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Long-term middle atmospheric influence of very large solar proton events

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[1] The Whole Atmosphere Community Climate Model (WACCM3) has been used to study the long-term (more than a few months) effects of solar proton events (SPEs). Extremely large solar proton events occurred in 1972, 1989, 2000, 2001, and 2003 and caused some longer-lasting atmospheric changes. The highly energetic solar protons produced odd hydrogen (HO_x) and odd nitrogen (NO_y), which then led to ozone variations. Some statistically significant long-term effects on mesospheric ozone were caused by the HO_x increases due to a very active time period for SPEs (years 2000–2004), even though the HO_x increases were short-lived (days). The long-term stratospheric ozone effects were caused by the NO_y enhancements. Very large NO_y enhancements lasted for months in the middle and lower stratosphere after a few of the largest SPEs. SPE-caused 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 5-year period (2000–2004). WACCM3-computed SPE-caused polar stratospheric ozone decreases of >10% continued for up to 5 months past the largest events; however, statistically significant ozone decreases were computed for only a relatively small fraction of this time in relatively limited altitudes in the lower mesosphere and upper stratosphere. Annually averaged model output showed statistically significant (to 95%) stratospheric ozone loss in the polar Northern Hemisphere for years 2000–2002. The computed annually averaged temperature and total ozone change in these years were not statistically significant.

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1. Introduction

[2] 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 HO_x, 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; *Seppälä et al.*, 2004, 2006; *López-Puertas et al.*, 2005a, 2005b; *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].

[3] 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 important in understanding the anthropogenic impact on ozone [*World Meteorological Organization*, 2007]. The SPE-caused impacts are largest in the polar regions in the mesosphere and upper stratosphere. Quantifying the downward and equatorward transport of this SPE-induced perturbation is one of the main objectives of this paper.

[4] SPEs lead to ionization and the production of the important constituent families of HO_x (H, OH, HO_2) and NO_y ($N(^4S)$, $N(^2D)$, NO, NO_2, NO_3, N_2O_5, HNO_3, HO_2NO_2, $ClONO_2$, $BrONO_2$). The SPE-produced HO_x constituents are relatively short-lived (approximately days) and lead to the destruction of ozone in the upper stratosphere and mesosphere (pressures less than about 2 hPa).

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Both short- and longer-term (approximately months) catalytic ozone destruction is caused by the SPE-produced NO_y constituents in the lower mesosphere and stratosphere (pressures greater than about 0.5 hPa) via the well-known NO_x ($NO + NO_2$) ozone loss cycle

$$NO + O_3 \rightarrow NO_2 + O_2,$$
$$\text{followed by } NO_2 + O \rightarrow NO + O_2,$$
$$\text{Net } O + O_3 \rightarrow O_2 + O_2.$$

[5] Some modeling studies have addressed the longer-term atmospheric influence of SPEs [e.g., *Jackman et al.*, 1990, 1995, 2000, 2005a, 2005b; *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_x lower mesospheric enhancements observed by ACE (Atmospheric Chemistry Experiment) in mid-February 2004, discussed by *Rinsland et al.* [2005], and found that the October/November 2003 SPEs were a relatively minor contributor compared with the auroral NO_x source.

[6] We studied the short- and medium-term (days to a few months) atmospheric constituent effects of very large SPEs [see *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 longer-term (more than a 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.

[7] This paper is divided into six sections, including this introduction. The solar proton flux and SPE-induced production of HO_x and 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 (HO_x) and Odd Nitrogen (NO_y) Production

[8] 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 to 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 to 2004 [*Jackman et al.*, 2005b].

[9] The proton flux data from the satellites were used to compute daily average ion pair production profiles using the energy deposition methodology discussed by *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 by *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 ($\sim$1 km) and 8×10^{-5} hPa ($\sim$115 km) at the SOLARIS (Solar Influence for SPARC) website (http://www.geo.fu-berlin.de/en/met/ag/strat/forschung/SOLARIS/Input_data/index.html).

[10] Odd hydrogen (HO_x) is produced through complex positive ion chemistry [*Solomon et al.*, 1981] by the SPEs. The SPE-produced 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 HO_x constituents have a relatively short lifetime (approximately hours) throughout most of the mesosphere, however, the ozone depletion can be very large during substantial SPEs [e.g., *Solomon et al.*, 1983; *Jackman et al.*, 2001; *Verronen et al.*, 2006] and thus do not influence the stratosphere on a long-term (more than a few months) time period. *Jackman et al.* [2007] calculated the ozone depletion and dynamics change from solar proton enhanced HO_x constituents with the Thermosphere Ionosphere Mesosphere Electrodynamic General Circulation Model (TIME-GCM). They found that mesospheric temperature and wind perturbations from SPE-produced HO_x were greatly diminished in just 4–6 weeks.

[11] Odd nitrogen is produced when the energetic charged particles (protons and associated secondary electrons) dissociate N_2. We assume that $\sim$1.25 N atoms are produced per ion pair and divide the proton impact of N atom production between ground state ($\sim$45% or $\sim$0.55 per ion pair) and excited state ($\sim$55% or $\sim$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 $N(^4S)$ per ion pair and 0.7 $N(^2D)$ atoms per ion pair.

[12] The period 1 January 1995 through 31 December 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 NO_y production from SPEs in the stratosphere and mesosphere in this time period. Although the solar UV-induced oxidation of nitrous oxide ($N_2O + O(^1D) \rightarrow NO + NO$) provides the largest source of NO_y in the middle atmosphere (52–58 gigamoles per year [*Vitt and Jackman*, 1996]), the SPE source of 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 NO_y due to N_2O oxidation. Table 1 shows the daily NO_y production from SPEs during the largest periods of proton fluxes in the 10-year period 1995–2004. SPE-produced NO_y greater than 1 gigamole was computed for 14–15 July 2000, 9 November 2000, 25–26 September 2001, 5–6 November 2001, 24 November 2001, and 29 October 2003.

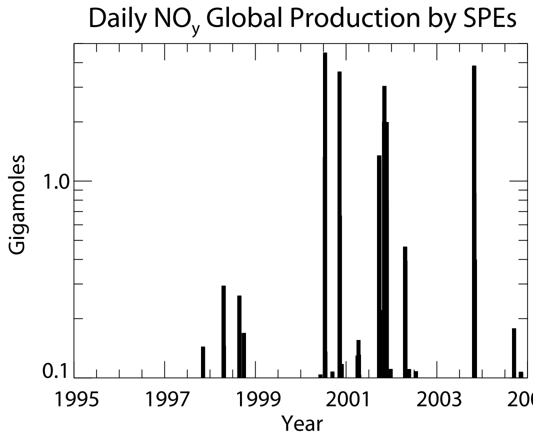

Figure 1. Daily column NO_y production in gigamoles (6.02×10^{32} molecules) as a function of time for 1 January 1995 to 31 December 2004.

[13] If the SPE-produced 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 several months or more. Downward transport of NO_y occurs mainly in the fall and winter time periods (October–November–December–January–February–March (ONDJFM) in the Northern Hemisphere; April–May–June–July–August–September (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)

[14] 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; *Kinnison et al.*, 2007; *Garcia et al.*, 2007; *Marsh et al.*, 2007; *Jackman et al.*, 2008]. The model has a domain from the surface to 4.5×10^{-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° and 5°, respectively. A more comprehensive description of WACCM3 is given by *Kinnison et al.* [2007].

[15] 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°N–90°N and 60°S–90°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. Any drift in the magnetic poles during the 1963–2004 period was not included in WACCM3 computations as the effects were likely minimal.

[16] 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, 1 January 1963 to 31 December 2004 (see Table 2). The ensembles of four simulations each [1(a,b,c,d) and 1(w,x,y,z)] are groups of model computations with identical boundary conditions, but slightly different initial conditions. The ensemble was created by taking four different Januaries from a perpetual simulation of year 1950. The simulations were essentially all originally started in January 1950 and run up to 1963, which is the starting year for all model computations shown in this paper. Simulations 1(w) and 1(a) have the same starting conditions, except simulation 1(w) is "without SPEs" and simulation 1(a) is

Table 1. Largest Daily NO_y Production From Solar Proton Events Between 1 January 1995 and 31 December 2004, Given in Chronological Order[a]

Date of SPE	Computed NO_y Daily Production In Middle Atmosphere (gigamoles[b])
14 July 2000	1.3
15 July 2000	4.5
9 November 2000	3.6[c]
10 November 2000	0.7[c]
25 September 2001	1.4
26 September 2001	1.3
5 November 2001	2.0
6 November 2001	3.0
23 November 2001	0.8
24 November 2001	2.0
21 April 2002	0.5
22 April 2002	0.4
28 October 2003	0.8
29 October 2003	3.9
30 October 2003	0.9
3 November 2003	0.4

[a]Only days with production greater than or equal to 0.4 gigamoles of NO_y are shown.
[b]Gigamole = 6.02×10^{32} atoms and molecules.
[c]The numbers reported for the November 2000 SPE add up to 4.3 gigamoles (GM) (2.6×10^{33} molecules), which is 0.5 GM larger than reported by *Jackman et al.* [2008], owing to different temporal averaging of the proton fluxes.

Table 2. Description of WACCM3 Simulations

Simulation Designation	Number of Realizations	Time Period	SPEs Included
1 (a, b, c, d)	4	1963–2004	Yes
1 (w, x, y, z)	4	1963–2004	No

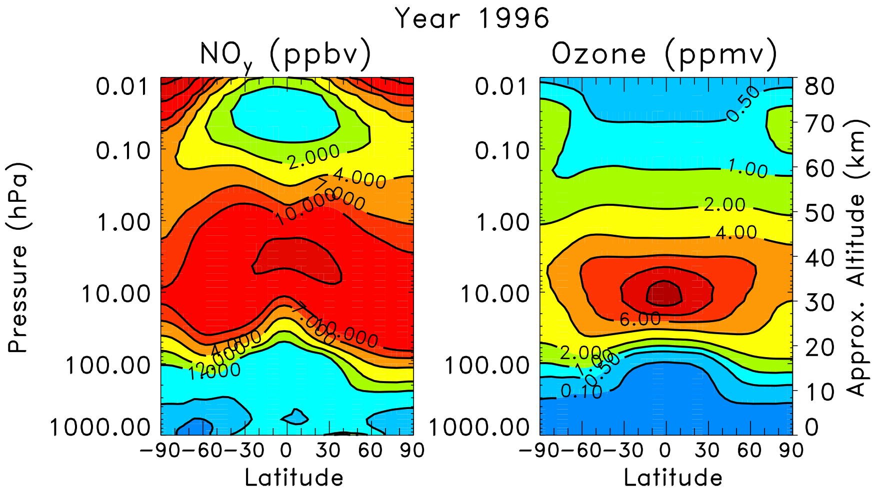

Figure 2. Annual mean of simulations "without SPEs" [1(w,x,y,z)] for year 1996. (left) NO_y with contour intervals 0.01, 0.1, 1, 2, 4, 7, 10, 20, and 40 ppbv. (right) Ozone with contour intervals 0.1, 0.5, 1, 2, 4, 6, 8, and 10 ppmv.

"with SPEs." Similar comments apply to paired simulations 1(x) and 1(b); 1(y) and 1(c); and 1(z) and 1(d).

4. SPE-Caused Long-Term Atmospheric Changes in Solar Cycle 23

[17] The influence of very large SPEs in the first 9 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, 2005b, 2008; *Jackman and McPeters*, 2004; *Jackman and Fleming*, 2008; *Seppälä et al.*, 2004; *Degenstein et al.*, 2005; *López-Puertas et al.*, 2005a, 2005b; *Orsolini et al.*, 2005; *von Clarmann et al.*, 2005; *Rohen et al.*, 2005].

[18] Some of the SPE-caused atmospheric perturbations were substantial during and shortly after very large SPEs. The October/November 2003 SPE period, known as the Halloween storms, caused NO_x enhancements measured by UARS HALOE that were reasonably well simulated in WACCM3 [*Jackman et al.*, 2008]. These NO_x increases were observed during and for a week past the SPEs. The changes in other nitrogen-containing gases during and shortly after these SPEs were not simulated as well, when compared to Envisat MIPAS measurements. Although qualitatively similar, WACCM3 predicted HNO_3 and $ClONO_2$ enhancements were smaller than measured and N_2O_5 enhancements were larger than measured. It is possible that particular ion chemistry reactions, not currently included in WACCM3, are responsible for such model/measurement disagreements.

[19] There is an indication that much of the longer-lived enhancement in nitrogen-containing constituents is in the form of NO_x several weeks after very large SPEs. For instance, *Randall et al.* [2001] showed evidence from HALOE observations of enhancements of about 15 ppbv in NO_x in the polar middle stratosphere two months after the July 2000 SPE in the Southern Hemisphere (SH). The NO_x increases in September/October 2000 are about a factor of 2–3 beyond the normal range in the polar middle stratosphere for years 1991–1999. We also computed NO_x enhancements in September/October 2000 with WACCM3 (simulation 1(a)) compared with years 1991–1999 [*Jackman et al.*, 2008, Figure 15], which were in reasonable agreement with HALOE observations. Similar results were found for WACCM3 simulations 1(b,c,d). This measurement/model agreement suggested that the very large NO_x peak in the year 2000 was caused by the July 2000 SPE.

[20] This measurement/model NO_x agreement in September/October 2000 also suggest that the measurement/model disagreements in HNO_3, $ClONO_2$, and N_2O_5, discussed earlier for the October/November 2003 SPEs, are relatively short-lived and last only a couple of weeks past SPEs. 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 6 months past the events?

4.1. SPE-Caused Annually Averaged Constituent Changes

[21] We focus most of our analyses on NO_y and ozone. As discussed before, the 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 NO_y family. We show the WACCM3-computed annual mean NO_y and ozone distribution in Figure 2 from the ensemble "without SPEs" [1(w, x, y, z)] for 1996, which is near solar minimum. As discussed by *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 NO_x being too low at this altitude by about 15% [*Eyring et al.*, 2006]. Both NO_y and ozone show peaks in the tropics with the NO_y maximum being about 5 km higher in altitude. Near the top of the altitude domain in Figure 2 (left), the NO_y clearly shows descent of NO_y-rich air from higher altitudes in both hemi-

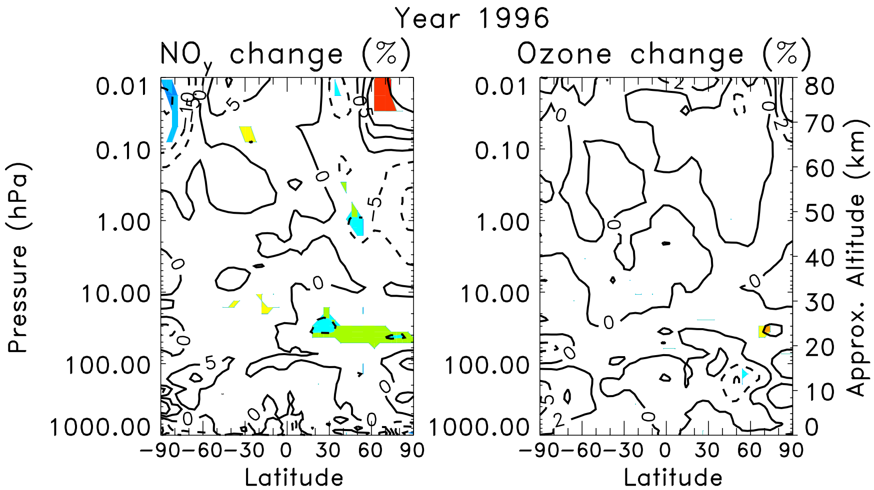

Figure 3. Annual average of solar proton event-caused change: Difference between average of simulations "with SPEs" [1(a,b,c,d)] and "without SPEs" [1(w,x,y,z)] for year 1996. (left) NO_y with contour intervals −20, −10, −5, 0, 5, 10, and 20%. (right) Ozone with contour intervals −5, −2, 0, 2, and 5%. Colored regions indicate 95% statistical significance with Student's *t* test.

spheres to the middle mesosphere (~0.04 hPa, 70 km) and lower.

[22] The annual zonal average computed change for 1996 caused by SPEs for NO_y and ozone is shown in Figure 3 as the percentage difference between the ensemble averages of simulations "with SPEs" [1(a,b,c,d)] and "without SPEs" [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 ±20% changes computed for 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 NO_x to the Southern Hemisphere stratosphere occurred during 1996. Also, virtually none of the computed ozone change is statistically significant.

[23] The annual zonal mean mesospheric NO_y for year 2000 from the ensemble "without SPEs" [1(w, x, y, z)] is somewhat larger than that computed for 1996. Compared with 1996, polar Southern Hemisphere NO_y was increased in 2000 by 120% near 0.01 hPa tapering to less than 5% by 0.3 hPa (not shown). Similarly, polar Northern Hemisphere NO_y was increased in 2000 beyond 1996 levels by 70% near 0.01 hPa tapering to 5% by 0.1 hPa (not shown). This larger source of NO_y in 2000 was caused by increased geomagnetic activity and high-energy photons near solar maximum. The SPEs further increased this already-enhanced NO_y in 2000, as described below.

[24] The computed annual zonal average change for 2000 caused by SPEs for NO_y and ozone is shown in Figure 4 (top) as the percentage difference between ensemble averages of simulations "with SPEs" [1(a,b,c,d)] and "without SPEs" [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 NO_y in the polar middle to lower mesosphere (Figure 4, top left) and a good portion of the computed polar NO_y enhancements

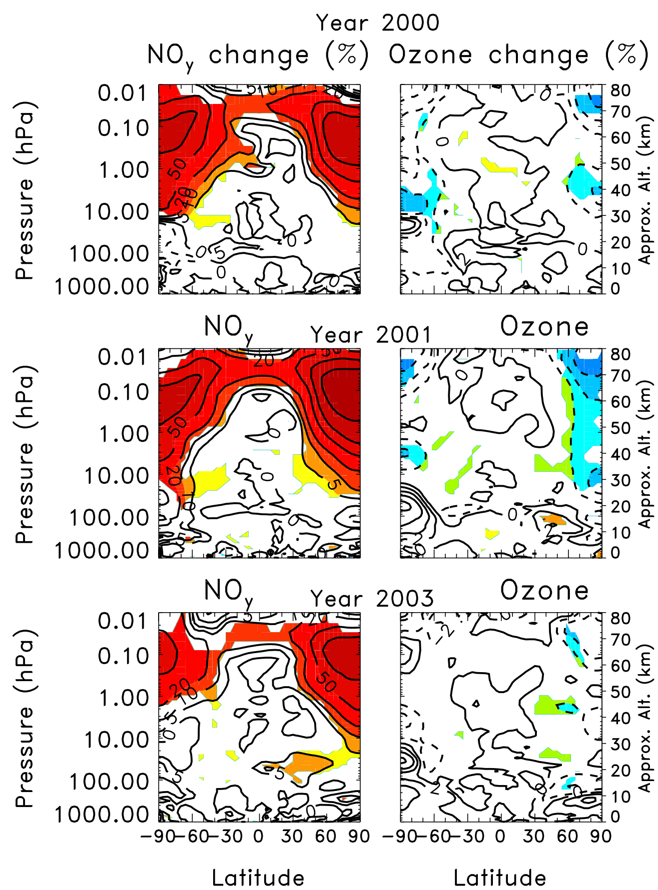

Figure 4. Annual average of changes caused by solar proton events for years (top) 2000, (middle) 2001, and (bottom) 2003: difference between average of simulations "with SPEs" [1(a,b,c,d)] and "without SPEs" [1(w,x,y,z)]. (left) NO_y with contour intervals −20, −10, −5, 0, 5, 10, 20, 50, 100, and 200%. (right) Ozone with contour intervals −10, −5, −2, 0, 2, 5, 10, and 20%. Colored regions indicate 95% statistical significance with Student's *t* test.

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 NO_y. Unlike 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$3 hPa) and in the NH upper stratosphere ($\sim$4 to $\sim$1 hPa). Even smaller regions in the mesosphere show a statistically significant ozone decrease (Figure 4, top right).

[25] 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 NO_y in regions of the polar middle atmosphere in both hemispheres (Figure 4, middle). The colored regions in the plots indicate 95% significance using Student's *t* test. A large portion of the computed polar 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.

[26] 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_x 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_x resulted in annual average NH polar ozone change from about -1% to -4%. The computed mesospheric ozone changes from SPE-produced NO_y were from near 0% to -1%. Thus the huge HO_x-caused ozone losses on SPE-active days in year 2001 [e.g., *Krivolutsky et al.*, 2003] did contribute to the overall annual average polar mesospheric ozone change and helped result in a statistically significant NH ozone signal.

[27] 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 (Figure 4, middle 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_y increases has been discussed before [*Jackman et al.*, 2000; *Jackman and Fleming*, 2008] and is related to the interference by NO_y constituents with the halogen (chlorine and bromine) catalytic cycles (discussed further in section 4.2).

[28] The annual zonal average change for the next 3 years (2002–2004) caused by SPEs for NO_y and ozone was computed, again as the percentage difference between the ensemble averages of simulations "with SPEs" [1(a,b,c,d)] and "without SPEs" [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 NO_y enhancement (not shown). Still, a good portion of the polar middle atmosphere had a statistically significant 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).

[29] 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 NO_y in the polar NH mesosphere and large maximum increases (>50%) were computed for NO_y in the polar SH mesosphere as a result of the SPEs (Figure 4, bottom left). A large portion of the computed polar NH NO_y enhancements from 30 to 0.01 hPa were statistically significant, whereas a more modest portion of the computed SH NO_y 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 (Figure 4, bottom right).

[30] Year 2004 was relatively quiet with no large SPEs. Modest statistically significant annual zonal average NO_y enhancements were computed in the polar mesosphere for both hemispheres and a statistically significant NO_y 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_y 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).

[31] 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 5-year average 2000–2004. The computed changes produced by SPEs for NO_y and ozone are shown in Figure 5 as the percentage difference between ensemble averages of simulations "with SPEs" [1(a,b,c,d)] and "without SPEs" [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_y in the polar NH and large maximum increases (>50%) were computed for the polar SH as a result of the SPEs (Figure 5, left). Very large regions of the computed polar NO_y 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_y 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 (Figure 5, right), which is related to the larger late fall and winter NO_y input. The statistically significant mesospheric NH ozone change was mainly caused by the SPE-produced HO_x.

4.2. SPE-Caused Polar Atmospheric Changes

[32] SPEs initially impact the polar cap regions (60°–90° geomagnetic latitude) of both hemispheres. The influence over longer periods beyond that initial disturbance is dependent on the amount of 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 NO_x. We examine

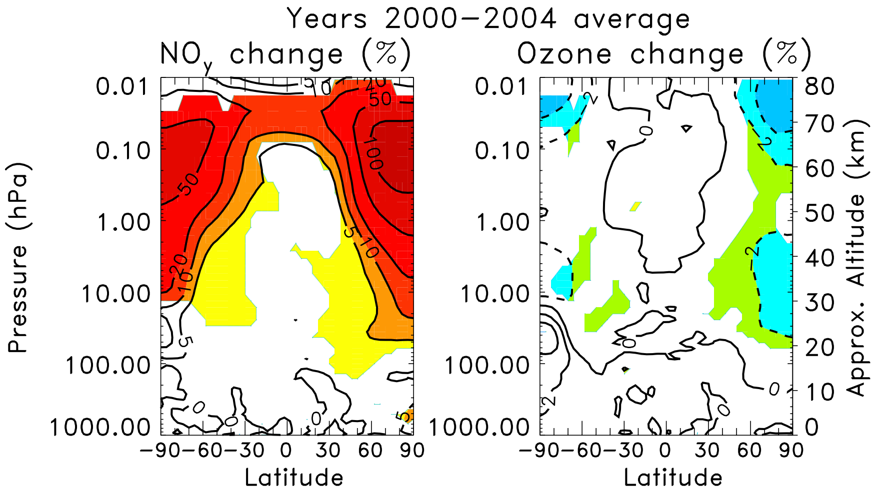

Figure 5. Solar proton event-caused change for average of years 2000–2004: difference between average of simulations "with SPEs" [1(a,b,c,d)] and "without SPEs" [1(w,x,y,z)]. (left) NO_y with contour intervals 0, 5, 10, 20, 50, and 100%. (right) Ozone with contour intervals −5, −2, 0, 2, and 5%. Colored regions indicate 95% statistical significance with Student's *t* test.

the WACCM3 SPE-caused changes at high latitudes in this section.

[33] We have investigated the polar changes over a particularly active 1-year period, 1 July 2000 to 30 June 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 NO_y and ozone in the latitude bands 70°S–90°S and 70°N–90°N are given in Figure 6. We use the monthly average output for the WACCM3 simulations and present the percentage difference between the ensemble averages of simulations "with SPEs" [1(a,b,c,d)] and "without SPEs" [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 *Jackman et al.*, 2008, Table 1], 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 NO_y enhancement in the 70°S–90°S band is enormous for the July 2000 SPE with maximum increases >500% (Figure 6, top left).

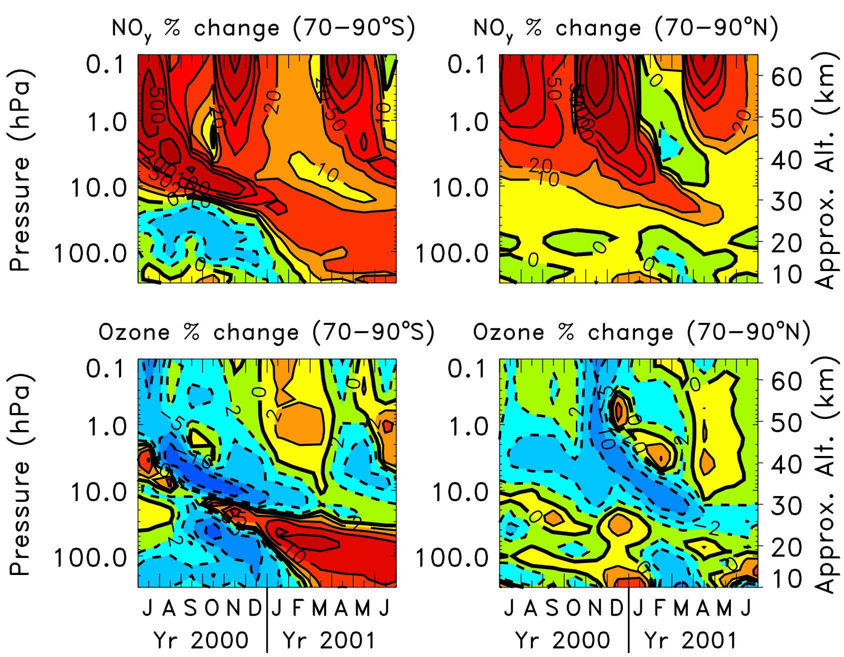

Figure 6. Solar proton event-caused change for July 2000 to June 2001 (left) in the SH polar latitude interval 70°S–90°S and (right) in the NH polar latitude interval 70°N–90°N: difference between average of simulations "with SPEs" [1(a,b,c,d)] and "without SPEs" [1(w,x,y,z)] for (top) NO_y with contour intervals −20, −10, 0, 10, 20, 50, 100, 200, 500, and 1000% and (bottom) ozone with contour intervals −20, −10, −5, −2, 0, 2, 5, 10, and 20%.

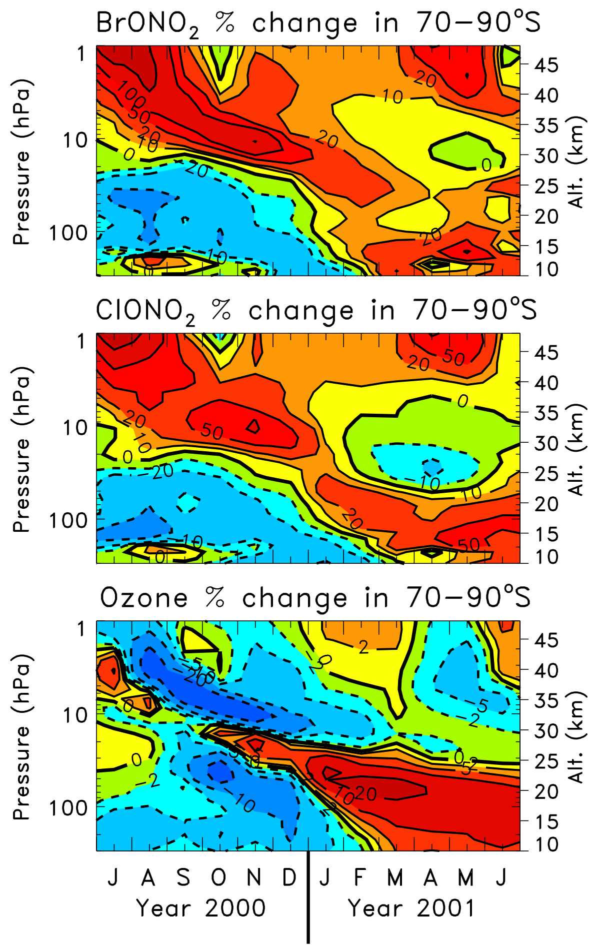

Figure 7. Solar proton event-caused change for July 2000 to June 2001 in the SH polar latitude interval 70°S–90°S: difference between average of simulations "with SPEs" [1(a,b,c,d)] and "without SPEs" [1(w,x,y,z)] – for (top) $BrONO_2$ with contour intervals −50, −20, −10, 0, 10, 20, 50, 100, and 200%; (middle) $ClONO_2$ with contour intervals −50, −20, −10, 0, 10, 20, 50, 100, and 200%; and (bottom) ozone with contour intervals −20, −10, −5, −2, 0, 2, 5, 10, and 20%.

This increase in NO_y, an indicator of the very large SPE, is transported slowly and steadily downward throughout the time period. The levels of increased NO_y were reduced owing to mixing with smaller amounts of NO_y from lower latitudes and altitudes, eventually reaching levels of just over 20% NO_y enhancement in the lower stratosphere (∼300 to 40 hPa) by 30 June 2001. The November 2000 SPE appears to have added somewhat to this huge NO_y SPE-caused signal in late 2000. The April 2001 SPE caused significant changes in NO_y, but its impact by 30 June 2001 was located in the middle to upper stratosphere (∼10 to 2 hPa).

[34] The longer-term computed ozone decrease connected with the July 2000 SPE is substantial, reaching levels >20% in the middle to upper stratosphere (∼10 to 2 hPa) between August and November, 2000 (Figure 6, bottom left). Note that we use WACCM3 monthly average output in these analyses, thus the much larger computed ozone decreases presented by *Jackman et al.* [2008] were not calculated. The larger computed ozone decreases of *Jackman et al.* [2008] were driven by the short-lived HO_x constituents and did not last beyond a couple of days after the SPEs. In the later part of this period (October 2000 to June 2001), ozone increases below ∼10 hPa were computed; these are presumably due to the fact that enhanced NO_y sequesters chlorine and bromine in the reservoir species ($ClONO_2$ and $BrONO_2$), resulting in reduced ozone loss in this region of the stratosphere. The close correlation between the enhancements of $ClONO_2$ and $BrONO_2$ and the enhanced lower stratospheric ozone is shown clearly in Figure 7. Both $ClONO_2$ and $BrONO_2$ are enhanced >10% throughout most of the region where the ozone is increased >10% starting in January 2001. The large enhancements in SH polar NO_y (Figure 6, top left) lead to increases in $ClONO_2$ (Figure 7, middle) and $BrONO_2$ (Figure 7, top) through the reactions

$$ClO + NO_2 + M \rightarrow ClONO_2 + M$$

$$BrO + NO_2 + M \rightarrow BrONO_2 + M.$$

[35] The chlorine and bromine reservoir species ($ClONO_2$ and $BrONO_2$) are produced at the expense of the reactive species (ClO and BrO) that drive the ClO_x and 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 at pressures greater than 20 hPa, especially after January 2001 (Figure 7, bottom).

[36] The computed SPE-caused NO_y enhancement in the latitude band 70°N–90°N is larger for the November 2000 SPE than for the July 2000 SPE with maximum increases >1000% (Figure 6, top right). The July 2000 SPE NH NO_y enhancement was substantially reduced owing to the summer sunlight increasing the loss process for NO_y via

$$NO + h\nu(<191\ \mathrm{nm}) \rightarrow N + O$$

followed by

$$N + NO \rightarrow N_2 + O.$$

[37] The levels of increased NO_y due to the November 2000 SPE were reduced owing to mixing with smaller amounts of NO_y from lower latitudes and altitudes, eventually reaching levels less than 10% NO_y enhancement in the middle stratosphere (∼40 to 10 hPa) by May 2001 (Figure 6, top right). The July 2000 and April 2001 SPEs caused significant changes in NO_y, but their impacts are primarily confined to pressures <10 hPa.

[38] 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 downward to the middle stratosphere and gradually diminished with decreases of <5% by the end of April 2001 (Figure 6, bottom right).

[39] We have also computed the polar changes over the longer period, 1 January 2000 to 31 December 2004, which included several very large SPEs (see Table 1). The changes caused by SPEs for NO_y, ozone, and temperature in the latitude band 70°S–90°S are given in Figure 8. Very large

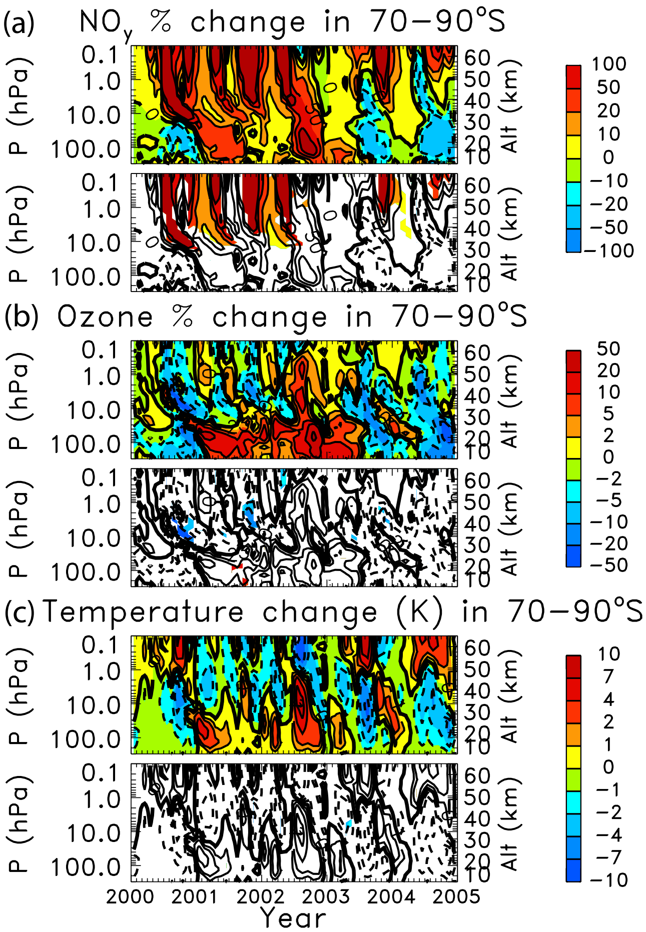

Figure 8. Solar proton event-caused change for the 5 years 2000–2004 in the SH polar latitude interval 70–90°S: Difference between simulations "with SPEs" [1(a,b,c,d)] and "without SPEs" [1(w,x,y,z)]. (a) NO_y change (%). (b) Ozone change (%). (c) Temperature change (K). The bottom plot in each part is the upper plot repeated with the colored areas indicating regions which are 95% statistically significant with the use of Student's *t* test.

enhancements of NO_y (Figure 8a) extending from the mesosphere to the middle stratosphere are apparent in 4 of the 5 years (e.g., 2000, 2001, 2002, and 2003). The 2 years 2000 and 2002 show the deepest penetration of the enhanced 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 (Figure 8b) follows the NO_y signal into the atmosphere and is generally anticorrelated with NO_y, except in the lower stratosphere (see Figure 7).

[40] 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 1 K nearly 3 months after the very large SPE of October 1989.

[41] The SH polar temperature stratospheric signal (Figure 8c) 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.

[42] The colored regions in the bottom plot of each part in Figure 8 indicate 95% statistical significance with the use of Student's *t* test. Much of the computed NO_y 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_y enhancement after May 2002 is statistically significant (Figure 8a, bottom). The areas of statistically significant ozone changes are much less than those for NO_y enhancements (Figure 8b, bottom). Four months in late 2000 (September–December) in the middle stratosphere, 3 months in late 2001 (October–December) in the upper stratosphere, and a few other small regions are statistically significant. Virtually none of the computed temperature changes are statistically significant (Figure 8c, bottom).

[43] The changes caused by SPEs for NO_y, ozone, and temperature in the latitude band 70°N–90°N are given in Figure 9. Very large tongues of SPE-produced NO_y

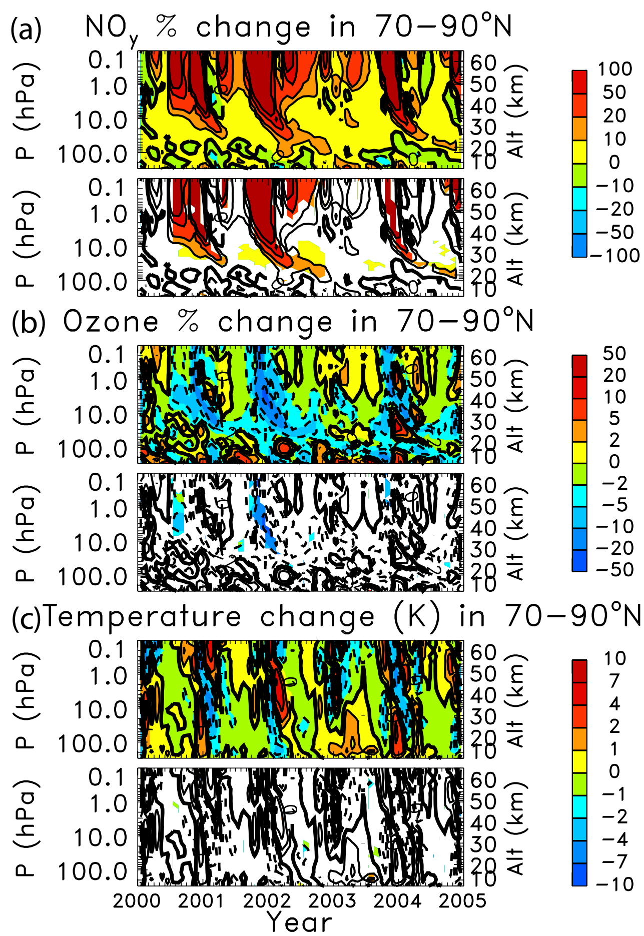

Figure 9. Same as Figure 8, but for the NH.

enhancement (Figure 9a) extending from the mesosphere to the middle stratosphere are apparent in three time periods: (1) November 2000 to April 2001; (2) November 2001 to December 2002; and (3) October 2003 to November 2004. The very large SPEs in November 2000, November 2001, and October 2003, and the large SPE in November 2003 drove most of these NO_y increases because they occurred during the fall season (see Table 1). These WACCM3 results are in line with our earlier study on the influence of SPEs on the middle atmosphere in years 2000–2003 using the Goddard Space Flight Center (GSFC) two-dimensional (2D) model [*Jackman et al.*, 2005b]. Both models show good qualitative agreement with the SPE-produced NO_y being transported to lower altitudes over several months, especially during the late fall and winter months.

[44] We compute a modest enhanced amount of upper stratospheric NO_y (5–15 ppbv) in the 2004 late winter and spring, which was caused by the October/November 2003 SPEs. These results support the conclusions of *Semeniuk et al.* [2005], who found that the huge upper stratospheric NO_y enhancements (>60 ppbv) observed by satellite instruments in mid-February to April 2004 [*Natarajan et al.*, 2004; *Randall et al.*, 2005; *Rinsland et al.*, 2005], were not caused by these SPEs. Rather, these measured upper stratospheric NO_y increases were likely a result of the intense aurorae associated with the October/November 2003 solar storms, which produced very large amounts of thermospheric NO_y that was subsequently transported to the stratosphere.

[45] The NH WACCM3 polar ozone signal (Figure 9b) follows the NO_y signal into the stratosphere and is anticorrelated with NO_y. Again, both WACCM3 and the *Jackman et al.* [2005b] SPE-caused ozone impact are fairly similar. The NH WACCM3 polar temperature changes (Figure 9c) 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.

[46] The colored regions in the bottom plot of each part in Figure 9 indicate 95% statistical significance with the use of Student's *t* test. Large regions of the computed SPE-caused NO_y enhancements are statistically significant. These statistically significant NO_y increases can extend from the mesosphere to the middle stratosphere (from 0.1 to $\sim$10 hPa; Figure 9a, bottom). As in the polar SH, the areas of statistically significant ozone changes are much smaller than those for NO_y enhancements (Figure 9b, bottom). A summer SPE (July 2000) helped contribute to nearly four months (July–October 2000) of statistically significant ozone decrease in the upper stratosphere. The furthest penetrating statistically significant ozone decrease occurred between September 2001 and January 2002, when the ozone signal followed the NO_y enhancement from the lower mesosphere/upper stratosphere down to the middle stratosphere. The large SPE in September 2001 and the two very large SPEs in November 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 December 2003. As in the SH, virtually none of the computed NH temperature changes are statistically significant (Figure 9c, bottom).

5. SPE-Caused Long-Term Polar Atmospheric Changes in the 1963–2004 Period

[47] The very large SPEs in solar cycle 23 caused fairly substantial statistically significant polar NO_y enhancements in both hemispheres in the period 1 January 2000 through 31 December 2004. Although the computed statistically significant ozone decreases covered much less of the domain in altitude and time than those computed for NO_y, 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

[48] Virtually no SPEs occurred in 1963–1964, 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°S–90°S is shown in Figures 10b and 10a, respectively, as the difference between the ensemble averages of simulations "with SPEs" [1(a,b,c,d)] and "without SPEs" [1(w,x,y,z)]. The relative size of SPE-caused annual middle atmospheric NO_y production is shown with "triangles" in Figure 10b (bottom) between 200 and 100 hPa to focus attention on any apparent correlations between SPEs and annually averaged ozone and temperature variations. The colored regions in the bottom plot of each part of Figure 10 indicate a 95% statistical significance with the use of Student's *t* test. Ozone variations up to ±10% and temperature variations up to ±2 K are computed. There are a few years with very strong SPE activity that showed a statistically significant stratospheric ozone change (1972, 1989, 2000, 2001) over a limited altitude region. The statistically significant temperature changes generally were not correlated with these statistically significant ozone changes.

[49] The NH model output was analyzed in a similar way (annual zonal average) and ozone and temperature change for the polar latitude region 70°N–90°N are given in Figures 11b and 11a, 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 ±1 K. The colored regions in the bottom plot of each part in Figure 11 indicate a 95% statistical significance with the use of Student's *t* test. A statistically significant ozone decrease is computed in the NH in the stratosphere and lower mesosphere 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 (July and November) and 2001 (September and two in November). However, there are no computed statis-

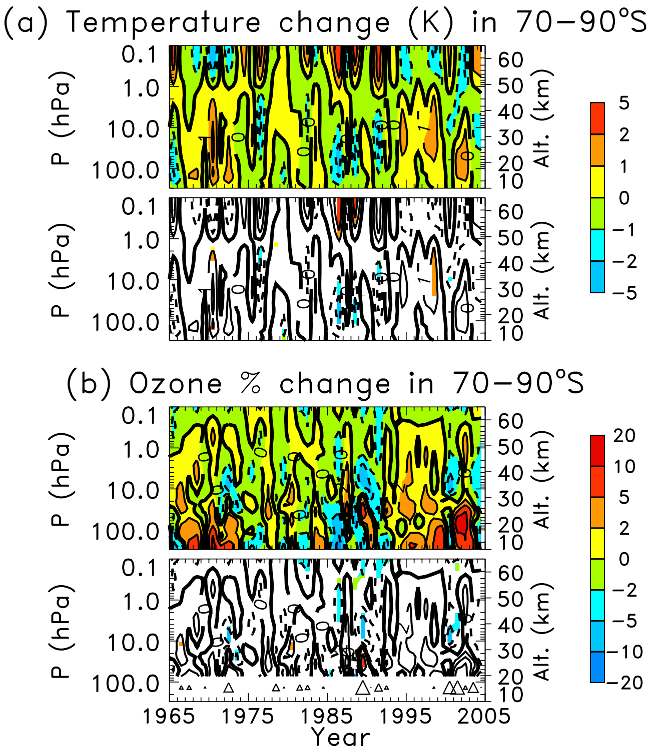

Figure 10. Annual mean solar proton event-caused change for the 40 years 1965 through 2004 in the SH polar latitude interval 70°S–90°S: difference between simulations “with SPEs” [1(a,b,c,d)] and “without SPEs” [1(w,x,y,z)]. (a) Temperature change (K). (b) Ozone change (%). The bottom plot in each part is the upper plot repeated with the colored areas indicating regions which are 95% statistically significant with the use of Student’s *t* test. The relative size of the SPE-caused annual middle atmospheric NO_y production is shown with “triangles” between 200 and 100 hPa in the bottom plot of Figure 10b (also, see Figure 12c).

tically significant temperature changes associated with this statistically significant ozone decrease.

5.2. SPE-Caused Total Ozone Changes

[50] A maximum polar ozone column decrease of about 1 to 4% due to very large SPEs has been computed in several previous two-dimensional modeling studies [e.g., *Jackman et al.*, 1990, 1995, 2000, 2005a, 2005b; *Reid et al.*, 1991; *Jackman and Fleming*, 2008]. These earlier analyses were unable to reveal any SPE-caused polar ozone column depletion in observations owing to the substantial intraseasonal and interannual variation in total ozone at high latitudes. We show the polar ozone column variations from the WACCM3 ensemble averages of simulations “without SPEs” [1(w,x,y,z)] and “with SPEs” [1(a,b,c,d)] compared with observations in Figure 12a for 70°S–90°S and Figure 12b for 70°N–90°N. The observations were differenced with respect to an average of the 2 years, 1979 and 1980, to allow for any quasi-biennial influences. The simulations were also differenced with respect to the ensemble average {1(w,x,y,z) or 1(a,b,c,d)} of 2 years, 1979 and 1980.

[51] A special analysis of total ozone measurements was needed to derive polar total ozone in Figure 12: 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 southernmost (or northernmost) 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 ∼40°–45°; however, these were not very large. The interspersing of the data interpolated in time and latitude appeared to reduce the artifacts.

[52] All WACCM3 simulations in Figure 12 contain time-dependent halogen loading, stratospheric aerosol loading, and solar cycle UV changes. These simulations are able to capture the general observed downward trend in polar ozone. This is especially true in the SH where the large ozone losses characteristic of the Antarctic ozone hole are driven by the atmospheric halogen loading. Total ozone in the polar regions is also strongly modulated by the year-to-year dynamical variability of the atmosphere, as seen in Figure 12. The WACCM3 simulations show interannual variability, however, the model is a free running GCM and cannot be expected to simulate the observed interannual behavior. Hence, there are large total ozone differences between the WACCM3 simulations and the observations, especially in the NH polar region where the interannual dynamical variability is especially large. For example, the reduced polar NH observed ozone values for 1993 and

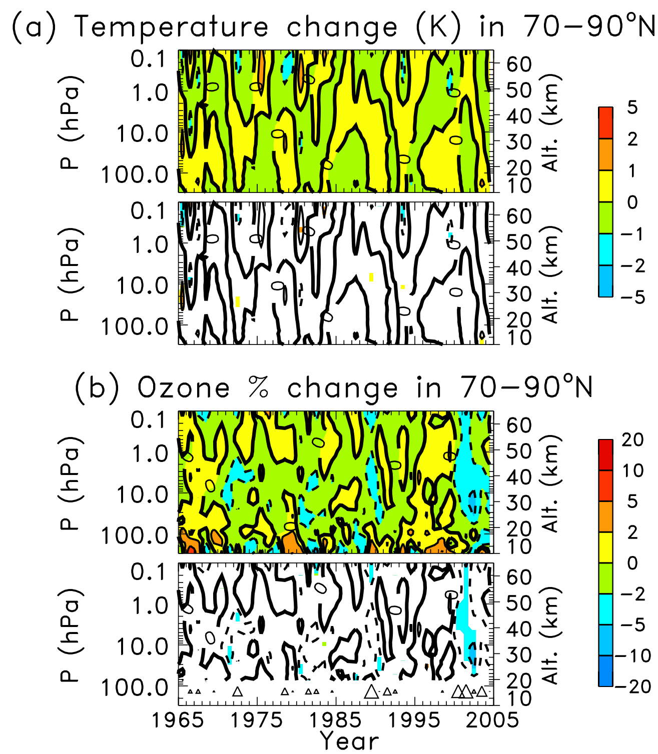

Figure 11. Same as Figure 10, but for the NH.

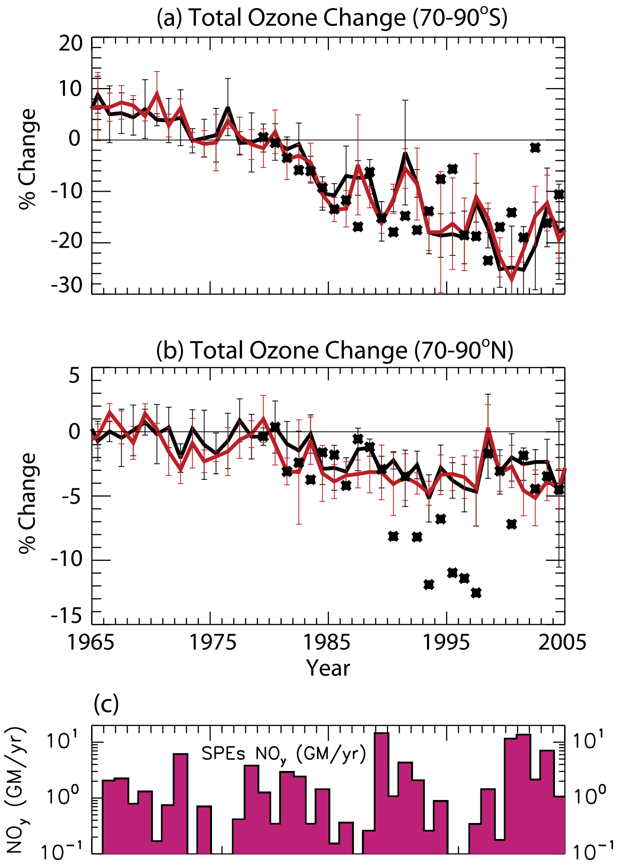

Figure 12. Total ozone change for the 40 years 1965 through 2004 in the (a) SH polar latitude interval 70°S–90°S and the (b) NH polar latitude interval 70°N–90°N. The black lines in Figures 12a and 12b show the average of WACCM3 simulations "without SPEs" [1(w,x,y,z)]. The red lines in Figures 12a and 12b show the average of WACCM3 simulations "with SPEs" [1(a,b,c,d)]. The computed standard deviations (one sigma) for the model results are also shown. The plotted WACCM3 results are the mean values minus the average for 1979–1980. The asterisks indicate observed total ozone (explained in section 5.2) minus the observed average for 1979–1980. (c) NO_y production per year (gigamoles, GM) by solar proton events.

1995–1997 have been attributed to the generally cooler temperatures and weaker poleward transport of ozone during the winter and spring [*Randel and Wu*, 1999; p. 4.4; *World Meteorological Organization*, 2007].

[53] Figure 12 reveals that there are only small differences in the WACCM3 simulations "with SPEs" (red lines) and "without SPEs" (black lines). There is some tendency for the simulations "with SPEs" to give slightly smaller ozone amounts during certain years, especially in the NH. There is some correlation of these changes with the yearly NO_y production from SPEs (Figure 12c); that is, larger NO_y results in slightly lower total ozone. However, these differences are significantly smaller than the very large impacts due to atmospheric halogen loading or the interannual dynamical variability.

[54] We then examined WACCM3 output to derive any statistically significant signal from SPEs in total polar ozone. Figure 12 includes the one standard deviation (one sigma) lines for both ensembles of simulations "with SPEs" [1(a,b,c,d)] and "without SPEs" [1(w,x,y,z)]. The SPE-caused total ozone change signal for most years for 70°N–90°N was found to be within one sigma of the average of the base simulations. The overall conclusion of Figure 12 is that the SPE influence on polar total ozone is very difficult to discern from observations given the large interannual dynamical variability inherent in the polar regions.

6. Conclusions

[55] WACCM3 has been used to study the long-term (more than a 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_y were initiated in years 2000, 2001, and 2003 after very large SPEs and lasted for up to a year past the events. A large 5-year average (2000–2004) statistically significant polar middle atmospheric NO_y signal was calculated in both hemispheres for the latest solar maximum period.

[56] 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 HO_x. The SPE-enhanced 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. Fairly small statistically significant stratospheric ozone signals were computed for the polar SH. The annually averaged polar temperature variations in both hemispheres temporally connected with SPEs, were not statistically significant. Very large SPEs did not lead to any statistically significant total ozone change.

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