

1 **Northern hemisphere atmospheric influence of the solar**
2 **proton events and ground level enhancement in January**
3 **2005**

4

5 **C. H. Jackman¹, D. R. Marsh², F. M. Vitt², R. G. Roble², C. E. Randall³, P. F.**
6 **Bernath⁴, B. Funke⁵, M. López-Puertas⁵, S. Versick⁶, G. P. Stiller⁶, A. J. Tylka⁷,**
7 **and E. L. Fleming^{1,*}**

8 [1]{NASA Goddard Space Flight Center, Greenbelt, Maryland}

9 [2]{National Center for Atmospheric Research, Boulder, Colorado}

10 [3]{University of Colorado, Boulder, Colorado}

11 [4]{University of York, York, United Kingdom}

12 [5]{Instituto de Astrofisica de Andalucia, Granada, Spain}

13 [6]{Karlsruhe Institute of Technology, Karlsruhe, Germany}

14 [7]{Naval Research Laboratory, Washington, DC}

15 [*]{also at: Science Systems and Applications Inc., Lanham, Maryland}

16 Correspondence to: C. H. Jackman (Charles.H.Jackman@nasa.gov)

17

18 **Abstract**

19 Solar eruptions in early 2005 led to a substantial barrage of charged particles on the Earth's
20 atmosphere during the January 16-21 period. Proton fluxes were greatly increased during
21 these several days and led to the production of HO_x (H, OH, HO₂) and NO_x (N, NO, NO₂),
22 which then caused the destruction of ozone. We focus on the Northern polar region, where
23 satellite measurements and simulations with the Whole Atmosphere Community Climate
24 Model (WACCM3) showed large enhancements in mesospheric HO_x and NO_x constituents,
25 and associated ozone reductions, due to these solar proton events (SPEs). The WACCM3
26 simulations show enhanced short-lived OH throughout the mesosphere in the 60-82.5°N
27 latitude band due to the SPEs for most days in the Jan. 16-21, 2005 period, in reasonable

1 agreement with the Aura Microwave Limb Sounder (MLS) measurements. Mesospheric HO₂
2 is also predicted to be increased by the SPEs, however, the modeled HO₂ results are
3 somewhat larger than the MLS measurements. These HO_x enhancements led to huge
4 predicted and MLS-measured ozone decreases of greater than 40% throughout most of the
5 Northern polar mesosphere during the SPE period. Envisat Michelson Interferometer for
6 Passive Atmospheric Sounding (MIPAS) measurements of hydrogen peroxide (H₂O₂) show
7 increases throughout the stratosphere with highest enhancements of about 60 pptv in the
8 lowermost mesosphere over the Jan. 16-18, 2005 period due to the solar protons. WACCM3
9 predictions indicate H₂O₂ enhancements over the same time period of more than twice that
10 amount. Measurements of nitric acid (HNO₃) by both MLS and MIPAS show an increase of
11 about 1 ppbv above background levels in the upper stratosphere during January 16-29, 2005.
12 WACCM3 simulations show only minuscule HNO₃ changes in the upper stratosphere during
13 this time period. However due to the small loss rates during winter, polar mesospheric
14 enhancements of NO_x are computed to be greater than 50 ppbv during the SPE period.
15 Computed NO_x increases, which were statistically significant at the 95% level, lasted about a
16 month past the SPEs. The SCISAT-1 Atmospheric Chemistry Experiment Fourier Transform
17 Spectrometer (ACE-FTS) NO_x measurements and MIPAS NO₂ measurements for the polar
18 Northern Hemisphere are in reasonable agreement with these predictions. An extremely large
19 ground level enhancement (GLE) occurred during the SPE period on January 20, 2005. We
20 find that protons of energies 300 to 20,000 MeV, not normally included in our computations,
21 led to enhanced lower stratospheric odd nitrogen concentrations of less than 0.1% as a result
22 of this GLE.

23

24 **1 Introduction**

25 Large solar eruptions during January 16-21, 2005 caused huge fluxes of high-energy solar
26 charged particles to reach Earth. The solar proton flux enhancement during this period has
27 been well documented and caused significant production of OH (Verronen et al. 2006;
28 Damiani et al. 2008) and destruction of ozone (Verronen et al. 2006; Seppälä et al. 2006;
29 Klekociuk et al. 2007; Damiani et al. 2008). The largest ground level enhancement (GLE) of
30 neutrons during solar cycle 23 also occurred in this period. A neutron monitor registered an
31 increase of about 270% on January 20, 2005 during the GLE (Gopalswamy et al. 2005).

1 We studied the short- and medium-term (days to a few months) atmospheric constituent
2 effects of the four largest solar proton events (SPEs) in the past 45 years (August 1972,
3 October 1989, July 2000, and October-November 2003) in Jackman et al. (2008) with version
4 3 of the Whole Atmosphere Community Climate Model (WACCM3). The present
5 investigation builds on that study and focuses on the short- and medium-term influences of
6 solar particles on the mesosphere and stratosphere in the time period January 1 through March
7 31, 2005. There was substantial solar activity in January 2005, which was also the period of
8 the eleventh largest SPE period in the past 45 years (Jackman et al., 2008). We include SPEs
9 in January 2005 and the highest energy protons leading to the GLE on January 20, 2005 in our
10 WACCM3 computations. Larger and longer-lasting impacts were expected in the Northern
11 winter polar region because of the diminished sunlight and general downward transport. We,
12 therefore, focus on the impact of the solar particles on constituents in the Northern polar
13 mesosphere and stratosphere. The highly energetic solar particles produced HO_x (H, OH,
14 HO₂) and NO_x (N, NO, NO₂), which then led to ozone variations. We compare the
15 WACCM3 predictions during this period with measurements from several platforms: Aura
16 Microwave Limb Sounder (MLS) of OH, HO₂, HNO₃, and ozone; Envisat Michelson
17 Interferometer for Passive Atmospheric Sounding (MIPAS) of H₂O₂, NO₂, and HNO₃; and
18 SCISAT-1 Atmospheric Chemistry Experiment (ACE) of NO_x.

19 This paper is divided into seven sections, including the Introduction. The charged particle
20 flux and ionization rate are discussed in Section 2. Odd hydrogen (HO_x) and odd nitrogen
21 (NO_y) production are discussed in Section 3. A description of WACCM3 is given in Section
22 4. The modeled and measured influences of the January 2005 SPEs over the January 1 –
23 March 31, 2005 period are shown in Section 5. The influence of the January 20, 2005 GLE is
24 shown in Section 6 and the conclusions are presented in Section 7.

25

26 **2 Charged particle flux and ionization rate**

27 Our WACCM3 computations with charged particle flux included: 1) the solar proton flux
28 (energies 1 to 300 MeV) over the January 1 – March 31, 2005 period; and 2) the highest
29 energy protons (300 to 20,000 MeV) associated with a GLE of neutrons on January 20, 2005.
30 We performed separate WACCM simulations with no charged particle flux, charged particles
31 described in 1), and charged particles described in both 1) and 2). These model simulations
32 are described in section 4.

1 The solar proton flux (energies 1 to 300 MeV) for 2005 was provided by the National Oceanic
2 and Atmospheric Administration (NOAA) Geostationary Operational Environmental Satellite,
3 GOES-11 (Jackman et al., 2008). The proton flux data from the satellites were used to
4 compute ion pair production profiles using the energy deposition methodology discussed in
5 Jackman et al. (1980), where the creation of one ion pair was assumed to require 35 eV
6 (Porter et al. 1976). The SPE-produced daily average ionization rates are given in Figure 1
7 for the eight day period, January 15-22, 2005, from 100 hPa (~16 km) to 0.001 hPa (~96 km).
8 There were two periods of SPEs in these eight days, January 16-18 and January 20-21. The
9 first period was the most intense with peak ionization above $1000 \text{ cm}^{-3} \text{ s}^{-1}$ for the 0.01 to 1
10 hPa region. The second period showed peak ionization above $500 \text{ cm}^{-3} \text{ s}^{-1}$ for the 0.2 to 10
11 hPa region.

12 We included the highest energy protons (300 to 20,000 MeV) associated with the GLE of
13 neutrons on January 20, 2005 in some computations with “SPEs+GLE”. This high energy
14 proton flux was taken from the spectrum given in Usoskin et al. (2009, 2010), which was
15 derived using methodology presented in Tylka and Dietrich (2009). The calculated GLE
16 ionization rate on Jan. 20, 2005 was added to the computed ionization rate from the GOES-11
17 measured protons for some of the model computations (see section 4.). Ionization rates on
18 January 20, 2005 between 10 and 100 hPa for “SPEs-only” and “SPEs+GLE” are compared
19 in Figure 2. At 10 hPa the ionization is primarily caused by the SPEs; ionization caused by
20 the GLE rapidly increases in importance below 10 hPa, and is more than an order of
21 magnitude larger than ionization by the SPEs at 40 hPa.

22

23 **3 Odd hydrogen (HO_x) and odd nitrogen (NO_y) production**

24 Charged particle precipitation results in the production of odd hydrogen (HO_x) through
25 complex positive ion chemistry (Solomon et al. 1981). The charged particle-produced HO_x is
26 a function of ion pair production and altitude and is included in WACCM3 simulations using
27 a lookup table from Jackman et al. (2005a, Table 1), which is based on the work of Solomon
28 et al. (1981). Even though the HO_x constituents have a relatively short lifetime (~hours)
29 throughout most of the mesosphere, the ozone depletion can be very large during substantial
30 SPEs (e.g., Solomon et al. 1983; Jackman et al. 2001; Verronen et al. 2006). This HO_x-
31 induced ozone depletion can have an influence on the mesospheric temperature and winds
32 over a relatively short period of time (~4-6 weeks), see Jackman et al. (2007).

1 Odd nitrogen (NO_y) is also produced when the energetic charged particles (protons and
2 associated secondary electrons) dissociate N_2 as they precipitate into the atmosphere. Here
3 we assume that ~ 1.25 N atoms are produced per ion pair and divide the proton impact of N
4 atom production between ground state $\text{N}(^4\text{S})$ ($\sim 45\%$ or ~ 0.55 per ion pair) and excited state
5 $\text{N}(^2\text{D})$ ($\sim 55\%$ or ~ 0.7 per ion pair) nitrogen atoms (Porter et al., 1976).

6 7 **4 Description of the Whole Atmosphere Community Climate Model** 8 **(WACCM3)**

9 WACCM3 has been used in several previous studies to investigate the impact of natural and
10 anthropogenic influences on the atmosphere from the troposphere through the middle
11 atmosphere to the lower thermosphere (Sassi et al., 2002, 2004; Forkman et al., 2003; Richter
12 and Garcia, 2006; Kinnison et al., 2007; Garcia et al., 2007; Marsh et al., 2007; Jackman et al.
13 2008, 2009). The model domain is from the surface to 4.5×10^{-6} hPa (about 145 km), with 66
14 vertical levels, and includes fully interactive dynamics, radiation, and chemistry. WACCM3
15 is based on the Community Atmosphere Model (CAM3) and includes modules from
16 Thermosphere-Ionosphere-Mesosphere-Electrodynamics General Circulation Model and the
17 Model for Ozone And Related chemical Tracers (MOZART-3) to simulate the dynamics and
18 chemistry of the Earth's atmosphere. The vertical resolution is ≤ 1.5 km between the surface
19 and about 25 km and increases slowly above 25 km to 2 km at the stratopause; it is 3.5 km in
20 the mesosphere and one half the local scale height above the mesopause. The version of
21 WACCM3 used here has latitude and longitude grid spacing of 4° and 5° , respectively. An
22 extensive description of WACCM3 is given in Garcia et al. (2007) and Kinnison et al. (2007).

23 WACCM3 was forced in all simulations with observed time-dependent sea surface
24 temperatures (SSTs), observed solar spectral irradiance and geomagnetic activity changes,
25 and observed concentrations of greenhouse gases and halogen species over the simulation
26 period (see Garcia et al., 2007). The geomagnetic activity included in all the WACCM3
27 simulations accounts for auroral precipitation, along with HO_x and NO_y production.
28 However, these auroral particles mostly deposit their energy in the lower thermosphere
29 (Marsh et al., 2007), whereas SPEs deposit most of their energy in the mesosphere and upper
30 stratosphere.

31 We have completed three 4-member ensemble WACCM3 simulations (described below) over
32 the January 1 – March 31, 2005 period: A) four realizations [A(1, 2, 3, 4)] without any daily

1 ionization rates from SPEs or the GLE; B) four realizations [B(1, 2, 3, 4)] with the daily
2 ionization rates from SPEs throughout the period; and C) four realizations [C(1, 2, 3, 4)] with
3 the daily ionization rates from SPEs throughout the period and the GLE on January 20. These
4 WACCM3 simulations are summarized in Table 1. The ionization rates, when included, were
5 applied uniformly over both polar cap regions (60-90°N and 60-90°S geomagnetic latitude) as
6 solar protons are guided by the Earth's magnetic field lines to approximately these areas
7 (Verronen et al., 2007; see, also McPeters et al., 1981 and Jackman et al., 2005a). Due to the
8 differing offsets of the geomagnetic and geographic poles in the two hemispheres, the effects
9 from the SPEs and GLE are not expected to be symmetric in the northern and southern
10 hemispheres.

11 WACCM3 is a free-running GCM and the realizations' starting conditions were each slightly
12 different from the other, initiated in January 1950. For all ensemble members WACCM3 was
13 run in its free-running mode with identical boundary conditions from January 1950 up to
14 January 1, 2005 (Garcia et al., 2007; Jackman et al., 2009), which is the starting date for all
15 model computations shown in this paper. Simulations A1, B1, and C1 have the same starting
16 conditions, except simulation A1 has “no SPEs and no GLE,” simulation B1 has “SPEs-
17 only,” simulation C1 has “SPEs+GLE.” Similar comments apply to grouped simulations A2,
18 B2, and C2; A3, B3, and C3; and A4, B4, and C4.

19

20 **5 Influences of the January 2005 SPEs**

21 The mesosphere was perturbed by the SPEs in January 2005 as seen in the measurements of
22 several satellite instruments and WACCM3 simulations. The short- (~days) as well as
23 medium- (~weeks) term changes due to these solar influences will be discussed in this
24 section.

25

26 **5.1 Short-term influences**

27 Since HO_x constituents have such short lifetimes (e.g., Solomon et al., 1981), a large
28 enhancement of HO_x caused by an influx of protons during an SPE will be relatively short-
29 lived (~days). MLS provided measurements of two HO_x constituents, OH and HO₂ (Pickett et
30 al., 2008). Previous papers have shown substantial HO_x and ozone impacts during the

1 January 2005 SPEs (Verronen et al., 2006, 2007; Seppälä et al., 2006; Klekociuk et al., 2007;
2 Damiani et al., 2008, 2009, 2010). We focus on the Northern polar latitudes, a geographic
3 region where HO_x constituents are at very small values in January due to minimal or no
4 sunlight. The HO_x constituents in the winter polar region are, therefore, especially sensitive
5 to solar proton impact in the mesosphere.

6 **5.1.1 Hydroxyl radical (OH)**

7 The Aura MLS OH measured enhancements due to the SPEs at 0.022 hPa for the Northern
8 Hemisphere are given in Figure 3 and were computed by subtracting the observations on
9 January 15, 2005 (before the SPE) from the observations on January 18, 2005 (during the
10 SPE). For added clarity, measurements are only shown northward of 42.5°N, however, no
11 MLS measurements are available in the band 82.5-90°N. MLS measurements were binned
12 into 30° longitude and 5° latitude bands. The polar cap edge (60°N geomagnetic latitude),
13 wherein the protons are predicted to interact with the atmosphere, is indicated by the white
14 circle. The MLS data shows that the SPE increased OH significantly: values greater than 4
15 ppbv are observed in a substantial part of the area poleward of 60°N geomagnetic latitude.

16 The WACCM3 OH predicted enhancements due to the SPEs at 0.022 hPa for the Northern
17 Hemisphere are given in Figure 4 and were computed from the B1 simulation (SPEs-only) by
18 subtracting the simulation results on January 15, 2005 (before the SPE) from the results on
19 January 18, 2005 (during the SPE). As in Jackman et al. (2008), we show results from only
20 one realization: the other realizations give similar results. For added clarity, the simulation
21 results are only shown from 44-90°N. As in Figure 3, the polar cap edge (60° geomagnetic
22 latitude) is indicated by the white circle. WACCM3 also predicted a significant increase in
23 OH: values greater than 4 ppbv are modeled in a substantial part of the area poleward of 60°N
24 geomagnetic latitude. Both the MLS measurements and WACCM3 predictions indicate
25 similar areas of enhanced OH as a result of the SPEs. The WACCM predictions do indicate a
26 slightly larger amount of OH change, when compared with MLS observations.

27 We compare the MLS OH measurements and WACCM3 model predictions for January 16-
28 23, 2005 in the latitude band 60-82.5°N in Figure 5. The first two weeks of January 2005
29 were relatively quiet and contained no SPEs. We thus used these first two weeks (January 1-
30 14) to construct an average quiescent OH profiles for both MLS and WACCM3, respectively.
31 This respective quiescent OH profile was subtracted from the OH observations or predictions

1 for January 16-23, 2005 and the results are given in Figure 5. The WACCM3 B1 simulation
2 (SPEs-only) was used for this figure.

3 Fairly substantial OH enhancements are shown in the MLS measurements (up to 4 ppbv) and
4 WACCM3 predictions (up to 6 ppbv) for the January 16-23 period. The OH increases were
5 largest on January 17-18, similar to the WACCM predictions. Similar to the comparisons
6 between Figures 3 and 4, the WACCM predictions of Figure 5 do indicate a slightly larger
7 peak OH change, when compared with MLS observations.

8 **5.1.2 Hydrogen dioxide (HO₂)**

9 The MLS instrument additionally provides HO₂ measurements during the January 2005
10 period. Such measurements are somewhat noisier than the OH observations, however, MLS
11 HO₂ does indicate enhancements above background levels (>0.1 ppbv) due to the January
12 2005 SPEs. Similar to Figure 5, Figure 6 was produced by averaging the HO₂ measurements
13 over the quiet (non-SPE) period January 1-14, 2005 and subtracting this average from the
14 HO₂ observations or predictions during January 16-23, 2005. Again, the WACCM3 B1
15 (SPEs-only) simulation was used for Figure 6.

16 As with OH, the WACCM predictions indicate a similar time frame for the HO₂ atmospheric
17 perturbation when compared with MLS observations. Also, a mostly larger HO₂ change is
18 predicted than measured in the January 16-21, 2005 time period. The cause of the
19 modeled/measured SPE-caused OH and HO₂ differences is not clear, but may be related to
20 problems in the modeled representation of HO_x chemistry (Canty et al., 2006).

21 **5.1.3 Ozone**

22 Besides these two HO_x constituents, MLS also measures ozone. Like Figures 5 and 6, Figure
23 7 was produced by averaging the ozone measurements over the quiet (non-SPE) period
24 January 1-14, 2005 and subtracting this average from the ozone observations or predictions
25 during January 16-23, 2005. As for OH and HO₂, the WACCM3 B1 simulation was used for
26 ozone in Figure 7.

27 The SPE-produced HO_x constituents are relatively short-lived (~days) and lead to the
28 destruction of ozone in the uppermost stratosphere and mesosphere. We have found that the
29 WACCM-predicted ozone change due to the SPEs for the time period plotted is confined to
30 pressures <1 hPa, similar to previous reported studies (e.g., Seppälä et al., 2006; Verronen et

1 al., 2006; Klekociuk et al., 2007). Ozone decreases (>40%) are measured and predicted for
2 the January 17-23, 2005 period at pressures <0.4 hPa. Although there is reasonable
3 agreement between WACCM and MLS, the model predictions indicate a slightly deeper
4 penetration of the SPE-caused ozone depletion signal. Changes in ozone for pressures >1 hPa
5 are likely not related to the SPEs, but are probably ongoing seasonal changes at this time of
6 year.

7 The measured and predicted ozone increases in a band between about 1 and 0.4 hPa could be
8 an indication of the “self-healing” effect (e.g., Jackman and McPeters, 1985), wherein large
9 ozone depletions above a certain level in the middle atmosphere are mitigated somewhat by
10 an ozone increase below the level. Such “self-healing” phenomena are most likely to occur at
11 the highest solar zenith angles and result from the enhanced ultraviolet flux caused by
12 mesospheric ozone depletion leading to more ozone production by molecular oxygen
13 photodissociation at lower atmospheric levels. However, the WACCM base simulation A1
14 (no SPEs and no GLE) also showed enhanced ozone in a similar band between about 1 and
15 0.4 hPa (not shown), suggesting that this increase might be normal seasonal behavior. Thus,
16 we cannot conclude that ozone “self-healing” is evident with the January 2005 SPEs.

17 **5.1.4 Hydrogen peroxide (H₂O₂)**

18 Envisat MIPAS has recently been shown to have the capability of observing hydrogen
19 peroxide (H₂O₂) (Versick, et al., 2009; Versick, 2010) and has provided these measurements
20 in January 2005 during the SPEs. We use here H₂O₂ data (version V4O_H2O2_304)
21 retrieved with the MIPAS level 2 processor developed and operated by the Institute of
22 Meteorology and Climate Research (IMK) in Karlsruhe together with the Instituto de
23 Astrofísica de Andalucía (IAA) in Granada. The main source for H₂O₂ is the HO₂ self-
24 reaction



26 with a smaller contribution from the three-body reaction



28 Thus, production of OH and HO₂ by the SPEs leads very rapidly to the production of H₂O₂.
29 Figure 8 (top) shows the polar (60-82.5°N) MIPAS observed 24-hour average H₂O₂ for three
30 days (Jan. 16-18, 2005) throughout most of the stratosphere and into the lowermost
31 mesosphere. H₂O₂ changes during the three days of the first January 2005 SPE (see section 2)

1 are minor at pressure levels greater than 30 hPa. At pressure levels less than 30 hPa, H₂O₂ is
2 measured to increase during these three days with the largest increases in the lowermost
3 mesosphere (~60 pptv).

4 Figure 8 (middle) shows the polar (60-82.5°N) WACCM3 predicted 24-hour average H₂O₂
5 for the same three days using the B1 simulation (SPEs-only). Generally, the modeled
6 amounts of H₂O₂ are substantially more than the measured values throughout the plotted
7 domain. Figure 8 (bottom) shows the enhanced H₂O₂ due to the SPEs and is the difference
8 between the A1 (no SPEs and no GLE) and the B1 (SPEs-only) simulations. H₂O₂ is
9 predicted to increase at all pressure levels during these three days as a result of the SPEs.
10 WACCM3 H₂O₂ increases about 140 pptv in the lowermost mesosphere, over a factor of two
11 larger than observed by MIPAS. For better direct comparisons, the MIPAS averaging kernel
12 (AK) was applied to the plotted WACCM3 results.

13 What is the reason behind the measurement and model H₂O₂ differences? Since the OH and
14 HO₂ predictions are higher than the MLS measurements, it does follow that H₂O₂ would
15 likely be overestimated, given the major production reaction (1). The major loss of H₂O₂
16 during daylight is through photolysis



18 During nighttime the reaction



20 is the major loss process for H₂O₂. Reaction (4) is especially important in the northern polar
21 latitudes in January, thus is most significant for this study. The HO_x production from SPEs is
22 in the form of OH and H (Solomon et al., 1981). These constituents can very rapidly lead to
23 HO₂ production through



26 The HO_x constituents are primarily lost through reactions



1 Other reactions, besides (1) through (9), are important as well and involve HO_x species with
2 other atmospheric constituents. All the neutral constituent photochemical reaction rates and
3 photodissociation cross sections are taken from Sander et al. (2006). It is unclear which
4 reaction (or reactions) may need to be modified to rectify the differences between MIPAS and
5 WACCM3 H₂O₂. These measurement/model disagreements may be related to the difficulties
6 in simulating OH and HO₂ (e.g., see Canty et al., 2006) and require further study.

7 **5.2 Medium-term influences**

8 SPE-produced NO_x constituents have longer lifetimes than HO_x constituents (e.g., Jackman et
9 al., 2008) and can cause atmospheric changes for several weeks or longer following such
10 events. López-Puertas et al. (2005a) has shown large Envisat MIPAS NO_x enhancements
11 caused by the October-November 2003 SPEs as well as associated ozone depletion over a two
12 and a half week period. The proton flux during the January 2005 SPEs was not quite as
13 significant as the proton flux during the October-November 2003 SPEs, however, the SPE-
14 induced NO_x change did occur in the middle of the NH winter when the impact can be
15 enhanced through a longer lifetime and downward transport (Jackman et al., 2000). We focus
16 on the Northern Hemisphere as any NO_x signal is most likely to last longer in the darker
17 hemisphere (e.g., Jackman et al., 2008). Quantifying the influence of the NO_x produced by
18 the January 2005 SPEs is one of the main objectives of this paper.

19 **5.2.1 Nitrogen dioxide (NO₂)**

20 Envisat MIPAS provided measurements for some days during the month of January 2005. In
21 particular, we show the four-day average (January 10-13, 2005) MIPAS NO₂ measurements
22 (IMK/IAA data version V4O_NO2_501) in Figure 9 (top) before any major SPE disturbance.
23 Although the measured NO₂ amounts are at modest levels (~4-10 ppbv) in the middle
24 latitudes (40-60°N), the observed polar middle mesosphere NO₂ can be quite substantial,
25 reaching peak amounts greater than 100 ppbv near 70 km (~0.03 hPa) at the highest Northern
26 latitudes.

27 WACCM3 predictions of NO₂ for the same time period are given in Figure 9 (bottom). The
28 model results do show relatively modest levels (~1-10 ppbv) in the middle latitudes, fairly
29 similar to MIPAS observations. However, WACCM3 only shows a peak of about 10 ppbv
30 near 70 km (~0.03 hPa) at the highest Northern latitudes, very different from MIPAS
31 measurements. It appears that MIPAS measurements are indicative of a very disturbed

1 mesosphere before the SPEs commence on January 16th. Seppälä et al. [2007] likewise
2 showed high NO₂ mixing ratios (>30 ppbv) in the northern hemisphere polar lower
3 mesosphere in early January 2005, measured by the GOMOS instrument.

4 We used our WACCM3 simulations to compute the NO₂ change over the January 16-23,
5 2005, period in Figure 10. This NO₂ change was computed by subtracting the four-day
6 average (Jan. 10-13, 2005) values from the Jan. 16-23 predictions for the B1 (SPEs-only)
7 simulation; results pertain to the average in the latitude band from 70°-90°N. Nitrogen
8 dioxide enhancements over 30 ppbv are computed in the 60-65 km (0.1-0.04 hPa) altitude
9 region for Jan. 18-20, 2005.

10 The MIPAS measurements are not shown over the Jan. 16-23, 2005, period due to its limited
11 coverage as the instrument was measuring in its upper troposphere/lower stratosphere mode
12 with an uppermost temperature retrieval of just 50 km. The temperature above 50 km was not
13 measured and the assumed temperature was too high in this region and greatly impacted the
14 NO₂. The TIMED Sounding of the Atmosphere using Broadband Emission Radiometry
15 (SABER) instrument took measurements during this time period and showed that the assumed
16 MIPAS temperatures were about 10-12 K too large in the 50-60 km region. Some
17 preliminary computations with temperatures more similar to SABER (i.e., decreased by 10-
18 12K) have resulted in enhanced MIPAS NO₂ values during the first SPE period (Jan. 16-18,
19 2005) of about 30 ppbv over the Jan. 10-13 levels. Thus, even though the MIPAS NO₂
20 observations before the SPEs are very different, the deduced MIPAS NO₂ increases as a result
21 of the first SPE are fairly similar to the WACCM3 predictions.

22 **5.2.2 Nitric acid (HNO₃)**

23 The SPE-caused impact on HNO₃ has been discussed before in relation to the October-
24 November 2003 SPEs (Orsolini et al., 2005; López-Puertas et al., 2005b; Jackman et al.,
25 2008, Verronen et al., 2008). Jackman et al. (2008) showed Envisat MIPAS measured HNO₃
26 enhancements of over 2 ppbv near 1 hPa as a result of the late October 2003 SPE, however,
27 the WACCM3 simulations predicted a smaller maximum enhancement of 0.8 ppbv near 1
28 hPa.

29 Klekociuk et al. (2007) demonstrated HNO₃ enhancements in Aura MLS measurements as
30 well as global model computations as a result of the January 2005 SPEs. We have analyzed
31 MLS HNO₃ measurements in a similar manner to Klekociuk et al. (2007) and show the results

1 in Figure 11 (top). Here, an average MLS HNO₃ for the period January 1-14, 2005, before
2 the first SPE, was subtracted from the MLS HNO₃ for January 16-29, 2005 in the 60-82.5°N
3 band. Envisat MIPAS HNO₃ measurements are also available in January 2005, but only for a
4 limited number of days (i.e., Jan. 10-13, 16-18, 27-28). Because of this limited dataset, the
5 four-day average of the MIPAS measurements before the first SPE (i.e., Jan. 10-13, 2005)
6 was subtracted from the Jan. 16-18 and 27-28, 2005 values (IMK/IAA data version
7 V4O_HNO3_201); this difference is given in Figure 11 (middle). The WACCM3 results are
8 presented in Figure 11 (bottom), where an average of the B1 (SPEs-only) simulated HNO₃ for
9 the period January 1-14, 2005 before the first SPE was subtracted from the modeled HNO₃ for
10 January 16-29, 2005 in the 60°-82.5°N band.

11 The MLS HNO₃ measurements indicated two enhanced regions (3-9 and 20-40 hPa) during
12 the Jan. 16-25, 2005 period (also, see Klekociuk et al. 2007) with a region of decreased HNO₃
13 in between. Also, MLS shows decreased HNO₃ between 40 and 100 hPa. The MIPAS HNO₃
14 observations show similarities to the MLS data for pressure levels less than 9 hPa and more
15 than 20 hPa, however, there is not an indication of the region of decreased HNO₃ between 9
16 and 20 hPa. The WACCM3 predicted HNO₃ change shows decreases between about 4 and 50
17 hPa during Jan. 16-23, 2005, with a slight increase at pressures less than 4 hPa. The
18 WACCM3 A1 simulation (no SPEs, no GLE), which is not shown, gives the same results as
19 those predicted with B1 (SPEs-only), the only difference being the small SPE-caused 0.1
20 ppbv enhanced HNO₃ contour near 3 hPa on Jan. 20 and a few days after. Thus, we are left
21 with the dilemma found in Jackman et al. (2008) whereby large increases in observed HNO₃
22 temporally connected to SPEs could not be properly simulated.

23 The creation of HNO₃ through the ion-ion recombination between H⁺ and NO₃⁻ cluster ions
24 was simulated during another solar proton event period, the Halloween storm episode in
25 October-November 2003, with the use of the Sodankylä Ion and Neutral Chemistry model in
26 Verronen et al. (2008). They showed that the HNO₃ production above 35 km as a result of
27 those large events could account for the extra HNO₃ observed by MIPAS in Oct./Nov. 2003.
28 It is likely that this ion chemistry, currently not included in WACCM3, could also explain the
29 MLS and MIPAS observed additional 0.5-1 ppbv HNO₃ above 35 km (pressures <20 hPa).

1 **5.2.3 Nitrogen oxides, NO_x (NO+NO₂)**

2 ACE-FTS (hereinafter referred to as ACE) (Bernath et al., 2005) provided measurements
3 during all of the SPE period. ACE measured both NO and NO₂ (e.g., see Rinsland et al.,
4 2005), and thus supplied NO_x (NO+NO₂) measurements at fairly high Northern latitudes for
5 January 1-31, 2005. These ACE observations are given in Figure 12 (a) and were taken in the
6 latitude range from ~57-66°N (see Figure 12, top). Large amounts of NO_x are observed at
7 pressures <0.01 hPa with evidence of some downward transport over this time period,
8 especially in the latter half of the month. We focus on pressures >0.01 hPa, where there is an
9 indication of a large perturbation around January 16. After that date the contour levels 20, 50,
10 100, and 200 ppbv show substantially more NO_x measured in the pressure range 0.02 to about
11 0.4 hPa (~55-75 km).

12 The ACE measurements (Fig. 12a) are compared with similar plots from our WACCM3
13 simulations in Figures 12 (b and c). The WACCM3 results are taken from the model
14 predictions for the 60-66°N latitude bins, approximately the latitude range for the ACE
15 Northern Hemisphere measurements after January 6, 2005. Figure 12 (c) shows WACCM3
16 NO_x predictions from an average of the four A realizations (no SPEs and no GLE). This plot
17 does not indicate much of a change in NO_x over the month. In fact, the predicted NO_x in the
18 pressure range 0.02 to 0.4 hPa after January 16th appears to show a slight decrease at most
19 levels. Figure 12 (b) shows WACCM3 NO_x predictions from an average of the four B
20 realizations (SPEs-only). These model predictions show a dramatic change after January 16th
21 with large NO_x increases indicated by changes in the slopes of contour levels 10, 20, 50, and
22 100 ppbv.

23 The NO_x variations over the three month time period (Jan. 1 – Mar. 31, 2005) are given in
24 Figure 13. Again, ACE measurements are shown in the top plot. There is a change in the
25 slopes of the NO_x contours after Day of Year (DoY) 32, when NO_x amounts tend to decrease
26 with time at virtually all levels above ~1 hPa. ACE observes at latitudes greater than 60°N up
27 through DoY 83 (March 24), thus this NO_x change is probably related more to a seasonal
28 effect, not related to the SPEs, than to the variation in ACE measurement latitudes during the
29 season. After DoY 83, the latitude observed by ACE varies rapidly from 60°N to 41°N by
30 DoY 90. These rapid changes in observed latitude help to explain the fast decrease of
31 observed NO_x in the last week plotted in Figure 13 (top). Downward transport of

1 thermospheric NO_x in the winter and early spring, not related to the SPEs, is much larger at
2 polar latitudes than middle latitudes (e.g., Randall et al., 2005, 2006).

3 MIPAS also measured NO_x for 16 days (e.g., DoYs 27-28, 38, 44-46, 48-49, 52-53, 61-62,
4 67-68, and 80-81) in this period over a limited altitude range on most days. We have found
5 that, generally, MIPAS observations are in reasonable agreement with ACE (not shown).

6 Figure 13 (middle) shows WACCM3 NO_x predictions (60-66°N) from an average of the B
7 simulations (SPEs-only), essentially an extension of Figure 12 (b) for another 59 days. There
8 are many similarities between these model computations and the ACE measurements. The
9 change in slope of the contour levels indicating a decrease in NO_x at virtually all levels above
10 ~1 hPa occurs in the model simulations at about DoY 25 (rather than the DoY 32 in the ACE
11 measurements), however, qualitatively the model results and ACE measurements are in
12 reasonable agreement.

13 We are able to compute the quantitative NO_x enhancement due to the SPEs by subtracting an
14 average of the A simulations (no SPEs and no GLE) from the average of the B simulations
15 (SPEs-only). These results are given in Figure 13 (bottom), where the colored regions
16 indicate 95% statistical significance with the use of Student's *t* test. The SPEs caused NO_x
17 increases > 50 ppbv in the middle to upper mesosphere. These NO_x enhancements
18 diminished over time to be less than 5 ppbv and no longer statistically significant by DoY 50.
19 Thus, the SPE-caused NO_x increases from the January 2005 SPEs lasted for about one month
20 past the beginning of the events.

21 **5.2.4 Ozone and temperature**

22 We computed the ozone change due to SPEs over the January 1 – March 31, 2005 period by
23 comparing the average of the B simulations (SPEs-only) relative to an average of the A
24 simulations (no SPEs and no GLE). The large ozone decreases shown in Figure 7 extended
25 another two days (through DoY 25), however, statistically significant (to 95%) NH polar
26 mesospheric ozone loss computed with Student's *t* test was evident only from DoY 17-23.
27 Ozone depletion less than 5% due to the SPEs was calculated for a couple of weeks past the
28 end of January. These results are consistent with the SPE-induced short-lived HO_x
29 enhancements causing most of the mesospheric ozone loss.

30 We also computed the temperature change due to SPEs over the January 1 – March 31, 2005
31 period by comparing the average of the B simulations (SPEs-only) relative to an average of

1 the A simulations (no SPEs and no GLE). These computed temperature changes were less
2 than 3 K during the time period of the large computed ozone losses (DoY 17-23) and were not
3 statistically significant. Such small temperature changes are consistent with Jackman et al.
4 (2007) and are not surprising in the limited sunlit polar region (NH) where less ozone heating
5 occurs.

6

7 **6 Influences of the January 20, 2005 GLE**

8 As discussed previously (sections 1 and 2), a very large GLE occurred on January 20, 2005,
9 during the SPE period. Although the flux of very energetic protons was extremely high, the
10 duration of this intense flux was fairly short (less than about 8 hours for the highest energy
11 protons, see NOAA GOES-11 data). Also, these very high energy protons primarily impacted
12 the middle to lower stratosphere (10 – 100 hPa, see Figure 2), thus the influence on this lower
13 region of the atmosphere is diluted by the increased number density of molecules (compared
14 to the mesosphere).

15 Since the NO_x family rapidly converts in the stratosphere to other constituents in the odd
16 nitrogen group

17 ($\text{NO}_y = \text{N}(^4\text{S}) + \text{N}(^2\text{D}) + \text{NO} + \text{NO}_2 + \text{NO}_3 + 2\text{N}_2\text{O}_5 + \text{HNO}_3 + \text{HO}_2\text{NO}_2 + \text{ClONO}_2 + \text{BrONO}_2$), it is
18 appropriate to concentrate on the NO_y impact due to the GLE. We have computed the
19 percentage change of NO_y at high Northern latitudes (60-90°N) over the January 19-23, 2005
20 period by subtracting the average of the C simulations (SPEs+GLE) from the average of the B
21 simulations (SPEs-only) and present these results in Figure 14. As a result of the GLE, odd
22 nitrogen is calculated to be enhanced by a maximum of about 0.09%, a very small increase.
23 These WACCM3 simulations indicate that inclusion of the GLE on January 20 leads to a very
24 small atmospheric perturbation.

25

26 **7 Conclusions**

27 The January 2005 SPEs caused large enhancements in the Northern polar mesospheric HO_x
28 and NO_x constituents, which were both observed and modeled. Aura MLS observations
29 indicated large mesospheric increases in OH (up to 4 ppbv) and HO_2 (>0.5 ppbv) as a result of
30 the SPEs during the time period January 16-21 in the 60-85°N latitude band. The WACCM3
31 simulations showed quantitatively similar enhancements in OH, however, the simulations

1 indicated somewhat larger HO₂ enhancements than measured by MLS. These large HO_x
2 enhancements led to considerable MLS-measured and predicted ozone decreases of greater
3 than 40% throughout most of the Northern polar mesosphere during the SPE period. MIPAS
4 measured H₂O₂ enhancements through the stratosphere into the lower mesosphere (reaching
5 ~60 pptv) from Jan. 16 to Jan. 18. WACCM3 also predicted H₂O₂ increases over the same
6 period, however, these predictions were about a factor of two or so larger than observed.

7 Nitric acid measured by both MLS and MIPAS increased in the upper stratosphere during Jan.
8 16-23 when compared with Jan. 1-14, 2005, however, WACCM3 predictions indicated only
9 minor enhancements in the same time period and altitude range, which suggests the model is
10 lacking ion chemical reactions responsible for the SPE-caused creation of HNO₃ (Verronen et
11 al., 2008). MIPAS observations showed large enhancements of polar middle mesospheric
12 NO₂ before the SPEs, which were likely the result of NO_x winter descent from higher
13 altitudes (also, see GOMOS measurements in Seppälä et al., 2007). However during the
14 SPEs, WACCM3 simulated a mesospheric NO₂ enhancement of greater than 30 ppbv in the
15 60-65 km (0.1-0.04 hPa) altitude region for Jan. 18-20, 2005 in the polar Northern
16 Hemisphere, which is in reasonable agreement with inferred MIPAS NO₂ increases over the
17 same altitude region. WACCM3 predictions are in reasonable agreement with SCISAT-1
18 ACE measurements of NO_x enhancements for the Northern Hemisphere. The observed and
19 predicted enhancements are considerable for the mesosphere and led to statistically significant
20 NO_x increases in polar Northern latitudes for about a month past the SPEs. We found that
21 protons of energies 300 to 20,000 MeV, not normally included in our computations, led to
22 enhanced stratospheric NO_y of less than 0.1% as a result of this GLE. Thus, protons with
23 energies less than 300 MeV had a much larger impact on the middle atmosphere in January
24 2005 than higher energy protons from the GLE.

25

26 **Acknowledgements**

27 We thank NASA Headquarters Living With a Star Targeted Research and Technology
28 Program for support during the time that this manuscript was written. CER was supported by
29 NASA LWS grants NNX08AU44G and NNX06AC05G. We thank Gordon J. Labow
30 (Science Systems and Applications, Inc.) for help with plotting Figures 3 and 4. We thank the
31 NOAA GOES team for providing the solar proton flux data over the Internet. The National
32 Center for Atmospheric Research is sponsored by the National Science Foundation.

1 WACCM3 results presented in this paper were generated using NASA's Columbia
2 supercomputer housed at the NASA Ames Research Center. The ACE mission is supported
3 primarily by the Canadian Space Agency. Some support was also provided by the UK
4 Natural Environment Research Council, NERC. MIPAS work presented in this paper was
5 funded by the project MANOXUVA within the DFG priority project 1176 CAWSES. The
6 IAA team was supported by the Spanish MICINN under project AYA2008-03498/ESP and
7 EC FEDER funds. MIPAS level-1b data have been provided by ESA.

8

1 **References**

- 2 Bernath, P.F., et al.: Atmospheric Chemistry Experiment (ACE): mission overview, *Geophys.*
3 *Res. Lett.*, 32, L15S01, doi:10.1029/2005GL022386, 2005. See <http://www.ace.uwaterloo.ca/>
- 4 Canty, T., Pickett, H. M., Salawitch, R. J., Jucks, K. W., Traub, W. A., and Waters, J. W.:
5 Stratospheric and mesospheric HOx: Results from Aura MLS and FIRS-2, *Geophys. Res.*
6 *Lett.*, 33, L12802, doi:10.1029/2006GL025964, 2006.
- 7 Damiani, A., Storini, M., Laurenza, M., and Rafanelli, C.: Solar particle effects on minor
8 components of the Polar atmosphere, *Ann. Geophys.*, 26, 361–370, 2008.
- 9 Damiani, A., Diego, P., Laurenza, M., Storini, M., and Rafanelli, C.: Ozone variability related
10 to SEP events occurring during solar cycle no. 23, *Adv. Space Res.*, 43, 28-40, 2009.
- 11 Damiani, A., Storini, M., Rafanelli, C., and Diego, P.: The hydroxyl radical as an indicator of
12 SEP fluxes in the high-latitude terrestrial atmosphere, *Adv. Space Res.*, 46, 1225-1235, 2010.
- 13 Forkman, P., Eriksson, P. Winnberg, A., Garcia, R. R., and Kinnison, D.: Longest continuous
14 ground-based measurements of mesospheric CO, *Geophys. Res. Lett.*, 30, 1532,
15 doi:10.1029/2003GL016931, 2003.
- 16 Garcia, R. R., Marsh, D. R., Kinnison, D. E., Boville, B. A., and Sassi, F.: Simulation of
17 secular trends in the middle atmosphere, 1950-2003, *J. Geophys. Res.*, 112, D09301
18 doi:10.1029/2006JD007485, 2007.
- 19 Gopalswamy, N., Xie, H., Yashiro, S., and Usoskin, I.: Coronal mass ejections and ground
20 level enhancements, 29th International Cosmic Ray Conference Pune, 00, 101-104, 2005.
- 21 Jackman, C. H., and McPeters, R. D.: The response of ozone to solar proton events during
22 solar cycle 21: A theoretical interpretation, *J. Geophys. Res.*, 90, 7955-7966, 1985.
- 23 Jackman, C. H., Frederick, J. E., and Stolarski, R. S.: Production of odd nitrogen in the
24 stratosphere and mesosphere: An intercomparison of source strengths, *J. Geophys. Res.*, 85,
25 7495-7505, 1980.
- 26 Jackman, C. H., Fleming, E. L., and Vitt, F. M.: Influence of extremely large solar proton
27 events in a changing stratosphere, *J. Geophys. Res.*, 105, 11659-11670, 2000.

- 1 Jackman, C. H., McPeters, R. D., Labow, G. J., Fleming, E. L., Praderas, C. J., and Russell, J.
2 M.: Northern hemisphere atmospheric effects due to the July 2000 solar proton event,
3 *Geophys. Res. Lett.*, 28, 2883-2886, 2001.
- 4 Jackman, C. H., DeLand, M. T., Labow, G. J., Fleming, E. L., Weisenstein, D. K., Ko, M. K.
5 W., Sinnhuber, M., and Russell, J. M.: Neutral atmospheric influences of the solar proton
6 events in October-November 2003, *J. Geophys. Res.*, 110, A09S27,
7 doi:10.1029/2004JA010888, 2005.
- 8 Jackman, C. H., Roble, R. G., and Fleming, E. L.: Mesospheric dynamical changes induced
9 by the solar proton events in October-November 2003, *Geophys. Res. Lett.*, 34, L04812,
10 doi:10.1029/2006GL028328, 2007.
- 11 Jackman, C. H., Marsh, D. R., Vitt, F. M., Garcia, R. R., Fleming, E. L., Labow, G. J.,
12 Randall, C. E., López-Puertas, M., von Clarmann, T., and Stiller, G. P.: Short- and medium-
13 term atmospheric constituent effects of very large solar proton events, *Atmos. Chem. Phys.*, 8,
14 765-785, 2008.
- 15 Jackman, C. H., Marsh, D. R., Vitt, F. M., Garcia, R. R., Randall, C. E., Fleming, E. L., and
16 Frith, S. M.: Long-term middle atmospheric influence of very large solar proton events, *J.*
17 *Geophys. Res.*, 114, D11304, doi:10.1029/2008JD011415, 2009.
- 18 Kinnison, D. E., Brasseur, G. P., Walters, S., Garcia, R. R., Marsh, D. R., Sassi, F., Harvey,
19 V. L., Randall, C. E., Emmons, L., Lamarque, J. F., Hess, P., Orlando, J. J., Tie, X. X.,
20 Randel, W., Pan, L. L., Gettelman, A., Granier, C., Diehl, T., Niemeier, U., and Simmons, A.
21 J.: Sensitivity of chemical tracers to meteorological parameters in the MOZART-3 chemical
22 transport model, *J. Geophys. Res.*, 112, D20302, doi:10.1029/2006JD007879, 2007.
- 23 Klekociuk, A. R., Bombardieri, D. J., Duldig, M. L., and Michael, K. J.: Atmospheric
24 chemistry effects of the 20 January 2005 solar proton event, *Advances in Geosciences* 14:
25 *Solar Terrestrial*. 305-319, 2007.
- 26 López-Puertas, M., Funke, B., Gil-López, S., von Clarmann, T., Stiller, G. P., Höpfner, M.,
27 Kellmann, S., Fischer, H., and Jackman, C. H.: Observation of NO_x enhancement and ozone
28 depletion in the Northern and Southern Hemispheres after the October-November 2003 solar
29 proton events, *J. Geophys. Res.*, 110, A09S43, doi:10.1029/2005JA011050, 2005a.
- 30 López-Puertas, M., Funke, B., Gil-López, S., von Clarmann, T., Stiller, G. P., Höpfner, M.,
31 Kellmann, S., Mengistu Tsidu, G., Fischer, H., and Jackman, C. H.: HNO₃, N₂O₅, and

1 ClONO₂ enhancements after the October-November 2003 solar proton events, *J. Geophys.*
2 *Res.*, 110, A09S44, doi:10.1029/2005JA011051, 2005b.

3 Marsh, D. R., Garcia, R. R., Kinnison, D. E., Boville, B. A., Sassi, F., Solomon, S. C., and
4 Matthes, K.: Modeling the whole atmosphere response to solar cycle changes in radiative and
5 geomagnetic forcing, *J. Geophys. Res.*, 112, D23306, doi:10.1029/2006JD008306, 2007.

6 McPeters, R. D., Jackman, C. H., and Stassinopoulos, E. G.: Observations of ozone depletion
7 associated with solar proton events, *J. Geophys. Res.*, 86, 12,071-12,081, 1981.

8 Orsolini, Y. J., Manney, G. L., Santee, M. L., and Randall, C. E.: An upper stratospheric layer
9 of enhanced HNO₃ following exceptional solar storms, *Geophys. Res. Lett.*, 32, L12S01,
10 doi:10.1029/2004GL021588, 2005.

11 Pickett, H. M., Drouin, B. J., Canty, T., Salawitch, R. J., Fuller, R. A., Perun, V. S., Livesey,
12 N. J., Waters, J. W., Stachnik, R. A., Sander, S. P., Traub, W. A., Jucks, K. W., and
13 Minschwaner, K.: Validation of Aura Microwave Limb Sounder OH and HO₂ measurements,
14 *J. Geophys. Res.* 113, D16S30, doi:10.1029/2007JD008775, 2008.

15 Porter, H. S., Jackman, C. H., and Green, A. E. S.: Efficiencies for production of atomic
16 nitrogen and oxygen by relativistic proton impact in air, *J. Chem. Phys.*, 65, 154-167, 1976.

17 Randall, C. E., Harvey, V. L., Manney, G. L., Orsolini, Y., Codrescu, M., Sioris, C., Brohede,
18 S., Haley, C. S., Gordley, L. L., Zawodny, J. M., and Russell, J. M.: Stratospheric effects of
19 energetic particle precipitation in 2003-2004, *Geophys. Res. Lett.*, 32, L05802,
20 doi:10.1029/2004GL022003, 2005.

21 Randall, C. E., Harvey, V. L., Singleton, C. S., Bernath, P. F., Boone, C. D., and Kozyra, J.
22 U.: Enhanced NO_x in 2006 linked to strong upper stratospheric Arctic vortex, *Geophys. Res.*
23 *Lett.*, 33, L18811, doi:10.1029/2006GL027160, 2006.

24 Richter, J. H., and Garcia, R. R.: On the forcing of the Mesospheric Semi-Annual Oscillation
25 in the Whole Atmosphere Community Climate Model, *Geophys. Res. Lett.*, 33, L01806,
26 doi:10.1029/2005GL024378, 2006.

27 Rinsland, C. P., Boone, C., Nassar, R., Walker, K., Bernath, P., McConnell, J. C., and Chiou,
28 L.: Atmospheric Chemistry Experiment (ACE) Arctic stratospheric measurements of NO_x
29 during February and March 2004: Impact of intense solar flares, *Geophys. Res. Lett.*, 32,
30 L16S05, doi:10.1029/2005GL022425, 2005.

1 Sassi, F., Garcia, R. R., Boville, B. A., and Liu, H.: On temperature inversions and the
2 mesospheric surf zone, *J. Geophys. Res.*, 107, 4380, doi:10.1029/2001JD001525, 2002.

3 Sassi, F., Kinnison, D., Boville, B. A., Garcia, R. R., and Roble, R.: Effect of El Nino-
4 Southern Oscillation on the dynamical, thermal, and chemical structure of the middle
5 atmosphere, *J. Geophys. Res.*, 109, D17108, doi:10.1029/2003JD004434, 2004.

6 Seppälä, A., Verronen, P. T., Sofieva, V. F., Tamminen, J., Kyrölä, E., Rodger, C. J., and
7 Clilverd, M. A.: Destruction of the tertiary ozone maximum during a solar proton event,
8 *Geophys. Res. Lett.*, 33, L07804, doi:10.1029/2005GL025571, 2006.

9 Seppälä, A., Verronen, P. T., Sofieva, Clilverd, M. A., Randall, C. E., Tamminen, J., Sofieva,
10 V., Backman, L., Kyrölä, E.: Arctic and Antarctic polar winter NO_x and energetic particle
11 precipitation in 2002-2006, *Geophys. Res. Lett.*, 34, L12810, doi:10.1029/2007GL029733,
12 2007.

13 Solomon, S., Rusch, D. W., Gerard, J.-C., Reid, G. C., and Crutzen, P. J.: The effect of
14 particle precipitation events on the neutral and ion chemistry of the middle atmosphere, 2,
15 *Odd hydrogen, Planet. Space Sci.*, 29, 885-892, 1981.

16 Solomon, S., Reid, G. C., Rusch, D. W., and Thomas, R. J.: Mesospheric ozone depletion
17 during the solar proton event of July 13, 1982, Part II. Comparison between theory and
18 measurements, *Geophys. Res. Lett.*, 10, 257-260, 1983.

19 Tylka, A. J., and Dietrich, W. F.: A new and comprehensive analysis of proton spectra in
20 ground-level enhanced (GLE) solar particle events, *Proceedings of the 31st ICRC, Lodz, 2009*
21 (see http://icrc2009.uni.lodz.pl/proc/html/index.php_id=2.html).

22 Usoskin, I. G., Tylka, A. J., and Kovaltsov, G. A.: Ionization effect of strong particle events:
23 Low-middle atmosphere, *Proceedings of the 31st ICRC, Lodz, 2009* (see
24 http://icrc2009.uni.lodz.pl/proc/html/index.php_id=2.html).

25 Usoskin, I. G., Kovaltsov, G. A., Mironova, I. A., Tylka, A. J., and Dietrich, W. F.: Ionization
26 effect of solar particle GLE events in low and middle atmosphere, *Atmos. Chem. Phys.*
27 *Discuss.*, 10, 30405-30451, 2010.

28 Verronen, P. T., Seppälä, A., Kyrola, E., Tamminen, J., Pickett, H. M., and Turunen, E.:
29 Production of odd hydrogen in the mesosphere during the January 2005 solar proton event,
30 *Geophys Res. Lett.*, 33, L24811, doi:10.1029/2006GL028115, 2006.

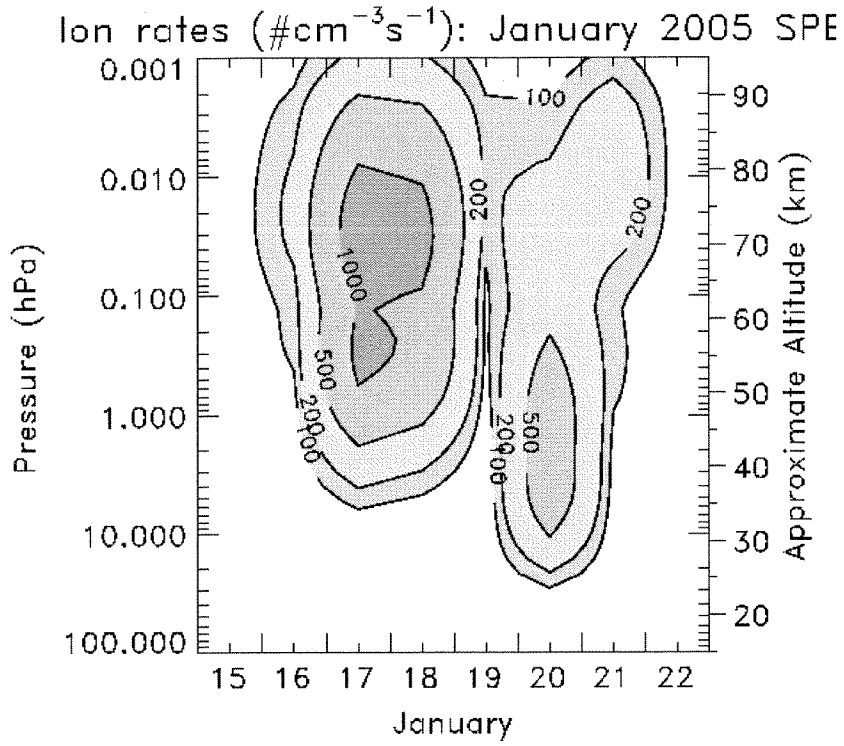
- 1 Verronen, P. T., Rodger, C. J., Clilverd, M. A., Pickett, H. M., and Turunen, E.: Latitudinal
2 extent of the January 2005 solar proton event in the Northern Hemisphere from satellite
3 observations of hydroxyl, *Ann. Geophys.*, 25, 2203-2215, 2007.
- 4 Verronen, P. T., Funke, B., López-Puertas, M., Stiller, G. P., von Clarmann, T., Glatthor, N.,
5 Enell, C.-F., Turunen, E., and Tamminen, J.: About the increase of HNO₃ in the stratopause
6 region during the Halloween 2003 solar proton event, *Geophys. Res. Lett.*, 35, L20809,
7 doi:10.1029/2008GL035312, 2008.
- 8 Versick, S., Stiller, G., Glatthor, N., Reddmann, T., Ruhnke, R., von Clarmann, T., Höpfner,
9 M., Kiefer, M., Linden, A., Kellmann, S., and Grabowski, U.: MIPAS-observations and
10 model results for H₂O₂ with focus on the SPEs 2003 and 2005, poster presented at the 2nd
11 International High Energy Particle Precipitation in the Atmosphere (HEPPA) Workshop,
12 Boulder, CO, held October 6-8, 2009.
- 13 Versick, S.: Ableitung von H₂O₂ aus MIPAS/ENVISAT-Beobachtungen und Untersuchung
14 der Wirkung von energetischen Teilchen auf den chemischen Zustand der mittleren
15 Atmosphäre, Ph.D. Dissertation (in German), Karlsruher Institute für Technologie, Karlsruhe,
16 Germany, 2010.
- 17

1 Table 1. Description of WACCM3 Simulations

Simulation designation	Number of realizations	Time Period	SPEs included	GLE included
A (1, 2, 3, 4)	4	Jan. 1 – Mar. 31, 2005	No	No
B (1, 2, 3, 4)	4	Jan. 1 – Mar. 31, 2005	Yes	No
C (1, 2, 3, 4)	4	Jan. 1 – Mar. 31, 2005	Yes	Yes

2

3



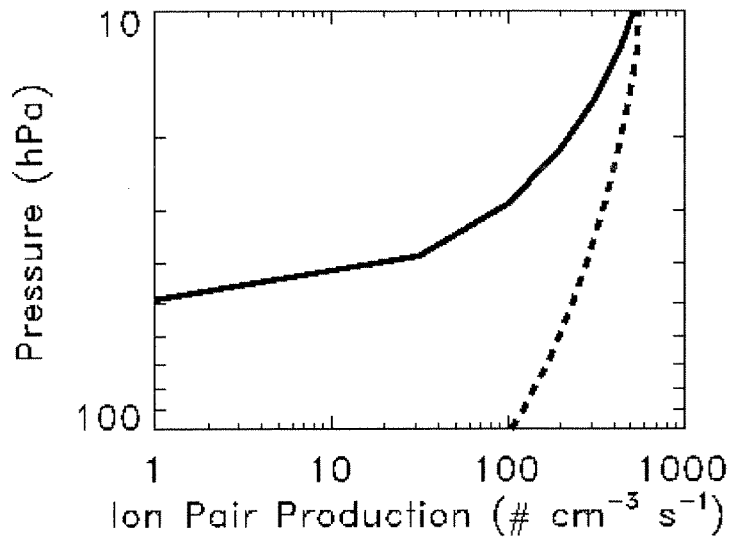
1

2 Figure 1. Daily average ion pair production rates ($\text{\#cm}^{-3}\text{s}^{-1}$) as a function of time for Jan. 15-

3 22, 2005.

4

IP Production – Jan. 20, 2005

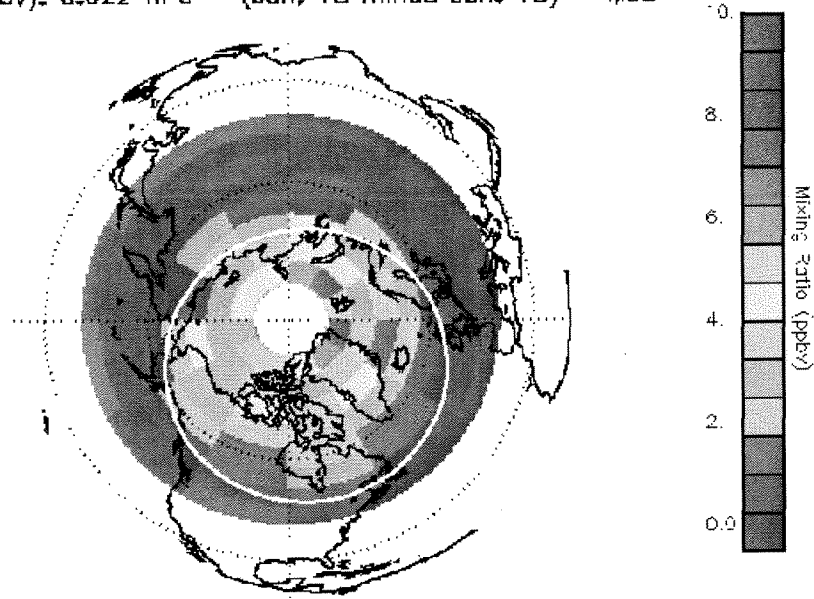


1

2 Figure 2. Daily average ion pair production rates ($\text{cm}^{-3} \text{ s}^{-1}$) computed for the "SPEs-only" case
3 (solid line) and the "SPEs+GLE" case (dashed line) on Jan. 20, 2005.

4

OH (ppbv): 0.022 hPa - (Jan, 18 minus Jan, 15) - MLS

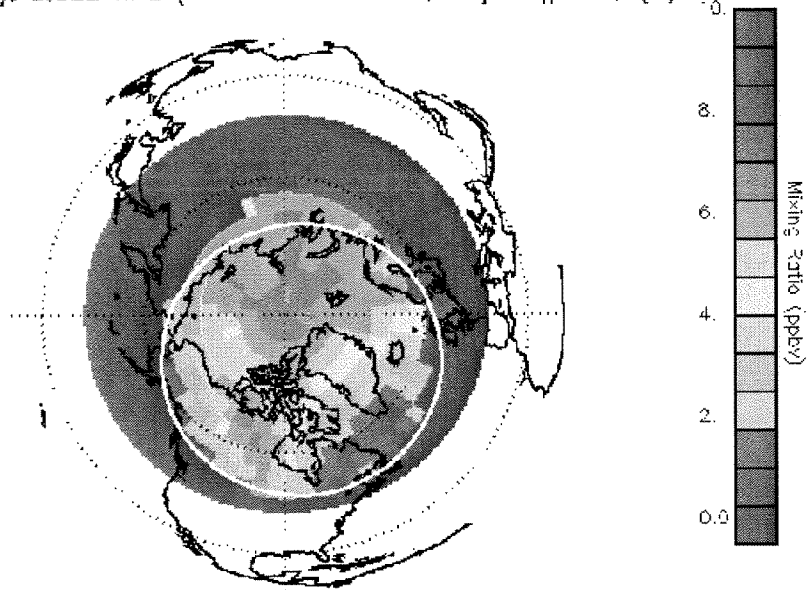


1

2 Figure 3. Aura MLS OH measurements at 0.022 hPa (~75 km) on January 18, 2005 (after
3 SPE) minus those on January 15, 2005 (before SPE). For added clarity, measurements are
4 only shown in the latitude range 42.5-82.5°N. No MLS measurements are available at 82.5-
5 90°N. The polar cap edge (60° geomagnetic latitude) is indicated by the white circle.

6

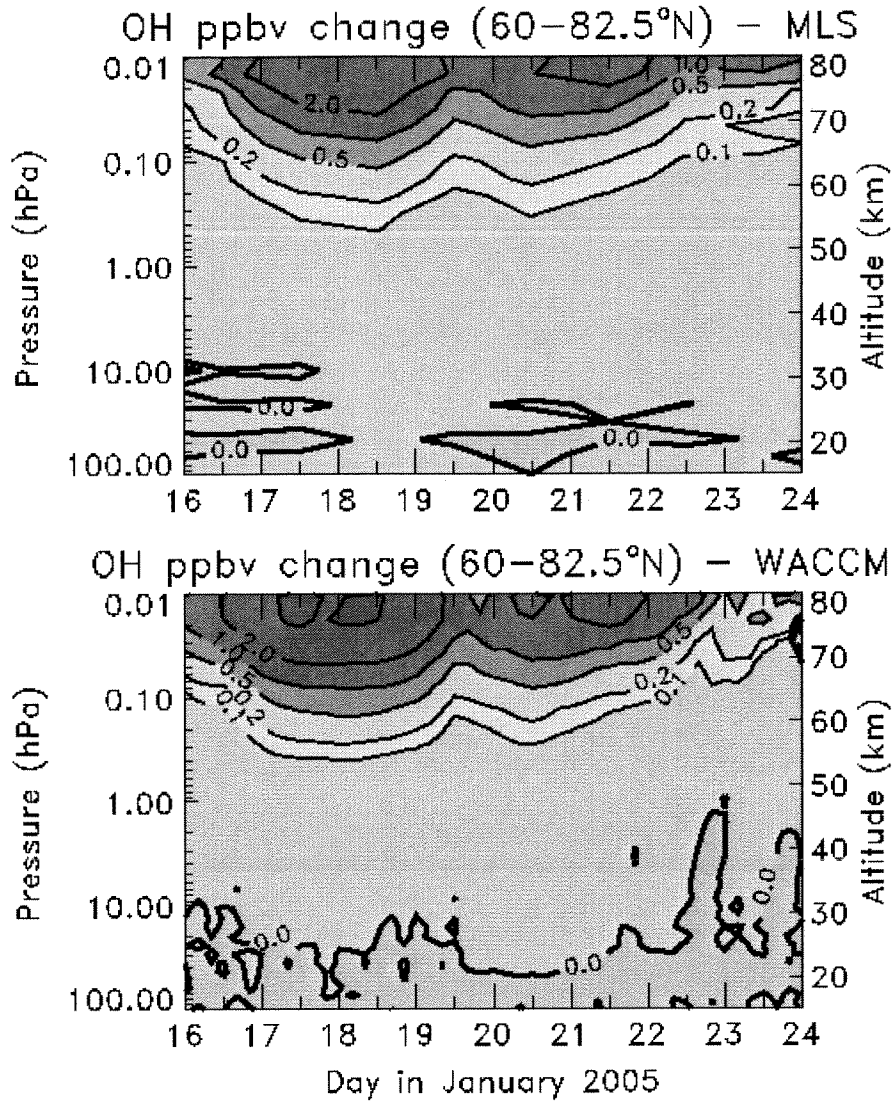
OH (ppbv): 0.022 hPa (Jan. 18 minus Jan. 15) - WACCM (B)



1

2 Figure 4. WACCM3 B1 OH predictions at 0.022 hPa (~75 km) on January 18, 2005 (after
3 SPE) minus those on January 15, 2005 (before SPE). For added clarity, the results from the
4 WACCM3 simulations are only shown from 44-90°N. The polar cap edge (60° geomagnetic
5 latitude) is indicated by the white circle.

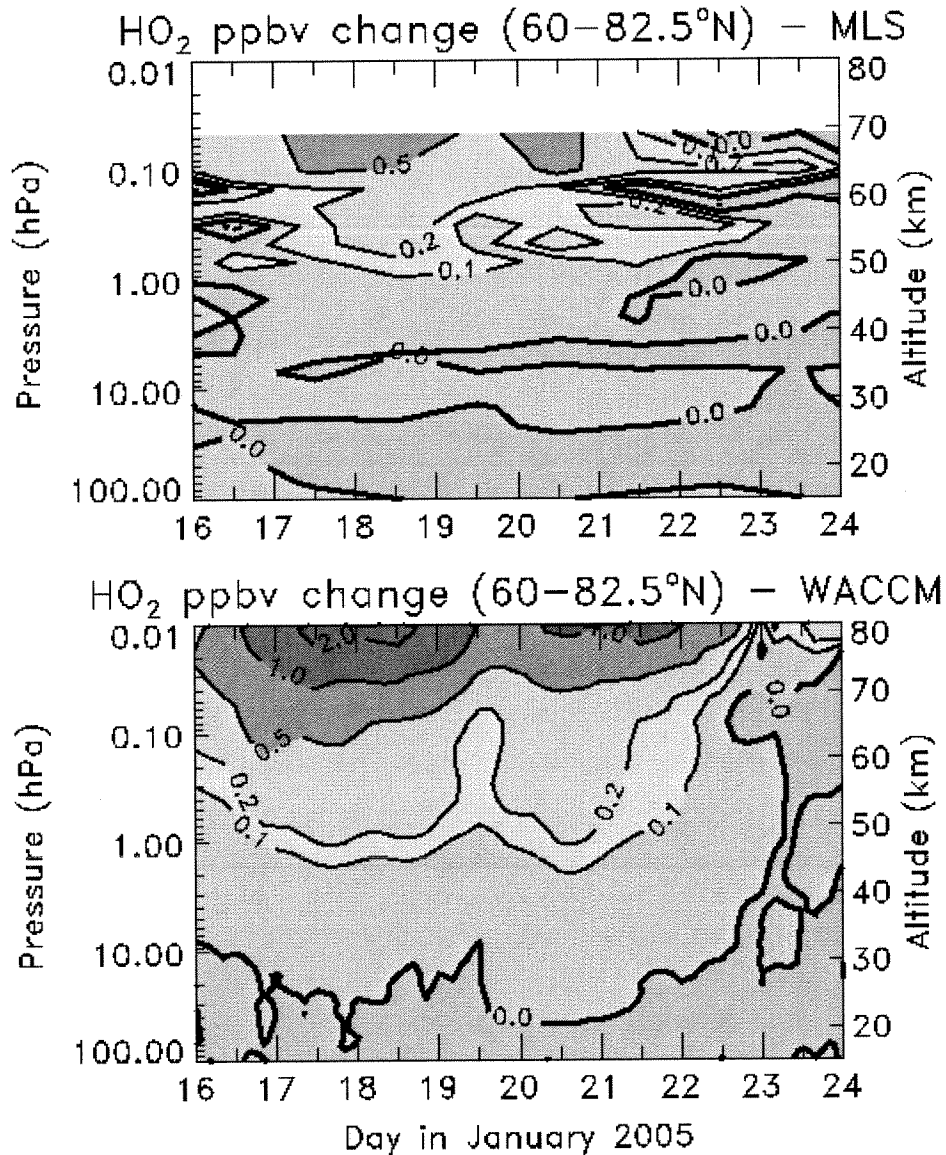
6



1

2 Figure 5. OH changes from Aura MLS measurements (top) and WACCM3 B1 predictions
 3 (bottom) for the 60-82.5°N band. An average observed (predicted) OH profile for the period
 4 January 1-14, 2005 was subtracted from the observed (predicted) OH values for the plotted
 5 days (January 16-23, 2005). The contour intervals for the OH differences are -0.2, -0.1, 0.0,
 6 0.1, 0.2, 0.5, 1, 2, and 5 ppbv.

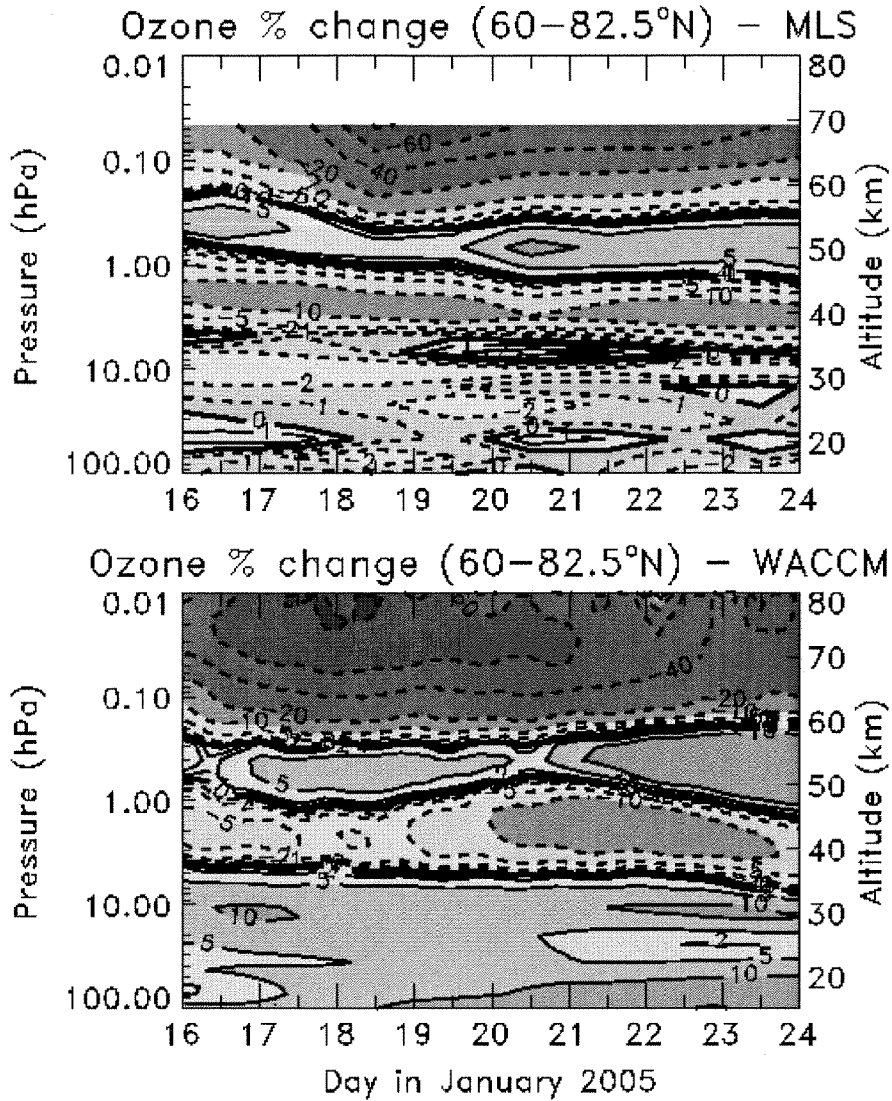
7



1

2 Figure 6. HO₂ changes from Aura MLS measurements (top) and WACCM3 B1 predictions
 3 (bottom) for the 60-82.5°N band. An average observed (predicted) HO₂ profile for the period
 4 January 1-14, 2005 was subtracted from the observed (predicted) HO₂ values for the plotted
 5 days (January 16-23, 2005). The contour intervals for the HO₂ differences are -0.1, 0.0, 0.1,
 6 0.2, 0.5, 1, and 2 ppbv.

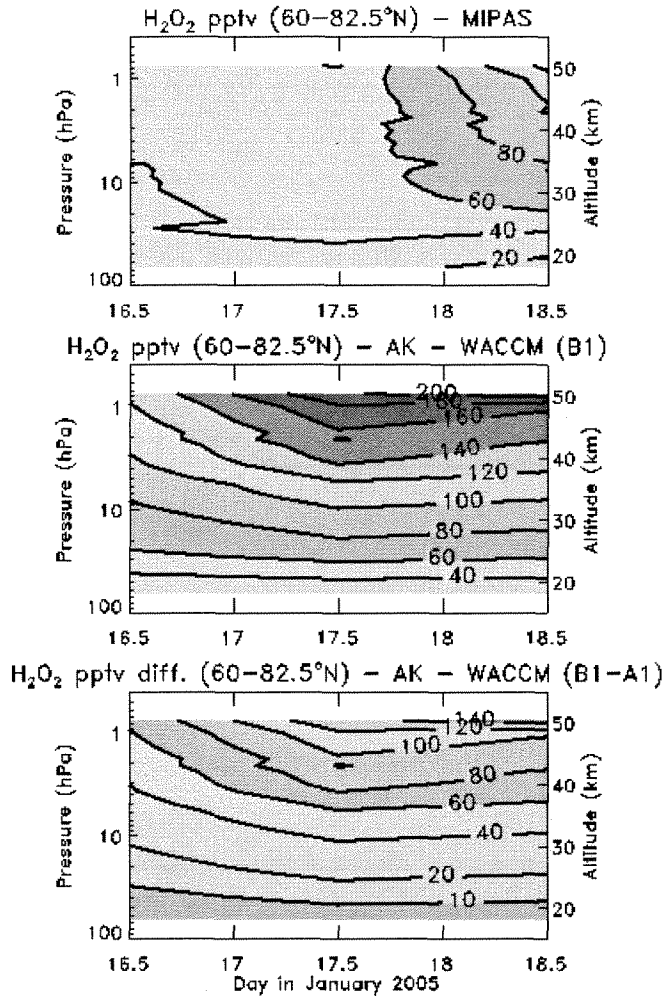
7



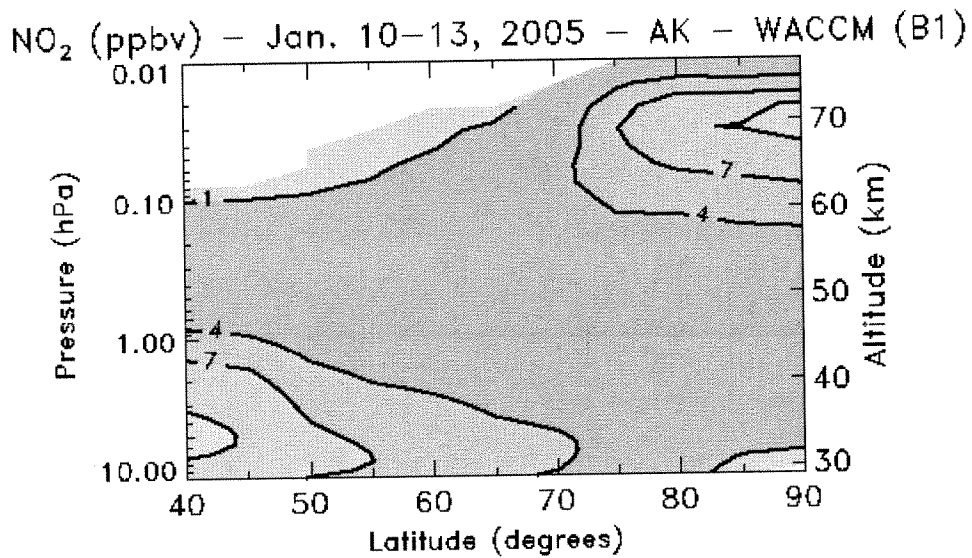
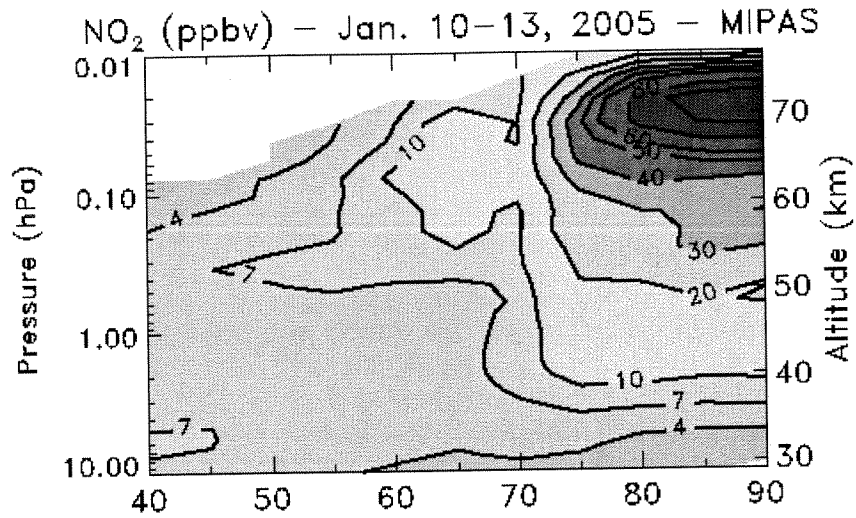
1

2 Figure 7. Ozone changes from Aura MLS measurements (top) and WACCM3 B1 predictions
 3 (bottom) for the 60-82.5°N band. An average observed (predicted) ozone profile for the
 4 period January 1-14, 2005 was subtracted from the observed (predicted) ozone values for the
 5 plotted days (January 16-23, 2005). The contour intervals for the ozone differences are -80, -
 6 60, -40, -20, -10, -5, -2, -1, 0, 1, 2, 5, and 10%.

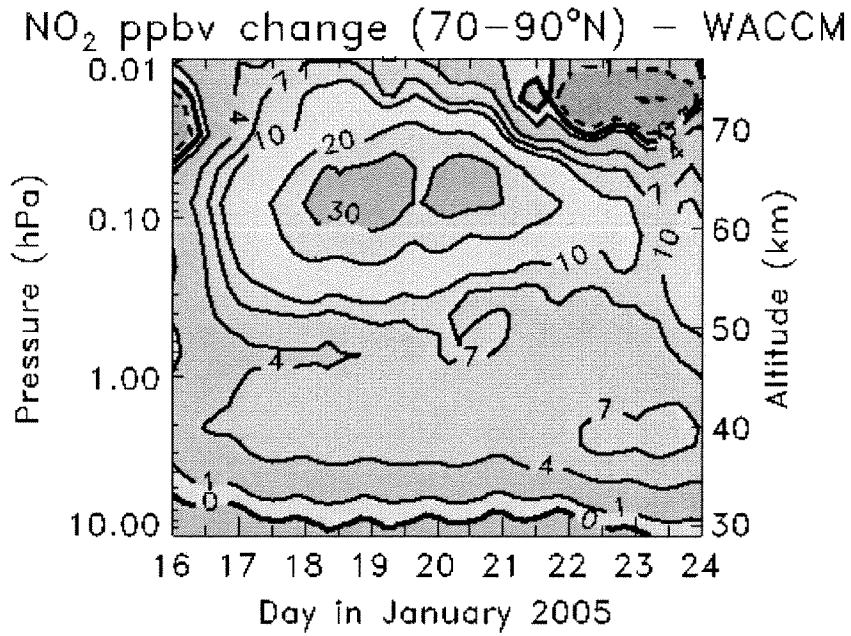
7



2 Figure 8. Hydrogen peroxide (H₂O₂): Envisat MIPAS measurements (top) and WACCM3
 3 predictions (middle, bottom) for January 16-18, 2005 in the 60-82.5°N band. The WACCM3
 4 results are from the B1 simulation (middle) and a difference between the B1 (SPes-only) and
 5 A1 (no SPes and no GLE) simulations (bottom). The MIPAS averaging kernel (AK) was
 6 used to sample the WACCM3 results. The contour intervals are 10, 20, 40, 60, 80, 100, 120,
 7 140, 160, 180, and 200 pptv.
 8



1
 2 Figure 9. Envisat MIPAS NO₂ measurements (top) and WACCM3 B1 simulation (bottom) for
 3 the four-day (January 10-13, 2005) average in the Northern Hemisphere. The MIPAS
 4 averaging kernel (AK) was used to sample the WACCM3 results. The contour intervals are
 5 1, 4, 7, 10, 20, 30, 40, 50, 60, 80, and 100 ppbv.
 6

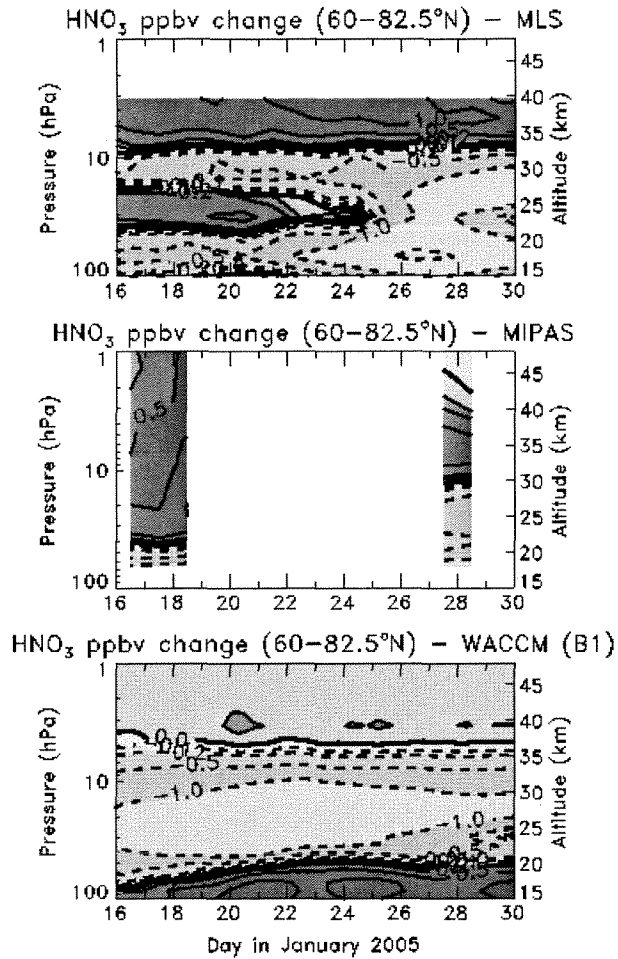


1

2 Figure 10. WACCM3 B1 (SPEs-only) simulation of NO₂ change from the four-day (January
 3 10-13, 2005) average for the 70°-90°N band. The contour intervals are -4, -1, 0, 1, 4, 7, 10,
 4 20, 30, 40, 50, 60, 80, and 100 ppbv.

5

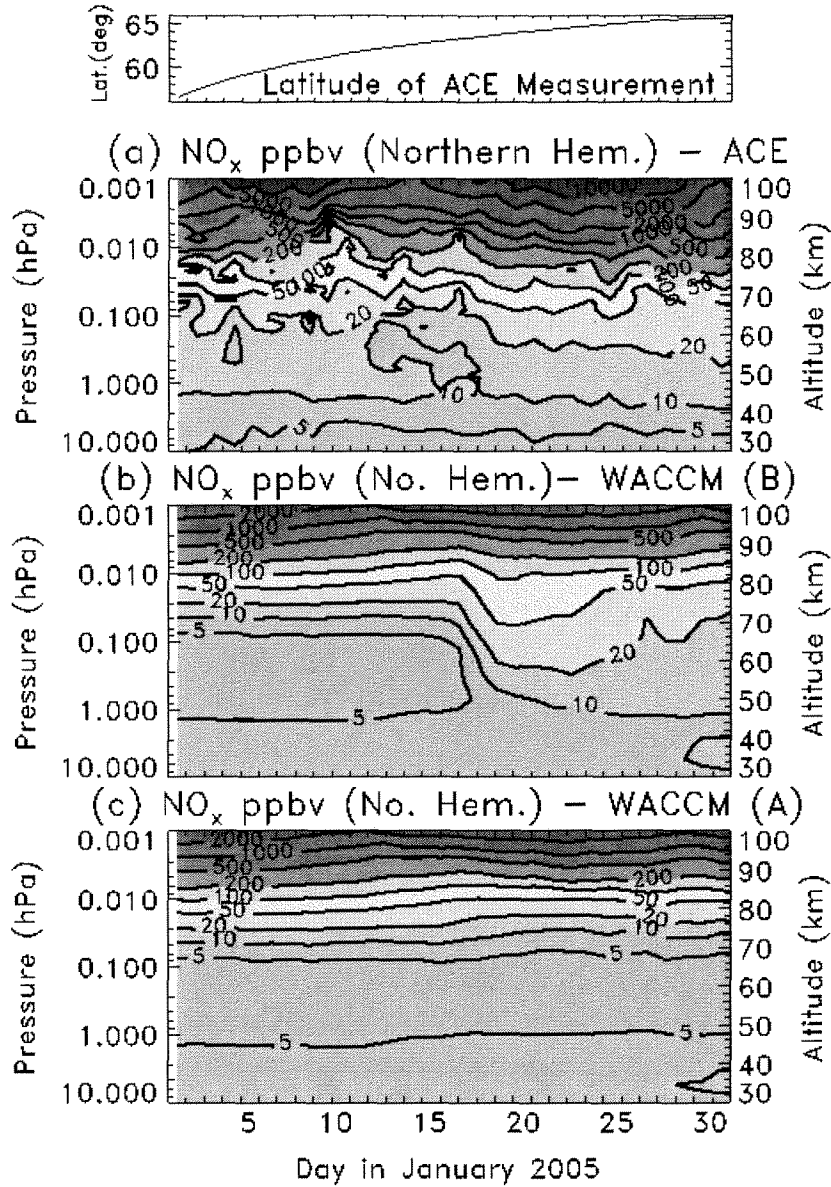
6



1

2 Figure 11. Nitric acid (HNO₃) change: Aura MLS measurements (top), Envisat MIPAS
 3 measurements (middle), and WACCM3 B1 (SPes-only) simulation (bottom) for January 16-
 4 29, 2005 in the 60°-82.5°N band. An average HNO₃ for the period January 1-14, 2005 was
 5 subtracted from the Aura MLS observed and WACCM3 B1 predicted values for the plotted
 6 days. Envisat MIPAS measurements were only available for January 10-13, 2005, and the
 7 average of these four days was subtracted from the January 16-18 and 27-28, 2005 values.
 8 The contour intervals are -2, -1, -0.5, -0.2, -0.1, 0, 0.1, 0.2, 0.5, and 1 ppbv.

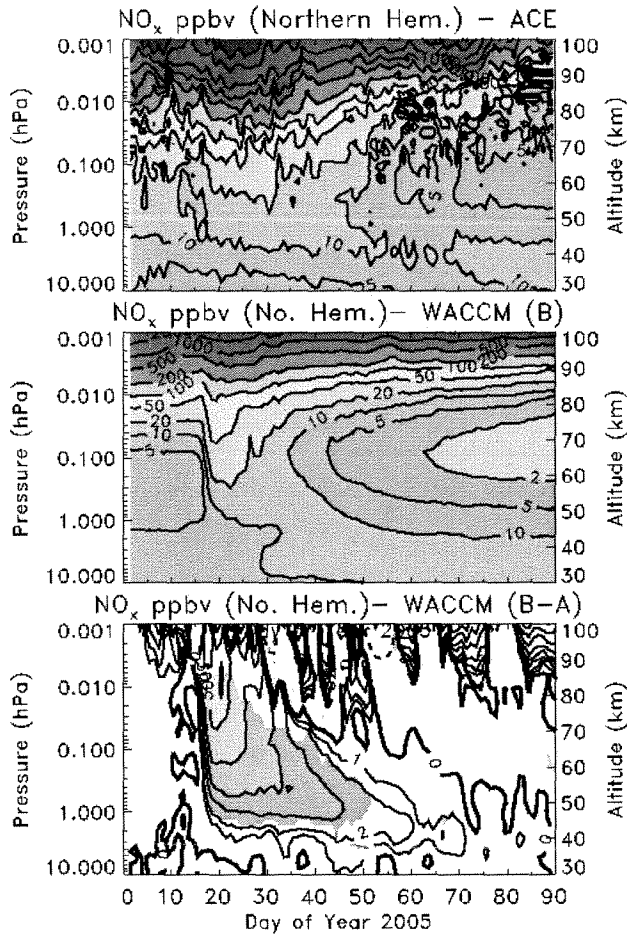
9



1

2 Figure 12. NO_x measurements (a) and predictions (b,c) for January 1-31, 2005 in the high
 3 latitude Northern Hemisphere (see section 5.2.3). The ACE NO_x measurements are given in
 4 (a). The WACCM3 NO_x predictions are from an average of the B simulations (SPEs-only, b)
 5 and the A simulations (no SPEs and no GLE, c). The contour intervals for NO_x are 1, 2, 5,
 6 10, 20, 50, 100, 200, 500, 1000, 2000, 5000, and 10000 ppbv. The latitudes of ACE
 7 measurements are given in the top plot.

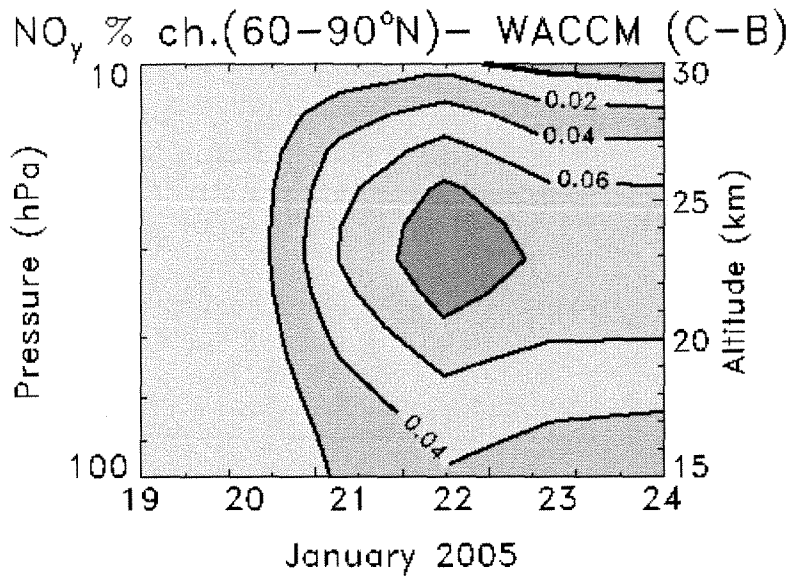
8



1

2 Figure 13. SCISAT-1 ACE measurements (top) and WACCM3 predictions (middle, bottom)
 3 for NO_x during the first 90 days of 2005 (January 1 - March 31) for the high latitude Northern
 4 Hemisphere. The WACCM3 NO_x predictions (middle) are from an average of the B
 5 simulations (SPEs-only) and the WACCM3 NO_x predictions (bottom) show the NO_x
 6 enhancement due to the SPEs [the average of the B simulations (SPEs-only) minus the
 7 average of the A simulations (no SPEs and no GLE)]. The colored regions indicate 95%
 8 statistical significance with the use of Student's t test. The contour intervals are 1, 2, 5, 10,
 9 20, 50, 100, 200, 500, 1000, 2000, 5000, and 10000 ppbv.

10



1

2 Figure 14. WACCM3 predicted of polar Northern Hemisphere (60-90°N) NO_y percentage
 3 enhancement due to the GLE [the average of the C simulations (SPEs+GLE) minus the
 4 average of the B simulations (SPEs-only)]. The contour intervals are 0.0, 0.02, 0.04, 0.06,
 5 and 0.08%.