deposition profiles can be calculated for each day from which the NO$_x$ source profiles for each hemisphere may be determined and used in the time dependent simulations extending from 920720 through 950515.

Some of the results of this approach are illustrated on Figure 1. The thin line on Figure 1 presents the total energy obtained in this way in NO$_x$ equivalents, i.e. the total number of molecules of NO$_x$ that could be formed were all of this energy used in the formation of ion pairs in the atmosphere. We use 35 ev of energy to form an ion pair and 1.2 NO$_x$ formed per ion pair (see Callis et al., [1991a] and references therein). As can be seen the energy fluctuations are sizeable.

Triangular symbols are the energy inputs to the atmosphere. In several instances, these energy inputs are shown to be higher (in terms of NO$_x$ equivalents) than the total NO$_x$ produced by the global oxidation of N$_2$O during a day. In most cases, the electron energy inputs are larger than the NO$_x$ formation due to the N$_2$O above 50° latitude. Daily varying NO$_x$ formation rate profiles are used as input in the 2-D simulations. We note that the formation rates above 80 km are underestimated since PET does not measure electrons with energies below 0.40 MeV. Consequently, we view these calculations as lower limits of the effect of electrons on stratospheric NO$_x$.

The HALOE, which is aboard the UARS (Upper Atmospheric Research Satellite) has been described by Russell et al. [1993]. Measurements of NO are made in occultation at sunrise and sunset. Normally, 15 profiles of each type are retrieved each day separated by about 24° in longitude. Vertical coverage extends from 15 to 130 km for NO. Validation of the NO data has been reported by Gordley et al. [1995]. For simulations 2 and 3, we use an average of the sunrise and sunset NO observed by HALOE at 98.7 km for 91014 through 940926 as the upper boundary condition for NO$_x$. From 90°N to 90°S in 11.25° increments, the NO mixing ratios used are 2.2, 2.0, 1.6, 1.2, 0.6, 0.4, 0.3, 0.3, 0.3, 0.3, 0.3, 0.3, 0.4, 0.6, 1.2, 1.6, 2.0, and 2.2. Units are 10$^4$ ppbv.

**Results and Discussion**

Simulation results for October 15 and November 15, 1994 are presented on Figures 2a-d. Panel 2a illustrates the October 15, 1994 NO$_x$ mixing ratio difference between simulations 3 and 1. The largest increases occur at high latitudes. The significant increases at high northern latitudes, and at the high altitudes, indicate descent of NO$_x$ which may occur during the northern fall, winter, and early spring.
Panel 2b illustrates the October 15, 1994 percentage changes in O₃ due to these NOₓ increases. The simulations indicate O₃ depletions of 22% in mid August near 65°S and 33 km. By mid September, the depletions have reached ≤30% at 30 km and extend polewards of 75°S. Depletion reaches ≥30-35% by mid October at 27 km and is polewards of 75°-80°S. Values below 25 km are not reliable since a parameterization is used to prescribe the O₃ depletion due to heterogeneous chemistry on PSCs.

An examination of the three simulations permits an examination of the relative contributions of the thermospheric source and the electron NO source to the stratospheric NOₓ increases. The ratio, R = (NOₓ,3 - NOₓ,2)/(NOₓ,3 - NOₓ,1), is calculated from the three simulations and represents the ratio of NOₓ increases between simulations 3 and 2 to the NOₓ increases from simulations 2 and 1 and is shown on Figure 2c. Polewards of 60°S and above 20 km 0.3 ≤ R ≤ 1.0 indicating that both sources of NO are important. Note, however, that the thermospheric NO observed by HALOE and used in the boundary condition also includes the effect of precipitating relativistic electrons [see Callis et al. 1996, this issue]. These effects cannot be separated out at this time. The parameter, R, will vary with time reflecting both relativistic electron variability, and thermospheric variations. Finally, panel 2d is similar to 2a except it is for November 15, 1994.

We turn now to the question of whether or not observational evidence exists to confirm the presence of the excess NOₓ (NOₓ beyond that due to N₂O oxidation) shown on Figures 2a, d. Siskind and Russell [1996] suggest that there is little evidence for significant transport of NOₓ from the mesosphere to the stratosphere. Our own analysis of HALOE CH₄ and NOₓ data is presented on Figures 3a-c. Figure 3a illustrates altitude-longitude contours of CH₄ mixing ratio near 75°S. Low values of CH₄, ≥0.2 to 0.3 ppmv above 25 km indicate air which has descended from the upper thermosphere or lower mesosphere.

Figure 3b shows a similar plot for the NOₓ. Between 25 to 30 km and within the vortex, the NOₓ of ≥11 ppbv are higher than would be expected to be associated with the relatively low CH₄. Figure 3c, for altitudes between 25 to 30 km, presents a scatter plot of NOₓ and CH₄ observed by HALOE. Increasing NOₓ with decreasing CH₄ at CH₄ less than 0.3 ppmv suggests a source of NOₓ above 1 mbar other than N₂O oxidation. We suggest that Siskind and Russell [1996] did not find significant evidence of downward transport with the HALOE data because first, wasn't available poleward of ≥50° during wintertime where the bulk of the NOₓ is descending (also see Plate 1 of Russell et al. [1984]) and, second, by equinox, high latitude upper stratospheric NOₓ originating in the mesosphere or above would have been photodissociated or dispersed.

Other independent evidence supports the presence of downward transport from the mesosphere. Rinsland et al. [1996] present ATMOS sunrise observations of total NOₓ made during the ATLAS-3 mission for November 3-12, 1994 at 64.5°S to 72.4°S. They present ΔN, a measure of NOₓ above or below that NOₓ expected due to the oxidation of N₂O both inside and outside the polar vortex. Inside the vortex, and at altitudes of ≥25-30 km, values of ΔN (which we call excess NOₓ) of up to 7 ppbv are found. These values are in excellent agreement, both in magnitude and location, with the results of the present simulation shown on Figure 2d and are consistent with the hypothesis that the positive
\(\Delta N\) shown by Rinsland et al. within the vortex, and the high levels of \(NO_x\) at low \(CH_4\) also within the vortex and shown on Figure 3, are due an \(NO_x\) source above the stratosphere followed by downward transport.

With regard to the October \(O_3\) declines of \(\approx 30-35\%\) calculated and shown on Figure 2b, we hypothesize that the local \(O_3\) reductions between 25-30 km reported by Hofmann et al. [1992] may be due to the same phenomenon (excess \(NO_x\)) occurring in 1991. Hofmann et al. conclude that these high altitude depletions extended over much, and possibly all of the vortex. Unfortunately, no low altitude electron measurements or thermospheric NO measurements are available for times which would permit a simulation for the 1991 period. Relative to the observations of Figure 3, we point out that \(O_3\) observed by HALOE within the vortex between 25-30 km is \(\approx 3\) ppmv while that outside is \(\approx 7\) ppmv.

We conclude by noting that the current simulations, using NO observed by HALOE near 100 km as an upper boundary condition for \(NO_x\), and using fluxes of relativistic electrons observed by SAMPEx suggest that downward transport of \(NO_x\) formed by thermospheric processes and by relativistic electron precipitation has significantly affected mid to lower stratospheric \(NO_x\) and \(O_3\) mixing ratios during the austral spring in 1984. These results also: (1) appear to be consistent with an analysis of \(NO_x\) and \(CH_4\) as observed by HALOE at 73°S in October 1994; (2) seem to offer an adequate explanation for the large positive levels of \(\Delta N\) observed by ATMOS in November 1994 and reported by Rinsland et al. [1996]; and, (3) suggest an explanation for the large local \(O_3\) depletions at 25-30 km observed in the antarctic spring and reported by Hofmann et al. [1992].

References


(received November 7, 1995; revised May 1, 1996; accepted May 21, 1996.)