Energetic Particle Influences on NO_y and Ozone in the Middle Atmosphere

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Natural variations in the middle atmosphere can result from the penetration of energetic protons and electrons. These energetic particles produce NO_x (N, NO, NO_2) through interactions with the background atmosphere, primarily at polar latitudes. The NO_x species then produce other odd nitrogen compounds, NO_y (N, NO, NO_2, NO_3, N_2O_5, HNO_3, HNO_4, $ClONO_2$), fairly quickly in the stratosphere (on time scales of seconds to days). The long lifetime of the NO_y family (up to several months in the middle atmosphere) as well as the NO_y species' significant influence on stratospheric ozone abundance make the charged particle increases of NO_y important. Galactic cosmic rays produce NO_y in the lower stratosphere, solar protons produce NO_y in the middle and upper stratosphere as well as the mesosphere, and relativistic electrons produce NO_y in the upper stratosphere and mesosphere, each affecting the NO_y middle atmosphere budget directly. Production of NO_y constituents by solar protons has been associated with an observed polar ozone depletion during and after the August 1972 and the August, September, and October 1989 solar proton events and a polar NO increase after the July 1982 solar proton event. Auroral electron and photoelectron production of NO_x in the thermosphere and its subsequent transport downwards to the polar mesosphere and upper stratosphere is thought to be an important component of the NO_y budget in the middle atmosphere in the wintertime at high latitudes. It has been suggested that relativistic electrons can have a significant impact on the middle atmosphere NO_y budget over a solar cycle time period. The absolute flux of relativistic electrons to the middle atmosphere needs to be quantified more thoroughly to verify this suggestion.

INTRODUCTION

Recent analyses [Stolarski et al. 1991] indicate that total ozone has decreased in the past decade by several percent in the northern and southern hemisphere middle latitudes and by even more in the spring-time southern hemisphere polar latitudes. Such a disturbing result suggests that humankind (anthropogenic) activity is the cause of this decline. The natural changes in ozone, whether caused by solar ultraviolet variations, interannual dynamical changes, or energetic particle fluxes, need to be understood more completely in order to clearly separate anthropogenically-caused from naturally-caused ozone changes. To help understand one part of these natural atmospheric variations, we consider the influence of energetic protons and electrons on the middle atmosphere in this review paper.

Energetic protons and electrons influence the background middle atmosphere primarily at high geomagnetic latitudes by perturbing the chemistry and constituents. These charged particles produce ions, radioactive isotopes, and HO_x (H, OH, HO_2) and NO_x (N, NO, NO_2) through interactions with the atmosphere.

The charged particle influences on ions, radioactive isotopes and HO_x are important, however, long-term changes in ozone are primarily influenced by the enhanced NO_x levels from charged particle interaction with the middle atmosphere. For this reason, we concentrate on the NO_x increases from charged particle precipitation which result in an overall enhancement in odd nitrogen compounds, NO_y (N, NO, NO_2, NO_3, N_2O_5, HNO_3, HNO_4, $ClONO_2$) in the middle atmosphere. The lifetime of the NO_y family in the middle atmosphere is from hours in the mesosphere to one to two years in the stratosphere. Some of these effects on the middle atmosphere caused by NO_y species can be large and long-lived such as the August 1972 and the August, September, and October 1989 solar proton event disturbances [Heath et al. 1977; McPeters et al. 1981; Jackman and McPeters, 1987; Reid et al. 1991], but these large perturbations are infrequent. Other charged particle effects on the middle atmospheric NO_y abundance are continuous, but variable, such as the perpetual flow of galactic cosmic rays [Legrand et al. 1989]. Jackman [1992] discussed and reviewed the state of knowledge of the effects of charged particles on the middle atmosphere recently, thus this review

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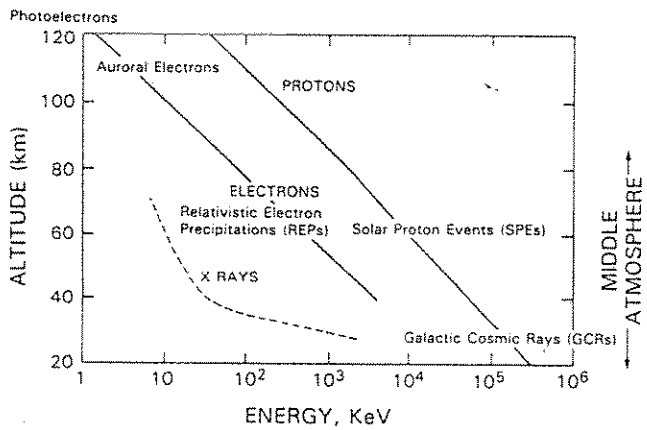

Fig. 1. Taken from Figure 1 of Jackman, Effects of energetic particles on minor constituents of the middle atmosphere, in press, *J. Geomag. and Geoelec.*, (1992), copyright by The Society of Geomagnetism and Earth, Planetary and Space Sciences. Altitude of penetration for protons, electrons, and X-rays vertically incident at the top of the atmosphere as a function of particle energy (adapted from Figure 2 of Thorne 1980).

will mainly consider developments in this field since that paper was written.

The middle atmosphere NO_y abundance is influenced directly by galactic cosmic rays which produce NO_y in the lower stratosphere, solar protons which produce NO_y in the stratosphere as well as the mesosphere, and relativistic electrons which produce NO_y in the upper stratosphere and mesosphere. Auroral electron and photoelectron production of NO_y in the thermosphere and its subsequent transport downwards to the mesosphere and upper stratosphere is thought to be important to the NO_y budget in the middle atmosphere for the wintertime. Both model results and measurements of charged particle influences on NO_y and ozone in the middle atmosphere will be discussed.

OVERVIEW OF CHARGED PARTICLE ENERGY DEPOSITION

. A schematic diagram of the areas of influence by the various categories of charged particles and their associated products is shown in Figure 1. This graph was created by modifying Figure 2 from Thorne [1980]. For a given energy, X-rays penetrate further than electrons and electrons penetrate further than protons (see Figure 1). The X-rays (bremsstrahlung) result from the slowing down of the energetic electrons. The magnitude of energy deposition by the X-rays is usually at least three orders of magnitude smaller than the energy deposition of the associated parent electrons [Berger et al. 1974].

Photoelectrons are produced by extreme ultraviolet (EUV) with energies up to a few hundred eV throughout the thermosphere. Primary electrons with energies less than 500 eV do not, in general, penetrate below 120 km (see Figure 1). Photoelectrons are more similar to secondary electrons than primary electrons because these particles are produced by the in situ EUV ionization of background atmospheric constituents. The major region of atmospheric influence by the photoelectrons is the thermosphere (see upper left corner of Figure 1). Since photoelectrons are produced by EUV, the only latitudinal dependence of the photoelectron energy deposition arises from changes in the solar zenith angle.

Some auroral electrons have energies capable of penetration to the mesosphere (electron energies < 100 keV) with associated bremsstrahlung reaching the upper to middle stratosphere. The higher energy electron fluxes are indicated as relativistic electron precipitations (electron energies > 100 keV) and are capable of depositing energy in the mesosphere and even the upper stratosphere with associated bremsstrahlung reaching the middle to lower stratosphere. Both auroral and relativistic electrons mainly deposit their energy in the subauroral region (geomagnetic latitudes between 60° and 70°) and their altitudes of deposition are indicated in Figure 1.

Solar protons deposit their energy in the mesosphere and stratosphere and generally in the polar cap region (geomagnetic latitudes greater than 60°). Galactic cosmic rays deposit most of their energy in the lower stratosphere and upper troposphere at high latitudes; however, penetration of the higher energy galactic cosmic rays is possible all the way to tropical latitudes thus latitude dependent energy deposition distributions are required. The major altitudes of influence for solar proton events and galactic cosmic rays are indicated in Figure 1.

Protons, electrons, and associated bremsstrahlung produce odd nitrogen (NO_y) constituents through dissociation or dissociative ionization processes in which N_2 is converted to $N(^4S)$, $N(^2D)$, or N^+. Rapid chemistry is initiated after N_2 dissociation and most of the atomic nitrogen is rapidly converted to NO and NO_2. Charged particle produced NO_y constituents can then deplete ozone through the following catalytic reaction cycle:

$$NO + O_3 \rightarrow NO_2 + O_2$$
$$NO_2 + O \rightarrow NO + O_2$$

Net: $$O_3 + O \rightarrow O_2 + O_2$$

GALACTIC COSMIC RAY INFLUENCE

The influence of galactic cosmic rays (GCRs) on the middle atmospheric NO_y abundance has been studied over the past two decades [Warneck 1972; Ruderman and Chamberlain, 1975; Nicolet 1975; Jackman et al. 1980; Thorne 1980; Garcia et al. 1984; Legrand et al. 1989]. The major production of NO_y results from nitrous oxide oxidation ($N_2O + O(^1D) \rightarrow NO + NO$), thus the GCR-related production of NO_y must be compared to that background source in order to put its NO_y budget contribution into perspective. Jackman et al. [1980] computed that GCRs were responsible for 2.7 to 3.7 x 10^{33} molecules of NO_y per year, whereas Crutzen and Schmailzl [1983] and Jackman et al. [1987] show that nitrous oxide oxidation produces about 2.3 to 2.6 x 10^{34} molecules of NO_y per year, respectively. Nitrous oxide oxidation produces odd

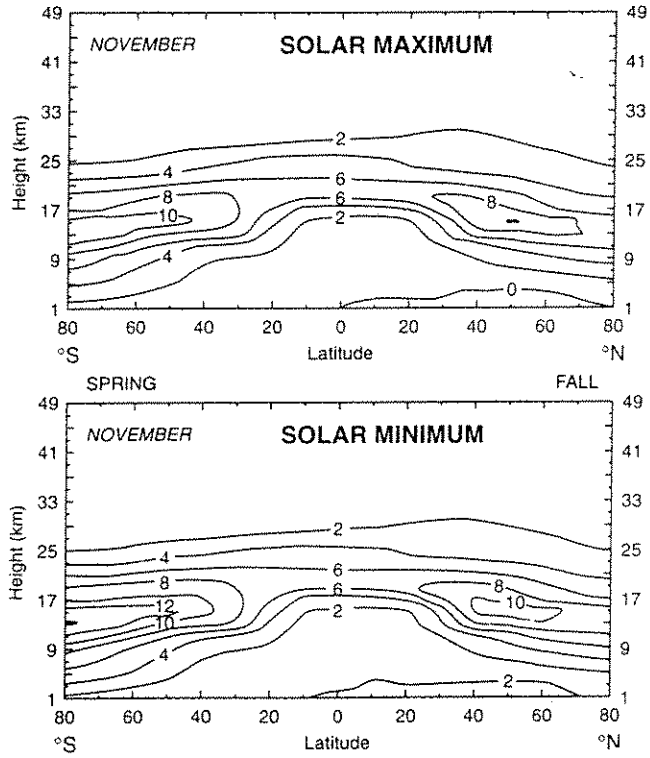

Fig. 2. Taken from Figure 4 of Legrand et al., A model study of the stratospheric budget of odd nitrogen, including effects of solar cycle variations, *Tellus, 41B*, 413-426, (1989), copyright by Munksgaard International Publishers Ltd. Computed percentage change in odd nitrogen from galactic cosmic rays at solar maximum and solar minimum.

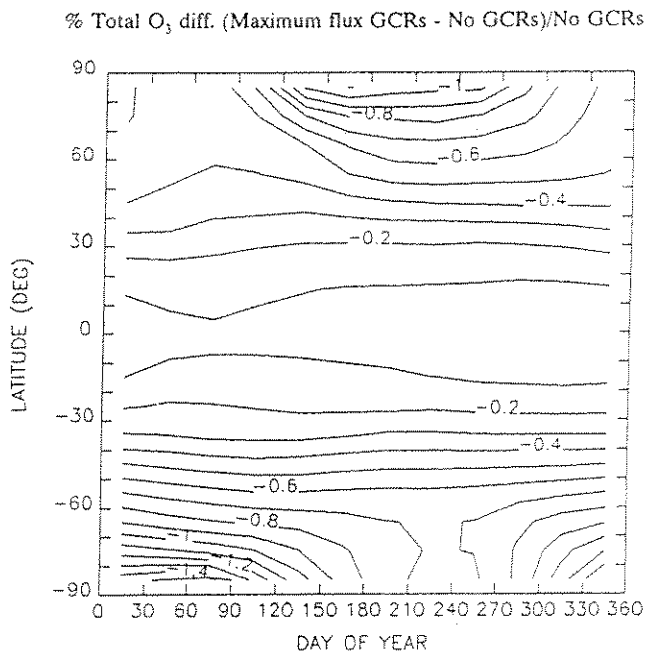

Fig. 3. Computed percentage total ozone change for a model simulation including galactic cosmic rays at solar minimum (maximum in GCRs) compared to a model simulation with no galactic cosmic rays. Contours range from -0.1% to -1.5% in 0.1% intervals.

nitrogen primarily in the middle stratosphere at low latitudes, while the GCR-induced odd nitrogen production peaks near the tropopause at high latitudes [see Figures 9a and 13 in Jackman et al. 1987].

Since the NO_y family has a lifetime of months in the middle and lower stratosphere, transport of NO_y created at higher altitudes and lower latitudes is significant. In Figure 2 [taken from Figure 4 of Legrand et al. 1989] we show the computed percentage change in NO_y which results from the GCR source of odd nitrogen. The lower stratospheric NO_y at high latitudes is calculated to be increased by about 10%. A solar cycle variation is apparent in the GCR flux with maximum flux during solar minimum and minimum flux during solar maximum.

The influence of GCRs on ozone over the 11-year solar cycle time period has been included in a two-dimensional (2D) model computation of Garcia et al. [1984]. Since the effects of ultraviolet (UV) and auroral flux variation were also included in that computation, no quantitative changes from only the GCRs were available. The Goddard Space Flight Center (GSFC) 2D model which extends from the ground to about 90 km [Jackman et al. 1990] was used to investigate the influence of GCRs on NO_y abundance and ozone amounts. The minimum flux in GCRs (solar maximum) from a GSFC model computation allows about 0.25% more total ozone near the poles than computed during the maximum flux in GCRs (solar minimum). Predictions from the GSFC model indicated about 1% less total ozone at polar latitudes for a model run including GCRs compared to a model run not including GCRs (see Figure 3). These model computations showed a seasonal as well as a strong latitudinal dependence with less than a 0.1% difference near the Equator between the two model runs just described.

SOLAR PROTON EVENT INFLUENCE

Direct constituent change in the middle atmosphere by particles has only been documented in the case of solar proton events (SPEs). SPEs are sporadic with durations up to several days and have a solar cycle dependence such that more SPEs occur closer to solar maximum.

The production of NO_y species by SPEs has been predicted since the mid-1970's [Crutzen et al. 1975]. The polar NO increase after the July 1982 SPE was inferred from the SBUV instrument to be about 6 X 10^{14} NO molecules cm^{-2} at polar latitudes [McPeters 1986], in good agreement with our calculated NO increase of 7 X 10^{14} NO molecules cm^{-2} in the polar cap [Jackman et al. 1990].

Polar ozone depletions associated with NO_y increases have been observed and modelled for the August 1972 SPE [Crutzen et al. 1975; Heath et al. 1977; Fabian et al. 1979; Maeda and Heath, 1980/81; McPeters et al. 1981; Solomon and Crutzen,

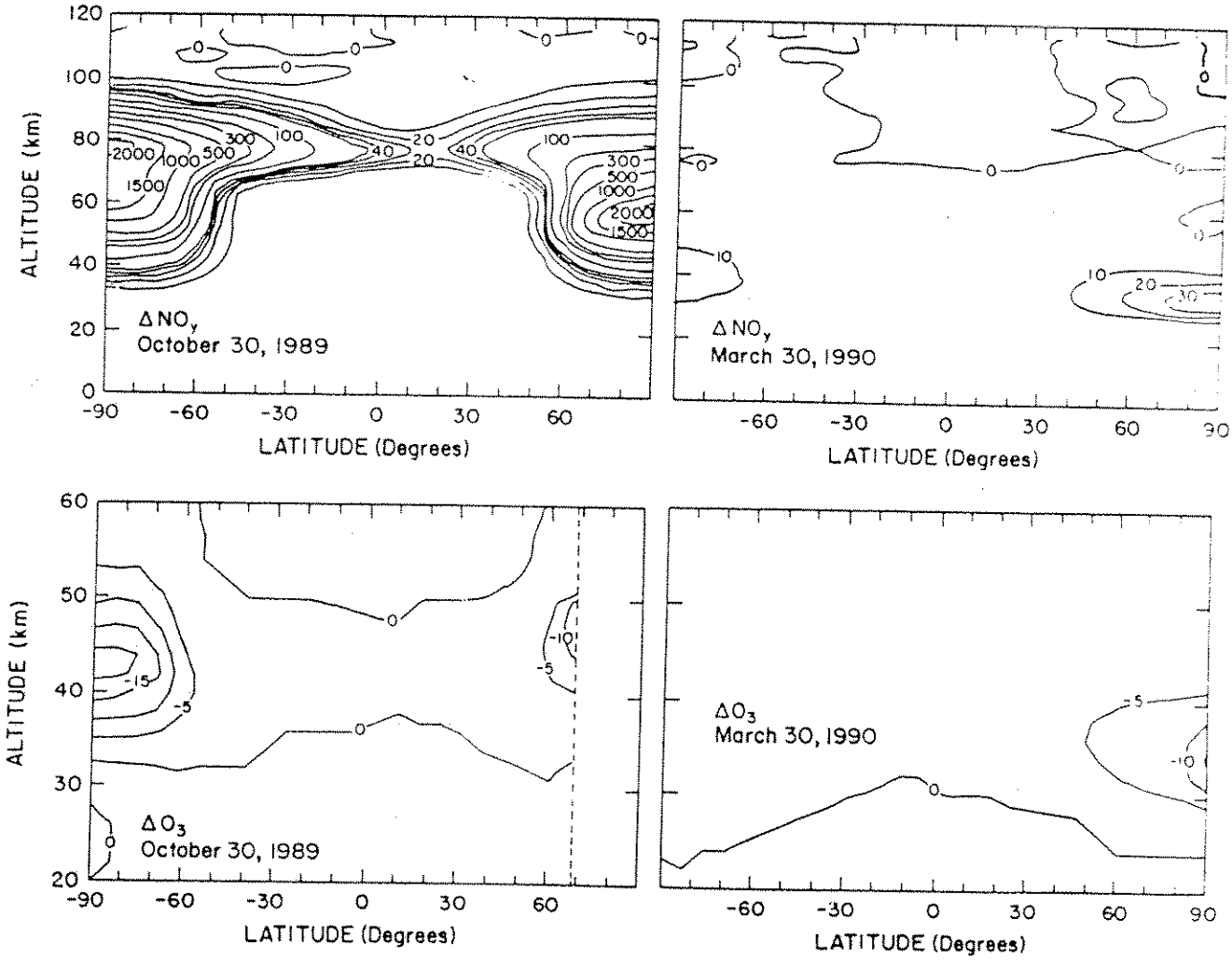

Fig. 4. Taken from Figure 4 of Reid et al., Response of the middle atmosphere to the solar proton events of August-December 1989, *Geophys. Res. Lett.*, *18*, 1019-1022, (1991), copyright by the American Geophysical Union. Contour plots of calculated percentage changes in NO_y and ozone at the end of October 1989 and March 1990 as compared to quiet geomagnetic conditions. Values of ozone percent change are shown only in the sunlit atmosphere.

1981; Reagan et al. 1981; Rusch et al. 1981; Jackman and McPeters, 1987; Jackman et al. 1990] and the August, September, and October 1989 SPEs [Reid et al. 1991]. These SPEs in 1972 and 1989 were very large events and substantial increases in NO_y have been computed to be associated with these events at polar latitudes in the middle to upper stratosphere [see, e.g., Jackman et al. 1990; Reid et al. 1991].

The model predicted percentage change for NO_y and the associated ozone depletion for October 30, 1989 and March 30, 1990 as a result of the August, September, and October 1989 SPEs are shown in Figure 4 [from Figure 4 of Reid et al. 1991]. NO_y on October 30, 1989 is predicted to increase by a maximum of over 20-fold in the polar mesosphere as a result of these SPEs. NO_y is also predicted to increase in the polar middle to upper stratosphere by 10% to 500% and the maximum associated ozone response is predicted to be a decrease of over 20%. By March 30, 1990, Reid et al. [1991] predicted increases in NO_y of over 30% with associated ozone decreases of over 10%.

Reid et al. [1991] used GOES-7 proton flux data in computing NO_y production rates. We have used IMP-8 proton flux measurements in our computations, which results in slightly higher NO_y production rates and, subsequently, a slightly larger ozone depletion. For example, on October 30, 1989, we predict a maximum ozone depletion of about 25% whereas Reid et al. [1991] predict a maximum ozone depletion of about 20% near 43 km and the Southern Hemisphere pole.

Our computations of NO_y and associated ozone percentage change at 75°N due to the 1989 SPEs are presented in Figure 5. The calculated NO_y changes are largest in the upper

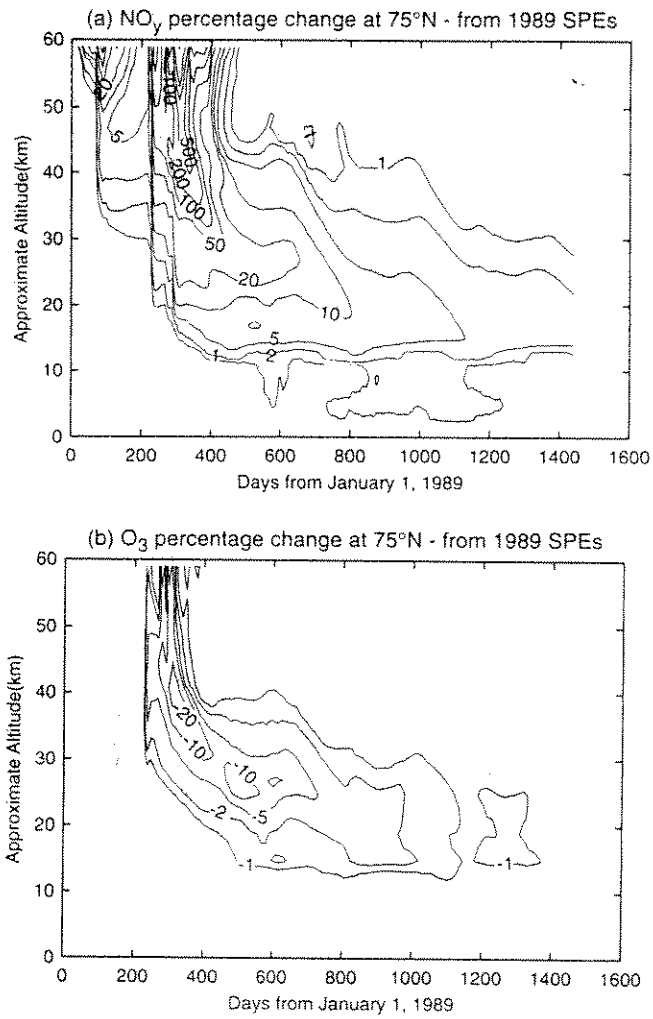

Fig. 5. Calculated percentage changes in (a) NO_y and (b) ozone at 75°N resulting from solar proton events in 1989 for four years starting from January 1, 1989. Contour intervals for NO_y are 1, 2, 5, 10, 20, 50, 100, 200, 500, 1000, 2000, and 5000%. Contour intervals for ozone are -1, -2, -5, -10, and -20%.

stratosphere and lower mesosphere with transport of the NO_y enhancements to lower altitudes occurring over the next couple of years. The simulated increases in NO_y of over 20% and associated decreases in ozone of over 5% persist for about a year and a half after the October 1989 SPEs.

Such predicted changes in profile ozone are reflected in predictions of total ozone amounts. We show our predicted percentage total ozone changes in 1990 as a result of the 1989 SPEs in Figure 6. Depletions of over 3% above 70° in the summer are predicted in both hemispheres. Due to the large variations in total ozone on days, weeks, seasons, and years time scales at the high latitudes, it would be unlikely that such SPE-related changes in total ozone could be measured unambiguously.

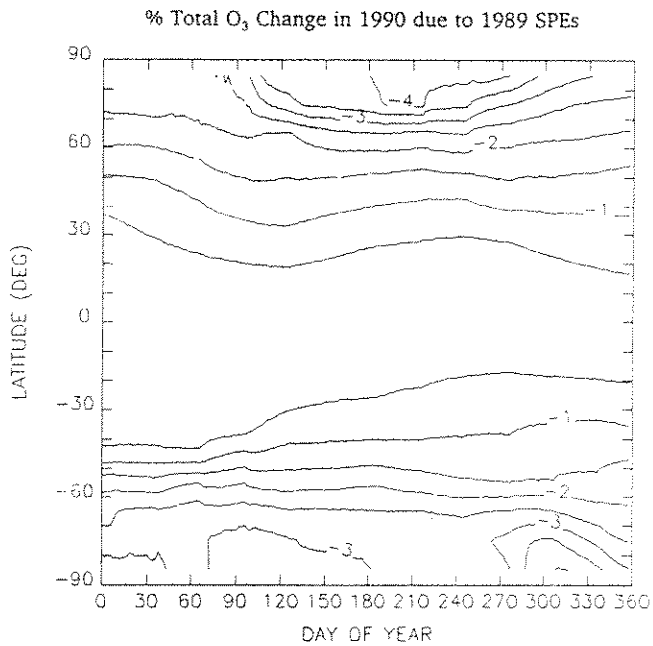

Fig. 6. Computed percentage total ozone change in 1990 which resulted from solar proton events in 1989. Contours range from -0.5% to -4% in 0.5% intervals

Measurements of ozone by SBUV-II (R. D. McPeters, private communication, 1991) and SAGE-II (J. M. Zawodny, private communication, 1991) have indicated a profile ozone decrease as a response to the 1989 SPEs, however, no published results are available. The SAGE-II ozone decreases appear to be correlated with substantial increases in NO_2, also measured by SAGE-II, after the 1989 SPEs (J. M. Zawodny, private communication, 1991). Zadorozhny et al. [1992], using rockets launched from a Soviet research vessel in the southern part of the Indian Ocean, measured large enhancements in NO as a result of the SPEs in October 1989. Since the 1989 SPEs were apparently large enough to cause an NO_y increase and an associated ozone decrease, it is important that the ozone decreases and NO_y increases be studied, as well as possible, through both measurements and model simulations to quantify the associated total ozone decrease.

RELATIVISTIC ELECTRON INFLUENCE

Relativistic electron precipitations (REPs) have been proposed in the past 15 years to be important in contributing to the polar NO_y budget of the mesosphere and upper stratosphere [Thorne 1977; Thorne 1980; Baker et al. 1987; Sheldon et al. 1988; Baker et al. 1988; Callis et al. 1991a]. The frequency and flux spectra of these REPs is still under discussion. Baker et al. [1987] show evidence of large fluxes of relativistic electrons at geostationary orbit measured by the Spectrometer for Energetic Electrons (SEE) instrument on board spacecraft 1979-053 and 1982-019. REPs, which are actually depositing energy into the middle atmosphere, have

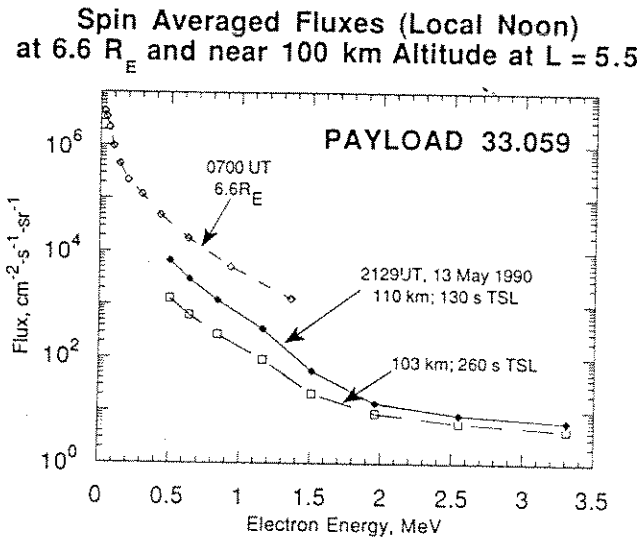

Fig. 7. Taken from Figure 6 of Herrero et al., Rocket measurements of relativistic electrons: New features in fluxes, spectra and pitch angle distributions, *Geophys. Res. Lett., 18*, 1481-1484, (1991), copyright by the American Geophysical Union. Comparison of spectra obtained with the high energy spectrometer to spectra obtained in-situ in geostationary orbit at 6.6R_E. At energies higher than about 2 MeV, the flux decreases to the cosmic background level.

been measured by instruments aboard sounding rockets [Goldberg et al. 1984; Herrero et al. 1991]. These rocket measurements have typically indicated much smaller fluxes of relativistic electrons than measured by the SEE instrument.

The coincident measurement of relativistic electrons at 6.6R_E on the GOES satellite with a sounding rocket between altitudes of 70 and 130 km is shown in Figure 7 (taken from Figure 6 of Herrero et al. 1991). The relativistic electron flux was about a factor of 5 to 25 at 6.6R_E compared with the flux at 110 km on May 13, 1990. Other coincident measurements of relativistic electrons at 6.6R_E on the 1982-019 satellite and the low altitude (170-280 km) S81-1 payload have also been reported [Imhof et al. 1991]. These measurements indicate a relativistic electron flux of about a factor of 10 larger at 6.6R_E compared to the flux between 170 and 280 km over the period May 28, 1982 to December 5, 1982.

Callis et al. [1991a] use a relativistic electron flux of one-third of the flux measured at 6.6R_E in order to compute NO_y production rates over the 1979 to 1988 time period. Their computations show enhancements in total NO_y of over 40% in the southern hemisphere and about 40% in the northern hemisphere over the solar cycle as a result of relativistic electrons [see Figure 20 of Callis et al. 1991a]. The associated calculated global total ozone decrease from these relativistic electrons is illustrated in Figure 8 [from Figure 16c of Callis et al. 1991b]. Figure 8 has four lines plotted: 1) The large dashed line shows a 2D model simulation which includes increasing CH_4, N_2O, and chlorine from chlorofluorocarbon and hydrochlorocarbon trace gases; 2) The dash-dot line illustrates a 2D simulation including the increasing trace gases as well as the solar ultraviolet (UV) variation; 3) The dash-with-three-dots line shows a 2D simulation with increasing trace gases, solar UV variation, and relativistic electron precipitations; and 4) The solid line illustrates a 2D simulation which includes the increasing trace gases, solar UV variation, REPs, and dilution of the Antarctic ozone hole. The REP's simulated global total ozone effects are clearly the largest of these simulated natural and anthropogenic components shown in Figure 8.

Since relativistic electrons are computed to cause such large variations in total ozone, it is reasonable to assume that a large REP event should cause a measurable decrease in ozone in the upper stratosphere. Callis et al. [1991b] argue that such ozone depletion has already been observed near 1 mbar for longitudes between 60°W and 60°E [see Plate 1 of Callis et al. 1991b]. Another investigation (Aikin, 1992) has failed to find any REP-caused ozone depletion. More work is necessary in order to definitively establish a strong correlation between REPs and middle atmosphere ozone depletion.

Relativistic electrons could be important in determining global ozone abundances if the electron flux entering the middle atmosphere is 10% or more of the flux measured at 6.6R_E. It is still not clear which REP events are more typical of REPs which deposit their energy in the earth's atmosphere, the large fluxes measured at geostationary orbit or the somewhat smaller fluxes measured by the sounding rockets. More coincident relativistic electron flux measurements between rocket and satellite instruments are necessary to determine the magnitude and frequency of REP events.

AURORAL ELECTRON AND PHOTOELECTRON INFLUENCE

The influence of auroral electrons and photoelectrons on the NO_y budget of the middle atmosphere through transport of NO_x from the thermosphere has been studied for the past two

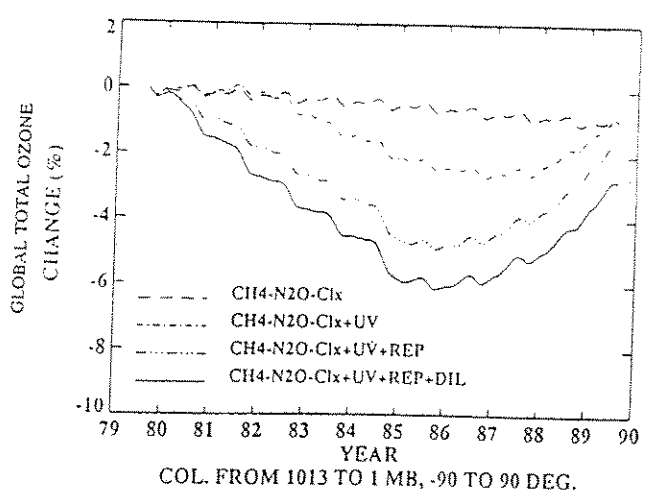

Fig. 8. Taken from Figure 16c of Callis et al., Ozone depletion in the high latitude lower stratosphere: 1979-1990, *J. Geophys. Res., 96*, 2921-2937, (1991b), copyright by the American Geophysical Union. Predicted percentage changes in global (90°S to 90°N) total column ozone as calculated for four scenarios explained in the text.

decades [Strobel 1971; McConnell and McElroy, 1973; Brasseur and Nicolet, 1973; Jackman et al. 1980; Solomon 1981; Solomon et al. 1982; Frederick and Orsini, 1982; Garcia et al. 1984; Solomon and Garcia, 1984; Russell et al. 1984; Brasseur 1984; Legrand et al. 1989]. Both auroral electrons and photoelectrons are capable of dissociating N_2 to form huge amounts of atomic nitrogen in the thermosphere. Transport of this NO_x to the mesosphere and upper stratosphere is possible, but certain conditions must be present.

The lifetime of NO_x in the thermosphere and mesosphere is short (less than a day) in the daytime and it is only during the long period of polar night at high latitudes, when several weeks of darkness is typical, that significant downward transport of NO_x is possible. Solomon et al. [1982] undertook a detailed 2D model study of the thermosphere - middle atmosphere coupling. Two of these model computations are shown in Figure 9 [taken from Figures 8 and 17 of Solomon et al., 1982]. They calculated enhancements of over an order of magnitude in the NO_x mixing ratio distribution in the upper mesosphere when auroral electron and photoelectron production of NO_x was included compared to a computation when both auroral electron and photoelectron production of NO_x were not included. These large enhancements of NO_x in the mesosphere caused by auroral electrons and photoelectrons are especially significant in the northern hemisphere which was most recently shrouded in polar night (see Figure 9).

Measurements by the limb infrared monitor of the stratosphere (LIMS) of one significant species of NO_x, NO_2, have also indicated that large enhancements of NO_x in the mesosphere (above 1 mbar) are possible during polar night [see Figure 5 in Russell et al. 1984]. Larger NO_2 values are indicated at higher latitudes, in qualitative agreement with model predictions.

Solomon and Garcia [1984] have computed the response of ozone in the middle atmosphere to changes in production of NO in the thermosphere over the course of an 11-year solar cycle. Their model simulations compare well with BUV and SBUV instrument measurements of ozone change at the 0.75 and 1.0 mbar levels. More study is required to determine how much of the thermospheric enhancement of NO_x is transported to lower altitudes and, also, how much of an effect is expected and measured in stratospheric ozone.

Bremsstrahlung from auroral electrons penetrate to the stratosphere and lower mesosphere and are also capable of producing NO_x directly in the middle atmosphere. Although the flux of bremsstrahlung is about three orders of magnitude smaller than the flux of auroral electrons, the in-situ production of middle atmospheric NO_x could be important for large auroral storms. Previous work [see, e.g., Goldberg et al. 1984] suggests that bremsstrahlung from auroral and relativistic electrons can be the dominate ion source in the upper stratosphere for the subauroral region during certain periods. The frequency and magnitude of this bremsstrahlung flux needs to be quantified to establish its regional and global contribution to the middle atmospheric NO_y budget.

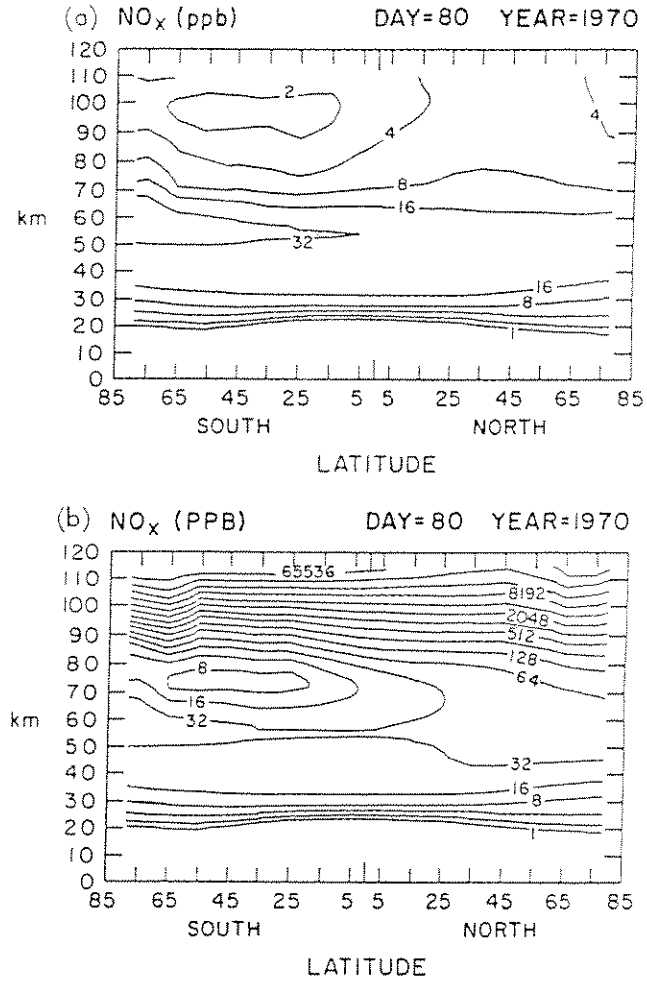

Fig. 9. Taken from Figures 8 and 17 of Solomon et al., Photochemical coupling between the thermosphere and the lower atmosphere, 1, Odd nitrogen from 50 to 120 km, *J. Geophys. Res.*, *87*, 7206-7220, (1982), copyright by the American Geophysical Union. (a) Computed NO_x mixing ratio distribution obtained after 80 days of integration assuming a zero flux at the upper boundary and neglecting production of odd nitrogen by thermospheric ion chemistry. (b) Calculated NO_x mixing ratio distribution obtained after 80 days of integration including photoelectrons and a parameterized model aurora.

CONCLUSIONS

Galactic cosmic rays are computed to cause changes in the NO_y abundance in the lower stratosphere by as much as 10% over a solar cycle. Infrequent large solar proton events, such as those observed in August 1972 and August, September and October 1989 are computed to cause substantial changes in the NO_y abundance in the middle and upper stratosphere. A long-lived ozone decrease during the August 1972 SPE has been calculated and verified by measurements to be important at polar latitudes in the middle to upper stratosphere. Relativistic electron precipitations could be important in modulating the NO_y abundance of the middle atmosphere but require more study and measurements of relativistic electrons in the earth's atmosphere. Auroral electrons and

photoelectrons cause enhancements in NO_x amounts in the thermosphere which then can be transported to the mesosphere during polar night. More study is required to quantify the stratospheric NO_y and ozone change from auroral electron and photoelectron precipitation. The importance of bremsstrahlung from auroral and relativistic electrons in producing NO_x in the middle atmosphere has not been clearly established and needs further investigation.

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