Temporal variability of the descent of high-altitude NOX inferred from ionospheric data

Mark A. Clilverd¹, Annika Seppälä², Craig J. Rodger³, Neil R. Thomson⁴, János Lichtenberger⁴, and Péter Steinbach⁵

Abstract. In this study we investigate periods of enhanced ionization in the mesosphere during northern hemisphere winter-times. Long-lasting ionization enhancements (days) are typically produced by solar proton events, or by the descent of thermospheric NOX during periods of sustained downwards vertical transport associated with a strong underlying polar vortex. Using a new application of ground-based low frequency radio wave remote sensing we study the mesospheric ionization conditions during the northern hemisphere winters spanning 2003-04, 2004-05, and 2005-06. The winter 2003-04 subionospheric radio wave propagation data from a transmitter in Iceland shows signatures of the descent of NOX through 80 km altitude starting on January 13, 2004, during the occurrence of a strong polar vortex, indicating a thermospheric source for the NOX. Similar analysis of radio wave propagation data in the northern hemisphere winter of 2004-05 does not show a NOX descent event passing through the mesosphere, due to a lack of downward vertical transport as a result of a weak underlying polar vortex, despite the occurrence of significant solar proton ionization during January 2005. In 2005-06 there were no significant ionization events and also no descent of significant amounts of thermospheric NOX, despite a strong polar vortex and strong vertical transport. We model the signature of the descent of NOX seen in the radio wave propagation data using the Sodankylä Ion Chemistry model, confirming that the levels of NOX in the mesosphere are ~100 times the usual background levels. The combination of strong NOX sources in the thermosphere and also a strong polar vortex is required for NOX to descend into the stratosphere with significant concentration levels.
1. Introduction

Winter-time polar odd nitrogen, NO\textsubscript{X} (NO + NO\textsubscript{2}), is produced at high altitudes in the thermosphere and the mesosphere. During periods of efficient vertical transport the NO\textsubscript{X} can descend to the stratosphere. In the upper mesosphere the NO\textsubscript{X} is mainly in the form of NO. As the NO\textsubscript{X} descends below 70 km it is converted to NO\textsubscript{2} [Solomon et al., 1982a; Brasseur and Solomon, 2005]. NO\textsubscript{X} plays a key role in the ozone balance of the middle atmosphere because it destroys odd oxygen (O+O\textsubscript{3}) through catalytic reactions [e.g., Brasseur and Solomon, 2005, pp. 336-355]. Hard energetic particle precipitation (EPP) into the mesosphere (that including a significant population of >100 keV electrons and >1 MeV protons), and softer EPP into the thermosphere (<100 keV electrons), generate in-situ enhancements in odd nitrogen. The mesospheric source is dominated by strong impulsive ionization episodes such as solar proton events [Verronen et al., 2005], while the thermospheric source is more continuous, being dominated by auroral ionization [Siskind, 2000]. During the dark polar winter, odd nitrogen can survive, and in the presence of strong polar vortex conditions, descend into the stratosphere [Solomon et al., 1982a; Siskind, 2000]. During the northern polar winter of 2003-2004 these conditions existed; Randall et al. [2005] reported unprecedented levels of spring-time stratospheric NO\textsubscript{X} as a result. Rinsland et al. [2005] also observed very high NO\textsubscript{X} mixing ratios at 40-50 km in February/March 2004 with the ACE experiment, detecting levels as high as 1365 ppbv. Randall et al. [2006] presented stratospheric data from 2003-2006, confirming the descent of NO\textsubscript{X} in spring 2004, and showing the descent of small amounts of NO\textsubscript{X} in the northern polar vortex during the spring of 2006, while none was observed in the spring of 2005.

Although several powerful solar storms occurred at the beginning of the 2003-04 winter period (October and November) the principle source of the NO\textsubscript{X} was uncertain because of the breakup of the stratospheric vortex in late December 2003. Clilverd et al. [2006a] used radio wave data to show that the primary source for the NO\textsubscript{X} was in the auroral zones in the thermosphere, and not a result of in situ production in the mesosphere. The descent of the NO\textsubscript{X} began a few days after the end of the stratospheric warming event at the end of December 2003. In that study it was unclear if the thermospheric reservoir of high-altitude NO\textsubscript{X} was generated by the large solar storms several months earlier, or by an accumulation of the effects of smaller storms as suggested by Siskind [2000]. Conversely, Renard et al. [2006] suggested that the source of descending NO\textsubscript{X} was from in-situ production at around 60 km caused by electrons of a few hundred keV, due to a geomagnetic storm that occurred on 22-25 January 2004.

One of the few experimental techniques which can probe the ionization at these altitudes uses very low-frequency (VLF) electromagnetic radiation, trapped between the lower ionosphere and the Earth [Barr et
The nature of the received radio waves is largely determined by propagation between these boundaries [e.g., Cummer, 2000], termed "subionospheric propagation". Subionospheric VLF propagation allows remote sensing of the upper atmosphere over large regions; these signals can be received thousands of kilometers from the source, where ionospheric modifications lead to changes in the received amplitude and phase. Man-made VLF transmitters provide a well defined modeling situation, due to the known transmitter-receiver locations and fixed frequency operation, generally operating near constantly [Barr et al., 2000]. By using multiple VLF communication transmitters we have previously gained good understanding of the daytime lower ionosphere [Thomson, 1993] and variations during transient changes (e.g., solar flares [Thomson et al., 2005], low-medium energy electron precipitation [Rodger et al., 2005], and solar proton precipitation [Clilverd et al., 2005]).

In this study we analyze ground-based ionospheric data from mid-high latitudes during the northern polar winters of 2003-2004, 2004-2005, and 2005-2006. Subionospheric VLF radio wave propagation is sensitive to ionization located below about 90 km, including that produced by the ionization of NOX by Lyman-α [Solomon et al., 1982b]. The effect of increased ionization on the propagating signals can be seen as either an increase or decrease in signal amplitude or phase depending on the modal mixture of each signal observed. Given the dynamic behavior of the polar vortex, and the mixing that occurs in it during the winter months, it is reasonable to expect the path integrated radio wave measurements to respond to any large-scale NOX changes that occur. From the changes in ionospheric propagation conditions during the winter period we determine the levels of mesospheric NOX either through in situ production or descent from the thermosphere. The three years studied showed significant differences in solar activity, and stratospheric vortex strength, allowing us to study the interplay between these two parameters. We compare the timing of the onset of the radio wave signatures of NOX in January 2004 with the NO2 data from GOMOS, which has the advantage over previous satellite observations of the descent of NOX in being able to measure NO2 at altitudes up to 70 km, and in the dark polar night conditions well inside the polar vortex.
2. Experimental setup

Here we use narrow band subionospheric LF data from a 37.5 kHz transmitter (call sign NRK, 64°N, 22°W, $L=5.6$) located in Iceland and received at two nearby sites. The receivers are located at: Ny Ålesund, Svalbard (79°N, 11°E, $L=18.3$), and Érd, Budapest (48°N, 19°E, $L=1.9$). These sites are part of the Antarctic-Arctic Radiation-belt Dynamic Deposition VLF Atmospheric Research Konsortia (AARDDVARK). Figure 1 shows the location of the transmitter site (cross), the receiver sites (diamonds), the Sodankylä Ion Chemistry (SIC) modeling location (70°N, 0°E, asterisk), and also indicates the great circle propagation paths between the transmitter and receivers. The dark circle at 60°N indicates an approximate location of the outer edge of the northern polar vortex. This is consistent with the 1700K sPV (potential vorticity) maps shown in Figure 4 of Manney et al. [2005] for a typical strong northern hemisphere upper stratospheric midwinter vortex.

Mesospheric ionization effects on VLF/LF wave propagation can be modeled using the Long Wave Propagation Code [LWPC, Ferguson and Snyder, 1990]. LWPC models VLF signal propagation from any point on Earth to any other point. Given electron density profile parameters for the upper boundary conditions, LWPC calculates the expected amplitude and phase of the VLF signal at the reception point. Thus it can be used to investigate the modification of the ionosphere as long as the induced changes to the electron density altitude-profiles are known. To provide this the Sodankylä Ion Chemistry model [SIC, version 6.8, Verronen et al., 2005] was used to determine the effects of the ionization of descending NO$_X$ on the mesospheric electron density profiles. The combination of LWPC and the SIC model to understand VLF observations has been reported in previous studies [e.g., Clilverd et al., 2005; 2006b].

In this paper we use NO$_2$ measurements from the GOMOS stellar occultation instrument [Bertaux et al., 2000; Bertaux et al., 2004; Kyrola et al., 2004] on board the Envisat satellite to investigate the timing of the onset of the radio wave signatures of NO$_X$ in January 2004. GOMOS has the advantage over previous satellite observations of the descent of NO$_X$ into the stratosphere in being able to measure NO$_2$ at altitudes up to 70 km [Hauchecorne et al., 2005], and in the dark polar night conditions well inside the polar vortex [Bracher et al., 2005; Hauchecorne et al., 2007]. The timing of the onset of the descent of NO$_X$ is important in defining the production mechanism, and high altitude observations also help clarify at which altitude it occurs. The GOMOS NO$_2$ measurements presented here are from the period January-February 2004. For this study we use only GOMOS dark limb (night-time) measurements from the Northern Hemisphere (GOMOS ground processing prototype, GOPR, version 6.0c) from occultations where the star temperature was $\geq 6000$ K for the optimal signal-to-noise ratio for data inversion at
upper stratospheric and mesospheric altitudes. In the current analysis we have included over 1800 GOMOS occultations from which nearly 1000 are from star 1 (Sirius, magnitude -1.44) and star 23 (21 Eps CMa, magnitude 1.5), and nearly 200 are from star 7 (19Bet Ori, magnitude 0.1). To improve both spatial and temporal coverage we have also included some stars with weaker magnitudes, for the latitudes from 65ºN to 90ºN the overall range of magnitudes is -1.44 to 2.58, while for 55ºN-65ºN it is 1.50 to 3.03. Night-time measurements of NO₂ are a good tracer for NOₓ in the stratosphere and the lower mesosphere, but not at higher altitudes where NOₓ is mainly in the form of NO [Brasseur and Solomon, 2005].


The winter of 2003-2004 was notable because of the unusually high strength of the upper stratospheric vortex in the period from mid-January 2004 to mid-March [Manney et al., 2005]. The stratospheric vortex allows mesospheric air to descend deep into the stratosphere during the polar night [Russell et al., 1993]. In 2003 the upper stratospheric vortex was apparent from the beginning of October, but was disrupted by a stratospheric warming event, reducing downward vertical transport, which lasted from 20 December 2003 to 10 January 2004. In our study all polar vortex dynamics observations are drawn from the NOAA stratospheric products website [http://www.cpc.ncep.noaa.gov/products/stratosphere/]. We make use of the zonal mean temperatures at 2 mb (~45 km altitude) to determine the timing and approximate intensity of the vortex dynamics in the upper stratosphere. In contrast with winter 2003-2004, in winter 2004-2005 the upper stratospheric vortex became less intense at the end of December 2004 and did not recover. The upper stratospheric vortex during the winter of 2005-2006 was similar in many respects to winter 2003-2004, in that an upper stratospheric warming event occurred in January, followed by a return to typical wintertime vortex conditions in February and March 2006.

Solar proton precipitation can produce high levels of NOₓ directly into the mesosphere and upper stratosphere because of the high proton particle energies which can penetrate deep into the atmosphere [see Seppälä et al, 2006, Figure 1, for a plot of atmospheric ionization rate from proton precipitation]. In winter 2003-2004 there was a large solar proton event (SPE) in late October 2003, followed by 4 smaller ones through November and December. In winter 2004-2005 there were two small SPEs in November 2004, followed by a large SPE in January 2005. In winter 2005-2006 no SPEs occurred.

High geomagnetic activity produces high levels of NOₓ through electron precipitation occurring at auroral altitudes (~120 km) and in some cases in situ in the mesosphere. In winter 2003-2004 there were 10 geomagnetic storms where the daily average Kₚ was greater than 4 in the 4-month period spanning 1 October to 31 January. A high proportion (8) of these occurred before the end of December. In winter 2004-
2005 there were 7 large geomagnetic storms, although most (4) occurred in January. In contrast, over winter 2005-2006 there were no large geomagnetic storms. Siskind [2000] used 4-month average $A_p$ to describe geomagnetic activity effects on high-altitude NOX production. The average $A_p$ between 1 October and 31 January ($<A_p>$) during the 2003-04 winter was 22.7, in winter 2004-2005 it was 17.3, and in winter 2005-2006 it was 6.9. The results of Siskind [2000] suggest that $<A_p> \sim 10$ is the lower threshold value above which significant concentrations of descending NOX are likely to be observed within the southern hemisphere polar vortex. We note that the level of $<A_p> \sim 10$ is our interpretation of the Siskind [2000] results.

In summary, the two winters with strong polar vortex conditions (2003-2004 and 2005-2006) have very different levels of solar and geomagnetic activity. Winter 2004-2005 has some periods of high geomagnetic activity, but differs in comparison with the other two winters in that it has weak polar vortex conditions after December.

4. Winter-time mesospheric NOX impact on radio wave observations, 2003-2006

The amplitude difference between midday and midnight for the NRK (Iceland) radio wave signal received at Ny Ålesund has been shown to be sensitive to changes in ionization conditions caused by the descent of NOX through VLF reflection heights, due to the production of additional ionization from solar Lyman-α radiation (and geo-coronal Lyman-α at night) which ionizes NO at altitudes of 65-95 km. [Clilverd et al., 2006a]. The effect of increased ionization on the propagating signals can be seen as either an increase or decrease in signal amplitude or phase depending on the modal mixture of the signal observed. The quiet day curve (QDC) exhibits around 7-8 dB lower amplitudes during the daylight hours (09-18 UT) than during the night, because of differences in the effective reflection height of the oblique-incidence radio waves and the effective electron density gradient with altitude at the reflection height.

In Figure 2 we plot the variation of the NRK amplitude difference (i.e., the difference between the midday amplitude value and the midnight amplitude in dB each day) for the winters of 2003-04, 2004-05, and 2005-06, with the plot starting at 01 October in each case, and day 1 = 01 January. The plots show periods where the difference amplitude is -8 dB, which is the value expected under normal quiet day curve (QDC) propagation conditions, and represented by a dot-dashed horizontal line. Vertical solid lines represent the times of known solar proton events (SPE) with the peak particle flux units (pfu) given in parentheses plotted close to the vertical solid lines. Vertical dotted lines indicate periods of subionospherically-observed increased ionization conditions, which do not correspond to SPE periods.
As was discussed in Clilverd et al. [2006a] the winter 2003-04 was marked by several large solar proton events in the early winter period, followed by the descent of NO\textsubscript{X} into the mesosphere from the thermosphere on January 13, 2004 (day 13 in Figure 2, top panel). This date coincides with the end of the warming event in the upper stratosphere and the beginning of a period with a strong polar upper stratospheric vortex [NOAA website] leading to strong downward vertical transport [Solomon, 1982a].

Enhanced ionization levels in the mesosphere detected through our subionospheric propagation measurements ended about February 19, 2004, lasting for 37 days in total. A less clear enhancement of mesospheric ionization occurred in October, 2003, where an increased difference amplitude was briefly observed, but quickly swamped by the large SPEs that occurred a few days later. We note here that the ionization increases from SPEs can be so large that they dominate the generation of ionization produced by the ionization of descending NO\textsubscript{X}, hence masking its detection through our technique.

In winter 2004-05 there were several notable SPEs, but no clear signature of the descent of NO\textsubscript{X} into the mesosphere. In contrast to the previous winter the strong polar vortex was weak in January-March 2005, and thus no strong downward vertical transport of NO\textsubscript{X} occurred. Two periods of subionospherically detected enhanced ionization, not associated with SPEs, are indicated by vertical dotted lines in the panel, one occurring in early December, 2004, and the other at the end of January, 2005. Both are short-lived (~5 days), and do not show the expected signature of significant NO\textsubscript{X} descent, in that the timescales are relatively short and not the ~30 days as observed previously. It is possible that the enhanced ionization at the end of January 2005 is due to the descent of thermospheric NO\textsubscript{X}, generated by the previous period of high geomagnetic activity, although there is no evidence for this in ACE data shown Fig. 1 in Randall et al. [2006], and downward vertical transport was unlikely to be significant because the underlying upper stratospheric vortex was not strong at this time.

In winter 2005-2006 no signatures of SPEs or other enhanced ionization periods were observed in the data, despite a strong polar vortex and downward vertical transport after January 25, 2006, lasting more than a month. This indicates that although strong downward vertical transport was present, the sources of NO\textsubscript{X} in the thermosphere and mesosphere were too small to produce significant ionization enhancements at the altitudes probed by VLF propagation. As the average A\textsubscript{p} in the 4-month winter period was 6 this observation is consistent with the threshold of >10 for observable NO\textsubscript{X} descending to 45 km, indicated by the results of Siskind [2000].

The contrast between the three winter periods (2003-06) shows a highly variable source of NO\textsubscript{X} in the thermosphere and mesosphere, due to particle precipitation. The appearance of NO\textsubscript{X} in the stratosphere,
from these high altitude sources, is further controlled by the presence of a strong polar vortex. ACE observations of NO$_2$ during these three winters show that no NO$_2$ was observed descending into the stratosphere in spring 2005, and only a small amount was observed in spring 2006 [Randall et al., 2006]. The results presented in this paper are consistent with those observations. Only when both source and transport are present will significant amounts of NO$_X$ descend into the stratosphere.

5. Modeling NO$_X$ in the mesosphere

In order to model the effect of descending NO$_X$ on the ionization levels as detected through radio wave propagation we employ the SIC model with varying NO$_X$ levels. The resultant electron density profiles are input into the Long Wave Propagation Code (LWPC) following the approach described by Clilverd et al. [2005, 2006b]. Changes in NO$_X$ are modeled by arbitrarily increasing the normal NO profile at a given altitude by factors of 10 - 1000. Figure 3 shows the effect on the electron density profile of a 15 km altitude-limited slab in which there is a 100 times enhancement of NO. The figure shows the effects of the slab centered on 90 km and 70 km, in comparison with the midday and midnight electron density profiles calculated with the SIC model. In all cases a low level proton flux is included in the SIC model runs to provide realistic non-disturbed conditions. At heights above ~70 km, the electron densities in the region of the NO enhancement are increased by a factor of 10, which is consistent with the earlier conclusions of Clilverd et al. [2006a] concerning periods of enhanced ionization due to NO$_X$ transport during the 2003-2004 winter. However at altitudes lower than ~70 km the NO$_X$ enhancement leads to no significant increase in electron density levels because most of the Lyman-$\alpha$ is absorbed at altitudes above the slab height.

Figure 4 shows the LWPC-calculated diurnal changes in the amplitude received from the NRK transmitter at Ny Ålesund, caused by the SIC-modeled NO$_X$ descent for January conditions. The electron density changes due to the NO enhancements have been represented in the LWPC model by exponential profile fits restricted over the altitude range 30-110 km [Wait and Spies, 1964], applied over the entire transmitter-receiver great circle path. In Figure 4 the nighttime amplitudes (00-08 UT, and 18-24 UT) are generally reduced as a result of the enhanced NO, while daytime amplitudes (09-17 UT) increase. The model results shown represent increases of NO by a factor of 10, 100, 300, and 1000 at 80 km altitudes. The plot shows that the nighttime amplitudes decrease with limited sensitivity to the absolute NO enhancement factor, but the daytime amplitudes change consistently with NO enhancement factor.

In Figure 5 we show the QDC from data recorded prior to, and just after, the NO descent period (solid line), as well as the average amplitude during descent period (16-20 January 2004, dashed line). Modeling results from SIC/LWPC are shown by diamonds (QDC, 1×NO), and asterisks (300×NO). As the observed QDC
amplitudes are un-calibrated they have been adjusted to the same level as the SIC/LWPC QDC levels which were arbitrarily based on a transmitter output power of 200 kW. We use 200 kW as the exact output power of the NRK transmitter is unknown to us. The 1×NO results agree well with the QDC propagation conditions observed in the data, while the 300×NO increase occurring at 80 km altitude produces behavior similar to that observed in the NRK data during the NOX descent period starting on January 13, 2004, i.e., a 3 dB decrease during the nighttime and higher daytime amplitudes than during nighttime, giving a day-night difference amplitude of +2.5 dB. Clearly the modeling indicates that the effect of increased NO is detectable on both the nighttime and the daytime radio wave data. In Figure 2 we plot the day-night difference, although we note here that it is possible to plot either day or night individually and still observe the effect. Plotting the day-night difference increases the NO effect on the data, however occasionally operational constraints only allows data collection at either one or the other – this is the case in the data used for section 6 below.

Figure 6 shows the change in day-night difference amplitude as the NO enhancement descends in altitude from 95 to 65 km with 5 km resolution steps. The plot shows the variation for three levels of NO enhancement – 10×NO, 100×NO, and 1000×NO, where 1×NO is the SIC model value for the model location, time of year, and time of day. SIC/LWPC results show that because the day-night difference peaks at 80 km the radio waves are primarily sensitive to the NOX enhancement as it passes through ~80 km, with little impact on the diurnal variation in amplitude once the NOX reaches 65 km and below. Hence the subionospheric propagation is sensitive to the early stages of NOX descent, and can also signal the end of a NOX descent period, but only if the NOX changes are significant, i.e., factor of ~10 changes. Satellite-based techniques tend to identify NOX enhancements at altitudes below 65 km [e.g., Randall et al., 2005; Rinsland et al., 2005]. Subionospheric VLF propagation therefore provides significant additional lead times, detecting NOX enhancements which are transported to the stratosphere ~6 weeks later.

6. Latitudinal extent of NOX descent

Up to this point we have shown the effects of the descent of NOX on the Iceland to Svalbard path (NRK to Ny Ålesund). This path is well inside the usual coverage of the polar vortex. In Figure 7 we show the variation of the 2003-04 winter-time average nighttime amplitude for the Iceland to Hungary path (NRK to Érd) in comparison with the NRK to Ny Ålesund difference amplitude for the same period. We use only nighttime data from Érd because of limited daytime recordings made during this period. However as shown previously in Figure 5 and 6 the nighttime-only data is sensitive to increased NOX. On the NRK to Érd path the location of the transmitter represents the highest latitude that could be affected by descending NOX, i.e., 64°N. The plot shows that the Érd nighttime amplitude responds to both SPEs and the descent of NOX.
horizontal and vertical lines are as Figure 2, with the addition of long-dashed vertical lines showing the end
of the descent of NO\textsubscript{X} in February 2004. The solar proton events of October/November 2003 show up
clearly as short-lived (days) increases in nighttime amplitude of \(-7 \text{ dB}\).

The descent of NO\textsubscript{X} in January 2004, is also observed on this lower latitude path, possibly starting
intermittently on January 6, 2004, and clearly by 12 January, lasting 42 days, but finishing prior to the end
of the NO\textsubscript{X}-produced enhanced ionization observed on the higher latitude path. This suggests that either
downward vertical transport begins \(-7 \text{ days}\) earlier at lower latitudes, and finishes earlier by \(-2 \text{ days}\), or that
the outer regions of the polar vortex have faster descent rates than the inner regions – detailed analysis of
the dynamics of the outer edge of the polar vortex is beyond the scope of this study, however, we note that
this interpretation is consistent with Callaghan and Salby [2002] who show that the maximum descent rate
in the polar vortex is not over the pole, but at 60°N. A recovery of the Érd nighttime amplitudes occurred
during early February 2004 (about day 40) suggesting that the downward transport faltered in the lower
latitude sector during this time.

These observations indicate that strong downward vertical transport occurs significantly equator-wards
of 64°N, increasing the ionization levels on at least part of the path to Érd. Clilverd et al. [2006b] showed
that changes in ionization over the first 500 km of this path caused by SPEs were able to produce
significant changes in received signal amplitude at Érd. SIC/LWPC model runs [not shown] with 100×NO
at 80 km altitude applied to the first 500 km of the path are able to produce the changes observed in Figure
7, i.e., the NO descent also occurs equator-wards of 64°N, which is consistent with previous satellite
observations [Randall et al. 2006].

GOMOS observations of NO\textsubscript{2} in January and February 2004 are shown in Figure 8. The observations are
averaged over two days to provide the results presented. The left-most panels show the NO\textsubscript{2} zonal mean number
density and mixing ratios for latitudes \(>65°\text{N}\). The right-most panels show the NO\textsubscript{2} zonal mean number density and
mixing ratios for latitudes 55°-65°N. Enhanced levels of NO\textsubscript{2} are seen descending from about 70 km starting on
January 10, 2004, reaching altitudes of 45 km by the end of February 2004. Each averaged data point is formed
from 15-30 measurements, only a small number of points have less than 15 measurements. Our key conclusions
from the GOMOS data (Figure 8 left) are based on nearly 500 occultations of Sirius, which is considered the best
star used in GOMOS occultations, providing excellent signal-to-noise for the measurement of NO\textsubscript{2}. 
The descending NO$_2$ enhancement is about 100 times that of the background density, and thus shows consistency with the radio wave modeling undertaken above. The enhancement passes below 60 km in early-February, at which time the modeling results indicate that the signature of the NO$_x$ descent should have ceased in the radio wave data. However, a further increase in NO$_2$ at altitudes >60 km occurs as a result of particle precipitation during the geomagnetic storm of 11-15 February, 2004. The NO$_2$ eventually descends below 60 km completely after 19 February, consistent with the radio wave observations.

In the right-most panels, representing lower latitude GOMOS observations (55°-65°N), similar signatures of descent of enhanced NO$_2$ can be seen during January and February, 2004. However, contrary to the radio wave observations from Érd there is no clear signature of the 11-15 February 2004 geomagnetic storm in the GOMOS data, suggesting that the radio wave propagation data is responding more sensitively to a lower latitude source of ionization that is not associated with the enhancement of significant levels of NO$_2$. Additionally there is no clearly observable onset date with which to confirm the NO$_x$ descent start date of 5 Jan 2004 suggested by the radio wave data, primarily because of a weaker descent feature in these lower latitude measurements.

In Figure 8 we plot the GOMOS data up to an altitude of 70 km. Even using the brightest stars GOMOS is only able to measure NO$_2$ from 20-70 km. Outside these altitude ranges GOMOS NO$_2$ data has to be considered with caution [Hauchecorne et al., 2005]. Figure 8 indicates that the descent of NO$_2$ began around 10 January 2004, about 2 weeks before the geomagnetic storm suggested by Renard et al. [2006] as the cause of the lower mesospheric NO$_2$ enhancement. In fact GOMOS data indicates that the 22-23 January 2004 geomagnetic storm created no observable enhancement of NO$_2$ in the 50-70 km altitude range. In contrast the storm of 11-12 February 2004 can be seen to produce significant amounts of NO$_2$ below 70 km, contributing to the body of NO$_2$ descending into the stratosphere. The solar wind speed and density increased more markedly during the 11 February storm than the 22 January storm, which is consistent with the enhanced production of NO$_x$ by particle precipitation [e.g., Rozanov et al., 2005]. The gradual increase in NO$_2$ number density from 10 January to 20 January at altitudes of about 65 km is consistent with the increasingly effective conversion of the descending NO to NO$_2$ at altitudes below 70 km through reaction with atomic oxygen [Brasseur and Solomon, 2005]. These observations argue strongly against the interpretation of Renard et al. [2006] where the source of descending NO$_x$ was suggested to come from in-situ production at around 60 km caused by electrons of a few hundred keV, due to a geomagnetic storm that occurred on 22-25 January 2004.
7. Discussion and Summary

In this study we have investigated periods of enhanced ionization in the mesosphere during northern hemisphere winter-times. Ionization enhancements lasting days are typically produced by solar proton events. In contrast, ionization enhancements lasting weeks are caused by the descent of thermospheric NO$_X$ during periods of sustained downwards vertical transport associated with a strong underlying polar vortex.

Using a new application of a well established radio wave propagation technique, we have shown that only during winters with both high geomagnetic activity (4-monthly mean $A_p>10$) and strong downward vertical transport, will large enhancements of polar NO$_X$ be transported down to the stratosphere. These conditions were met in winter 2003-2004, but not in 2004-2005, or 2005-2006 as only high geomagnetic activity or strong downward transport occurred, but not both simultaneously.

In winter 2003-2004 the descent of polar NO$_X$ began on 13 January 2004, a few days after the onset of strong downward transport associated with a strong underlying upper stratospheric polar vortex. The start date is not associated with any specific geomagnetic storm, but rather due to the formation of a strong polar vortex giving rise to strong downward vertical transport of a pre-existing thermospheric reservoir of NO$_X$. The existence of a significant thermospheric NO$_X$ reservoir is dependent on the average $A_p>10$ over the preceding 4 months, which agrees with the results of Siskind [2000]. This finding is consistent with the impact of geomagnetically enhanced auroral electron precipitation, leading to a significantly enhanced thermospheric NO$_X$ population in the polar winter.

Both our radio propagation data and independent GOMOS observations indicate significant NO$_X$ descent into the stratosphere from latitudes equator-wards of 65°N. Ground-based measurements made in Hungary indicated that the descent started a few days earlier at lower latitudes than at high latitudes.

The lack of influence of any specific geomagnetic storm argues against an in-situ NO$_X$ production mechanism at altitudes of around 60 km. The results of propagation modeling using the SIC calculations, undertaken to simulate our radio wave observations, are also consistent with a source population descending from >90 km, i.e., auroral altitudes in the thermosphere. This is consistent with previous work indicating that the auroral zone can produce significant quantities of NO$_X$ as a result of geomagnetic disturbances [Baker et al., 2001; Barth 2003]
Acknowledgments. The authors would like to thank Frank Skogen of the Kings Bay Company for overseeing the collection and return of the Ny Ålesund data. We would also like to thank the GOMOS team for their useful and informative discussions regarding this work.
References

Baker, D. N., C. A. Barth, K. E. Mankoff, S. G. Kanekal, S. G. Bailey, G. M. Mason, and J. E. Mazur
(2001), Relationships between precipitating auroral zone electrons and lower thermospheric nitric oxide


Barth, C. A., K. D. Mankoff, S. M. Bailey, and S. C. Solomon, Global observations of nitric oxide in the

Bertaux, J. L., E. Kyrölä, and T. Wehr (2000), Stellar occultation technique for atmospheric ozone


Company, Dordrecht.

Bracher, A., et al. (2005), Cross comparisons of O3 and NO2 measured by the atmospheric ENVISAT

Callaghan, P. F., and M. L. Salby (2002), Three-dimensionality and forcing of the Brewer-Dobson
circulation, *J. Atmospheric Sciences*, 59, 976-991.

Modeling a large solar proton event in the southern polar atmosphere, *J. Geophys. Res.*, 110, A09307,


Clilverd, M. A., A. Seppälä, C. J. Rodger, N. R. Thomson, P. T. Verronen, E. Turunen, Th. Ulich, J. Lichtenberger, and P. Steinbach (2006b), Modeling polar ionospheric effects during the October-

Transactions on Antennas and Propagation*, 48(9), 1420-1429.

Ferguson, J. A., and F. P. Snyder (1990), Computer programs for assessment of long wavelength radio


---

1M. A. Clilverd, Physical Sciences Division, British Antarctic Survey, High Cross, Madingley Road, Cambridge CB3 0ET, England, U.K. (e-mail: macl@bas.ac.uk)

2A. Seppala, Finnish Meteorological Institute, Helsinki, Finland (email: annika.seppala@fmi.fi)

3C. J. Rodger and N.R. Thomson, Department of Physics, University of Otago, P.O. Box 56, Dunedin, New Zealand. (email: crodger@physics.otago.ac.nz and n_thomson@physics.otago.ac.nz)

4J. Lichtenberger, Space Research Group, Eötvös University, Budapest H 1518, Hungary. (email: spacerg@sas.elte.hu)

5P. Steinbach, Research Group for Geoinformatics and Space Sciences, MTA-ELTE, Budapest, Hungary. (email: spacerg@sas.elte.hu)

(Received N x, 2006; N x 27, 2006; accepted N x, 2007.)

CLILVERD ET AL.: NO₃ SOURCES AND TRANSPORT
Figure 1. The location of subionospheric propagation paths from Iceland to the AARDDVARK receiver sites at Ny Ålesund, and Érd. The Sodankylä Ion Chemistry (SIC) modeling location is also shown.

Figure 2. The day-night difference amplitudes for the Iceland transmitter (NRK) received at Ny Ålesund (NYA) during three winters. Normal values are indicated by the horizontal dashed line. Times of identified solar proton events are given by solid vertical lines with the peak particle flux units (pfu) are given in parentheses plotted close to the vertical solid lines, while similar behavior with no identified proton event is shown by the vertical dotted lines.

Figure 3. The midday (12 UT, bold line) and midnight (00 UT, lighter line) electron density profiles from SIC showing the effect of enhanced NO centered about 90 km (circles), and 70 km (crosses).

Figure 4. The modeled diurnal amplitude of the Iceland transmitter received at Ny Ålesund, Svalbard, under the influence of 10, 100, 300, 1000 × NO enhancements centered on 80 km altitude, compared with the normal diurnal curve (1xNO).

Figure 5. The diurnal amplitude of the Iceland transmitter received at Ny Ålesund, Svalbard, (solid line) compared with the average amplitude during 16-20 January 2004 (dashed line). SIC/LWPC model results are shown for the QDC (1×NO, diamonds) and for a 300×NO enhancement (asterisks) centered at 80 km.

Figure 6. The winter-time differences in day-night amplitude for the Iceland transmitter (NRK) received at Ny Ålesund (NYA) during the descent of a layer of different enhancement factors (10, 100, and 1000). The 1 x NO case is plotted as a dot-dashed line at -3 dB.

Figure 7. The winter-time average nighttime amplitude for the Iceland transmitter received at Érd, Hungary, during winter-time 2003-04 compared with the NRK to Ny Ålesund difference amplitude for the same period. The horizontal and vertical lines are as Figure 2 where the peak particle flux units (pfu) are given in parentheses plotted close to the vertical solid lines, with the addition of long-dashed vertical lines showing the end of the descent of NOX in February 2004.

Figure 8. (Left top) The GOMOS nighttime NO₂ number density for latitudes >65°N during the period January and February 2004. (Left bottom) The NO₂ mixing ratio over the same period. (Right top) The NO₂ number density for latitudes 55-65°N from January and February 2004. (Right bottom) The NO₂ mixing ratio over the same period. The nighttime NO₂ measurements are used as an estimation of the total NOₓ (NO + NO₂).
Figure 1. The location of subionospheric propagation paths from Iceland to the AARDDVARK receiver sites at Ny Ålesund, and Érd. The Sodankylä Ion Chemistry (SIC) modeling location is also shown.
Figure 2. The day-night difference amplitudes for the Iceland transmitter (NRK) received at Ny Ålesund (NYA) during three winters. Normal values are indicated by the horizontal dashed line. Times of identified solar proton events are given by solid vertical lines with the peak particle flux units (pfu) are given in parentheses plotted close to the vertical solid lines, while similar behavior with no identified proton event is shown by the vertical dotted lines.
Figure 3. The midday (12 UT, bold line) and midnight (00 UT, lighter line) electron density profiles from SIC showing the effect of enhanced NO centered about 90 km (circles), and 70 km (crosses).
Figure 4. The modeled diurnal amplitude of the Iceland transmitter received at Ny Ålesund, Svalbard, under the influence of 10, 100, 300, 1000 × NO enhancements centered on 80 km altitude, compared with the normal diurnal curve (1xNO).
Figure 5. The diurnal amplitude of the Iceland transmitter received at Ny Ålesund, Svalbard, (solid line) compared with the average amplitude during 16-20 January 2004 (dashed line). SIC/LWPC model results are shown for the quiet day curve (QDC, 1×NO, diamonds) and for a 300×NO enhancement (asterisks) centered at 80 km.
Figure 6. The winter-time differences in day-night amplitude for the Iceland transmitter (NRK) received at Ny Ålesund (NYA) during the descent of a layer of NO with different enhancement factors (10, 100, and 1000). The 1 x NO case is plotted as a dot-dashed line at -3 dB.
Figure 7. The winter-time average nighttime amplitude for the Iceland transmitter received at Érd, Hungary, during winter-time 2003-04 compared with the NRK to Ny Ålesund difference amplitude for the same period. The horizontal and vertical lines are as Figure 2 where the peak particle flux units (pfu) are given in parentheses plotted close to the vertical solid lines, with the addition of long-dashed vertical lines showing the end of the descent of NOX in February 2004.
Figure 8. (Left top) The GOMOS nighttime NO₂ number density for latitudes >65°N during the period January and February 2004. (Left bottom) The NO₂ mixing ratio over the same period. (Right top) The NO₂ number density for latitudes 55-65°N from January and February 2004. (Right bottom) The NO₂ mixing ratio over the same period. The nighttime NO₂ measurements are used as an estimation of the total NOₓ (NO + NO₂).
NRK at NYA: Diurnal amplitude

Amplitude (dB)

Time (UT)

QDC

16-20 Jan 04