### **Old Dominion University**

### **ODU Digital Commons**

Chemistry & Biochemistry Faculty Publications

Chemistry & Biochemistry

2012

# Atmospheric Effects of Energetic Particle Precipitation in the Arctic Winter 1978-1979 Revisted

L. A. Holt University of Colorado at Boulder

C. E. Randall University of Colorado at Boulder

V. L. Harvey University of Colorado at Boulder

E. E. Remsberg
NASA Langley Research Center

G. P. Stiller
Karlsruhe Institute of Technology

See next page for additional authors

Follow this and additional works at: https://digitalcommons.odu.edu/chemistry\_fac\_pubs

Part of the Chemistry Commons, Oceanography and Atmospheric Sciences and Meteorology Commons, and the Other Earth Sciences Commons

### **Original Publication Citation**

Holt, L. A., Randall, C. E., Harvey, V. L., Remsberg, E. E., Stiller, G. P., Funke, B., Bernath, P. F., & Walker, K. A. (2012) Atmospheric effects of energetic particle precipitation in the Arctic winter 1978-1979 revisited. *Journal of Geographical Research: Atmospheres, 117*(5) 1-14, Article D05315. https://doi.org/10.1029/2011JD016663

This Article is brought to you for free and open access by the Chemistry & Biochemistry at ODU Digital Commons. It has been accepted for inclusion in Chemistry & Biochemistry Faculty Publications by an authorized administrator of ODU Digital Commons. For more information, please contact digitalcommons@odu.edu.

Authors L. A. Holt, C. E. Randa	l, V. L. Harvey, E. E. Remsberg, G. P. Stiller, B. Funke, P. F. Bernath, and K. A. Walker

## Atmospheric effects of energetic particle precipitation in the Arctic winter 1978–1979 revisited

L. A. Holt, <sup>1,2</sup> C. E. Randall, <sup>1,2</sup> V. L. Harvey, <sup>1</sup> E. E. Remsberg, <sup>3</sup> G. P. Stiller, <sup>4</sup> B. Funke, <sup>5</sup> P. F. Bernath, <sup>6</sup> and K. A. Walker <sup>7</sup>

Received 2 August 2011; revised 12 January 2012; accepted 13 January 2012; published 15 March 2012.

[1] The Limb Infrared Monitor of the Stratosphere (LIMS) measured polar stratospheric enhancements of NO<sub>2</sub> mixing ratios due to energetic particle precipitation (EPP) in the Arctic winter of 1978–1979, Recently reprocessed LIMS data are compared to more recent measurements from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) and the Atmospheric Chemistry Experiment Fourier transform spectrometer (ACE-FTS) to place the LIMS measurements in the context of current observations. The amount of  $NO_x$  (NO + NO<sub>2</sub>) entering the stratosphere that has been created by EPP in the mesosphere and lower thermosphere (EPP-NO<sub>x</sub>) has been quantified for the 1978–1979 and 2002–2003 through 2008–2009 Arctic winters. The NO<sub>2</sub> enhancements in the LIMS data are similar to those in MIPAS and ACE-FTS data in the Arctic winters of 2002–2003, 2004–2005, 2006–2007, and 2007–2008. The largest enhancement by far is in 2003–2004 ( $\sim$ 2.2 Gmol at 1500 K), which is attributed to a combination of elevated EPP and unusual dynamics that led to strong descent in the upper stratosphere/lower mesosphere in late winter. The enhancements in 2005–2006 and 2008–2009, during which large stratospheric NO<sub>x</sub> enhancements were caused by a dynamical situation similar to that in 2003–2004, are larger than in all the other years (except 2003–2004) at 3000 K. However, by 2000 K the enhancements in 2005-2006 (2008-2009) are on the same order of magnitude as (smaller than) all other years. These results highlight the importance of the timing of the descent in determining the potential of EPP-NO<sub>x</sub> for reaching the middle stratosphere.

Citation: Holt, L. A., C. E. Randall, V. L. Harvey, E. E. Remsberg, G. P. Stiller, B. Funke, P. F. Bernath, and K. A. Walker (2012), Atmospheric effects of energetic particle precipitation in the Arctic winter 1978–1979 revisited, *J. Geophys. Res.*, 117, D05315, doi:10.1029/2011JD016663.

### 1. Introduction

[2] The key catalytic cycle responsible for ozone loss in the stratosphere between about 25 and 40 km is the  $NO_x$  (NO +  $NO_2$ ) catalytic cycle [e.g., *Crutzen*, 1970; *Garcia and Solomon*, 1994; *Watson et al.*, 1986]. The primary source of  $NO_x$  in the stratosphere is NO produced from the oxidation of  $N_2O$  of tropospheric origin [e.g., *Bates and Hays*, 1967; *Crutzen*, 1971; *McElroy and McConnell*, 1971; *Nicolet*, 1971]. Another source of stratospheric  $NO_x$  is NO

produced by energetic particle precipitation (EPP). EPP ionizes the atmosphere, resulting in formation of NO at an altitude dependent upon the energy of the precipitating particles [e.g., *Crutzen et al.*, 1975; *Gylvan Meira*, 1971; *Narcisi et al.*, 1972; *Rusch et al.*, 1981]. The  $NO_x$  so produced is referred to as EPP- $NO_x$ .

[3] NO in the polar winter thermosphere above 100 km is produced mainly from routine precipitation of low-energy (auroral) electrons (energy < 30 keV) and protons (energy < 1 MeV) and subsequent reaction of excited N(<sup>2</sup>D) and O<sub>2</sub> [Thorne, 1980]. Regular precipitation of medium energy electrons (30-300 keV) results in NO production at mesospheric altitudes [Codrescu et al., 1997]. NO is rapidly photodissociated in the sunlit mesosphere and thermosphere, but in the polar night region can be transported to the stratosphere. Once NO reaches the lower mesosphere, where O<sub>3</sub> concentrations become significant, it can react with O<sub>3</sub> to produce NO<sub>2</sub> [e.g., Cohen and Murphy, 2003]. The process by which NO<sub>x</sub> created in the upper atmosphere is transported to the stratosphere is called the EPP indirect effect (EPP IE), and was predicted by Solomon et al. [1982] using a 2-D model. Satellite evidence of this phenomenon was first

Copyright 2012 by the American Geophysical Union. 0148-0227/12/2011JD016663

**D05315** 1 of 14

<sup>&</sup>lt;sup>1</sup>Laboratory for Atmospheric and Space Physics, University of Colorado at Boulder, Boulder, Colorado, USA.

<sup>&</sup>lt;sup>2</sup>Department of Atmospheric and Oceanic Sciences, University of Colorado at Boulder, Boulder, Colorado, USA.

<sup>&</sup>lt;sup>3</sup>NASA Langley Research Center, Hampton, Virginia, USA.

<sup>&</sup>lt;sup>4</sup>Institute for Meteorology and Climate Research, Karlsruhe Institute of Technology, Karlsruhe, Germany.

<sup>&</sup>lt;sup>5</sup>Instituto de Astrofísica de Andalucía, CSIC, Granada, Spain.

<sup>&</sup>lt;sup>6</sup>Department of Chemistry, University of York, York, UK.

<sup>&</sup>lt;sup>7</sup>Department of Physics, University of Toronto, Toronto, Ontario, Canada.

obtained from the Limb Infrared Monitor of the Stratosphere (LIMS) during the Northern Hemisphere (NH) winter of 1978–1979 [Russell et al., 1984]. It has since been observed a number of times, along with evidence for the destruction of O<sub>3</sub> by EPP-NO<sub>x</sub> [e.g., Callis et al., 1996, 1998a, 1998b; Funke et al., 2005a; López-Puertas et al., 2005; Randall et al., 1998, 2001, 2005, 2007, 2009; Rinsland et al., 1996, 1999; Seppälä et al., 2004, 2007]. NO is also produced in situ when high-energy electrons (E > 300 keV) and protons (E > 30 MeV) deposit their energy in the stratosphere, but this happens sporadically during periods of strong geomagnetic activity.

- [4] Measurements of Southern Hemisphere (SH) NO<sub>x</sub> and tracers show that variations in the amount of EPP-NO<sub>x</sub> descending to the SH stratosphere depend mainly on the level of geomagnetic activity. That is, the interannual variability of SH stratospheric NO<sub>x</sub> correlates well with the Ap index and auroral and medium energy electron hemispheric power. For example, Randall et al. [2007, hereinafter R07] documented this correlation using data from solar occultation instruments, and showed that in years with high geomagnetic activity the SH EPP IE contributed up to 40% of the annual source of polar stratospheric NO<sub>x</sub>. That the correlation was so strong was attributed in part to the fact that variability in SH dynamics is small, and thus has little effect on interannual variations in the amount of EPP-NO<sub>x</sub> transported downward to the stratosphere. However, the same correlation between the Ap index or energetic particle hemispheric power and the EPP IE is not found in the NH. This hemispherical discrepancy has been substantiated by NO<sub>x</sub> measurements from the Halogen Occultation Experiment (HALOE) and attributed to dynamical variability in the NH through a 2-D chemical transport model [Siskind et al., 1997]. Using data from the Global Ozone Monitoring by Occultation of Stars (GOMOS) instrument, Seppälä et al. [2007] found that NH stratospheric EPP-NO<sub>x</sub> enhancements from 46 to 56 km correlated well with the Ap index from 2002 to 2003 through 2005-2006, but these calculations included only the months of October through January.
- [5] A salient discovery in recent years is that even in periods of minimal geomagnetic activity enhancements in stratospheric NO<sub>x</sub> mixing ratios due to the EPP IE can be as large as in years with high geomagnetic activity. This was first illustrated in the NH winter of 2005–2006. Although geomagnetic activity was low, observations showed large enhancements of EPP-NO<sub>x</sub> in the stratosphere [Randall et al., 2006; Siskind et al., 2007]. A similar scenario was observed again in 2009 when EPP-NO<sub>x</sub> enhancements in the uppermost stratosphere were up to 50 times higher than average in spite of low geomagnetic activity [Randall et al., 2009]. These enhancements were attributed to unusual dynamical conditions caused by a remarkable recovery from a major sudden stratospheric warming (SSW). This recovery resulted in strong descent in the mesosphere and upper stratosphere, as indicated by an elevated stratopause, and a strong upper stratospheric vortex that sequestered air in the polar region. The precise phenomena that trigger these unusual conditions are not yet understood. Once initiated, however, the reversal of the zonal winds during the SSW prevents the upward propagation of planetary waves, which allows the vortex to reform with strong westerly winds that favor the propagation of gravity waves. Gravity waves then act to

decelerate the westerly flow, which creates a poleward meridional flow leading to enhanced downward motion at the winter pole [e.g., *Hauchecorne et al.*, 2007].

- [6] Following the LIMS measurements in 1978-1979 until the launch of the Upper Atmosphere Research Satellite (UARS) in 1991, there were few reports of stratospheric EPP-NO<sub>x</sub> enhancements; it is not known, however, if this is due to the fact that such enhancements did not occur, or to the lack of appropriate data in the polar winter. This raises the question of why the EPP-NO<sub>x</sub> enhancements were seen so clearly in the LIMS data: Were they observed because enhanced EPP or unusual meteorology led to higherthan-normal stratospheric NO<sub>x</sub> mixing ratio enhancements, as in 2004, 2006, and 2009? Or were the mixing ratio enhancements typical for NH winters, but easily observed because of the capability of LIMS to view the polar night? In this study, we compare the EPP-NO<sub>x</sub> enhancements in the LIMS data to more recent satellite data from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) and Atmospheric Chemistry Experiment Fourier transform spectrometer (ACE-FTS) to determine whether the LIMS enhancements are unusual in the context of current observations. We quantify the amount of EPP-NO<sub>x</sub> entering the stratosphere in the 1978–1979 Arctic winter and in the winters observed by MIPAS and ACE-FTS.
- [7] The paper is structured as follows. Section 2 gives a review of the satellite observations that we use. In section 3 we compare the temporal evolution of  $NO_x$  between the different instruments. We then discuss the dynamics and geomagnetic activity for each of the years. In section 4 we quantify the amount of EPP- $NO_x$  descending past the 1500 K, 2000 K and 3000 K isentropic surfaces. Section 5 includes the summary and conclusions.

2156202d, 2012, DS, Downloaded from https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2011JD016663 by Old Dominion University, Wiley Online Library on [0908/2023]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Cerative Commons

### 2. Observations

### 2.1. LIMS

[8] LIMS was a thermal infrared limb scanning radiometer with six channels between 6.2 and 15.0 microns [Gille and Russell, 1984]. It measured vertical radiance profiles from which temperature, water vapor (H<sub>2</sub>O), O<sub>3</sub>, NO<sub>2</sub> and nitric acid (HNO<sub>3</sub>) were retrieved. LIMS was launched on the NIMBUS 7 spacecraft into a near polar, Sun-synchronous orbit with an inclination of 99.1° and an altitude of 955 km on 24 October 1978, and operated until 28 May 1979. It collected data day and night with near global coverage from 64°S to 84°N every 6 days, returning more than 7000 radiance profiles each day. The current study utilizes level 2 of the reprocessed or Version 6 (V6) LIMS data set [Remsberg] et al., 2004, 2010]. The vertical resolution of the V6 NO<sub>2</sub> is approximately 3.7 km and the nighttime NO<sub>2</sub> mixing ratios extend from approximately 50 hPa to the lower mesosphere, at least in the polar night. The precision (accuracy) of the updated V6 NO<sub>2</sub> is approximately 3% (18%) from 3 to 10 hPa, 7% (30%) at 30 hPa and 14% (30%) at 1 hPa [Remsberg et al., 2010]. The accuracy of LIMS V6 NO<sub>2</sub> in the upper stratosphere is better than the previous versions because of improved spectral line parameters for the NO<sub>2</sub> forward radiance model.

#### 2.2. MIPAS

[9] MIPAS can measure NO<sub>2</sub> and NO in the polar night stratosphere, and is thus well suited for comparing to the LIMS measurements. MIPAS is a high-resolution Fourier transform spectrometer operating in the midinfrared (685-2410 cm<sup>-1</sup>) aboard the Environmental Satellite (ENVISAT), which was launched into a Sun-synchronous orbit with an inclination of 98.55° and an altitude of 800 km on 1 March 2002 [European Space Agency, 2000; Fischer et al., 2008]. It is a limb emission instrument that collects data day and night with global coverage, and returns up to 72 scans in each of its daily 14.3 orbits. MIPAS has a vertical resolution of 3 km in the altitude range of 6-68 km. Reprocessed version 4.61/4.62 of the operational European Space Agency (ESA) NO<sub>2</sub> data was used here for comparison to LIMS. Wetzel et al. [2007] reported an overall accuracy of approximately 10%-20% and a precision of around 5%-15% for version 4.61 MIPAS NO<sub>2</sub> below about 45 km. They compared MIPAS NO2 to observations from balloons, satellites, and ground-based measurements. In addition to NO<sub>2</sub>, we used ESA CH<sub>4</sub> [e.g., Raspollini et al., 2006] as a tracer of vertical motion. We have also used a special scientific version of the MIPAS data from the Institute for Meteorology and Climate Research/Instituto de Astrofísica de Andalucía (IMK/IAA) (MIPAS-IMK/IAA) [Funke et al., 2005b; von Clarmann et al., 2003]. MIPAS-IMK/IAA has both NO and NO2. MIPAS-IMK/IAA NO2 and NO data were compared to ACE-FTS data [Kerzenmacher et al., 2008], resulting in typical differences for NO of 20% in the range 42–60 km, 10% in the range 15–42 km, and 20% for NO<sub>2</sub> in the range 28–44 km.

[10] Results from the MIPAS instrument have been detailed in several papers to date, many of which include studies of the EPP IE. A handful of the most relevant are outlined here. López-Puertas et al. [2005] looked at MIPAS data to study the stratospheric NO<sub>x</sub> enhancements and subsequent O<sub>3</sub> depletion in the polar regions resulting from solar proton events (SPEs) during the solar storms in October and November of 2003. Funke et al. [2005a, hereinafter F05] used MIPAS data to quantify the amount of EPP-NO<sub>x</sub> transported to the polar stratosphere in the 2003 Antarctic winter. In their study they used CH<sub>4</sub> and CO as dynamical tracers to determine the origin of the NO<sub>x</sub>-enhanced air. Stiller et al. [2005] looked at vertical profiles of stratospheric HNO<sub>3</sub> from MIPAS. They reported a second maximum in HNO<sub>3</sub> at approximately 34 km, which they attributed to ion cluster chemistry and/or heterogeneous chemistry made possible by N2O5 produced by a large EPP IE between May and August 2003. López-Puertas et al. [2006] investigated stratospheric and mesospheric NO<sub>v</sub> in the polar winters of both hemispheres from 2002 to 2004. They found very high interannual as well as interhemispheric variability, which they examined in terms of dynamics and solar activity.

### 2.3. ACE-FTS

[11] ACE-FTS is a 0.02 cm<sup>-1</sup> resolution Fourier transform spectrometer operating in the midinfrared (750–4400 cm<sup>-1</sup>) aboard SCISAT-1, which was launched into a circular low Earth orbit with an inclination of 74° and an altitude of 650 km on 12 August 2003 [Bernath et al., 2005]. Using

solar occultation it measures over 20 atmospheric constituents, including NO, NO<sub>2</sub> and CH<sub>4</sub>, in two latitude circles (at satellite sunrise and sunset) between 85°S and 85°N each day. Since it is a solar occultation instrument, it does not take measurements in the polar night. Version 2.2 of the ACE-FTS data was used in this study [Boone et al., 2005].

[12] Initial validation of ACE-FTS NO<sub>x</sub> by McHugh et al. [2005] showed agreement of ACE-FTS NO<sub>x</sub> with the Halogen Occultation Experiment (HALOE) to within 20% between 22 and 55 km. They also showed that ACE-FTS CH<sub>4</sub> was about 10% higher than HALOE throughout the stratosphere. Kar et al. [2007] showed that ACE-FTS NO<sub>2</sub> agrees with the Measurements of Aerosol Extinction in the Stratosphere and Troposphere Retrieved by Occultation (MAESTRO) instrument, also aboard SCISAT-1, to within approximately 10%–15% from 15 to 40 km for sunrise measurements and from 22 to 35 km for sunset measurements. Kerzenmacher et al. [2008] compared ACE-FTS version 2.2 NO and NO<sub>2</sub> to several satellite, balloon-borne, and ground-based measurements. They found agreement of ACE-FTS NO<sub>2</sub> with other satellite data sets to within about 20% between 25 and 40 km. ACE-FTS NO agreed with HALOE mixing ratios to within 8% from 22 to 64 km. De Mazière et al. [2008] compared ACE-FTS CH<sub>4</sub> to correlative satellite, balloon-borne, and ground-based Fourier transform infrared remote sensing data and concluded that the accuracy of ACE-FTS CH<sub>4</sub> is within 25% throughout the stratosphere.

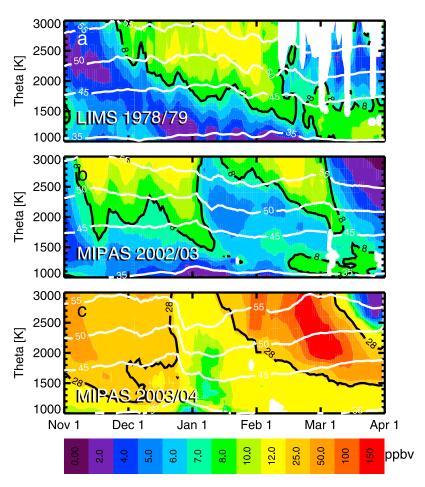
### 3. Arctic Winter NO<sub>x</sub> Evolution in LIMS, MIPAS, and ACE

2156202d, 2012, DS, Downloaded from https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2011JD016663 by Old Dominion University, Wiley Online Library on [0908/2023]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Cerative Commons

### 3.1. LIMS Nighttime NO<sub>2</sub> Versus MIPAS Nighttime NO<sub>2</sub>

[13] Figure 1 shows LIMS NO<sub>2</sub> from the 1978–1979 Arctic winter compared to MIPAS NO<sub>2</sub> from the 2002–2003 and 2003–2004 Arctic winters. Each image shows nighttime (solar zenith angle  $> 90^{\circ}$ ) NO<sub>2</sub> mixing ratios averaged inside the vortex from 1 November to 31 March. To determine the extent of the polar vortex, we used the method described by Harvey et al. [2002]; meteorological parameters were taken from the 40 year ECMWF reanalysis (ERA-40) data set [Uppala et al., 2005] and the Met Office (MetO) Unified Model [Swinbank and O'Neill, 1994]. Using this method, we were only able to determine the vortex edge up to approximately 2000 K. Above 2000 K, we have assumed that the vortex edge is the value at 2000 K. The data is smoothed in time with a 3 day running average. NO at LIMS measurement altitudes is quickly converted to NO<sub>2</sub> after sunset. During the night NO<sub>2</sub> begins to decrease, however, as it reacts with O3 to form NO3, which then reacts with another NO2 molecule to form N2O5 [e.g., Brasseur and Solomon, 2005]. We use nighttime NO<sub>2</sub> as a reasonable approximation for NO<sub>x</sub> with the caveat that it is most likely an underestimate since some of the NO<sub>2</sub> will have been converted to N<sub>2</sub>O<sub>5</sub> during the night.

[14] LIMS measurements (Figure 1a) clearly show NO<sub>2</sub> descending with time in the polar stratosphere. The temporal behavior of NO<sub>2</sub> is emphasized with the black contour line, which shows the 8 ppbv mixing ratio contour descending with time. Since the only source of NO<sub>x</sub> in the mesosphere during the polar winter is EPP, these high NO<sub>2</sub> mixing ratios



**Figure 1.** Nighttime NO<sub>2</sub> averaged in the vortex for (a) LIMS in 1978–1979, (b) MIPAS in 2002–2003, and (c) MIPAS in 2003–2004. White regions indicate missing data. White lines indicate altitude in kilometers. Black contour lines at 8 and 28 ppbv are shown to highlight the descent of NO<sub>2</sub> over time.

are unambiguously identified as being caused by EPP. NO2 mixing ratios reached a maximum of 18 ppbv in the upper stratosphere near 2600 K ( $\sim$ 52 km) at the end of January. Upper atmosphere EPP-NO<sub>x</sub> had already descended to 3000 K by at least mid-November and reached 1500 K by the beginning of February. According to *Dunkerton* [1991], a pair of minor warmings took place on 26 January and 8 February; this might explain the rapid decrease in NO<sub>2</sub> mixing ratios above 1500 K near 1 February and again about a week later. Because of missing data, it is difficult to say what happened to the NO<sub>2</sub> above 2000 K after 8 February, but there is little indication of further descent or substantially enhanced NO<sub>x</sub> at these altitudes. A major SSW occurred around 23 February; it was caused by the propagation of a planetary wave 2 of exceptional strength from the troposphere into the stratosphere, which occurred two weeks after the amplification of wave 1 [Labitzke, 1981]. At this time mixing with extra-vortex polar air caused NO<sub>2</sub> mixing ratios near 1500 K to decrease slightly. After the vortex recovered, however, NO<sub>2</sub> descent continues into March, reaching below 1000 K by the end of March. Some of the increase in NO<sub>2</sub> mixing ratios in March below 1500 K can be attributed to the conversion of reservoir species back into NO<sub>2</sub> as sunlight returns to the polar region.

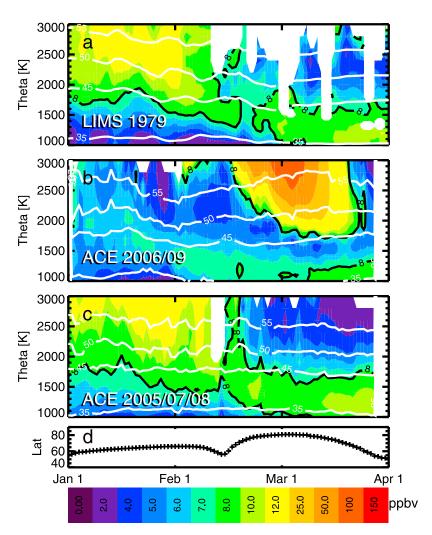
[15] The MIPAS data in the Artic winter of 2002-2003 (Figure 1b) also shows the downward transport of  $NO_2$  into

the stratosphere, and has been discussed in detail by F05. Again the temporal evolution is emphasized by the black contour line. Elevated NO<sub>2</sub> mixing ratios appeared below 3000 K by 1 November and reached the 1500 K level at the end of December; descent from above was abruptly cut short by a major SSW at the beginning of January. Mixing ratios reached a maximum of 26 ppbv on 18 November in the upper stratosphere (3000 K). Note that the occurrence of this midwinter SSW is a major difference between the winters of 2002-2003 and 1978-1979, but is not unusual for NH winters. Using CH<sub>4</sub> as a tracer, F05 showed that the increase in NO<sub>2</sub> around 1500 K after 1 February was due to mixing of extra-vortex midlatitude air. Downward transport of NO2 continued again after the major SSW, but F05 point out that it was confined to the upper stratosphere since the vortex below 2000 K did not regain strength following the warming. Overall, meteorology during the 2002–2003 Arctic winter was typical of NH winters prior to 2004, and EPP levels were near average.

onlinelibrary.wiley.com/doi/10.1029/2011JD016663 by Old Dominion

University, Wiley Online Library on [09/08/2023]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons

[16] In the 2003–2004 Arctic winter, the MIPAS NO<sub>2</sub> (Figure 1c) shows two distinct enhancements in stratospheric NO<sub>2</sub>. The first took place in November and December, and has been attributed to in situ production from the exceptional SPEs that occurred in October and November as well as downward transport following the events [*López-Puertas et al.*, 2005]. The high NO<sub>2</sub> mixing ratios



**Figure 2.** (a) Nighttime LIMS  $NO_2$  in the vortex in 1979. (b) The average of ACE-FTS  $NO_x$  in 2006 and 2009. (c) The average of ACE-FTS  $NO_x$  in 2005, 2007, and 2008. (d) ACE-FTS measurement latitudes. White regions indicate missing data. Black contour lines at 8 ppbv are shown to highlight the descent of  $NO_2$  over time.

were affected by a major SSW in late December. At this time, the downward transport was interrupted, and the NO<sub>2</sub>rich air in the vortex was diluted by mixing with air from lower latitudes; this explains the low mixing ratios that separate the two periods of enhanced NO<sub>2</sub>. Following the major SSW, the vortex rapidly recovered in the upper stratosphere to become the strongest on record in February and March [Manney et al., 2005]. The second enhancement was first observed by MIPAS in early January, and descended to 1500 K by mid-March; maximum NO2 mixing ratios exceeded those observed by LIMS in 1978-1979 and MIPAS in 2002–2003 by a factor of 20 and 14, respectively, in the upper stratosphere. Because of operational problems, MIPAS data are not available in 2004 after late March. Analyses of other data sets, however, show that the EPP-NO<sub>x</sub> enhancements observed in March-April 2004 exceeded 600 ppbv at 48 km [Randall et al., 2006] and were the largest on record for both hemispheres [Randall et al., 2005; Rinsland et al., 2005]. Moderately elevated EPP levels as well as peculiar dynamical conditions played a large role in the enhancements seen in 2004 [Clilverd et al., 2006;

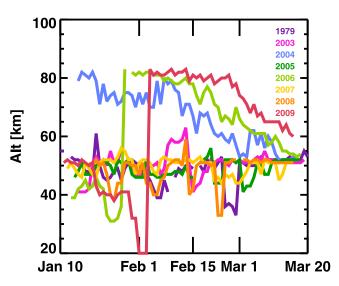
Hauchecorne et al., 2007; Jin et al., 2005; Natarajan et al., 2004; Randall et al., 2005; Rinsland et al., 2005].

[17] The stratospheric NO<sub>2</sub> enhancements in the LIMS data are similar in magnitude to those in the MIPAS data during the Arctic winter of 2002–2003, but much smaller than in MIPAS during 2003–2004. The primary differences between LIMS and MIPAS in 2002–2003 can largely be attributed to the different timing of the NO<sub>2</sub> descent in each winter. On the other hand, the differences between LIMS and MIPAS in 2003–2004, which indicate that MIPAS NO<sub>2</sub> was up to 50 times larger than LIMS NO<sub>2</sub>, arise because of the unusually high levels of particle activity and extraordinary meteorology of the 2003–2004 winter. Neither exceptional EPP levels nor exceptional meteorological conditions were present in 1978–1979, as described more below.

-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons

### 3.2. LIMS Nighttime NO<sub>2</sub> Versus ACE-FTS NO<sub>x</sub>

[18] Figure 2 compares the nighttime LIMS  $NO_2$  mixing ratios in the vortex to  $NO_x$  mixing ratios from ACE-FTS. Figure 2b is the average of 2006 and 2009 ACE-FTS  $NO_x$  mixing ratios for January through March. Figure 2c is the



**Figure 3.** Zonal mean LIMS and SABER stratopause heights from 10 January to 20 March, averaged from 75°N to 80°N. Breaks in the lines indicate missing data.

average of 2005, 2007, and 2008 ACE-FTS NO<sub>x</sub> mixing ratios for January through March. The average of 2006 and 2009 represents those years in which extraordinary EPP-NO<sub>x</sub> enhancements have been observed under conditions of low EPP [*Randall et al.*, 2006, 2009]. We chose to exclude ACE-FTS NO<sub>x</sub> in 2004 from the average since there is no data for January and part of February 2004. The measurement latitudes of ACE-FTS are shown in Figure 2d.

[19] ACE-FTS NO<sub>v</sub> mixing ratios in 2006 and 2009 exceed 150 ppbv in the upper stratosphere in February and March. Mixing ratios reach maxima of 30 ppbv, 20 ppbv, and 14 ppbv in 2005, 2007, and 2008, respectively, around 2500–3000 K ( $\sim$ 55–60 km) in January. The average of the maximum mixing ratios for these three years is about 21.3 ppbv, which is similar to the maximum LIMS mixing ratios for 1979 of 18 ppbv. In 2006 and 2009, unusual dynamics were observed; however, the level of geomagnetic activity was low in these years. The LIMS NO2 much more closely resembles ACE-FTS NO<sub>x</sub> in 2005, 2007, and 2008, and MIPAS data for 2002–2003. Interestingly, the primary EPP-induced NO<sub>x</sub> enhancements in both 2006 and 2009 occurred in February-March, significantly later than the primary NO<sub>x</sub> enhancements in 2005, 2007, and 2008 or in 1979. Since ACE-FTS samples latitudes equatorward of 65 N during January to mid February, the observed differences in this period between the years 2006 and 2009 and years 2005, 2007, and 2008 might be related to different vortex extensions and do not necessarily imply stronger mesospheric midwinter NO<sub>x</sub> intrusions during the latter years. In support of this speculation, MIPAS averages poleward of 60 N do not show such pronounced midwinter differences in NO<sub>x</sub>.

## 3.3. LIMS Temperature Versus Sounding of the Atmosphere Using Broadband Radiometry (SABER) Temperature

[20] One of the signatures of the unusual meteorology that led to the large  $NO_x$  enhancements in 2004, 2006 and 2009

is a strongly elevated stratopause height, which is indicative of enhanced adiabatic descent causing warming at altitudes that are normally in the mesosphere [Hauchecorne et al., 2007; Siskind et al., 2007]. Manney et al. [2008] pointed out that during the vortex breakup in early 2004 the stratopause was virtually isothermal; and upon recovery of the vortex a cool stratopause reformed above 75 km, which is much higher than the typical winter stratopause height of 50 km. They also found that in 2006 both the evolution of the SSW and the vortex recovery were very similar to those in 2004. An exceptionally strong and protracted SSW took place in January 2006, followed by the rapid recovery in early February of the upper stratospheric vortex and formation of an elevated stratopause around 80 km. In 2009, the strongest and most prolonged SSW on record took place in January [Manney et al., 2009]. As in 2004 and 2006, a strong upper stratospheric vortex reformed following the warming and the stratopause reformed around 80 km.

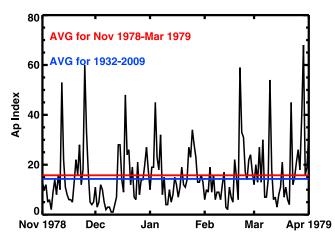
[21] Figure 3 shows that an elevated stratopause was not present in 1979. The stratopause heights are derived from LIMS (1979) and the SABER instrument (2003–2009) zonal mean temperature averaged from 75 to 80 N from 10 January to 12 March. The stratopause reforms at an altitude of 80 km after major SSW events in 2004, 2006 and 2009 (in 2004 the SSW itself was not observed by SABER because it was viewing in the opposite hemisphere at that time). The temperature of the 80 km stratopause is between 230 and 240 K in these three years, which is much cooler than typical winter stratopause temperatures. In contrast, in 1979, 2005, 2007 and 2008 the stratopause height remains around 50 km throughout most of the season, with temperatures between 250 and 260 K; occasional dips of the stratopause height are caused by major or minor warmings in these years, but an elevated stratopause is never present. The other dynamical feature of note, a strong vortex that reformed after the major SSWs in 2004, 2006, and 2009 [Manney et al., 2009], was also absent in 1979. Although the conditions for a major SSW were met on 23 February 1979 [Labitzke, 1981], a strong vortex did not reform in the upper stratosphere.

21562202d, 2012, D5, Downloaded from https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2011D016663 by Old Dominion University, Wiley Online Library on [09/08/2023]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms/

-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons

### 3.4. Geomagnetic Activity During LIMS Observing Period

[22] Figure 4 shows the geomagnetic Ap index from 1 November 1978 through 31 March 1979. The level of geomagnetic activity was not unusual during the 1978-1979 Arctic winter. The average of the Ap index for this time period was 15.8, whereas the average of the entire record from 1932 to 2009 was 14.3. There were also no significant solar proton events during the Arctic winter of 1978–1979. Using a two-dimensional model, Jackman and Meade [1988] investigated a solar proton event (SPE) that occurred in September 1978 to determine whether the SPE could have accounted for the elevated NO<sub>2</sub> mixing ratios in the LIMS NO<sub>2</sub>. They found that an increase in excess of 20 ppbv of NO<sub>2</sub> above 1 mbar following the SPE diminished to only a few ppbv by 1 December 1978. They concluded that the SPE did not significantly contribute to the LIMS NO<sub>2</sub> measurements in the polar night. In addition, they reported that no significant SPEs took place during the time that LIMS operated. That the geomagnetic Ap index was average during the 1979 Arctic winter indicates that the



**Figure 4.** Geomagnetic Ap index from 1 November 1978 through 31 March 1979. The average of the Ap index from 1 November 1978 through 31 March 1979 is 15.8 (red line), whereas the average of the Ap index for the entire record from 1932 to 2009 is 14.3 (blue line).

LIMS  $EPP-NO_x$  enhancements cannot be attributed to unusually high EPP levels.

### 4. Quantification of EPP-NO<sub>x</sub> in LIMS, MIPAS, and ACE-FTS

#### 4.1. LIMS and MIPAS EPP-NO<sub>x</sub>

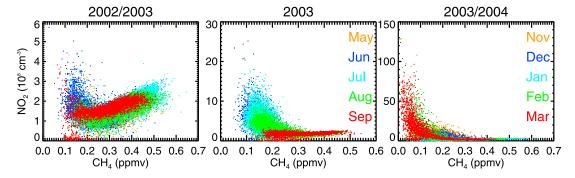
[23] In this section we describe the method used to quantify the absolute amount of EPP-NO<sub>x</sub> in the LIMS data, and compare this estimate with values for other years. Owing to the absence of a tracer species in the LIMS data, we were unable to directly correlate the high NO<sub>2</sub> mixing ratios with the descent of air from above as in previous studies [e.g., F05; R07; Siskind and Russell, 1996; Siskind et al., 1997]. Nevertheless, with observations in the polar night throughout the winter, we can directly infer the amount of descending EPP-NO<sub>x</sub>. This requires the assumption, however, that the increase in NO<sub>2</sub> in the polar stratospheric vortex is entirely due to the EPP IE.

[24] We estimated the amount of EPP-NO $_{\rm x}$  that crossed the 1500 K, 2000 K and 3000 K potential temperature surfaces over the entire season. This is consistent with the

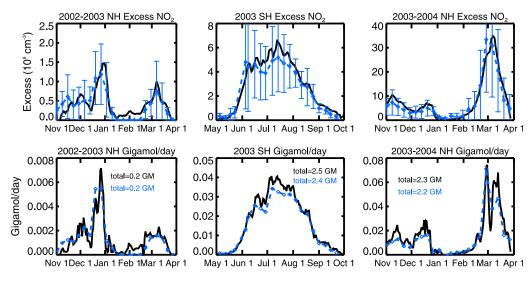
method used by R07 to estimate the amount of EPP-NO<sub>x</sub> entering the SH stratosphere from 1992 to 2005. Whereas R07 used only the 2000 K surface, we performed the calculation at additional levels because, unlike the SH, the NH is highly variable and the 2000 K surface does not necessarily provide a comprehensive picture. Note that the subtraction method is not valid below 1500 K because of difficulties tracing excess NO<sub>x</sub> to upper atmospheric origin. As above, we have used nighttime  $NO_2$  as a proxy for  $NO_x$ . Our method assumes that NO<sub>2</sub> has not yet been converted to reservoir gases nor has it been lost by photochemical destruction. Also, we include only data inside the vortex in our calculations. These assumptions lead to an underestimate in the results reported below, although this might be at least partially balanced by the assumption that all increases in vortex NO<sub>2</sub> are due to the EPP IE. For each day on each isentropic surface the average NO<sub>2</sub> density inside the vortex minus the amount in the vortex before the enhancement was multiplied by the area enclosed by the vortex. This calculation produces the excess number of NO<sub>2</sub> molecules on each isentropic surface, which was multiplied by the descent rate to arrive at the number of molecules crossing the isentropic surface per day as a function of time. The flux of molecules across each isentropic surface as a function of time was summed over the entire winter to obtain the total number of molecules crossing each level. Henceforth, we will refer to this method as the subtraction method; results are presented below in units of gigamoles.

21562202d, 2012, D5, Downloaded from https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2011D016663 by Old Dominion University, Wiley Online Library on [09/08/2023]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms/

[25] We tested our method by applying it at 2000 K to MIPAS data for the 2002-2003 Arctic winter, the 2003 Antarctic winter and the 2003–2004 Arctic winter and comparing our results to the results obtained using CH<sub>4</sub> as a tracer and to previous studies. Figure 5 shows scatterplots of MIPAS nighttime NO<sub>2</sub> versus CH<sub>4</sub> at 2000 K for each of the three winters. Measurements at all latitudes for the pertinent winter hemisphere are included in each plot. In the winter upper stratosphere NO<sub>x</sub> and CH<sub>4</sub> are generally positively correlated, with the lowest values of CH<sub>4</sub> indicating transport of air from above; in the absence of EPP-induced enhancements this air is depleted in NO<sub>x</sub>. Because the only source of NO<sub>x</sub> in the polar winter mesosphere is EPP, increasing values of NO<sub>x</sub> with decreasing CH<sub>4</sub> indicate the presence of EPP-induced enhancements in the air transported from above [see Siskind et al., 1997]. We search



**Figure 5.** MIPAS nighttime  $NO_2$  versus  $CH_4$  at 2000 K for the (left) 2002–2003 Northern Hemisphere (NH), (middle) 2003 Southern Hemisphere (SH), and (right) 2003–2004 NH winters. The months are colored as shown in the middle plot for the SH and the right plot for the NH. Note the different vertical scales in each plot.



**Figure 6.** (top) Excess nighttime  $NO_2$  densities in the vortex at 2000 K as a function of time for the 2002–2003 Arctic winter, the 2003 Antarctic winter, and the 2003–2004 Arctic winter. (bottom) The number of gigamoles crossing 2000 K per day as a function of time. In the top and bottom graphs, blue diamonds with dashed blue lines represent the  $CH_4$  method, and the black lines represent the subtraction method. Error bars on the  $CH_4$  method indicate 1-sigma standard deviations in the calculated excess  $NO_2$ . The total number of gigamoles integrated over each time period is given in Figure 6 (bottom): blue for the  $CH_4$  method and black for the subtraction method.

for anticorrelations between CH<sub>4</sub> and NO<sub>2</sub> using CH<sub>4</sub> < 0.27 ppmv as the requirement to indicate descent from above the stratosphere; this is consistent with R07. When such anticorrelations are present, we quantify the deviation from the positive correlation using the method described in R07. labeling the deviations as "excess" NO<sub>2</sub>. One modification to R07 is that the excess NO2 was calculated in weekly instead of 2 week time periods because MIPAS has daily global coverage, and thus significantly more data than HALOE, which sampled only a single latitude in each hemisphere on any given day. The excess NO<sub>2</sub> densities are mainly seen in November, December, January and March for the 2002-2003 NH winter, June to August in the 2003 SH winter and February and March for the 2003-2004 NH winter. The largest excess is seen in the 2003-2004 NH winter, with densities peaking at over  $100 \times 10^8$  cm<sup>-3</sup>. Peak densities are near  $30 \times 10^8$  cm<sup>-3</sup> in the 2003 SH winter and  $5 \times 10^{8} \text{ cm}^{-3}$  in the 2002–2003 NH winter.

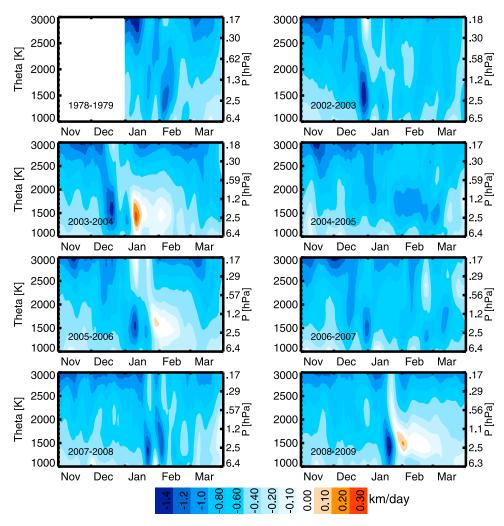
[26] Figure 6 compares the subtraction and CH<sub>4</sub> methods applied to the MIPAS data. Figure 6 (top) shows the quantification of excess nighttime NO2 densities in the vortex at the 2000 K level as a function of time for the 2002-2003 Arctic winter, the 2003 Antarctic winter, and the 2003–2004 Arctic winter. The solid, black lines show the amount of EPP-produced NO2 inferred from the subtraction method. The diamonds with the dashed lines show the amount of excess NO2 using CH4 as a tracer as described in the previous paragraph. Each diamond represents an average of all of the points deemed to be excess NO<sub>2</sub> for each respective 1 week time period. Error bars show the 1- $\sigma$  standard deviation excess NO<sub>2</sub> with the CH<sub>4</sub> method, where  $1-\sigma$  refers to one standard deviation above or below the mean value of excess NO<sub>2</sub>. In general, there is very good agreement between the subtraction method of approximating excess  $NO_2$  and the  $CH_4$  method.

[27] Figure 6 (bottom) shows the number of Gmol/d crossing 2000 K as a function of time for the same time periods as in the top graphs with the subtraction (black lines) and CH<sub>4</sub> (diamonds with dashed blue lines) methods. We approximated the descent rates by the vertical component of the residual circulation, which we calculated using the method of Solomon et al. [1986]. Diabatic heating rates and temperatures required for this calculation were obtained from the Modern Era Retrospective-Analysis for Research and Applications (MERRA) [Rienecker et al., 2011]. Figure 7 shows the vertical component of the residual circulation ( $\bar{w}^*$ ) for the 1978–1979 and 2002–2003 through 2008–2009 Arctic winters. The residual circulation is by definition a zonal mean quantity, so obtaining a vortex average is not possible. Instead we have averaged the residual circulation poleward of the equivalent latitude of the vortex. This assumption is most appropriate when the vortex is pole-centered and circular, but will introduce some error when the vortex is distorted and/or offset from the pole.

onlinelibrary. wiley. com/doi/10.1029/2011JD016663 by Old Dominion University, Wiley Online Library on [09/08/2023]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms

--and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons

[28] In all winters descent increases monotonically with increasing altitude early in the season. Later in the season this pattern changes, so that maximum descent rates might occur anywhere from about 1000–2000 K, but with substantial interseasonal variability in the timing and magnitude of the maximum. One obvious feature of note in Figure 7 is the interruption of downwelling by major SSWs in the winters of 2003–2004, 2005–2006, and 2008–2009 followed by strong descent into late winter in the lower mesosphere. However, this strong late winter descent might be underestimated with MERRA because the temperature structure in the upper stratosphere is not captured well by MERRA during these events. Measuring vertical velocities in the upper stratosphere remains a difficult problem, but on the basis of the overall agreement of our approximation with



**Figure 7.** Vertical descent rates for the 1978–1979 and 2002–2003 through 2008–2009 NH winters. The descent rates are derived using diabatic heating rates and temperature from Modern Era Retrospective-Analysis for Research and Applications (MERRA) data. For reference, global mean pressure is shown on the right axis.

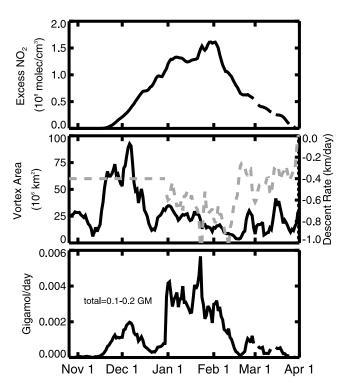
published estimates of -400 to -500 m/d in the upper stratosphere [F05; *López-Puertas et al.*, 2005], and the fact that the results are in agreement with our understanding of the basic physics, we believe that using the vertical component of the calculated residual circulation is a reasonable approach.

[29] The total number of gigamoles integrated over each time period is noted in Figure 6 (bottom) for both the subtraction (black lines) and CH<sub>4</sub> (dashed blue lines) methods. The total amount of EPP-NO<sub>x</sub> crossing the 2000 K level was found to be 0.2 (0.2) Gmol for the 2002–2003 Arctic winter, 2.5 (2.4) Gmol for the 2003 Antarctic winter and 2.3 (2.2) Gmol for the 2003–2004 Arctic winter with the subtraction (CH<sub>4</sub>) method. Clearly, the results from the subtraction method compare well with the results from the CH<sub>4</sub> method, supporting the validity of applying the subtraction method to satellite measurements of nighttime NO<sub>2</sub>. Our results from the subtraction method also compare reasonably well with previous estimates based on measurements of NO+NO<sub>2</sub> and NO<sub>2</sub>. F05 calculated the net deposition of NO<sub>y</sub> below 3000 K to be 2.4 Gmol for the 2003 Antarctic winter using

MIPAS data, which agrees very well with our estimate of 2.5 Gmol. We note that the ratio of NO<sub>x</sub> to NO<sub>y</sub> is above 0.9 in the upper stratosphere, so it is reasonable to use these results for comparison to our results with the caveat that NO<sub>x</sub> will be a slight underestimate. Our estimate also falls within the range of 1.1–2.6 Gmol that R07 estimated was deposited below 2000 K in the 2003 Antarctic winter using solar occultation data. Using a 3-D model together with MIPAS data, Reddmann et al. [2010] estimated the amount of excess NO<sub>v</sub> in the stratosphere from July 2002 to March 2004. They estimated the excess NO<sub>v</sub> deposited below 55 km for the 2003 Antarctic winter to be 1.4 Gmol. Their estimate of excess NO<sub>v</sub> for the 2002–2003 Arctic winter is 0.4 Gmol, while their estimate for the 2003-2004 Arctic winter is 2.0 Gmol. This is in fairly good agreement with our estimates of 0.2 Gmol and 2.3 Gmol for the 2002-2003 and 2003–2004 Arctic winters, respectively. On the basis of the overall agreement of our subtraction method with previous studies and with the CH<sub>4</sub> method, we conclude that our approach is valid and is reasonable to apply to LIMS data.

.wiley.com/doi/10.1029/2011JD016663 by Old Dominion

on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons

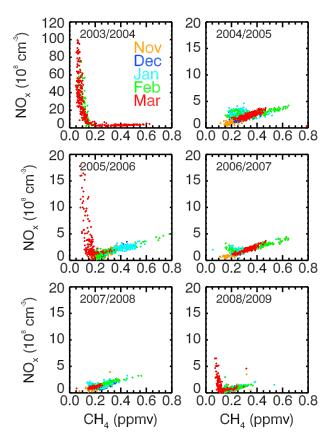


**Figure 8.** (top) Excess nighttime  $NO_2$  densities in the vortex at 2000 K in the 1978–1979 Arctic winter as a function of time, calculated from LIMS data. (middle) The vortex area (black line, left axis) and the descent rate (gray dashed line, right axis). (bottom) The inferred number of gigamoles crossing 2000 K per day as a function of time. The total amount for the entire season is also given in the bottom graph.

[30] Figure 8 shows the excess nighttime NO<sub>2</sub> densities at 2000 K in the vortex for the Arctic winter of 1978–1979 (Figure 8, top), the vortex area and descent rate at 2000 K (Figure 8, middle), and the number of Gmol/d crossing 2000 K (Figure 8, bottom) as a function of time. The descent rates for the 1978–1979 Arctic winter were also estimated by the vertical component of the residual circulation, as described above. MERRA data is only available after January 1979, so we were unable to calculate the descent rates before that time. The total amount of EPP-NO<sub>x</sub> crossing the 2000 K level from January-March of 1979 was found to be 0.1 Gmol. Figure 7 shows that the descent rates earlier in the winter were generally less than about 400 m/d. Using 400 m/d as an upper estimate, we calculate the total amount of EPP-NO<sub>x</sub> crossing the 2000 K level in November-December of 1978 to be 0.1 Gmol. Thus the total amount of EPP-NO<sub>x</sub> entering the stratosphere as seen in the LIMS data ranges from about 0.1 to 0.2 Gmol. This is in better agreement, quantitatively, with the amount of EPP-NO<sub>x</sub> entering the stratosphere in the 2002-2003 Arctic winter than with the 2003-2004 Arctic winter, as was shown qualitatively in section 3.1.

### 4.2. ACE-FTS EPP-NO<sub>x</sub>

[31] Scatterplots of ACE-FTS  $NO_x$  versus  $CH_4$  for the 2003–2004 through 2008–2009 NH winters are shown in Figure 9. Excess  $NO_x$  is defined in the same way as for

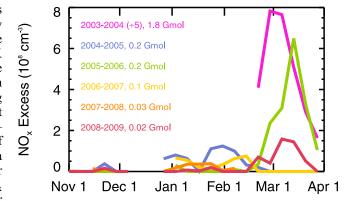


onlinelibrary.wiley.com/doi/10.1029/2011JD016663 by Old Dominion University, Wiley Online Library on [09/08/2023]. See the Terms

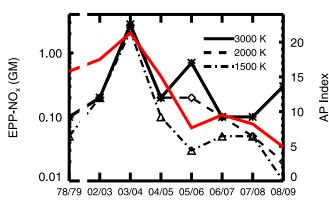
of use; OA articles are governed by the applicable Creative Common

**Figure 9.** ACE-FTS  $NO_x$  versus  $CH_4$  at 2000 K for the 2003–2004 through 2008–2009 Arctic winters. The months are colored as shown in the 2003–2004 plot. Note the different vertical scale in the 2003–2004 plot.

MIPAS (Figure 5). A great deal of excess  $NO_x$  can be seen in 2003–2004, 2005–2006 and 2008–2009. The largest excess is seen in 2003–2004 with densities peaking around  $100\times10^8~\text{cm}^{-3}$ . Peak densities are near  $20\times10^8~\text{cm}^{-3}$  in 2005–2006 and  $7\times10^8~\text{cm}^{-3}$  in 2008–2009. Figure 10



**Figure 10.** Average excess  $NO_x$  at 2000 K for ACE-FTS using the  $CH_4$  method for the 2003–2004 through 2008–2009 NH winters. The 2003–2004 line is reduced by a factor of 5 to fit on the plot. Breaks in the line indicate no data, since ACE-FTS did not sample inside the vortex at these times. The sum over each season is shown in gigamoles (Gmol).



**Figure 11.** Amount of EPP-NO $_x$  descending across the 1500, 2000, and 3000 K potential temperature surfaces deduced from LIMS data in the Arctic winter of 1978–1979, MIPAS data in the Arctic winters of 2002–2003 and 2003–2004, and ACE-FTS data in the winters of 2004–2005 through 2008–2009 (black lines, left axis). The red line shows the Ap index for each of the winters (right axis).

shows the quantification of ACE-FTS excess  $NO_x$  densities at 2000 K as a function of time for the 2003–2004 through 2008–2009 NH winters using the CH<sub>4</sub> method. As expected from *Randall et al.* [2009], maximum values are largest in 2003–2004, followed by 2005–2006 and 2008–2009, the years with unusual meteorology.

[32] The total amount of EPP-NO<sub>x</sub> crossing the 2000 K level in ACE-FTS is given in the Figure 10 legend. As noted earlier, no ACE-FTS data are available prior to 21 February 2004. Thus for comparison, we recalculated the total amount of EPP-NO<sub>x</sub> from MIPAS data using only the time period from 21 February through 31 March 2004; the result was 1.4 Gmol, which is similar to the ACE-FTS result of 1.8 Gmol. Contrary to the maximum values, the total amount of EPP-NO<sub>x</sub> descending across the 2000 K level in 2005– 2006 and 2008–2009 was not outstanding. Although the unusual meteorology in these winters led to extraordinary enhancements in the descent of EPP-NO<sub>x</sub>, these effects occurred too late in the winter for substantial amounts of excess NO<sub>x</sub> to descend past 2000 K [e.g., Salmi et al., 2011]. This highlights the fact that the transient effects at 2000 K of the unusual meteorology in these years were not large enough to make up for the fact that descent of EPP-NO<sub>x</sub> earlier in the winter was significantly less than in other years.

#### 4.3. EPP IE in the NH

[33] Figure 11 quantifies the EPP IE, as measured by the amount of EPP-NO $_{\rm x}$  descending across the 1500, 2000, and 3000 K potential temperature surfaces, for the Arctic winter of 1978–1979 and the Arctic winters of 2002–2003 through 2008–2009. Because ACE-FTS data did not start until February of 2004, MIPAS results are presented for the 2002–2003 and 2003–2004 winters, and ACE-FTS results are presented for the 2004–2005 through 2008–2009 winters. The calculated EPP IE for each NH winter is also summarized in Table 1, which includes results from all calculations performed, including LIMS and ACE-FTS data as well as the MIPAS data from both ESA and IMK/IAA.

[34] As shown in Figure 11, the largest EPP IE observed in the NH at all levels during this time period was the 2003–2004 winter. Table 1 shows that the estimate for 2003–2004 from MIPAS-IMK/IAA compares well with ACE-FTS at 2000 K for the time period during which both instruments were observing: 1.9 Gmol and 1.8 Gmol for MIPAS-IMK/IAA and ACE-FTS, respectively. Both the MIPAS-IMK/IAA and ACE-FTS estimates are slightly larger than the MIPAS-ESA estimate at 2000 K, which is most likely a reflection of the difference between using nighttime NO $_2$  (MIPAS-ESA) and NO $_x$  (ACE-FTS and MIPAS-IMK/IAA) for the calculation.

[35] We expect that some of the EPP-NO<sub>x</sub> will mix out to lower latitudes as air descends, and that air that reaches 3000 K late in the winter might not reach 2000 K and below by the end of the winter. Consistent with this expectation, the amounts of EPP-NO<sub>x</sub> descending across 3000 K for the 2005-2006 and 2008-2009 Arctic winters were larger than at 2000 K, by over a factor of 3 and 20, respectively. The MIPAS-IMK/IAA data show similar values at 1500 K (2.2 Gmol), 2000 K (2.7 Gmol) and 3000 K (2.8 Gmol) in 2003–2004, suggesting that very little mixing to lower latitudes occurred during the descent. In fact, 2003-2004 is the only year for which the signal of upper atmospheric NO<sub>x</sub> is evident at or below 1000 K (the NO<sub>x</sub>-CH<sub>4</sub> relationship in 2003-2004 shows evidence of EPP-NO<sub>x</sub> down to 800 K). That the amount for the 2003-2004 winter is less at 3000 K than at 2000 K for ACE-FTS can be explained by the fact that ACE-FTS did not have observations before 21 February when the enhancements would have been present at 3000 K. Again we compared MIPAS-IMK/IAA and ACE-FTS calculations for 21 February through 31 March 2004, the time period that was 2156202d, 2012, D5, Dowlonded from https://agupubs.onlinelbrary.wiley.com/doi/10.1029/2011JD016663 by Old Dominion University, Wiley Online Library on [09082023], See the Terms and Conditions (https://onlinelbrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Cerative Commons

**Table 1.** The Total Number of Gigamoles of EPP-NO<sub>x</sub> Crossing 1500, 2000, and 3000 K for LIMS, MIPAS, and ACE-FTS for the Relevant Arctic Winters<sup>a</sup>

	1979	2002-2003	2003-2004	2004-2005	2005-2006	2006-2007	2007-2008	2008-2009
LIMS (1500 K)	0.05-0.1	-	-	-	-	-	-	-
MIPAS-ESA (1500 K)	-	0.2	2.2 (0.8)	-	-	-	-	-
ACE-FTS (1500 K)	-	-	0.8	0.1	0.03	0.05	0.05	0.01
LIMS (2000 K)	0.1 - 0.2	-	-	-	-	-	-	-
MIPAS-ESA (2000 K)	-	0.2	2.3 (1.4)	-	-	-	-	-
MIPAS-IMK (2000 K)	-	-	2.7 (1.9)	-	-	-	-	-
ACE-FTS (2000 K)	-	-	1.8	0.2	0.2	0.1	0.05	0.02
LIMS (3000 K)	0.1 - 0.2	-	-	-	-	-	-	-
MIPAS-IMK (3000 K)	-	0.2	2.8 (0.3)	-	-	-	-	-
ACE-FTS (3000 K)	-	-	0.4	0.2	0.7	0.1	0.1	0.3

 $<sup>^{</sup>a}$ The amount shown in parentheses is the amount after 21 February, for comparison to the ACE-FTS data. Nighttime NO<sub>2</sub> is used for the first three rows of Table 1, and NO<sub>x</sub> is used for the subsequent rows.

observed by both instruments. The results, 0.3 Gmol using MIPAS-IMK/IAA data and 0.4 Gmol using ACE-FTS data, were very similar.

[36] Also shown in Figure 11 is the Ap index corresponding to each season (average Ap index from 1 October through 31 March). The correlation between the Ap index and the amount of EPP-NO<sub>x</sub> descending across a given potential temperature surface is not obvious, although the correlation coefficient for 2000 K (3000 K) is 0.66 (0.55). With only eight years of data, we cannot say with confidence if this is significant. Nevertheless, the lack of strong correlation is consistent with the fact that both geomagnetic activity and dynamics influence the amount of EPP-NO<sub>x</sub> reaching the stratosphere. If we exclude the years with unusual dynamics, the correlation coefficient between the Ap index and the amount of EPP-NO<sub>x</sub> crossing both 2000 and 3000 K becomes 0.8; again, we emphasize that this is based on only five years of data.

### 5. Summary and Conclusions

[37] In this paper we have compared LIMS NO<sub>2</sub> data from 1978 to 1979 to more recent data from MIPAS and ACE-FTS from 2002 to 2009. The magnitude and timing of the LIMS NO<sub>2</sub> enhancements in the 1978–1979 Arctic winter are similar to enhancements in the Arctic winters of 2002–2003, 2004–2005, 2006–2007 and 2007–2008 in the MIPAS and ACE-FTS data. We calculated that when integrated over the entire winter, approximately 0.1 Gmol of EPP-NO<sub>x</sub> descended past the 1500 K, 2000 K and 3000 K surfaces in 1978–1979. This is similar to the amount of EPP-NO<sub>x</sub> in the winters just mentioned, For reference, 0.1 Gmol is approximately 0.5% of the annual average contribution to Arctic (>50° latitude) NO<sub>v</sub> from the oxidation of N<sub>2</sub>O that Vitt and Jackman [1996] calculated using a 2-D photochemical transport model averaged over 20 years from 1974 to 1993.

[38] The largest EPP IE on record was the 2003–2004 winter, when approximately 2.2 Gmol of EPP-NO<sub>x</sub> descended across the 1500 K potential temperature surface. An extraordinary EPP IE has been reported previously for the 2005–2006 and 2008–2009 winters [e.g., Randall et al., 2009]. During these winters, however, the unusually strong EPP IE occurred later in the season than in 2003–2004, and was primarily confined to altitudes above 2000 K. We report here that in 2005–2006 and 2008–2009, 0.2 (0.1) and 0.02 (0.01) Gmol of NO<sub>x</sub>, respectively, descended across the 2000 K (1500 K) surface. These numbers are on the same order as, or in the case of 2008-2009 smaller than, the numbers for all other years analyzed. On the other hand, significantly more EPP-NO<sub>x</sub> was observed at 3000 K in 2005–2006 and 2008–2009, so that these years ranked second and third, respectively, after 2003–2004, in the total amount of EPP-NO<sub>x</sub> crossing the 3000 K surface. The differences between the results at 1500 K, 2000 K and 3000 K reflect the fact that the unusually strong enhanced descent in 2006 and 2009 occurred late enough in the season that the enhancements largely dissipated before reaching the 2000 K surface.

[39] Overall, the results reported here confirm that in the NH, the total amount of EPP-NO<sub>x</sub> transported to the stratosphere depends on both the level of EPP and the prevailing dynamics. That more total EPP-NO<sub>x</sub> was observed to

descend into the stratosphere in 2003–2004 than in any other winter is because that particular winter had a moderately high level of EPP and favorable dynamical conditions to transport NO<sub>x</sub>-rich air from the MLT to the stratosphere. In 2005–2006 and in 2008–2009, substantially more EPP-NO<sub>x</sub> was observed to descend across the 3000 K surface than in any other year aside from 2003 to 2004, even though the level of EPP was low. But the timing of the enhanced descent was such that the total amount of EPP-NO<sub>x</sub> crossing the 2000 K surface was not unusual. The LIMS results from 1978 to 1979 are similar both morphologically and quantitatively to the results from winters in which there was an average or low level of EPP and no unusual dynamics, consistent with conditions in the 1978–1979 winter itself.

[40] The estimation of descent rates is a primary source of uncertainty in our calculations of the integrated amount of EPP-NO<sub>x</sub> descending across different potential temperature surfaces. There is likely high variability in the descent rates at different locations inside the polar vortex, yet we use a single descent rate on any given day. Also, since the residual circulation is by definition a zonal average quantity, our method essentially assumes a pole-centered vortex. Another source of uncertainty is that there is no tracer available in the LIMS data set, so we assumed that all of the NO<sub>2</sub> in the vortex was EPP-NO<sub>x</sub> from above. This assumption will not be valid if there are intrusions of NO<sub>x</sub>-rich air from lower latitudes during SSWs at the altitudes that the calculation is performed at. However, in the years discussed here we showed that this is a reasonable assumption by comparing both the subtraction and CH<sub>4</sub> methods with the MIPAS data. Yet a third source of uncertainty is that we assumed that the vortex at 3000 K was the same as the vortex at 2000 K. This is likely a conservative estimate since the vortex generally grows larger with increasing height, so the estimates at 3000 K are likely underestimates. This could especially be true following major SSWs when the vortex at higher altitudes is sometimes much larger in area than at lower altitudes [e.g., F05; Mengistu Tsidu et al., 2005].

2156202d, 2012, DS, Downloaded from https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2011JD016663 by Old Dominion University, Wiley Online Library on [0908/2023]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Cerative Commons

[41] The NO<sub>x</sub> enhancements in the 1978–1979 Arctic winter, although relatively small, were very evident in the LIMS data because of its nighttime measurement capability, enabling it to measure NO2 throughout the polar night. Between 1979 and 2002, when MIPAS was launched, the only satellite instruments capable of such measurements were the Improved Stratospheric and Mesospheric Sounder (ISAMS), which had a limited lifetime, and the Cryogenic Limb Array Etalon Spectrometer (CLAES), both aboard UARS. Thus, the lack of reported EPP-NO<sub>x</sub> descending to the NH stratosphere in the 1980s and 1990s was most likely due to a lack of appropriate measurements. Nighttime measurements of NO<sub>2</sub> are a useful proxy for total NO<sub>x</sub> in the stratosphere and lower mesosphere [e.g., Seppälä et al., 2007]. An instrument like LIMS would not be able to capture NO<sub>x</sub> in the mesosphere and higher, however, since above about 65-70 km NO<sub>x</sub> is in the form of NO. To fully characterize the EPP-NO<sub>x</sub> production and descent, observations of NO<sub>x</sub> from the stratosphere to the lower thermosphere throughout the polar winter are required.

[42] **Acknowledgments.** Support for this work was provided by the NSF CEDAR program, grant AGS 0940124, and by the NASA Living With a Star program, grants NNX10AQ54G and NNX06AC05G. The ACE mission is supported primarily by the Canadian Space Agency. Support was

also provided by the UK Natural Environment Research Council (NERC). We thank the National Geophysical Data Center for the *Ap* index and the Space Environment Center for SPE data.

#### References

- Bates, D. R., and P. B. Hays (1967), Atmospheric nitrous oxide, *Planet. Space Sci.*, 15, 189–197, doi:10.1016/0032-0633(67)90074-8.
- Bernath, P. F., et al. (2005), Atmospheric Chemistry Experiment (ACE): Mission overview, *Geophys. Res. Lett.*, 32, L15S01, doi:10.1029/2005GL022386.
- Boone, C. D., et al. (2005), Retrievals for the Atmospheric Chemistry Experiment Fourier-transform spectrometer, *Appl. Opt.*, *44*, 7218–7231, doi:10.1364/AO.44.007218.
- Brasseur, G. P., and S. Solomon (2005), Aeronomy of the Middle Atmosphere: Chemistry and Physics of the Stratosphere and Mesosphere, 3rd ed., Springer, Dordrecht, Netherlands.
- Callis, L. B., D. N. Baker, M. Natarajan, J. B. Blake, R. A. Mewaldt, R. S. Selesnick, and J. R. Cummings (1996), A 2-D model simulation of downward transport of NO<sub>y</sub> into the stratosphere: Effects on the 1994 austral spring O<sub>3</sub> and NO<sub>y</sub>, *Geophys. Res. Lett.*, 23, 1905–1908, doi:10.1029/96GL01788.
- Callis, L. B., M. Natarajan, D. S. Evans, and J. D. Lambeth (1998a), Solar atmospheric coupling by electrons (SOLACE): 1. Effects of the May 12, 1997 solar event on the middle atmosphere, *J. Geophys. Res.*, 103, 28,405–28,419, doi:10.1029/98JD02408.
- Callis, L. B., M. Natarajan, J. D. Lambeth, and D. N. Baker (1998b), Solar-atmospheric coupling by electrons (SOLACE): 2. Calculated stratospheric effects of precipitating electrons, 1979–1988, J. Geophys. Res., 103, 28,421–28,438, doi:10.1029/98JD02407.
- Clilverd, M. A., A. Seppälä, C. J. Rodger, P. T. Verronen, and N. R. Thomson (2006), Ionospheric evidence of thermosphere-to-stratosphere descent of polar NO<sub>x</sub>, *Geophys. Res. Lett.*, 33, L19811, doi:10.1029/2006GL026727.
- Codrescu, M. V., T. J. Fuller-Rowell, R. G. Roble, and D. S. Evans (1997), Medium energy particle precipitation influences on the mesosphere and lower thermosphere, *J. Geophys. Res.*, 102, 19,977–19,987, doi:10.1029/97JA01728.
- Cohen, R. C., and J. G. Murphy (2003), Photochemistry of NO<sub>2</sub> in Earth's stratosphere: Constraints from observations, *Chem. Rev.*, 103, 4985–4998, doi:10.1021/cr020647x.
- Crutzen, P. J. (1970), The influence of nitrogen oxides on the atmospheric ozone content, *Q. J. R. Meteorol. Soc.*, *96*, 320–325, doi:10.1002/qj.49709640815.
- Crutzen, P. J. (1971), Ozone production rates in oxygen-hydrogen-nitrogen oxide atmosphere, *J. Geophys. Res.*, 76, 7311–7327, doi:10.1029/JC076i030p07311.
- Crutzen, P. J., I. S. A. Isaksen, and G. C. Reid (1975), Solar proton events: Stratospheric sources of nitric oxide, *Science*, 189, 457–459, doi:10.1126/science.189.4201.457.
- De Mazière, M., et al. (2008), Validation of ACE-FTS v2.2 methane profiles from the upper troposphere to the lower mesosphere, *Atmos. Chem. Phys.*, 8, 2421–2435, doi:10.5194/acp-8-2421-2008.
- Dunkerton, T. J. (1991), LIMS (Limb Infrared Monitor of the Stratosphere) observation of traveling planetary waves and potential vorticity advection in the stratosphere and mesosphere, *J. Geophys. Res.*, 96, 2813–2834, doi:10.1029/90JD02340.
- European Space Agency (2000), Envisat, MIPAS: An instrument for atmospheric chemistry and climate research, Eur. Space Agency Spec. Publ., ESA SP-1229, 1–124.
- Fischer, H., et al. (2008), MIPAS: An instrument for atmospheric and climate research, *Atmos. Chem. Phys.*, 8, 2151–2188, doi:10.5194/acp-8-2151-2008.
- Funke, B., M. López-Puertas, S. Gil-López, T. von Clarmann, G. P. Stiller, H. Fischer, and S. Kellmann (2005a), Downward transport of upper atmospheric NO<sub>x</sub> into the polar stratosphere and lower mesosphere during the Antarctic 2003 and Arctic 2002/2003 winters, *J. Geophys. Res.*, 110, D24308, doi:10.1029/2005JD006463.
- Funke, B., et al. (2005b), Retrieval of stratospheric NO<sub>x</sub> from 5.3 and 6.2 μm nonlocal thermodynamic equilibrium emissions measured by Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) on Envisat, J. Geophys. Res., 110, D09302, doi:10.1029/2004JD005225.
- Garcia, R. R., and S. Solomon (1994), A new numerical model for the middle atmosphere: 2. Ozone and related species, *J. Geophys. Res.*, 99, 12,937–12,951, doi:10.1029/94JD00725.
- Gille, J. C., and J. M. Russell III (1984), The limb infrared monitor of the stratosphere: Experiment description, performance, and results, *J. Geophys. Res.*, 89, 5125–5140, doi:10.1029/JD089iD04p05125.

- Gylvan Meira, L., Jr. (1971), Rocket measurements of upper atmospheric nitric oxide and their consequences to the lower ionosphere, *J. Geophys. Res.*, 76, 202–212, doi:10.1029/JA076i001p00202.
- Harvey, V. L., R. B. Pierce, T. D. Fairlie, and M. H. Hitchman (2002), A climatology of stratospheric polar vortices and anticyclones, *J. Geophys. Res.*, 107(D20), 4442, doi:10.1029/2001JD001471.
- Hauchecorne, A., J.-L. Bertaux, F. Dalaudier, J. M. Russell III, M. G. Mlynczak, E. Kyrola, and D. Fussen (2007), Large increase of NO<sub>2</sub> in the north polar mesosphere in January–February 2004: Evidence of a dynamical origin from GOMOS/ENVISAT and SABER/TIMED data, *Geophys. Res. Lett.*, *34*, L03810, doi:10.1029/2006GL027628.
- Jackman, C. H., and P. E. Meade (1988), Effect of solar proton events in 1978 and 1979 on the odd nitrogen abundance in the middle atmosphere, J. Geophys. Res., 93, 7084–7090, doi:10.1029/JD093iD06p07084.
- Jin, J. J., et al. (2005), Co-located ACE-FTS and Odin-SMR stratospheric-mesospheric CO 2004 measurements and comparison with a GCM, Geophys. Res. Lett., 32, L15S03, doi:10.1029/2005GL022433.
- Kar, J., et al. (2007), Initial comparison of ozone and NO<sub>2</sub> profiles from ACE-MAESTRO with balloon and satellite data, *J. Geophys. Res.*, 112, D16301, doi:10.1029/2006JD008242.
- Kerzenmacher, T., et al. (2008), Validation of NO<sub>2</sub> and NO from the Atmospheric Chemistry Experiment (ACE), *Atmos. Chem. Phys.*, 8, 5801–5841, doi:10.5194/acp-8-5801-2008.
- Labitzke, K. (1981), The amplification of height wave 1 in January 1979: A characteristic precondition for the major warming in February, *Mon. Weather Rev.*, 109, 983–989, doi:10.1175/1520-0493(1981)109<0983: TAOHWI>2.0.CO;2.

21562202d, 2012, DS, Downloaded from https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2011D016663 by Old Dominion University, Wiley Online Library on [09/08/2023]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms

∻and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Common:

- López-Puertas, M., B. Funke, S. Gil-López, T. von Clarmann, G. P. Stiller, M. Höpfner, S. Kellmann, H. Fischer, and C. H. Jackman (2005), Observation of NO<sub>x</sub> enhancement and ozone depletion in the Northern and Southern Hemispheres after the October–November 2003 solar proton events, *J. Geophys. Res.*, 110, A09S43, doi:10.1029/2005JA011050.
- López-Puertas, M., B. Funke, T. von Clarmann, H. Fischer, and G. P. Stiller (2006), The stratospheric and mesospheric NOy in the 2002–2004 polar winters as measured by MIPAS/Envisat, *Space Sci. Rev.*, 125, 403–416, doi:10.1007/s11214-006-9073-2.
- Manney, G. L., et al. (2005), The remarkable 2003–2004 winter and other recent warm winters in the Arctic stratosphere since the late 1990s, *J. Geophys. Res.*, *110*, D04107, doi:10.1029/2004JD005367.
- Manney, G. L., et al. (2008), The evolution of the stratopause during the 2006 major warming: Satellite data and assimilated meteorological analyses, *J. Geophys. Res.*, 113, D11115, doi:10.1029/2007JD009097.
- Manney, G. L., M. J. Schwartz, K. Krüger, M. L. Santee, S. Pawson, J. N. Lee, W. H. Daffer, R. A. Fuller, and N. J. Livesey (2009), Aura Microwave Limb Sounder observations of dynamics and transport during the record-breaking 2009 Arctic stratospheric major warming, *Geophys. Res. Lett.*, 36, L12815, doi:10.1029/2009GL038586.
- McElroy, M. B., and J. C. McConnell (1971), Nitrous oxide: A natural source of stratospheric NO, *J. Atmos. Sci.*, 28, 1095–1098, doi:10.1175/1520-0469(1971)028<1095:NOANSO>2.0.CO;2.
- McHugh, M., B. Magill, K. A. Walker, C. D. Boone, P. F. Bernath, and J. M. Russell III (2005), Comparison of atmospheric retrievals from ACE and HALOE, *Geophys. Res. Lett.*, 32, L15S10, doi:10.1029/2005GL022403.
- Mengistu Tsidu, G., et al. (2005), NO<sub>y</sub> from Michelson Interferometer for Passive Atmospheric Sounding on Environmental Satellite during the Southern Hemisphere polar vortex split in September/October 2002, *J. Geophys. Res.*, 110, D11301, doi:10.1029/2004JD005322.
- Narcisi, R. S., C. R. Philbrick, F. C. Ulwick, and M. E. Gardner (1972), Mesospheric nitric-oxide concentrations during a PCA, *J. Geophys. Res.*, 77, 1332–1336, doi:10.1029/JA077i007p01332.
- Natarajan, M., E. E. Remsberg, L. E. Deaver, and J. M. Russell III (2004), Anomalously high levels of NO<sub>x</sub> in the polar upper stratosphere during April, 2004: Photochemical consistency of HALOE observations, *Geophys. Res. Lett.*, 31, L15113, doi:10.1029/2004GL020566.
- Nicolet, M. (1971), Aeronomic reactions of hydrogen and ozone, in *Mesospheric Models and Related Experiments*, edited by G. Fiocco, pp. 1–51, D. Reidel, Dordrecht, Netherlands.
- Randall, C. E., D. W. Rusch, R. M. Bevilacqua, K. W. Hoppel, and J. D. Lumpe (1998), Polar Ozone and Aerosol Measurement (POAM) II stratospheric NO<sub>2</sub>, 1993–1996, *J. Geophys. Res.*, 103, 28,361–28,371, doi:10.1029/98JD02092.
- Randall, C. E., D. E. Siskind, and R. M. Bevilacqua (2001), Stratospheric NO<sub>x</sub> enhancements in the Southern Hemisphere vortex in winter/spring of 2000, *Geophys. Res. Lett.*, 28, 2385–2388, doi:10.1029/2000GL012746.
- Randall, C. E., et al. (2005), Stratospheric effects of energetic particle precipitation in 2003–2004, Geophys. Res. Lett., 32, L05802, doi:10.1029/2004GL022003.

- Randall, C. E., V. L. Harvey, C. S. Singleton, P. F. Bernath, C. D. Boone, and J. U. Kozyra (2006), Enhanced NO<sub>x</sub> in 2006 linked to strong upper stratospheric Arctic vortex, *Geophys. Res. Lett.*, *33*, L18811, doi:10.1029/2006GL027160.
- Randall, C. E., et al. (2007), Energetic particle precipitation effects on the Southern Hemisphere stratosphere in 1992–2005, *J. Geophys. Res.*, 112, D08308, doi:10.1029/2006JD007696.
- Randall, C. E., V. L. Harvey, D. E. Siskind, J. France, P. F. Bernath, C. D. Boone, and K. A. Walker (2009), NO<sub>x</sub> descent in the Arctic middle atmosphere in early 2009, *Geophys. Res. Lett.*, 36, L18811, doi:10.1029/2009GL039706.
- Raspollini, P., et al. (2006), MIPAS level 2 operational analysis, *Atmos. Chem. Phys.*, 6, 5605–5630, doi:10.5194/acp-6-5605-2006.
- Reddmann, T., R. Ruhnke, S. Versick, and W. Kouker (2010), Modeling disturbed stratospheric chemistry during solar-induced NO<sub>x</sub> enhancements observed with MIPAS/ENVISAT, *J. Geophys. Res.*, 115, D00I11, doi:10.1029/2009JD012569.
- Remsberg, E. E., L. L. Gordley, B. T. Marshall, R. E. Thompson, J. Burton, P. Bhatt, V. L. Harvey, G. S. Lingenfelser, and M. Natarajan (2004), The Nimbus 7 LIMS version 6 radiance conditioning and temperature retrieval methods and results, *J. Quant. Spectrosc. Radiat. Transfer*, 86, 395–424, doi:10.1016/j.jqsrt.2003.12.007.
- Remsberg, E. E., M. Natarajan, B. T. Marshall, L. L. Gordley, R. E. Thompson, and G. Lingenfelser (2010), Improvements in the profiles and distributions of nitric acid and nitrogen dioxide with the LIMS version 6 dataset, *Atmos. Chem. Phys.*, 10, 4741–4756, doi:10.5194/acp-10-4741-2010.
- Rienecker, M. M., et al. (2011), MERRA: NASA's Modern-Era Retrospective Analysis for Research and Applications, *J. Clim.*, 24, 3624–3648, doi:10.1175/JCLI-D-11-00015.1.
- Rinsland, C. P., et al. (1996), ATMOS measurements of H<sub>2</sub>O + 2CH<sub>4</sub> and total reactive nitrogen in the November 1994 Antarctic stratosphere: Dehydration and denitrification in the vortex, *Geophys. Res. Lett.*, 23, 2397–2400, doi:10.1029/96GL00048.
- Rinsland, C. P., R. J. Salawitch, M. R. Gunson, S. Solomon, R. Zander, E. Mahieu, A. Goldman, M. J. Newchurch, F. W. Irion, and A. Y. Chang (1999), Polar stratospheric descent of NO<sub>y</sub> and CO and Arctic denitrification during winter 1992–1993, *J. Geophys. Res.*, 104, 1847–1861, doi:10.1029/1998JD100034.
- Rinsland, C. P., C. Boone, R. Nassar, K. Walker, P. Bernath, J. C. McConnell, and L. Chiou (2005), Atmospheric Chemistry Experiment (ACE) Arctic stratospheric measurements of NO<sub>x</sub> during February and March 2004: Impact of intense solar flares, *Geophys. Res. Lett.*, 32, L16S05, doi:10.1029/2005GL022425.
- Rusch, D. W., L. C. Gerard, S. Solomon, P. J. Crutzen, and G. C. Reid (1981), The effect of particle precipitation events on the neutral and ion chemistry of the middle atmosphere—1. Odd nitrogen, *Planet. Space Sci.*, 29, 767–774, doi:10.1016/0032-0633(81)90048-9.
- Russell, J. M., III, S. Solomon, L. L. Gordley, E. E. Remsberg, and L. B. Callis (1984), The variability of stratospheric and mesospheric NO<sub>2</sub> in the polar winter night observed by LIMS, *J. Geophys. Res.*, 89, 7267–7275, doi:10.1029/JD089iD05p07267.
- Salmi, S.-M., P. T. Verronen, L. Thölix, E. Kyrölä, L. Backman, A. Y. Karpechko, and A. Seppälä (2011), Mesosphere-to-stratosphere descent of odd nitrogen in February-March 2009 after sudden stratospheric warming, *Atmos. Chem. Phys.*, 11, 4645–4655, doi:10.5194/ acp-11-4645-2011.
- Seppälä, A., P. T. Verronen, E. Kyrölä, S. Hassinen, L. Backman, A. Hauchecorn, J. L. Bertaux, and D. Fussen (2004), Solar proton events of October-November 2003: Ozone depletion in the Northern Hemisphere polar winter as seen by GOMOS/Envisat, *Geophys. Res. Lett.*, 31, L19107, doi:10.1029/2004GL021042.

- Seppälä, A., P. T. Verronen, M. A. Clilverd, C. E. Randall, J. Tamminen, V. Sofieva, L. Backman, and E. Kyrölä (2007), Arctic and Antarctic polar winter NO<sub>x</sub> and energetic particle precipitation in 2002–2006, *Geophys. Res. Lett.*, 34, L12810, doi:10.1029/2007GL029733.
- Siskind, D. E., and J. M. Russell III (1996), Coupling between middle and upper atmospheric NO: Constraints from HALOE observations, *Geophys. Res. Lett.*, *23*, 137–140, doi:10.1029/95GL03782.
- Siskind, D. E., J. T. Becmeister, M. E. Summers, and J. M. Russell III (1997), Two-dimensional model calculations of nitric oxide transport in the middle atmosphere and comparison with Halogen Occultation Experiment data, J. Geophys. Res., 102, 3527–3545, doi:10.1029/96JD02970.
- Siskind, D. E., S. D. Eckermann, L. Coy, J. P. McCormack, and C. E. Randall (2007), On recent interannual variability of the Arctic winter mesosphere: Implications for tracer descent, *Geophys. Res. Lett.*, 34, L09806, doi:10.1029/2007GL029293.
- Solomon, S., P. J. Crutzen, and R. G. Roble (1982), Photochemical coupling between the thermosphere and the lower atmosphere: 1. Odd nitrogen from 50 to 120 km, *J. Geophys. Res.*, 87, 7206–7220, doi:10.1029/JC087iC09p07206.
- Solomon, S., J. T. Kiehl, R. R. Garcia, and W. Grose (1986), Tracer transport by the diabatic circulation deduced from satellite observations, *J. Atmos. Sci.*, 43, 1603–1617, doi:10.1175/1520-0469(1986)043<1603: TTBTDC>2.0.CO;2.
- Stiller, G. P., et al. (2005), An enhanced HNO<sub>3</sub> second maximum in the Antarctic midwinter upper stratosphere 2003, *J. Geophys. Res.*, 110, D20303, doi:10.1029/2005JD006011.
- Swinbank, R., and A. O'Neill (1994), A stratosphere-troposphere data assimilation system, *Mon. Weather Rev.*, 122, 686–702, doi:10.1175/1520-0493(1994)122<0686:ASTDAS>2.0.CO;2.
- Thorne, R. M. (1980), The importance of energetic particle precipitation on the chemical composition of the middle atmosphere, *Pure Appl. Geophys.*, 118, 128–151, doi:10.1007/BF01586448.
- Uppala, S. M., et al. (2005), The ERA-40 re-analysis, *Q. J. R. Meteorol. Soc.*, 131, 2961–3012, doi:10.1256/qj.04.176.

2156202d, 2012, D5, Dowloaded from https://agupubs.onlinelibrary.wiley.com/doi/10.1029/20111D016663 by Old Dominion University, Wiley Online Library on [09/08/2023], See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of uses OA articles

are governed by the applicable Creative Commons

- Vitt, F. M., and C. H. Jackman (1996), A comparison of sources of odd nitrogen production from 1974 through 1993 in the Earth's middle atmosphere as calculated using a two-dimensional model, *J. Geophys. Res.*, 101, 6729–6739, doi:10.1029/95JD03386.
- von Clarmann, T., et al. (2003), Retrieval of temperature and tangent altitude pointing from limb emission spectra recorded from space by the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS), *J. Geophys. Res.*, 108(D23), 4736, doi:10.1029/2003JD003602.
- Watson, R. T., M. A. Geller, R. S. Stolarski, and R. F. Hampson (1986), Present State of Knowledge of the Upper Atmosphere: An Assessment Report, NASA Ref. Publ., 1162, 20 pp.
- Wetzel, G., et al. (2007), Validation of MIPAS-ENVISAT NO<sub>2</sub> operational data, *Atmos. Chem. Phys.*, 7, 3261–3284, doi:10.5194/acp-7-3261-2007.
- P. F. Bernath, Department of Chemistry, University of York, Heslington, York YO10 5DD, UK.
- B. Funke, Instituto de Astrofísica de Andalucía, CSIC, E-18008 Granada, Spain
- V. L. Harvey, L. A. Holt, and C. E. Randall, Laboratory for Atmospheric and Space Physics, University of Colorado at Boulder, Boulder, CO 80303, USA. (holt@lasp.colorado.edu)
- E. E. Remsberg, NASA Langley Research Center, 21 Langley Blvd., Mail Stop 401B, Hampton, VA 23681, USA.
- G. P. Stiller, Institute for Meteorology and Climate Research, Karlsruhe Institute of Technology, D-76344 Karlsruhe, Germany.
- K. A. Walker, Department of Physics, University of Toronto, Toronto, ON M5S 1A1, Canada.