

# **The Long-term Middle Atmospheric Influence of Very Large Solar Proton Events**

**Charles H. Jackman, Daniel R. Marsh, Francis M. Vitt, Rolando R. Garcia,  
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## **Brief, Popular Summary of the Paper:**

Long-term variations in ozone have been caused by both natural and humankind related processes. The humankind or anthropogenic influence on ozone originates from the chlorofluorocarbons and halons (chlorine and bromine) and has led to international regulations greatly limiting the release of these substances. Certain natural ozone influences are also important in polar regions and are caused by the impact of solar charged particles on the atmosphere. Such natural variations have been studied in order to better quantify the human influence on polar ozone.

Large-scale explosions on the Sun near solar maximum lead to emissions of charged particles (mainly protons and electrons), some of which enter the Earth's magnetosphere and rain down on the polar regions. "Solar proton events" have been used to describe these phenomena since the protons associated with these solar events sometimes create a significant atmospheric disturbance.

We have used the National Center for Atmospheric Research (NCAR) Whole Atmosphere Community Climate Model (WACCM) to study the long-term (> few months) influences of solar proton events from 1963 through 2004 on stratospheric ozone and temperature. There were extremely large solar proton events in 1972, 1989, 2000, 2001, and 2003. These events caused very distinctive polar changes in layers of the Earth's atmosphere known as the stratosphere (12-50 km; ~7-30 miles) and mesosphere (50-90 km; 30-55 miles). The solar protons connected with these events created hydrogen- and nitrogen-containing compounds, which led to the polar ozone destruction. The nitrogen-containing compounds, called odd nitrogen, lasted much longer than the hydrogen-containing compounds and led to long-lived stratospheric impacts. An extremely active period for these events occurred in the five-year period, 2000-2004, and caused increases in odd nitrogen which lasted for several months after individual events. Associated stratospheric ozone decreases of >10% were calculated to last for up to five months past the largest events. However, the computed total column ozone and stratospheric temperature changes connected with the solar events were not found to be statistically significant. Thus, solar proton events do not likely contribute significantly to measured total column ozone fluctuations and stratospheric temperature changes.

# **The Long-term Middle Atmospheric Influence of Very Large Solar Proton Events**

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**Abstract:** The Whole Atmosphere Community Climate Model (WACCM3) has been used to study the long-term (> few months) effects of solar proton events (SPEs). Extremely large solar proton events occurred in 1972, 1989, 2000, 2001, and 2003 and caused long-lasting atmospheric changes. The highly energetic solar protons produced odd hydrogen ( $\text{HO}_x$ ) and odd nitrogen ( $\text{NO}_y$ ), which then led to ozone variations. The long-term effects on ozone were caused by the  $\text{NO}_y$  enhancements since the  $\text{HO}_x$  increases were short-lived (days). Very large  $\text{NO}_y$  enhancements lasted for months in the middle and lower stratosphere after a few of the largest SPEs. SPE-caused  $\text{NO}_y$  increases computed with WACCM3 were statistically significant at the 95% level throughout much of the polar stratosphere and mesosphere in the recent solar maximum five-year period (2000-2004). WACCM3-computed SPE-caused polar stratospheric ozone decreases of >10% continued for up to five months past the largest events. WACCM3 also computed SPE-caused polar lower stratospheric ozone increases >10% in the Southern Hemisphere (SH). These SH impacts were due to interference by enhanced  $\text{NO}_y$  constituents with the chlorine and bromine catalytic cycles for ozone, leading to a long-lived increase in ozone several months after the very large July 2000 SPE. Annually averaged SPE-caused polar total ozone and temperature changes from WACCM3 were not found to be statistically significant.

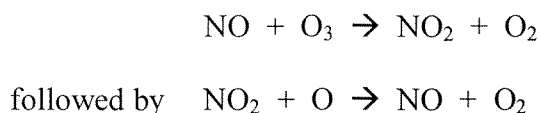
## 1. Introduction

Large solar eruptions can cause huge fluxes of high-energy solar protons that reach Earth, especially near solar maximum. Such periods of intense solar proton flux are known as solar proton events (SPEs) and tend to be infrequent. These SPEs typically last for a few days and lead to polar atmospheric changes through ionization, dissociation, dissociative ionization, and excitation processes. Some of the larger SPEs have caused a significant change in chemical constituents such as  $\text{HO}_x$ ,  $\text{NO}_y$ , and ozone [e.g., Heath et al., 1977; Thomas et al., 1983; McPeters and Jackman, 1985; McPeters, 1986; Zadorozhny et al., 1992; Jackman et al., 1995, 2001, 2005a, 2008; Randall et al., 2001; Seppala et al., 2004, 2006; Lopez-Puertas et al., 2005a,b; von Clarmann et al., 2005; Orsolini et al., 2005; Degenstein et al., 2005; Rohen et al., 2005; Verronen et al., 2005, 2006]. Since SPEs affect radiatively active ozone, they have been computed to cause middle atmospheric temperature and other dynamical changes [Reagan et al., 1981; Jackman and McPeters, 1985; Roble et al., 1987; Reid et al., 1991; Zadorozhny et al., 1994; Jackman et al., 1995; Krivolutsky et al., 2006; Jackman et al., 2007].

The magnitude and longevity of the SPE-caused atmospheric constituent and temperature changes has a direct relationship to the long-term stratospheric trends, which are so important in understanding the anthropogenic impact on ozone [WMO, 2007]. The SPE-caused impacts are largest in the polar regions in the mesosphere and upper stratosphere. Quantifying the downward and Equator-ward transport of this SPE-induced perturbation is one of the main objectives of this paper.

SPEs lead to ionization and the production of the important constituent families of  $\text{HO}_x$  (H, OH,  $\text{HO}_2$ ) and  $\text{NO}_y$  ( $\text{N}(^4\text{S})$ ,  $\text{N}(^2\text{D})$ , NO,  $\text{NO}_2$ ,  $\text{NO}_3$ ,  $\text{N}_2\text{O}_5$ ,  $\text{HNO}_3$ ,  $\text{HO}_2\text{NO}_2$ ,  $\text{ClONO}_2$ ,  $\text{BrONO}_2$ ). The SPE-produced  $\text{HO}_x$  constituents are relatively short-lived ( $\sim$ days) and lead to the

destruction of ozone in the upper stratosphere and mesosphere (pressures less than about 2 hPa). Both short- and longer-term (~months) catalytic ozone destruction is caused by the SPE-produced NO<sub>y</sub> constituents in the lower mesosphere and stratosphere (pressures greater than about 0.5 hPa) via the well-known NO<sub>x</sub> (NO+NO<sub>2</sub>) ozone loss cycle



Some modeling studies have addressed the longer-term atmospheric influence of SPEs [e.g., Jackman et al., 1990, 1995, 2000, 2005a,b; Reid et al., 1991; Semeniuk et al., 2005; Jackman and Fleming, 2008]. Only one of these previous studies [Semeniuk et al., 2005] used a general circulation model (GCM). In that study, Semeniuk et al. [2005] focused on the huge polar NO<sub>x</sub> lower mesospheric enhancements observed by ACE (Atmospheric Chemistry Experiment) in mid-February 2004, discussed in Rinsland et al. [2005], and found that the Oct./Nov. 2003 SPEs were a relatively minor contributor compared with the auroral NO<sub>x</sub> source.

We studied the short- and medium-term (days to a few months) atmospheric constituent effects of very large SPEs in Jackman et al. [2008] with version 3 of the Whole Atmosphere Community Climate Model (WACCM3). The present investigation complements that study with model simulations and analyses of the long-term (>few months) stratospheric constituent changes caused by SPEs. WACCM3 is a general circulation model with complete interactive photochemistry. The model has a domain that extends from the ground to the lower thermosphere, which allows study of the detailed time-dependent 3-D atmospheric response to a

variety of perturbations. We will focus on the years 2000-2004, in which six of the largest ten SPEs in the past 45 years have occurred [Jackman et al., 2008], but will also address the SPE-caused changes over the longer 1963-2004 period.

This paper is divided into six sections, including the Introduction. The solar proton flux and SPE-induced production of  $\text{HO}_x$  and  $\text{NO}_y$  are discussed in Section 2. A description of WACCM3 is given in Section 3. WACCM3 model results for SPE-caused long-term constituent changes in solar cycle 23 (years 1996-2004) are shown in Section 4 while WACCM3 model results for SPE-caused long-term constituent changes over the period 1963 through 2004 are discussed in Section 5. The conclusions are presented in Section 6.

## **2. Proton Fluxes; Odd Hydrogen ( $\text{HO}_x$ ) and Odd Nitrogen ( $\text{NO}_y$ ) Production**

Several satellites in interplanetary space or in orbit around the Earth have measured solar proton fluxes. The National Aeronautics and Space Administration (NASA) Interplanetary Monitoring Platform (IMP) series of satellites provided measurements of proton fluxes from 1963-1993 [Jackman et al., 1990; Vitt and Jackman, 1996]. The National Oceanic and Atmospheric Administration (NOAA) Geostationary Operational Environmental Satellites (GOES) provided observed proton fluxes from 1994-2004 [Jackman et al., 2005b].

The proton flux data from the satellites were used to compute daily average ion pair production profiles using the energy deposition methodology discussed in Jackman et al. [1980] and Vitt and Jackman [1996]. The deposition of energy by all the protons and associated secondary electrons is included in the scheme. The creation of one ion pair was assumed to require 35 eV [Porter et al. 1976]. Details about the source of the proton fluxes for the various time periods are given in Jackman et al. [2008]. The daily averaged SPE-produced ionization

rates from 1963 through 2004 were calculated for use in WACCM3 and are provided as functions of pressure between 888 hPa (~1 km) and  $8 \times 10^{-5}$  hPa (~115 km) at the SOLARIS (Solar Influence for SPARC) website (<http://strat-www.met.fu-berlin.de/~matthes/sparc/inputdata.html>).

Odd hydrogen ( $\text{HO}_x$ ) is produced through complex ion chemistry [Solomon et al. 1981] by the SPEs. The SPE-produced  $\text{HO}_x$  is a function of ion pair production and altitude and is included in WACCM3 simulations using a lookup table from Jackman et al. [2005a, Table 1], which is based on the work of Solomon et al. [1981]. The  $\text{HO}_x$  constituents have a relatively short lifetime (~hours) throughout most of the mesosphere, and thus do not influence the stratosphere on a long-term (> few months) time period. Jackman et al. [2007] calculated the ozone depletion and dynamics change from solar proton enhanced  $\text{HO}_x$  constituents with the Thermosphere Ionosphere Mesosphere Electrodynamic General Circulation Model (TIME-GCM). They showed that mesospheric temperature and wind perturbations from SPE-produced  $\text{HO}_x$  were greatly diminished in just 4-6 weeks.

Odd nitrogen is produced when the energetic charged particles (protons and associated secondary electrons) dissociate  $\text{N}_2$ . We assume that ~1.25 N atoms are produced per ion pair and divide the proton impact of N atom production between ground state (~45% or ~0.55 per ion pair) and excited state (~55% or ~0.7 per ion pair) nitrogen atoms [Porter et al., 1976]. Thus in our model simulations we use a production of 0.55 ground state  $\text{N}(^4\text{S})$  per ion pair and 0.7  $\text{N}(^2\text{D})$  atoms per ion pair.

The period January 1, 1995 through December 31, 2004 included some very quiet periods with minor or no SPEs and some very large SPEs in 2000, 2001, and 2003. Figure 1 shows a time series of our computed daily averaged global  $\text{NO}_y$  production from SPEs in the stratosphere

and mesosphere in this time period. Although the solar UV-induced oxidation of nitrous oxide ( $\text{N}_2\text{O} + \text{O}(^1\text{D}) \rightarrow \text{NO} + \text{NO}$ ) provides the largest source of  $\text{NO}_y$  in the middle atmosphere (52-58 gigamoles per year; Vitt and Jackman [1996]), the SPE source of  $\text{NO}_y$  can be significant on certain days. This applies particularly at polar latitudes where the transport from lower latitudes and the larger solar zenith angles result in a somewhat smaller local source of  $\text{NO}_y$  due to  $\text{N}_2\text{O}$  oxidation. Table 1 shows the daily  $\text{NO}_y$  production from SPEs during the largest periods of proton fluxes in the ten year period 1995-2004. SPE-produced  $\text{NO}_y$  greater than 1 gigamole was computed for July 14-15, 2000; November 9, 2000; September 25-26, 2001; November 5-6, 2001; November 24, 2001; and October 29, 2003.

If the SPE-produced  $\text{NO}_y$  is transported to the middle and lower stratosphere [e.g., Randall et al., 2001; Jackman et al., 2005a], it has a lifetime of months to years. Downward transport of  $\text{NO}_y$  occurs mainly in the fall and winter time periods [Oct.-Nov.-Dec.-Jan.-Feb.-Mar. (ONDJFM) in the Northern Hemisphere; Apr.-May-Jun.-Jul.-Aug.-Sep. (AMJJAS) in the Southern Hemisphere], thus SPEs in these time periods cause longer-lasting effects on the stratosphere.

### **3. Description of the Whole Atmosphere Community Climate Model (WACCM3)**

WACCM3 is a useful tool for investigating the coupling among the various atmospheric regions from the troposphere through the middle atmosphere to the lower thermosphere [Sassi et al., 2002, 2004; Forkman et al., 2003; Richter and Garcia, 2006; Garcia et al., 2007; Marsh et al., 2007; Jackman et al. 2008]. The model has a domain from the surface to  $4.5 \times 10^{-6}$  hPa (about 145 km), with 66 vertical levels, and includes fully interactive dynamics, radiation, and chemistry. Modules from the Community Atmospheric Model (CAM3), the Thermosphere-



Ionosphere-Mesosphere-Electrodynamics General Circulation Model (TIME-GCM), and the Model for Ozone And Related chemical Tracers (MOZART-3) are incorporated into WACCM3 to simulate the dynamics and chemistry of the Earth's atmosphere. The vertical resolution is  $\leq 1.5$  km between the surface and about 25 km. Above that altitude, vertical resolution increases slowly to 2 km at the stratopause and 3.5 km in the mesosphere; beyond the mesopause, the vertical resolution is one half the local scale height. The version of WACCM3 used here has latitude and longitude grid spacing of  $4^\circ$  and  $5^\circ$ , respectively.

WACCM3 was forced with observed time-dependent sea surface temperatures (SSTs), observed solar spectral irradiance and geomagnetic activity changes, and observed concentrations of greenhouse gases and halogen species over the simulation periods [see Garcia et al., 2007]. We have completed eight WACCM3 simulations, four with the daily ionization rates from SPEs and four without. The ionization rates, when included, were applied uniformly over both polar cap regions ( $60\text{-}90^\circ\text{N}$  and  $60\text{-}90^\circ\text{S}$  geomagnetic latitude) as solar protons are guided by the Earth's magnetic field lines to these areas [e.g., McPeters et al., 1981; Jackman et al., 2005a]. There are differing offsets of the geomagnetic and geographic poles in the two hemispheres thus the effects are not expected to be symmetric in the northern and southern hemispheres. The four simulations [1(a, b, c, d)] *with* the daily ionization rates from SPEs and the four simulations [1(w, x, y, z)] *without* the daily ionization rates were each performed over the 42 year period, Jan. 1, 1963 – Dec. 31, 2004 [see Table 2].

#### **4. SPE-caused Long-term Atmospheric Changes in Solar Cycle 23**

The influence of very large SPEs in the first nine years of solar cycle 23 (years 1996 – 2004) caused some significant documented changes in atmospheric composition, primarily

during and within several months of the events [e.g., Randall et al., 2001; Jackman et al., 2001, 2005a,b, 2008; Jackman and McPeters, 2004; Jackman and Fleming, 2008; Seppala et al., 2004; Degenstein et al., 2005; López-Puertas et al., 2005a,b; Orsolini et al., 2005; von Clarmann et al., 2005; Rohen et al., 2005]. For instance, Randall et al. [2001] showed evidence from HALOE observations of enhancements of about 15 ppbv in  $\text{NO}_x$  in the polar middle stratosphere two months after the July 2000 SPE in the Southern Hemisphere (SH), see Figure 2 (left). The  $\text{NO}_x$  increases in Sep./Oct. 2000 are about a factor of 2-3 beyond the normal range in the polar middle stratosphere for years 1991-1999. We also computed  $\text{NO}_x$  enhancements in Sep./Oct. 2000 with WACCM3 (simulation 1(a)) compared with years 1991-1999, see Figure 2 (right).

Although both WACCM3 predictions and HALOE measurements show reasonable agreement, there are some differences between the WACCM3 predictions and HALOE measurements such as a somewhat differently shaped interannual variability and a sharper  $\text{NO}_x$  peak in year 2000. These disparities point to differences between WACCM3 dynamics and atmospheric dynamics, a coarser model grid for WACCM3 than HALOE, and other differences discussed in more detail in Jackman et al. [2008]. In spite of these model/measurement differences, WACCM3 computed a very large  $\text{NO}_x$  peak in the year 2000, which is similar to that observed in HALOE and was caused by the July 2000 SPE. We therefore analyze the WACCM3 output for longer periods beyond the several very large SPEs in solar cycle 23 and focus on longer-term atmospheric changes. Such analyses should address the primary question: Do very large SPEs significantly influence constituents, particularly ozone, in the middle atmosphere beyond six months past the events?

#### **4.1 SPE-caused Annually Averaged Constituent Changes**

We focus most of our analyses on  $\text{NO}_y$  and ozone. As discussed before, the  $\text{NO}_y$  family has a long lifetime in the polar stratosphere and can be greatly enhanced by very large SPEs. Stratospheric ozone is extremely important for life on Earth and its abundance is partly controlled by the  $\text{NO}_y$  family. We show the WACCM3-computed annual mean  $\text{NO}_y$  and ozone distribution in Figure 3 for 1996 from the ensemble 1(w, x, y, z), which does not include SPEs and is near solar minimum. As discussed in Garcia et al. [2007] the WACCM3 ozone is in general agreement with HALOE data. There is a modest difference near 32 km in the tropics where WACCM3 is high by about 0.5 ppmv, which has been attributed to the WACCM3  $\text{NO}_x$  being too low at this altitude by about 15% [Eyring et al., 2006]. Both  $\text{NO}_y$  and ozone show peaks in the tropics with the  $\text{NO}_y$  maximum being about 5 km higher in altitude. Near the top of the altitude domain in Figure 3 (left), the  $\text{NO}_y$  clearly shows descent of  $\text{NO}_y$ -rich air from higher altitudes in both hemispheres to the middle mesosphere ( $\sim 0.04$  hPa, 70 km) and lower.

The annual zonal average computed change for 1996 caused by SPEs for  $\text{NO}_y$  and ozone is shown in Figure 4 as the percentage difference between the ensemble averages of simulations 1(a,b,c,d) and 1(w,x,y,z). The colored regions in the plots indicate 95% statistical significance using Student's t-test. Note that there are up to  $\pm 20\%$  changes computed for  $\text{NO}_y$  in the middle to upper mesosphere; however, such changes are mostly not statistically significant. This is not surprising given the fact that 1996 was a very quiet year with no substantial SPEs. Consistent with this, Randall et al. [1998; 2007] inferred from HALOE and POAM measurements that only minimal, if any, descent of mesospheric  $\text{NO}_x$  to the southern hemisphere stratosphere occurred during 1996. Also, virtually none of the computed ozone change is statistically significant.

The WACCM3-computed annual zonal mean  $\text{NO}_y$  and ozone distribution is shown in Figure 5 for 2000 from the ensemble 1(w, x, y, z), which does not include SPEs and is near solar

maximum. Near the top of the altitude domain in Figure 5 (left), the  $\text{NO}_y$  shows descent of  $\text{NO}_y$ -rich air from higher altitudes in both hemispheres to the lower mesosphere ( $\sim 0.1$  hPa, 64 km). The descent of  $\text{NO}_y$  in the polar SH in Figure 5 (left) is clearly larger than that seen in Figure 3 (left) and is caused by the larger source of  $\text{NO}_y$  near solar maximum from increased geomagnetic activity and high energy photons [also, see Figure 9 of Marsh et al., 2007].

The computed annual zonal average change for 2000 caused by SPEs for  $\text{NO}_y$  and ozone is shown in Figure 6 as the percentage difference between ensemble averages of simulations 1(a,b,c,d) and 1(w,x,y,z). The colored regions in the plots indicate 95% statistical significance using Student's t-test. There are over 100% maximum increases computed for  $\text{NO}_y$  in the polar middle to lower mesosphere (Fig. 6, left) and a good portion of the computed polar  $\text{NO}_y$  enhancements from 10 to 0.01 hPa are statistically significant. The very large SPEs in July and November, 2000, account for most of this increase in  $\text{NO}_y$ . Unlike  $\text{NO}_y$ , only small regions of the computed polar ozone changes are statistically significant. Computed ozone decreases of -5 to -2 % are statistically significant in the SH polar middle to upper stratosphere ( $\sim 10$  to  $\sim 2$  hPa) and in the NH upper stratosphere ( $\sim 4$  to  $\sim 1$  hPa). Even smaller regions in the mesosphere show a statistically significant ozone decrease.

Year 2001 was particularly active with three very large SPEs (one in September and two in November, see Table 1); thus maximum increases  $>100\%$  were computed for annual averaged SPE-impacted  $\text{NO}_y$  in regions of the polar middle atmosphere in both hemispheres (Fig. 7, left). The colored regions in the plots indicate 95% significance using Student's t-test. A large portion of the computed polar  $\text{NO}_y$  enhancements from 30 to 0.01 hPa are statistically significant. Most of the computed Northern Hemisphere (NH) polar ozone decrease above 20 hPa is statistically

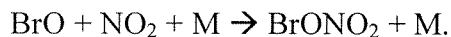
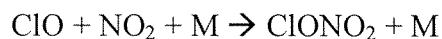
significant, however, only small portions of the SH polar ozone decreases are statistically significant.

The extent of the statistically significant NH ozone decrease throughout much of the polar mesosphere is somewhat surprising. SPE-caused mesospheric ozone decrease tends to be dominated by the HO<sub>x</sub> increases, which are short-lived [e.g., Jackman et al., 2001, Verronen et al., 2006]. Analysis of our model results show that the SPE-produced HO<sub>x</sub> resulted in annual average NH polar ozone change from about -1% to -4%. The computed mesospheric ozone changes from SPE-produced NO<sub>y</sub> were from near 0% to -1%. Thus the huge HO<sub>x</sub>-caused ozone losses in the very SPE-active year 2001 did contribute to the overall annual average polar mesospheric ozone change and helped result in a statistically significant NH ozone signal.

The two very large SPEs in November 2001 occurred at a near-ideal time to maximize a significant NH impact due to the prevailing downward transport in the fall, which continued for another few months. The other very large SPE in September 2001 occurred near the end of the SH winter, thus a reduced statistically significant ozone impact was computed for the SH (Fig. 7, right). Although not statistically significant, we computed a modest ozone increase in the SH polar lower stratosphere (maximum >20%) as a result of the SPEs. This type of ozone behavior connected with large SPE-induced NO<sub>y</sub> increases has been discussed before [Jackman et al., 2000; Jackman and Fleming, 2008] and is related to the interference by NO<sub>y</sub> constituents with the halogen (chlorine and bromine) catalytic cycles.

In order to elucidate the WACCM3 result of SPE-caused ozone enhancements in the SH polar lower stratosphere, we focus on a smaller region of the atmosphere in Figure 8 for year 2001. The annual zonal average change caused by SPEs for NO<sub>y</sub>, ozone, BrONO<sub>2</sub>, and ClONO<sub>2</sub> was computed and is shown over the globe from the ground to 10 hPa as the percentage

difference between the ensemble averages of simulations 1(a,b,c,d) and 1(w,x,y,z). The large enhancements in SH polar  $\text{NO}_y$  (Fig. 8, top left) between 300 and 10 hPa lead to increases in  $\text{ClONO}_2$  (Figure 8, bottom right) and  $\text{BrONO}_2$  (Fig. 8, bottom left) increases through the reactions



The chlorine and bromine reservoir species ( $\text{ClONO}_2$  and  $\text{BrONO}_2$ ) are produced at the expense of the reactive species ( $\text{ClO}$  and  $\text{BrO}$ ) that drive the  $\text{ClO}_x$  and  $\text{BrO}_x$  catalytic cycles. The ozone loss rates due to chlorine and bromine are then reduced in the SH lower polar stratosphere and ozone is increased (Fig. 8, top right).

The annual zonal average change for the next three years (2002-2004) caused by SPEs for  $\text{NO}_y$  and ozone was computed, again as the percentage difference between the ensemble averages of simulations 1(a,b,c,d) and 1(w,x,y,z). A large SPE occurred in April 2002 (see Table 1), however, this event was much smaller than the very large SPEs of 2000 and 2001 and caused a smaller annual average maximum  $\text{NO}_y$  enhancement (not shown). Still, a good portion of the polar middle atmosphere had a statistically significant  $\text{NO}_y$  enhancement due to the SPEs in 2002. The statistically significant ozone impact of these SPEs was mainly confined to the NH polar middle stratosphere region in 2002 (not shown).

Year 2003 had a very active period with a very large SPE in late October and a large SPE in early November (see Table 1), connected with the “Halloween Storms” of 2003. Very large maximum increases (>100 %) were computed for  $\text{NO}_y$  in the polar NH mesosphere and large maximum increases (>50%) were computed for  $\text{NO}_y$  in the polar SH mesosphere as a result of the SPEs (Fig. 9, left). A large portion of the computed polar NH  $\text{NO}_y$  enhancements from 30 to

0.01 hPa were statistically significant, whereas a more modest portion of the computed SH NO<sub>y</sub> enhancements were statistically significant in the same pressure range. There were only a few small regions that showed a statistically significant ozone change due to SPEs in 2003 (Fig. 9, right).

Year 2004 was relatively quiet with no large SPEs. Modest statistically significant annual zonal average NO<sub>y</sub> enhancements were computed in the polar mesosphere for both hemispheres and a statistically significant NO<sub>y</sub> increase was calculated for the polar NH middle to lower stratosphere (not shown). This feature was probably a result of the downward transport of the NO<sub>y</sub> signal in 2003 to lower atmospheric regions. There were only very small regions that showed a statistically significant ozone change due to SPEs (not shown).

Given the several very large SPEs that occurred in years 2000, 2001, and 2003, we have also investigated the SPE-caused atmospheric changes in the five-year average 2000-2004. The computed changes produced by SPEs for NO<sub>y</sub> and ozone are shown in Figure 10 as the percentage difference between ensemble averages of simulations 1(a,b,c,d) and 1(w,x,y,z). The colored regions in the plots indicate 95% significance using Student's t-test. For the period 2000-2004 period, very large maximum increases (>100 %) were computed for NO<sub>y</sub> in the polar NH and large maximum increases (>50%) were computed for the polar SH as a result of the SPEs (Fig. 10, left). Very large regions of the computed polar NO<sub>y</sub> enhancements are statistically significant in both hemispheres; however, the statistically significant region in the NH extends much farther into the lower stratosphere. This is a reflection of the larger NO<sub>y</sub> input to the NH during fall and winter (ONDJFM) of 20.1 gigamoles versus the SH input of 9.4 gigamoles during corresponding seasons (AMJJAS). There are also more regions in the polar NH that show a statistically significant ozone change due to SPEs in the 2000-2004 period (Fig.

10, right), which is related to the larger late fall and winter  $\text{NO}_y$  input. The statistically significant mesospheric NH ozone change was mainly caused by the SPE-produced  $\text{HO}_x$ .

#### **4.2 SPE-caused Polar Atmospheric Changes**

SPEs initially impact the polar cap regions ( $60\text{-}90^\circ$  geomagnetic latitude) of both hemispheres. The influence over longer periods beyond that initial disturbance is dependent on the amount of  $\text{NO}_y$  produced and the strength of the downward transport. Therefore, the largest computed SPE impacts are at polar latitudes, which is consistent with Hood and Soukharev [2006], who inferred from HALOE data that there is no statistically significant signature of a solar cycle in low-latitude stratospheric  $\text{NO}_x$ . We examine the WACCM3 SPE-caused changes at high latitudes in this section.

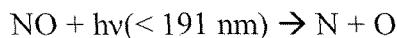
We have investigated the polar changes over a particularly active one year period, July 1, 2000 – June 30, 2001. The time dependent atmospheric changes were computed, some of which are a result of the initial SPE input and some of which are a result of transport. The changes caused by SPEs for  $\text{NO}_y$  and ozone in the latitude bands  $70\text{-}90^\circ\text{S}$  and  $70\text{-}90^\circ\text{N}$  are given in Figure 11. We use the monthly average output for the WACCM3 simulations and present the percentage difference between the ensemble averages of simulations 1(a,b,c,d) and 1(w,x,y,z). This period included the very large July 2000 SPE, the third largest SPE period in the past 45 years [see Table 1 of Jackman et al., 2008], and the very large November 2000 SPE, the sixth largest SPE period in the past 45 years, as well as another moderately large SPE, which occurred in April 2001. The computed  $\text{NO}_y$  enhancement in the  $70\text{-}90^\circ\text{S}$  band is enormous for the July 2000 SPE with maximum increases  $>500\%$  (Fig. 11, left top). This increase in  $\text{NO}_y$ , an indicator of the very large SPE, is transported slowly and steadily downwards throughout the time period.



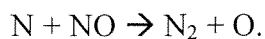
The levels of increased  $\text{NO}_y$  were reduced due to mixing with smaller amounts of  $\text{NO}_y$  from lower latitudes and altitudes, eventually reaching levels of just over 20%  $\text{NO}_y$  enhancement in the lower stratosphere ( $\sim 300$  to 40 hPa) by June 30, 2001. The November 2000 SPE appears to have added somewhat to this huge  $\text{NO}_y$  SPE-caused signal in late 2000. The April 2001 SPE caused significant changes in  $\text{NO}_y$ , but its impact by June 30, 2001 was located in the middle to upper stratosphere ( $\sim 10$  to 2 hPa).

The longer-term computed ozone decrease connected with the July 2000 SPE is substantial, reaching levels  $>20\%$  in the middle to upper stratosphere ( $\sim 10$  to 2 hPa) between August and November, 2000 (Fig. 11, left bottom). Note that we use WACCM3 monthly average output in these analyses, thus the much larger computed ozone decreases presented in Jackman et al. [2008] were not calculated. The larger computed ozone decreases of Jackman et al. [2008] were driven by the short-lived  $\text{HO}_x$  constituents and did not last beyond a couple of days after the SPEs. In the later part of this period (October 2000 – June 2001), ozone increases below  $\sim 10$  hPa were computed; these are presumably due to the fact that enhanced  $\text{NO}_y$  sequesters chlorine and bromine in the reservoir species ( $\text{ClONO}_2$  and  $\text{BrONO}_2$ ), resulting in reduced ozone loss in this region of the stratosphere. The close correlation between the enhancements of  $\text{ClONO}_2$  and  $\text{BrONO}_2$  and the enhanced lower stratospheric ozone is shown clearly in Figure 12. Both  $\text{ClONO}_2$  and  $\text{BrONO}_2$  are enhanced  $>10\%$  throughout most of the region where the ozone is increased  $>10\%$  starting in January 2001.

The computed SPE-caused  $\text{NO}_y$  enhancement in the latitude band  $70\text{--}90^\circ\text{N}$  is larger for the November 2000 SPE than for the July 2000 SPE with maximum increases  $>1000\%$  (Fig. 11, right top). The July 2000 SPE NH  $\text{NO}_y$  enhancement was substantially reduced due to the summer sunlight increasing the loss process for  $\text{NO}_y$  via



followed by



The levels of increased  $\text{NO}_y$  due to the November 2000 SPE were reduced due to mixing with smaller amounts of  $\text{NO}_y$  from lower latitudes and altitudes, eventually reaching levels less than 10%  $\text{NO}_y$  enhancement in the middle stratosphere ( $\sim 40$  to 10 hPa) by May 2001 (Fig. 11, right top). The July 2000 and April 2001 SPEs caused significant changes in  $\text{NO}_y$ , but their impacts are primarily confined to pressures  $< 10$  hPa.

The largest longer-term computed ozone decrease in the polar NH appears to be connected with the November 2000 SPE and reached levels  $> 10\%$  in the mesosphere and upper stratosphere in November 2000. The signal of ozone decrease was slowly transported downwards to the middle stratosphere and gradually diminished with decreases of  $< 5\%$  by the end of April 2001 (Fig. 11, right bottom).

We have also computed the polar changes over the longer period, January 1, 2000 – December 31, 2004, which included several very large SPEs (see Table 1). The changes caused by SPEs for  $\text{NO}_y$ , ozone, and temperature in the latitude band  $70\text{--}90^\circ\text{S}$  are given in Figure 13. Very large enhancements of  $\text{NO}_y$  (Fig. 13, top) extending from the mesosphere to the middle stratosphere are apparent in four of the five years (e.g., 2000, 2001, 2002, and 2003). The two years 2000 and 2002 show the deepest penetration of the enhanced  $\text{NO}_y$  signal as a very large SPE and a large SPE in those years occurred in the fall or winter (July 2000 and April 2002, see Table 1). The SH polar ozone signal (Fig. 13, middle) follows the  $\text{NO}_y$  signal into the atmosphere and is generally anti-correlated with  $\text{NO}_y$ , except in the lower stratosphere (see Figure 12).

Earlier papers have addressed the possibility of temperature changes resulting from SPEs [Reagan et al., 1981; Jackman and McPeters, 1985; Roble et al., 1987; Reid et al., 1991; Zadorozhny et al., 1994; Jackman et al., 1995; Krivolutsky et al., 2006; Jackman et al., 2007]. Temperature changes (mostly decreases) of 1-10 K were computed to follow from very large SPEs, although most of these computed influences occurred within hours to days of very large SPEs. Reid et al. [1991] did compute a fairly long-lasting upper stratospheric temperature decrease of about 1K nearly three months after the very large SPE of Oct. 1989.

The SH polar temperature stratospheric signal (Fig. 13, bottom) is generally well-correlated with the ozone change. A fairly high degree of correlation was computed for the pressure range 30-200 hPa, where the correlation coefficient between temperature and ozone change was found to be greater than 0.75.

The colored regions in Figure 13 indicate 95% statistical significance with the use of Student's t-test. Much of the computed NO<sub>y</sub> enhancement greater than about 20% in the middle stratosphere and above in the first two and a half years of the plot is statistically significant, however, very little of the computed NO<sub>y</sub> enhancement after May 2002 is statistically significant (Fig. 13, top). The areas of statistically significant ozone changes are much less than those for NO<sub>y</sub> enhancements (Fig. 13, middle). Four months in late 2000 (Sep.-Dec.) in the middle stratosphere, three months in late 2001 (Oct.-Dec.) in the upper stratosphere, and a few other small regions are statistically significant. Virtually none of the computed temperature changes are statistically significant (Fig. 13, bottom).

The changes caused by SPEs for NO<sub>y</sub>, ozone, and temperature in the latitude band 70-90°N are given in Figure 14. Very large tongues of SPE-produced NO<sub>y</sub> enhancement (Fig. 14, top) extending from the mesosphere to the middle stratosphere are apparent in three time periods:

1) Nov. 2000 – Apr. 2001; 2) Nov. 2001 – Dec. 2002; and 3) Oct. 2003 – Nov. 2004. The very large SPEs in Nov. 2000, Nov. 2001, and Oct. 2003, and the large SPE in Nov. 2003 drove most of these NO<sub>y</sub> increases because they occurred during the fall season (see Table 1). The NH polar ozone signal (Fig. 14, middle) follows the NO<sub>y</sub> signal into the stratosphere and is anti-correlated with NO<sub>y</sub>. The NH polar temperature changes do not show the same correlation with ozone that was computed in the SH polar region. The correlation coefficient between temperature and ozone change in the NH polar stratosphere was computed to be less than 0.5 for pressures less than 200 hPa.

The colored regions in Figure 14 indicate 95% statistical significance with the use of Student's t-test. Large regions of the computed SPE-caused NO<sub>y</sub> enhancements are statistically significant and the statistically significant NO<sub>y</sub> increases can extend from the mesosphere to the middle stratosphere (from 0.1 to ~10 hPa; Fig. 14, top). As in the polar SH, the areas of statistically significant ozone changes are much smaller than those for NO<sub>y</sub> enhancements (Fig. 14, middle). A summer SPE (July 2000) helped contribute to nearly four months (Jul. – Oct., 2000) of statistically significant ozone decrease in the upper stratosphere. The furthest penetrating statistically significant ozone decrease occurred between Sep. 2001 and Jan. 2002, when the ozone signal followed the NO<sub>y</sub> enhancement from the lower mesosphere/upper stratosphere down to the middle stratosphere. The large SPE in Sep. 2001 and the two very large SPEs in Nov. 2001 contributed to most of this strong signal. A lesser statistically significant signal of ozone decrease was connected with the “Halloween storms” in late 2003, but the signal was in the mesosphere and did not extend beyond Dec. 2003. As in the SH, virtually none of the computed NH temperature changes are statistically significant (Fig. 14, bottom).

## **5. SPE-caused Long-term Polar Atmospheric Changes in the 1963-2004 Period**

The very large SPEs in solar cycle 23 caused fairly substantial statistically significant polar NO<sub>y</sub> enhancements in both hemispheres in the period Jan. 1, 2000 through Dec. 31, 2004. Although the computed statistically significant ozone decreases covered much less of the domain in altitude and time than those computed for NO<sub>y</sub>, a few of the very large SPEs in solar cycle 23 managed to create long-lasting statistically significant polar ozone signals. In general, the computed polar temperature changes were not statistically significant over the years 2000-2004. Our analysis will be extended in this section to include years 1963-2004. We will focus on a quantification of the long-term impact of SPEs on ozone and temperature.

### **5.1 SPE-caused Temperature and Ozone Profile Changes**

Virtually no SPEs occurred in 1963-64, thus we focused on the WACCM3 output over the 40 year period 1965-2004, to determine if any signals in ozone and temperature were statistically significant. There were large and very large SPEs throughout the 40 year period, which included solar cycles 20-23. An annual zonal average SPE-caused ozone and temperature change for the polar SH latitude region 70-90°S is shown in Figure 15 (middle and top, respectively) as the difference between the ensemble averages of simulations 1(a,b,c,d) and 1(w,x,y,z). The SPE-caused NO<sub>y</sub> production (in gigamoles) per year is shown in the bottom plot of Figure 15 to focus attention on any apparent correlations between SPEs and annually averaged ozone and temperature variations. The colored regions indicate a 95% statistical significance with the use of Student's t-test. Ozone variations up to +/- 10% and temperature variations up to +/- 2K are computed. The main conclusion from these analyses is that there is not much

statistically significant atmospheric change from SPEs in the SH, when evaluating the annual average ozone and temperature variations.

The NH model output was analyzed in a similar way (annual zonal average) and ozone and temperature change for the polar latitude region 70-90°N are given in Figure 16 (middle and top, respectively). The computed ozone and temperature variations are smaller in the NH than the SH with ozone fluctuations of up to +/- 5% and temperature variations up to +/- 1K. The colored regions indicate a 95% statistical significance with the use of Student's t-test. A small statistically significant ozone change is computed in the NH in the middle to upper stratosphere for years 2000-2001-2002 (slightly different altitudes for each year), probably caused by the large and very large SPEs which occurred in years 2000 (Jul. and Nov.) and 2001 (Sep. and two in Nov.). However, there are no computed statistically significant temperature changes.

## **5.2 SPE-caused Total Ozone Changes**

A maximum polar ozone column decrease of about 1 to 4% has been computed in several previous two-dimensional modeling studies due to very large SPEs [e.g., Jackman et al., 1990, 1995, 2000, 2005a,b; Reid et al., 1991; Jackman and Fleming, 2008]. These earlier analyses were unable to reveal any SPE-caused observed polar ozone column depletion due to the substantial intra-seasonal and inter-annual variation in total ozone at high latitudes. We show the polar ozone column variations from the individual WACCM3 ensemble of simulations 1(a,b,c,d) compared with observations in Figure 17 (top, 70-90°S and bottom, 70-90°N). The observations were differenced with respect to an average of the two years, 1979 and 1980, to allow for any quasi-biennial influences. The simulations were differenced with respect to the ensemble average {1(a,b,c,d)} of two years, 1979 and 1980.

A special analysis of total ozone measurements was needed to derive polar total ozone: Following the methodology of Fioletov et al. [2002], the gaps at various latitudes in the monthly-mean merged ozone data set [Stolarski and Frith, 2006] were interpolated in time for each latitude band as long as the nearest good measurement on either side of the missing point was within 3 months. The total ozone data were extrapolated in latitude by duplicating the northernmost (or southernmost) good measurement to the next missing poleward bin. This time interpolation was repeated and the extrapolation was redone in latitude until all the points were filled in. There were some artifacts at high latitudes during periods where the real data cut off occurred at  $\sim 40\text{-}45^\circ$ ; however, these were not very large. The interspersing of the data interpolated in time and latitude appeared to reduce the artifacts.

The polar total ozone for  $70\text{-}90^\circ\text{S}$  from WACCM3 with SPEs and observations show similar trend behavior and magnitude of interannual variability (Fig. 17, top). The polar total ozone for  $70\text{-}90^\circ\text{N}$  from WACCM3 and observations does indicate a similar trend between 1979/1980 and 2004 (Fig. 17, bottom); however, WACCM3 simulated significantly higher total ozone in 1989 and in 1991 through 1995. Could the very large SPEs in Aug.-Sep.-Oct. 1989 [see Table 1, Jackman et al., 2008] have contributed to the substantial change (decrease) from 1988 to 1989 for the  $70\text{-}90^\circ\text{N}$  band? Given the modest predicted profile ozone decreases ( $<5\%$ , Fig. 16), this seems unlikely. Also, there were not any very large SPEs in 1991-1995, which may have contributed to the lower total ozone values in those years. The Mt. Pinatubo eruption occurred in 1991 and likely contributed to lower total ozone levels in 1991 and 1992, similar to the effect at mid-latitudes [e.g., Solomon et al., 1996]. Mostly cooler winter and spring temperatures have also been linked to the observed reduced polar NH ozone values from 1991 to 1995 [Randel and Wu, 1999; p. 4.4, WMO, 2007].

We next examined WACCM3 output to derive any statistically significant signal from SPEs in total polar ozone. For this analysis, we computed the annually averaged SPE-caused total ozone change from WACCM3 simulations by differencing the ensemble averages of simulations 1(a,b,c,d) and 1(w,x,y,z). These differences are compared in Figure 18 for the polar regions, 70-90°S (top) and 70-90°N (bottom). The thick solid line indicates the difference of the simulations and the light dashed line indicates the one sigma values for the base ensemble of simulations 1(w,x,y,z). The SPE-caused total ozone change signal for most years is within one sigma of the average of the base simulations. A Student's t-test analysis indicates that the signal in 1966 for the NH is the only one that is statistically significant at the 95% level. Although there was a very large SPE in September 1966 [see Table 1, Jackman et al. 2008], the computed total ozone change was positive in 1966. Year 1966 had minimal halogen loading [WMO, 2007], thus an ozone depletion would be expected from solar protons rather than an ozone increase. Hence this positive total ozone change in 1966 is not likely related to SPEs.

## 6. Conclusions

WACCM3 has been used to study the long-term (>few months) constituent changes caused by SPEs over the 1963-2004 time period. The most pronounced atmospheric effects were caused by the very largest SPEs in this period and were concentrated in the polar regions. Substantial statistically significant signals in NO<sub>y</sub> were computed for years 2000, 2001, and 2003 after very large SPEs and lasted for up to a year past the events. A large five-year average (2000-2004) statistically significant polar middle atmospheric NO<sub>y</sub> signal was calculated in both hemispheres for the latest solar maximum period.



Only modest statistically significant signals in mesospheric and stratospheric ozone were computed for the same events and these signals lasted only a few months, at most. The statistically significant NH mesospheric ozone signal for years 2000-2004 appears to be mainly caused by SPE-enhanced  $\text{HO}_x$ . The SPE-enhanced  $\text{NO}_y$  is the primary cause of the computed stratospheric ozone change. Analysis of annually averaged WACCM3 output showed statistically significant stratospheric ozone signals of some note in the polar NH for years 2000-2002. Very small statistically significant stratospheric ozone signals were computed for the polar SH. An ozone increase connected with SPEs was found in the lower stratosphere and was driven by the enhanced production of  $\text{ClONO}_2$  and  $\text{BrONO}_2$ , which reduced the halogen-caused ozone loss. However, this SPE-caused enhanced ozone was not statistically significant. The annually averaged polar temperature and total ozone variations connected with SPEs were also not statistically significant.

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**Table 1.** Largest daily NO<sub>y</sub> production from solar proton events between Jan. 1, 1995 and Dec. 31, 2004, given in chronological order. Only days with production greater than or equal to 0.4 gigamoles of NO<sub>y</sub> are shown.

<i>Date of SPE</i>	<i>Computed NO<sub>y</sub> Daily Production In Middle Atmosphere (Gigamoles<sup>1</sup>)</i>
July 14, 2000	1.3
July 15, 2000	4.5
November 9, 2000	3.6
November 10, 2000	0.7
September 25, 2001	1.4
September 26, 2001	1.3
November 5, 2001	2.0
November 6, 2001	3.0
November 23, 2001	0.8
November 24, 2001	2.0
April 21, 2002	0.5
April 22, 2002	0.4
October 28, 2003	0.8
October 29, 2003	3.9
October 30, 2003	0.9
November 3, 2003	0.4
<sup>1</sup> Gigamole = 6.02 x 10 <sup>32</sup> atoms and molecules	

**Table 2.** Description of WACCM3 simulations

<i>Simulation designation</i>	<i>Number of realizations</i>	<i>Time period</i>	<i>SPEs included</i>
1 (a, b, c, d)	4	1963-2004	Yes
1 (w, x, y, z)	4	1963-2004	No



## Figure Captions

**Figure 1.** Daily column  $\text{NO}_y$  production in gigamoles ( $6.02 \times 10^{32}$  molecules) as a function of time for Jan. 1, 1995 – Dec. 31, 2004.

**Figure 2.** Taken from Fig. 15 of Jackman et al. (2008). Left plot is an adaptation of Fig. 5a of Randall et al. (2001) showing Southern Hemisphere (SH) polar vortex HALOE  $\text{NO}_x$  (ppbv) profiles in September/October for years 1991-2000. Right plot shows WACCM3 simulation 1(a) predicted SH polar vortex  $\text{NO}_x$  (ppbv) profiles for the same periods.

**Figure 3.** Annual mean of simulations 1(w,x,y,z) for year 1996 – **Left:**  $\text{NO}_y$  with contour intervals 0.01, 0.1, 1, 2, 4, 7, 10, 20, and 40 ppbv. **Right:** Ozone with contour intervals 0.1, 0.5, 1, 2, 4, 6, 8, and 10 ppmv.

**Figure 4.** Annual average of solar proton event-caused change: Difference between average of simulations 1(a,b,c,d) and 1(w,x,y,z) for year 1996. **Left:**  $\text{NO}_y$  with contour intervals -20, -10, -5, 0, 5, 10, and 20%. **Right:** Ozone with contour intervals -5, -2, 0, 2, and 5%. Colored regions indicate 95% statistical significance with Student's t-test.

**Figure 5.** Annual mean of simulations 1(w,x,y,z) for year 2000 – **Left:**  $\text{NO}_y$  with contour intervals 0.01, 0.1, 1, 2, 4, 7, 10, 20, 40, 70, and 100 ppbv. **Right:** Ozone with contour intervals 0.1, 0.5, 1, 2, 4, 6, 8, and 10 ppmv.

**Figure 6.** Annual average of changes caused by solar proton events for year 2000: Difference between average of simulations 1(a,b,c,d) and 1(w,x,y,z) for year 2000 – **Left:** NO<sub>y</sub> with contour intervals -20, -10, -5, 0, 5, 10, 20, 50, and 100%. **Right:** Ozone with contour intervals -10, -5, -2, 0, 2, and 5%. Colored regions indicate 95% statistical significance with Student's t-test.

**Figure 7.** Annual average of changes caused by solar proton events for year 2001: Difference between average of simulations 1(a,b,c,d) and 1(w,x,y,z) for year 2001 – **Left:** NO<sub>y</sub> with contour intervals -20, -10, -5, 0, 5, 10, 20, 50, and 100%. **Right:** Ozone with contour intervals -10, -5, -2, 0, 2, and 5%. Colored regions indicate 95% statistical significance with Student's t-test.

**Figure 8.** Changes caused by solar proton events for year 2001. Difference between average of simulations 1(a,b,c,d) and 1(w,x,y,z) – **Left, top:** NO<sub>y</sub> with contour intervals -5, 0, 5, 10, 20, and 50%; **Right, top:** Ozone with contour intervals -5, 0, 5, 10, and 20%; **Left, bottom:** BrONO<sub>2</sub> with contour intervals -10, -5, 0, 5, 10, 20, and 50%; **Right, bottom:** ClONO<sub>2</sub> with contour intervals -10, -5, 0, 5, 10, and 20%.

**Figure 9.** Annual average of changes caused by solar proton events for year 2003: Difference between average of simulations 1(a,b,c,d) and 1(w,x,y,z) for year 2003 – **Left:** NO<sub>y</sub> with contour intervals -20, -10, -5, 0, 5, 10, 20, 50, and 100%. **Right:** Ozone with contour intervals -10, -5, -2, 0, 2, and 5%. Colored regions indicate 95% statistical significance with Student's t-test.

**Figure 10.** Solar proton event-caused change for average of years 2000-2004: Difference between average of simulations 1(a,b,c,d) and 1(w,x,y,z) - **Left:** shows NO<sub>y</sub> with contour intervals 0, 5, 10, 20, 50, and 100%; **Right:** Ozone with contour intervals -5, -2, 0, 2, and 5%. Colored regions indicate 95% statistical significance with Student's t-test.

**Figure 11.** Solar proton event-caused change for July 2000 to June 2001 in the SH polar latitude interval 70-90°S (**Left**) and in the NH polar latitude interval 70-90°N (**Right**): Difference between average of simulations 1(a,b,c,d) and 1(w,x,y,z) for NO<sub>y</sub> with contour intervals -20, -10, 0, 10, 20, 50, 100, 200, 500, and 1000% (**Top plots**); and ozone with contour intervals -20, -10, -5, -2, 0, 2, 5, 10, and 20% (**Bottom plots**).

**Figure 12.** Solar proton event-caused change for July 2000 to June 2001 in the SH polar latitude interval 70-90°S: Difference between average of simulations 1(a,b,c,d) and 1(w,x,y,z) – for BrONO<sub>2</sub> with contour intervals -50, -20, -10, 0, 10, 20, 50, 100, and 200% (**Top**); ClONO<sub>2</sub> with contour intervals -50, -20, -10, 0, 10, 20, 50, 100, and 200% (**Middle**); and ozone with contour intervals -20, -10, -5, -2, 0, 2, 5, 10, and 20% (**Bottom**).

**Figure 13.** Solar proton event-caused change for the five years 2000-2004 in the SH polar latitude interval 70-90°S: Difference between simulations 1(a,b,c,d) and 1(w,x,y,z). Colored regions indicate 95% statistical significance with the use of Student's t-test. – **Top:** NO<sub>y</sub> with contour intervals -20, -10, 0, 10, 20, 50, and 100%. **Middle:** Ozone with contour intervals -20, -

10, -5, -2, 0, 2, 5, 10, and 20%. **Bottom:** Temperature with contour intervals -7, -4, -2, -1, 0, 1, 2, 4, and 7K.

**Figure 14.** Solar proton event-caused change for the five years 2000-2004 in the NH polar latitude interval 70-90°N: Difference between simulations 1(a,b,c,d) and 1(w,x,y,z). Colored regions indicate 95% statistical significance with the use of Student's t-test. – **Top:** NO<sub>y</sub> with contour intervals -20, -10, 0, 10, 20, 50, and 100%. **Middle:** Ozone with contour intervals -20, -10, -5, -2, 0, 2, 5, 10, and 20%. **Bottom:** Temperature with contour intervals -7, -4, -2, -1, 0, 1, 2, 4, and 7K.

**Figure 15.** Solar proton event-caused change for the forty years 1965 through 2004 in the SH polar latitude interval 70-90°S: Difference between simulations 1(a,b,c,d) and 1(w,x,y,z). Colored regions indicate 95% statistical significance with the use of Student's t-test. – **Top:** Temperature with contour intervals -2, -1, 0, 1, and 2 K. **Middle:** Ozone with contour intervals -10, -5, -2, 0, 2, 5, and 10%. **Bottom:** NO<sub>y</sub> production per year (GigaMoles, GM) by solar proton events.

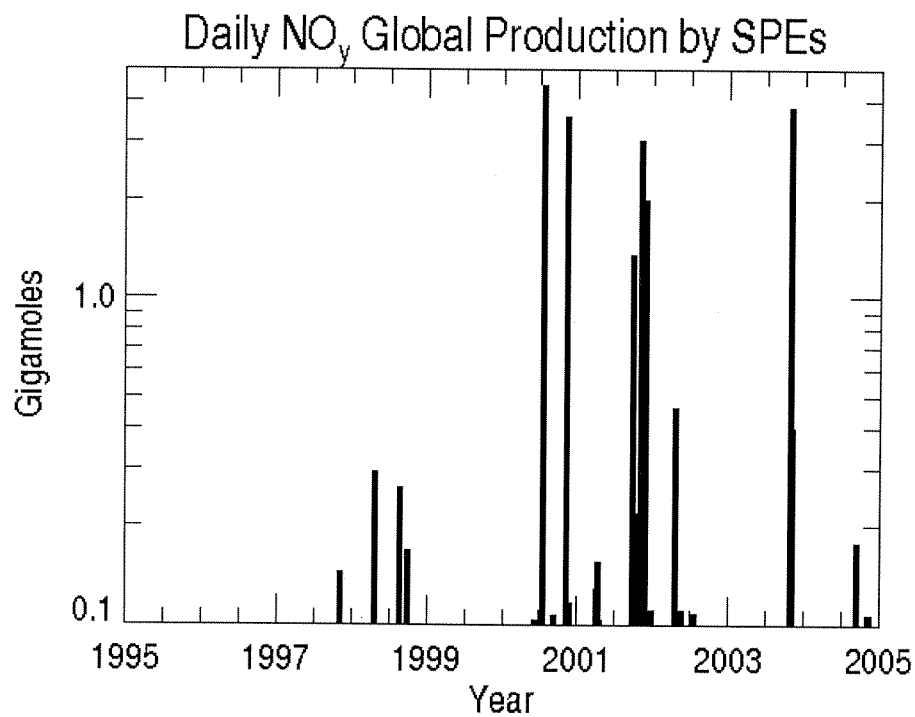
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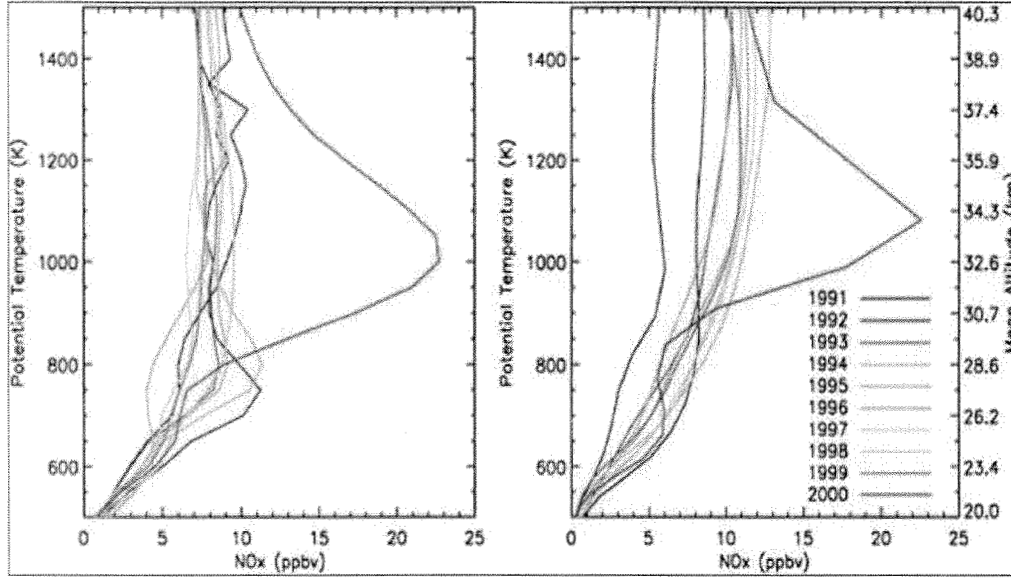
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**Figure 18.** Solar proton event-caused change in annually averaged total ozone for the forty years 1965 through 2004 in the SH polar latitude interval 70-90°S (**Top**) and the NH polar latitude interval 70-90°N (**Bottom**). The thick solid line indicates the difference between the average of simulations 1(a,b,c,d) and 1(w,x,y,z). The light dashed lines indicate the one sigma values for the base simulations 1(w,x,y,z).

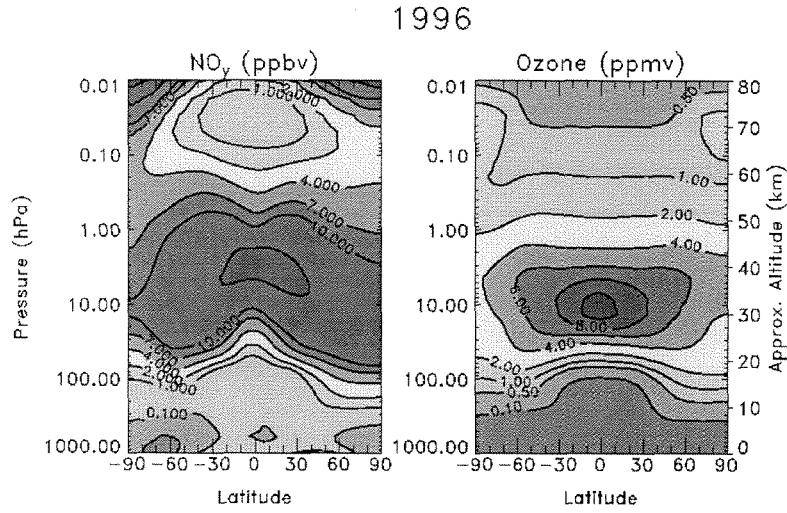
## Figures



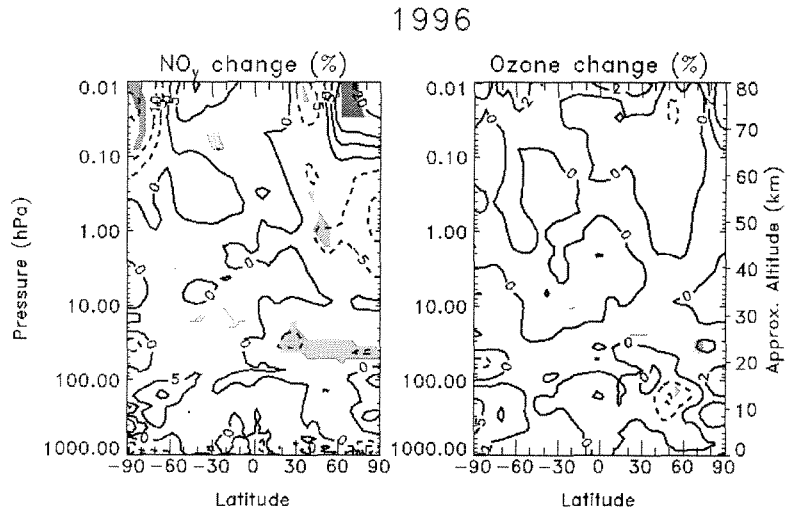
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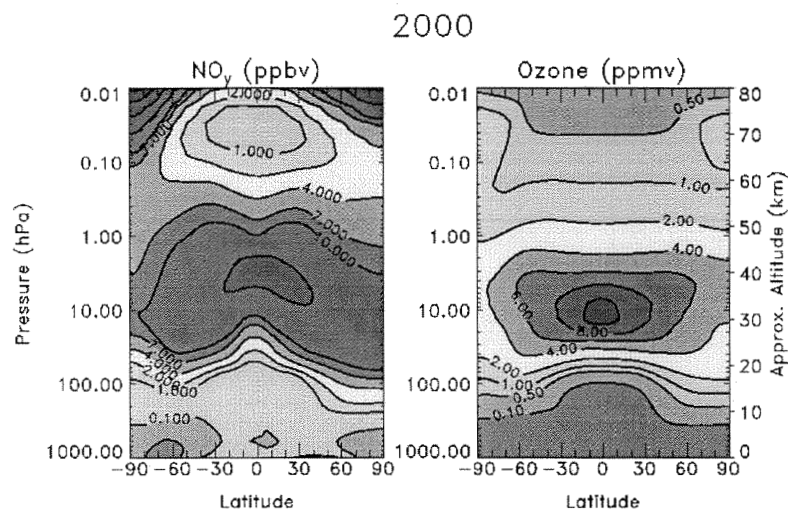


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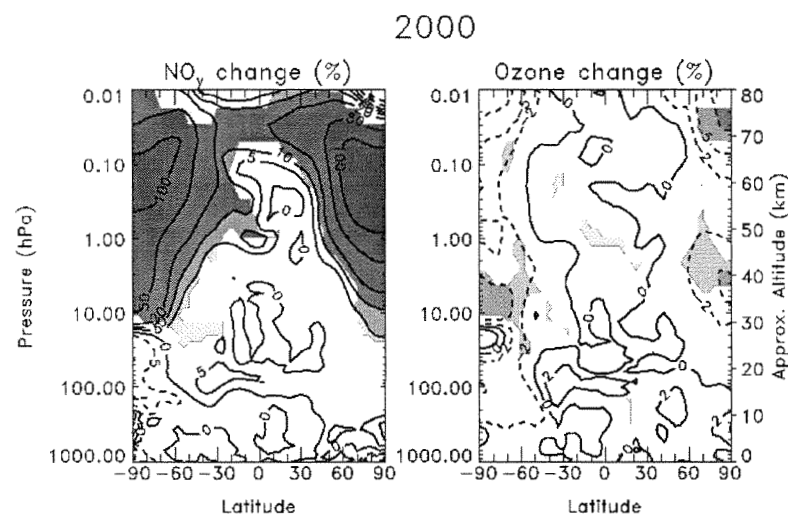


**Figure 4.** Annual average of solar proton event-caused change: Difference between average of simulations 1(a,b,c,d) and 1(w,x,y,z) for year 1996. **Left:**  $\text{NO}_y$  with contour intervals -20, -10, -5, 0, 5, 10, and 20%. **Right:** Ozone with contour intervals -5, -2, 0, 2, and 5%. Colored regions indicate 95% statistical significance with Student's t-test.



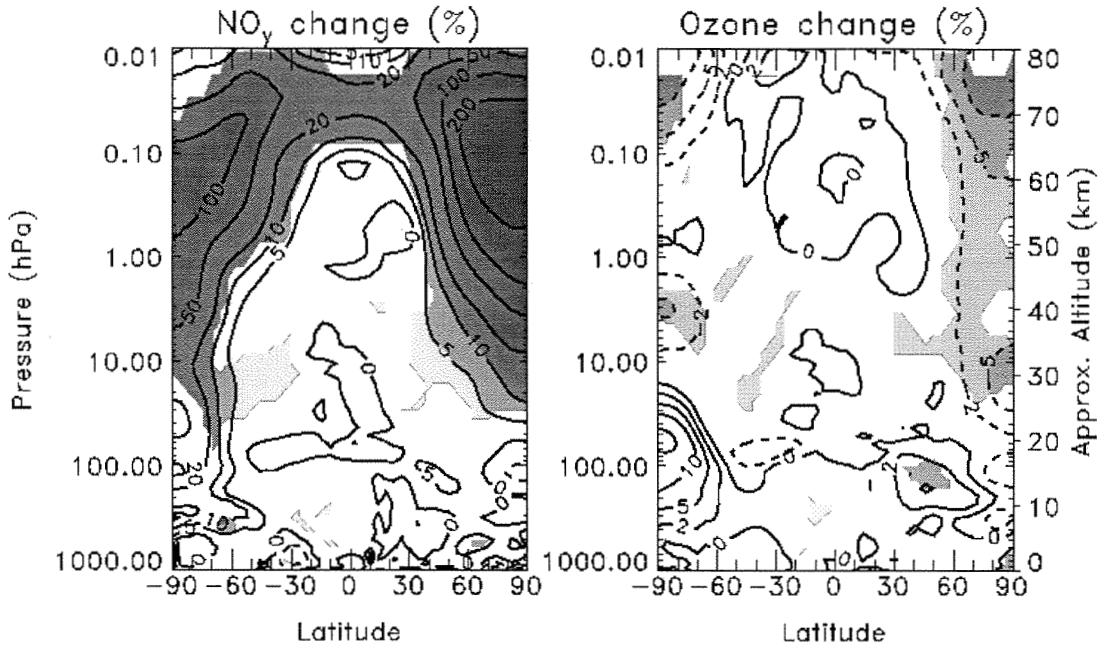


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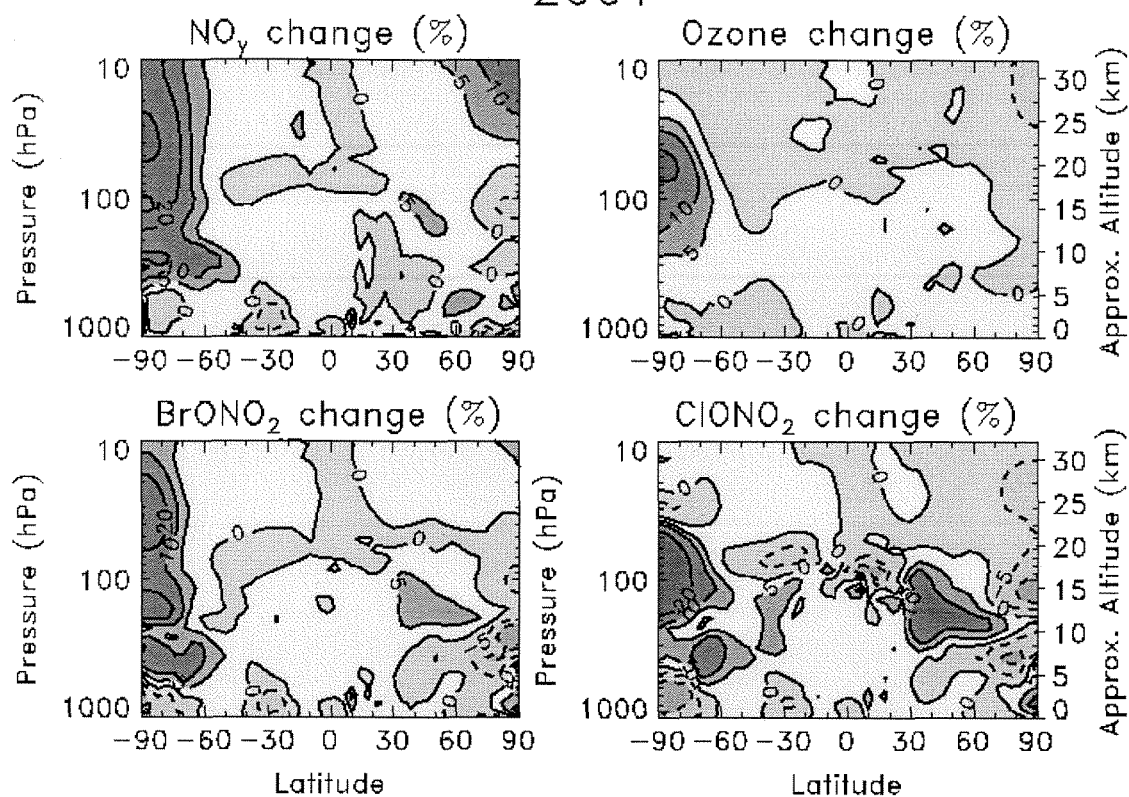
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2001



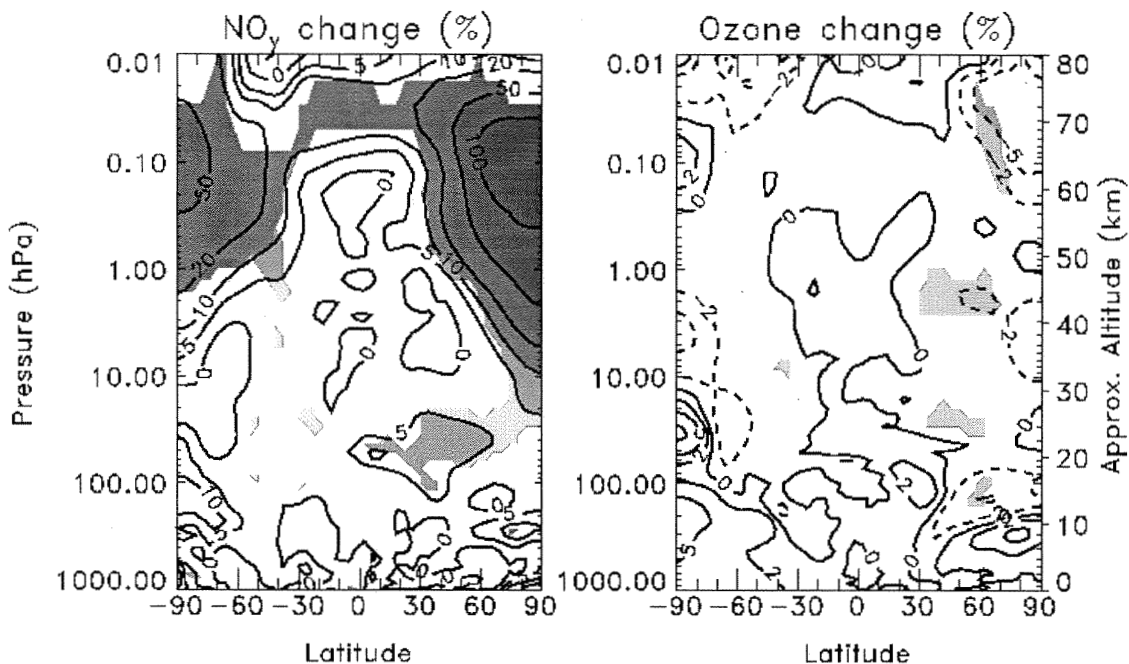
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2001

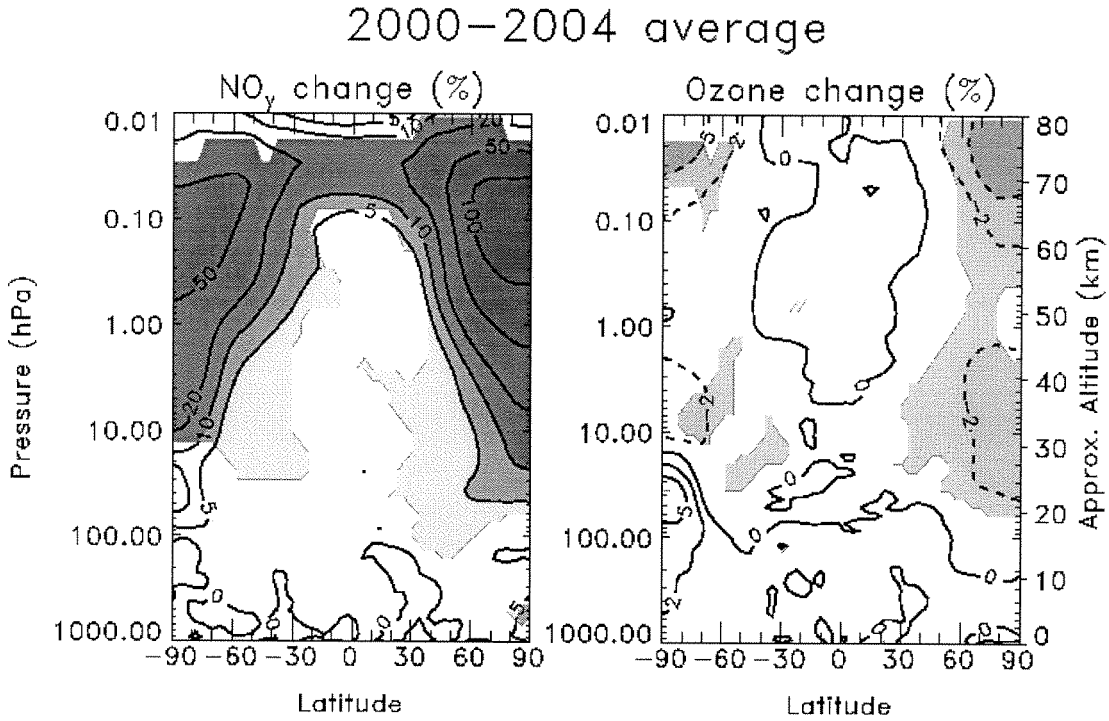


**Figure 8.** Changes caused by solar proton events for year 2001. Difference between average of simulations 1(a,b,c,d) and 1(w,x,y,z) – **Left, top:** NO<sub>y</sub> with contour intervals -5, 0, 5, 10, 20, and 50%; **Right, top:** Ozone with contour intervals -5, 0, 5, 10, and 20%; **Left, bottom:** BrONO<sub>2</sub> with contour intervals -10, -5, 0, 5, 10, 20, and 50%; **Right, bottom:** ClONO<sub>2</sub> with contour intervals -10, -5, 0, 5, 10, and 20%.

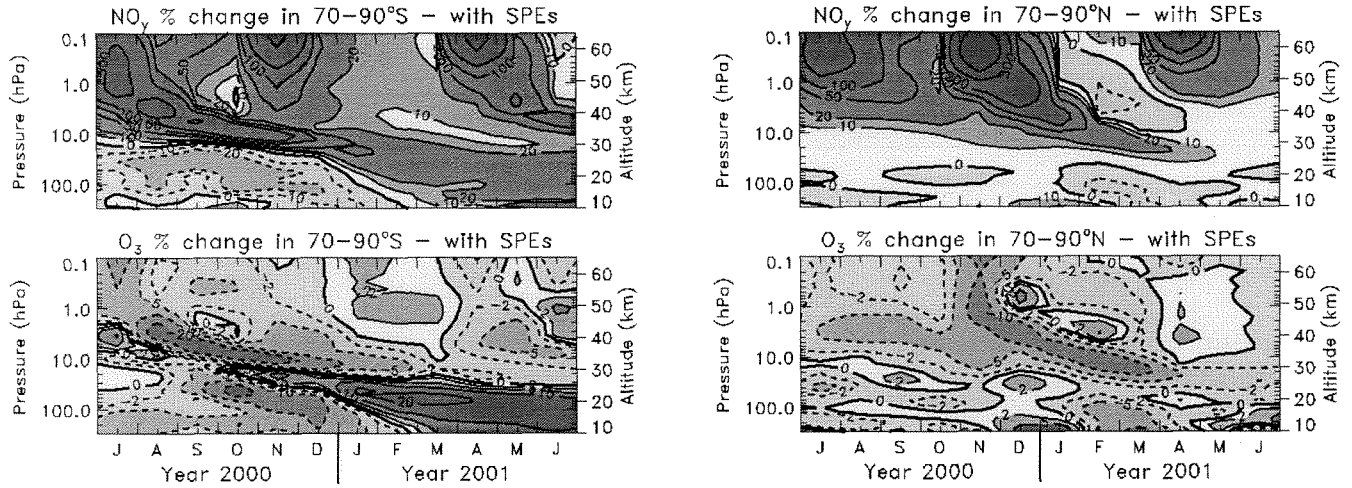
2003



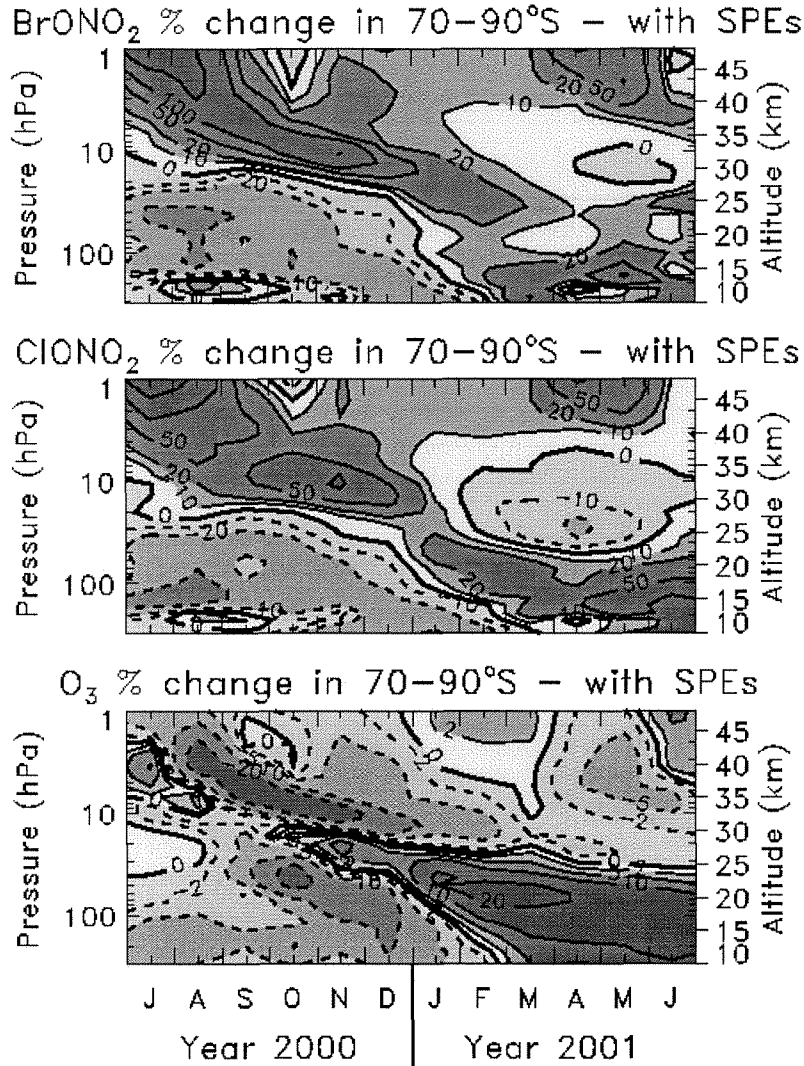
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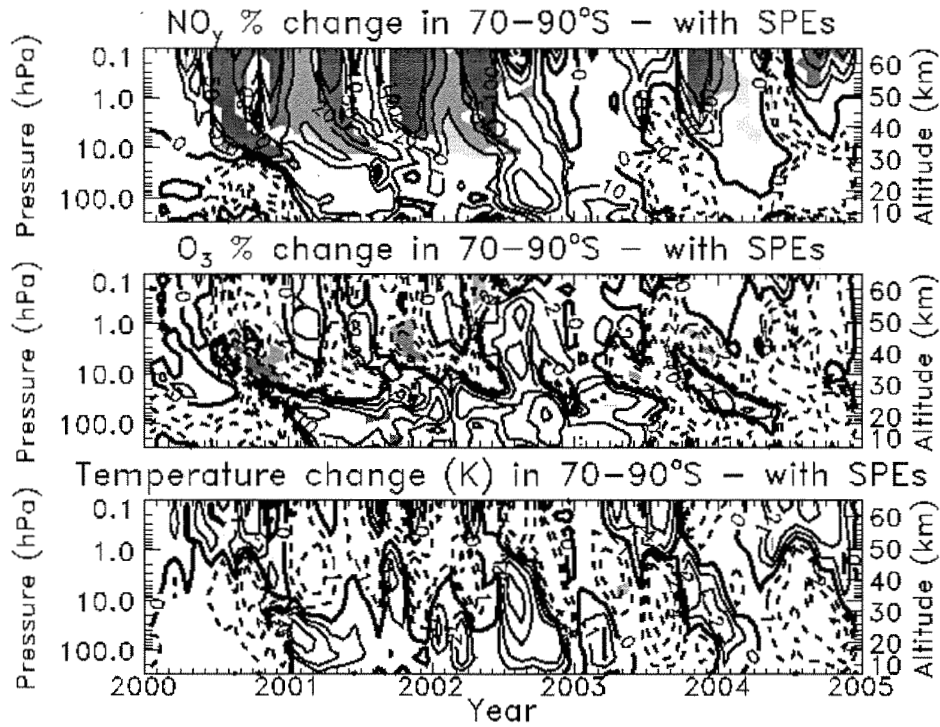
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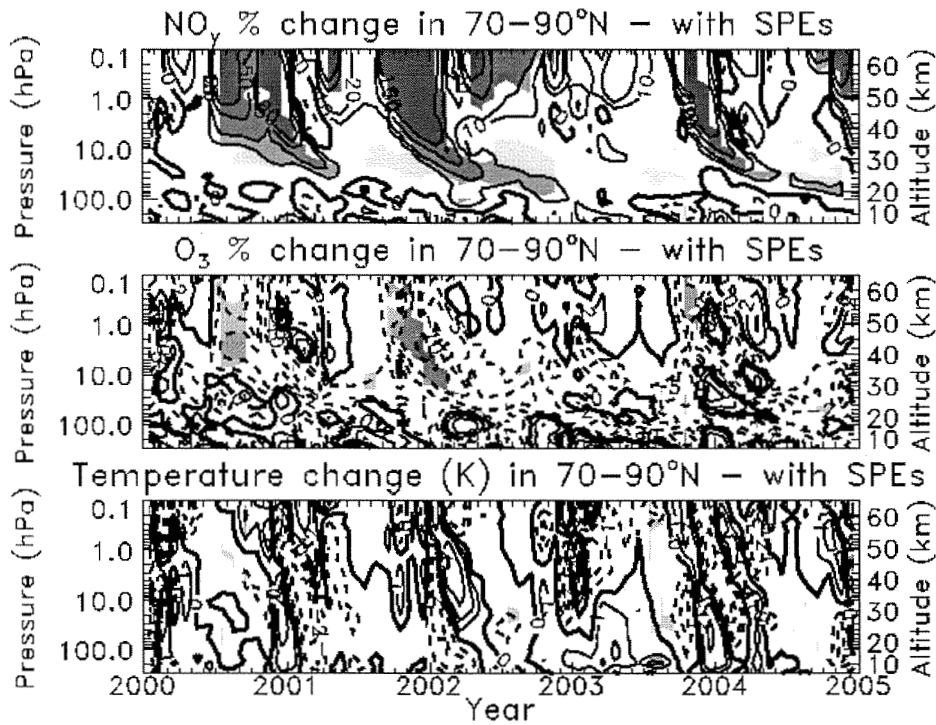


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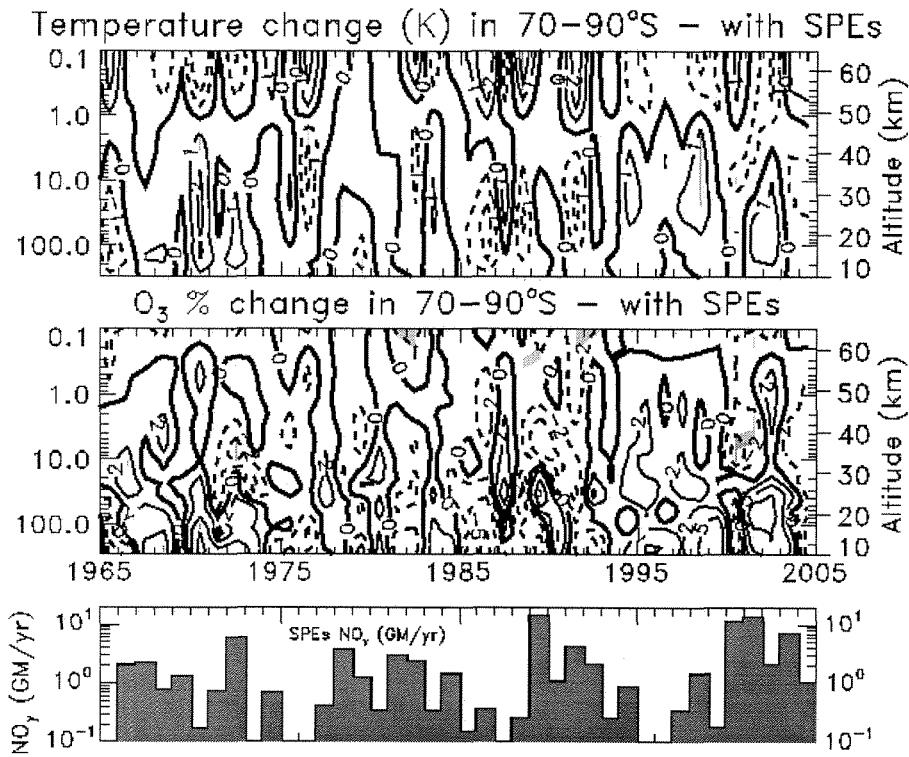


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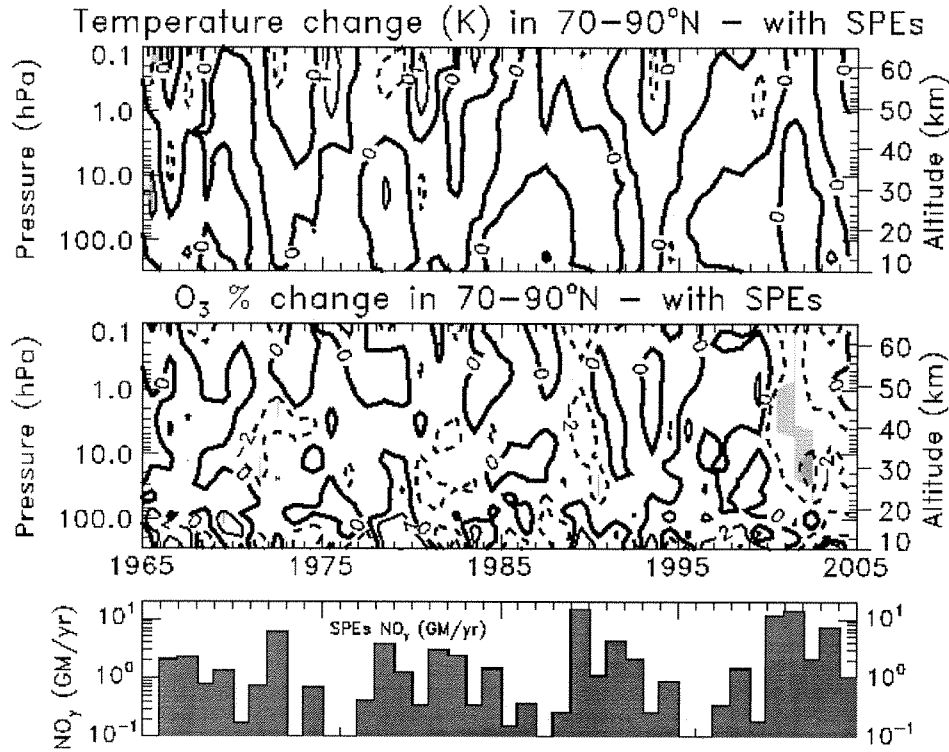




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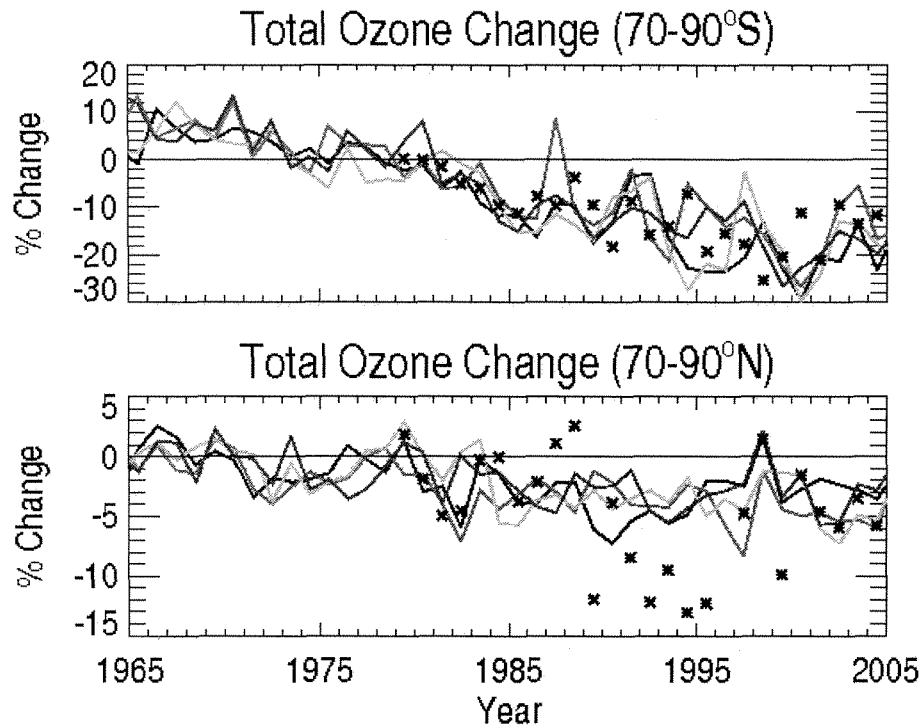
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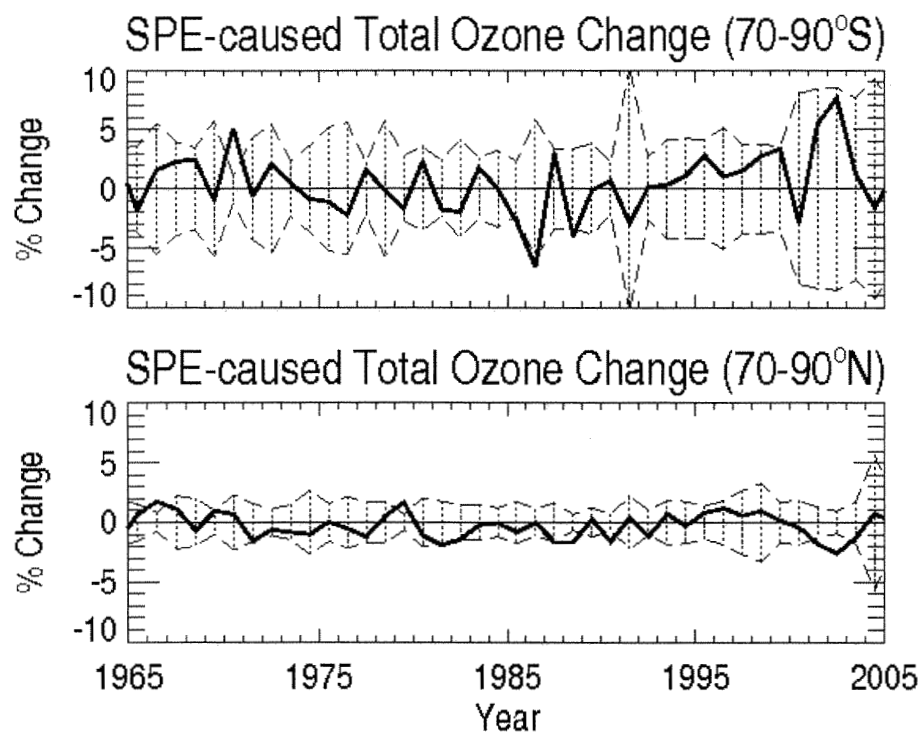
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Colored regions indicate 95% statistical significance with the use of Student's t-test. – **Top:**

Temperature with contour intervals -2, -1, 0, 1, and 2 K. **Middle:** Ozone with contour intervals -10, -5, -2, 0, 2, 5, and 10%. **Bottom:** NO<sub>y</sub> production per year (GigaMoles, GM) by solar proton events.



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