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

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[1] The Limb Infrared Monitor of the Stratosphere (LIMS) measured polar stratospheric enhancements of NO2 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 NOx (NO + NO2) entering the stratosphere that has been created by EPP in the mesosphere and lower thermosphere (EPP-NOx) has been quantified for the 1978–1979 and 2002–2003 through 2008–2009 Arctic winters. The NO2 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 (~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 NOx 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-NOx for reaching the middle stratosphere.


1. Introduction

[2] The key catalytic cycle responsible for ozone loss in the stratosphere between about 25 and 40 km is the NOx (NO + NO2) catalytic cycle [e.g., Crutzen, 1970; Garcia and Solomon, 1994; Watson et al., 1986]. The primary source of NOx in the stratosphere is NO produced from the oxidation of N2O of tropospheric origin [e.g., Bates and Hays, 1967; Crutzen, 1971; McIntyre and McConnell, 1971; Nicolet, 1971]. Another source of stratospheric NOx 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 NOx so produced is referred to as EPP-NOx.

[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(2D) and O2 [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 O3 concentrations become significant, it can react with O3 to produce NO2 [e.g., Cohen and Murphy, 2003]. The process by which NOx 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
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 O3 by EPP-NOx [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 > 30 keV) and protons (E > 30 MeV) deposit their energy in the stratosphere, but this happens sporadically during periods of strong geomagnetic activity.

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Funke et al. 1999; 300 keV) and protons al.

LIMS during the Northern Hemisphere (NH) winter of 1978–1979 [LIMS] in 1991, there were few reports of stratospheric EPP-NOx 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-NOx enhancements were seen so clearly in the LIMS data: Were they observed because enhanced EPP or unusual meteorology led to higher-than-normal stratospheric NOx 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-NOx 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-NOx entering the stratosphere in the 1978–1979 Arctic winter and in the winters observed by MIPAS and ACE-FTS.

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 NOx 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-NOx descending past the 1500 K, 2000 K and 3000 K isentropic surfaces. Section 5 includes the summary and conclusions.

2. Observations

2.1. LIMS

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 (H2O), O3, NO and nitric acid (HNO3) 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 NO2 is approximately 3.7 km and the nighttime NO2 mixing ratios extend from approximately 50 hPa to the lower mesosphere, at least in the polar night. The precision (accuracy) of the updated V6 NO2 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 NO2 in the upper stratosphere is better than the previous versions because of improved spectral line parameters for the NO2 forward radiance model.
2.2. MIPAS

MIPAS can measure NO2 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⁻¹) 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) NO2 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 NO2 below about 45 km. They compared MIPAS NO2 to observations from balloons, satellites, and ground-based measurements. In addition to NO2, we used ESA CH4 [e.g., Raspolini 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 NO2 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 NOx enhancements and subsequent O3 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-NOx transported to the polar stratosphere in the 2003 Antarctic winter. In their study they used CH4 and CO as dynamical tracers to determine the origin of the NOx-enriched air. Stiller et al. [2005] looked at vertical profiles of stratospheric HNO3 from MIPAS. They reported a second maximum in HNO3 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 NOx 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

ACE-FTS is a 0.02 cm⁻¹ resolution Fourier transform spectrometer operating in the midinfrared (750–4400 cm⁻¹) 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, NO2, and CH4, 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].

[11] Initial validation of ACE-FTS NOx by McHugh et al. [2005] showed agreement of ACE-FTS NOx with the Halogen Occulation Experiment (HALOE) to within 20% between 22 and 55 km. They also showed that ACE-FTS CH4 was about 10% higher than HALOE throughout the stratosphere. Kar et al. [2007] showed that ACE-FTS NO2 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 NO2 to several satellite, balloon-borne, and ground-based measurements. They found agreement of ACE-FTS NO2 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 CH4 to correlative satellite, balloon-borne, and ground-based Fourier transform infrared remote sensing data and concluded that the accuracy of ACE-FTS CH4 is within 25% throughout the stratosphere.

3. Arctic Winter NOx Evolution in LIMS, MIPAS, and ACE

3.1. LIMS Nighttime NO2 Versus MIPAS Nighttime NO2

[13] Figure 1 shows LIMS NO2 from the 1978–1979 Arctic winter compared to MIPAS NO2 from the 2002–2003 and 2003–2004 Arctic winters. Each image shows nighttime (solar zenith angle > 90°) NO2 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 NO2 after sunset. During the night NO2 begins to decrease, however, as it reacts with O3 to form NO3, which then reacts with another NO2 molecule to form N2O3 [e.g., Brasseur and Solomon, 2005]. We use nighttime NO2 as a reasonable approximation for NO3 with the caveat that it is most likely an underestimate since some of the NO3 will have been converted to N2O3 during the night.

[14] LIMS measurements (Figure 1a) clearly show NO2 descending with time in the polar stratosphere. The temporal behavior of NO2 is emphasized with the black contour line, which shows the 8 ppbv mixing ratio contour descending with time. Since the only source of NOx in the mesosphere during the polar winter is EPP, these high NO2 mixing ratios
are unambiguously identified as being caused by EPP. NO$_2$ mixing ratios reached a maximum of 18 ppbv in the upper stratosphere near 2600 K (2452 km) at the end of January. Upper atmosphere EPP-NO$_x$ 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$_2$ 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$_2$ above 2000 K after 8 February, but there is little indication of further descent or substantially enhanced NO$_2$ 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$_2$ mixing ratios near 1500 K to decrease slightly. After the vortex recovered, however, NO$_2$ descent continues into March, reaching below 1000 K by the end of March. Some of the increase in NO$_2$ mixing ratios in March below 1500 K can be attributed to the conversion of reservoir species back into NO$_2$ as sunlight returns to the polar region.

Figure 1. Nighttime NO$_2$ 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$_2$ over time.

In the 2003–2004 Arctic winter, the MIPAS NO$_2$ (Figure 1c) shows two distinct enhancements in stratospheric NO$_2$. 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$_2$ mixing ratios
were affected by a major SSW in late December. At this time, the downward transport was interrupted, and the NO$_2$-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$_2$. 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 NO$_2$ 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$_x$ 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].

3.2. LIMS Nighttime NO$_2$ Versus ACE-FTS NO$_x$

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
is a strongly elevated stratopause height, which is indicative of enhanced adiabatic descent causing warming at altitudes that are normally in the mesosphere [Haukecorne 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.

### 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$_2$ mixing ratios in the LIMS NO$_2$. They found that an increase in excess of 20 ppbv of NO$_2$ 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$_2$ 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
LIMS EPP-NOx enhancements cannot be attributed to unusually high EPP levels.

4. Quantification of EPP-NOx in LIMS, MIPAS, and ACE-FTS

4.1. LIMS and MIPAS EPP-NOx

In this section we describe the method used to quantify the absolute amount of EPP-NOx 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 NO2 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-NOx. This requires the assumption, however, that the increase in NO2 in the polar stratospheric vortex is entirely due to the EPP IE.

We estimated the amount of EPP-NOx 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-NOx 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 NOx to upper atmospheric origin. As above, we have used nighttime NO2 as a proxy for NOx. Our method assumes that NO2 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 NO2 are due to the EPP IE. For each day on each isentropic surface the average NO2 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 NO2 molecules on each 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.

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 CH4 as a tracer and to previous studies. Figure 5 shows scatterplots of MIPAS nighttime NO2 versus CH4 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 NOx and CH4 are generally positively correlated, with the lowest values of CH4 indicating transport of air from above; in the absence of EPP-induced enhancements this air is depleted in NOx. Because the only source of NOx in the polar winter mesosphere is EPP, increasing values of NOx with decreasing CH4 indicate the presence of EPP-induced enhancements in the air transported from above [see Siskind et al., 1997].
for anticorrelations between CH₄ and NO₂ using CH₄ < 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” NO₂. 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 NO₂ 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 × 10⁸ cm⁻³. Peak densities are near 30 × 10⁸ cm⁻³ in the 2003 SH winter and 5 × 10⁸ cm⁻³ in the 2002–2003 NH winter.

[26] Figure 6 compares the subtraction and CH₄ 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. (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 CH₄ method, and the black lines represent the subtraction method. Error bars on the CH₄ method indicate 1-sigma standard deviations in the calculated excess NO₂. The total number of gigamoles integrated over each time period is given in Figure 6 (bottom): blue for the CH₄ method and black for the subtraction 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 CH₄ (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 (üw) 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.

[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
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$_4$ (dashed blue lines) methods. The total amount of EPP-NO$_x$ 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$_4$) method. Clearly, the results from the subtraction method compare well with the results from the CH$_4$ method, supporting the validity of applying the subtraction method to satellite measurements of nighttime NO$_2$. Our results from the subtraction method also compare reasonably well with previous estimates based on measurements of NO+NO$_2$ and NO$_x$. F05 calculated the net deposition of NO$_x$ 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$_x$ to NO$_2$ 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$_x$ 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$_x$ in the stratosphere from July 2002 to March 2004. They estimated the excess NO$_x$ deposited below 55 km for the 2003 Antarctic winter to be $1.4$ Gmol. Their estimate of excess NO$_x$ 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$_4$ method, we conclude that our approach is valid and is reasonable to apply to LIMS data.

![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.](image-url)
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.

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.

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).

4.2. ACE-FTS EPP-NO$_x$

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 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 $100 \times 10^8$ cm$^{-3}$. Peak densities are near $20 \times 10^8$ cm$^{-3}$ in 2005–2006 and $7 \times 10^8$ cm$^{-3}$ in 2008–2009. Figure 10
The Total Number of Gigamoles of EPP-NOx Crossing 1500, 2000, and 3000 K for LIMS, MIPAS, and ACE-FTS for the Relevant Arctic Winters

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<td>LIMS (2000 K)</td>
<td>- 0.1–0.2</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>MIPAS-ESA (2000 K)</td>
<td>- 0.2</td>
<td>- 2.3 (1.4)</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>MIPAS-IMK (2000 K)</td>
<td>- -</td>
<td>- 2.7 (1.9)</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>ACE-FTS (2000 K)</td>
<td>- -</td>
<td>- 1.8</td>
<td>- 0.2</td>
<td>- 0.2</td>
<td>- 0.1</td>
<td>- 0.05</td>
<td>- 0.02</td>
<td>-</td>
</tr>
<tr>
<td>LIMS (3000 K)</td>
<td>- 0.1–0.2</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>MIPAS-IMK (3000 K)</td>
<td>- 0.2</td>
<td>- 2.8 (0.3)</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>ACE-FTS (3000 K)</td>
<td>- -</td>
<td>- 0.4</td>
<td>- 0.2</td>
<td>- 0.7</td>
<td>- 0.1</td>
<td>- 0.1</td>
<td>0.1</td>
<td>0.3</td>
</tr>
</tbody>
</table>

*The amount shown in parentheses is the amount after 21 February, for comparison to the ACE-FTS data. Nighttime NO2 is used for the first three rows of Table 1, and NOx 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$_x$ 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$_x$ reaching the stratosphere. If we exclude the years with unusual dynamics, the correlation coefficient between the $Ap$ index and the amount of EPP-NO$_x$ 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$_2$ 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$_2$ 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$_x$ descended past the 1500 K, 2000 K and 3000 K surfaces in 1978–1979. This is similar to the amount of EPP-NO$_x$ in the winters just mentioned. For reference, 0.1 Gmol is approximately 0.5% of the annual average contribution to Arctic (>50° latitude) NO$_x$ from the oxidation of N$_2$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$_x$ 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$_x$, 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$_x$ 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$_x$ 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$_x$ transported to the stratosphere depends on both the level of EPP and the prevailing dynamics. That more total EPP-NO$_x$ 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$_x$-rich air from the MLT to the stratosphere. In 2005–2006 and in 2008–2009, substantially more EPP-NO$_x$ 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$_x$ 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$_x$ 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$_2$ in the vortex was EPP-NO$_x$ from above. This assumption will not be valid if there are intrusions of NO$_x$-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$_4$ 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].

[41] The NO$_x$ 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 NO$_2$ 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$_x$ descending to the NH stratosphere in the 1980s and 1990s was most likely due to a lack of appropriate measurements. Nighttime measurements of NO$_2$ are a useful proxy for total NO$_x$ in the stratosphere and lower mesosphere [e.g., Seppälä et al., 2007]. An instrument like LIMS would not be able to capture NO$_x$ in the mesosphere and higher, however, since above about 65–70 km NO$_2$ is in the form of NO. To fully characterize the EPP-NO$_x$ production and descent, observations of NO$_x$ from the stratosphere to the lower thermosphere throughout the polar winter are required.

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