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Arctic and Antarctic polar winter NO_x and energetic particle precipitation in 2002–2006

Annika Seppälä,[1] Pekka T. Verronen,[1] Mark A. Clilverd,[2] Cora E. Randall,[3]
Johanna Tamminen,[1] Viktoria Sofieva,[1] Leif Backman,[1] and Erkki Kyrölä[1]

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[1] We report GOMOS nighttime observations of middle atmosphere NO_2 and O_3 profiles during eight recent polar winters in the Arctic and Antarctic. The NO_2 measurements are used to study the effects of energetic particle precipitation and further downward transport of polar NO_x. During seven of the eight observed winters NO_x enhancements occur in good correlation with levels of enhanced high-energy particle precipitation and/or geomagnetic activity as indicated by the A_p index. We find a nearly linear relationship between the average winter time A_p index and upper stratospheric polar winter NO_2 column density in both hemispheres. In the Arctic winter 2005–2006 the NO_x enhancement is higher than expected from the geomagnetic conditions, indicating the importance of changing meteorological conditions.

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1. Introduction

[2] Precipitating charged particles with high energies produce odd nitrogen, NO_x ($NO + NO_2$) in the Earth's atmosphere. During polar winters when little or no sunlight is available in the polar regions, the photochemical lifetime of NO_x increases significantly from days to months and the NO_x distribution becomes subject to dynamical control. The polar vortex formed near the winter pole isolates the polar air (and hence the generated NO_x) and hinders latitudinal mixing. This allows the NO_x produced at higher altitudes to be transported downward along with the descending vortex air. The NO_x produced by energetic particles can have a significant impact on the polar stratospheric ozone content because in the upper stratosphere the O_x ($O + O_3$) balance is largely determined by catalytic NO_x cycles. Recently, several studies have presented observed NO_x enhancements in the middle atmosphere as a result of solar storms or downward transport from the upper atmosphere [*Seppälä et al.*, 2004; *Randall et al.*, 2005, 2007; *Funke et al.*, 2005].

[3] Several new instruments, like the three atmospheric chemistry instruments on board the Envisat satellite, provide improved information on the middle atmosphere. Many of these new instruments started to operate quite near the maximum of the solar cycle 23. As geomagnetic activity, and thus particle precipitation, is known to peak during the declining phase of a solar cycle [e.g., *Vennerstrom and Friis-Christensen*, 1996], these new instruments have had an excellent opportunity to make observations of the effect that enhanced energetic particle precipitation (EPP) has on the atmosphere. Particularly interesting are the new observations revealing the composition of the dark polar night middle atmosphere. This region has been difficult to observe as many of the earlier instruments measured either attenuated or scattered solar light. One of the instruments observing the polar night atmosphere is GOMOS (Global Ozone Monitoring by Occultation of Stars). Flying on board the Envisat satellite, GOMOS measures vertical profiles of several species with up to 600 occultations per day with good global coverage [*Kyrölä et al.*, 2004]. By year 2006 the instrument had performed approximately 350,000 successful occultation measurements.

[4] In this paper we report GOMOS nighttime observations of polar middle atmosphere NO_2 and O_3 from November 2002 until the end of September 2006. This is the first time both NO_x and O_3 nighttime observations from a single instrument during several consecutive Arctic and Antarctic winters are presented. We will focus on the connection of the observed NO_x enhancements with the precipitation of energetic electrons and solar protons, and downward transport into the middle atmosphere.

2. GOMOS Measurements

[5] In this study we have used all available GOMOS measurements from the Northern Hemisphere (NH) and Southern Hemisphere (SH) polar areas from 2002 to 2006. We have selected stars with temperature $\geq$6000 K and required the solar zenith angle at the tangent point to be $\geq 107°$, and $>90°$ at the satellite point to avoid stray light conditions. The selected occultations (total >16,000) are all located in the polar area, at latitudes poleward of 60°N/S, with a good daily latitudinal and longitudinal coverage.

[6] The accuracy of profiles retrieved from GOMOS measurements depends on stellar magnitude and temperature. For ozone the valid altitude range is 10–100 km and best accuracy is achieved from bright and hot stars with typical uncertainties for the whole altitude range on the order of 5% around both the primary and secondary maximum of ozone [*Kyrölä et al.*, 2006]. For NO_2 the valid altitude range is typically 20–50 km because for low mesospheric NO_2 concentration the signal-to-noise ratio can be poor. During times when the amount of NO_x exceeds the typical values [see, e.g., *Hauchecorne et al.*, 2005], the

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[1] Earth Observation, Finnish Meteorological Institute, Helsinki, Finland.
[2] Physical Sciences Division, British Antarctic Survey, Cambridge, UK.
[3] Laboratory for Atmospheric and Space Physics and Department of Atmospheric and Oceanic Sciences, University of Colorado, Boulder, Colorado, USA.

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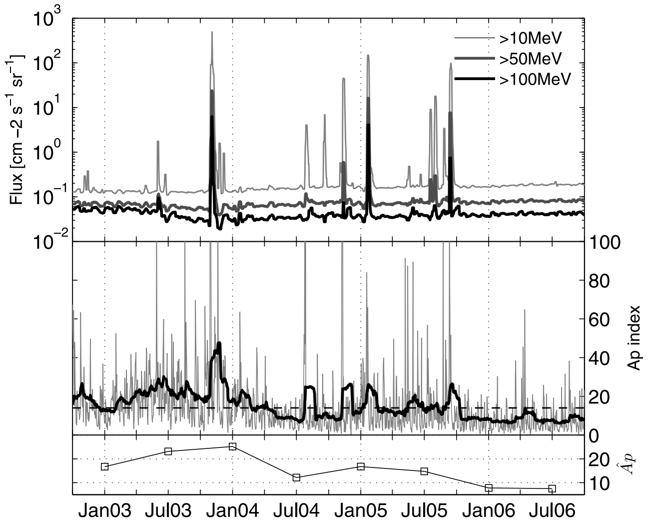

Figure 1. (top) GOES integral proton measurements. Seven-day average at three threshold energies >10 MeV, >50 MeV and >100 MeV. (middle) Daily mean A_p index (grey, values above 100 not shown) and 4-week moving average A_p (black). The dashed horizontal line is the mean A_p value (14.0) for the given period. (bottom) Four-month average A_p index ($\hat{A}_p$) (Oct–Jan, May–Aug).

upper altitude limit rises and NO_2 profiles can be measured even in the mesosphere (70 km). An example of a situation like this is NO_x production from Solar Proton Events (SPEs).

[7] In the stratosphere NO_x gases are in photochemical balance during daytime. After sunset NO is quickly converted into NO_2 by reaction with O_3, and thus the nighttime NO_2 measurements used in this study are a good representation of stratospheric NO_x.

3. Solar Storms and Geomagnetic Activity 2002–2006

[8] To estimate the forcing from energetic charged particles associated with SPEs we have used the proton flux measurements from the GOES satellites available through the Space Physics Interactive Data Resource (http://spidr.ngdc.noaa.gov). The 7-day average of the measured fluxes at three different threshold energies (1, 50, and 100 MeV) are shown in Figure 1. A 7-day average was used to filter out transient changes and to focus on times when the fluxes stayed elevated for extended periods. The averaged fluxes indicate that there is enhanced high-energy proton precipitation (E >100 MeV) in Oct–Nov 2003, Jan 2005 and Sept 2005. Precipitation of lower energy protons that mainly affect the mesosphere is enhanced in Jul–Aug 2002, Nov 2004, and Jul 2005.

[9] We use the A_p index to estimate geomagnetic activity and auroral particle precipitation (http://spidr.ngdc.noaa.gov). The daily A_p index as well as a 4-week running average are shown in Figure 1. A_p values ≥ 16 refer to active geomagnetic conditions, ≥ 30 to geomagnetic storm conditions. The mean A_p index for Jan 2002–Sept 2006 is 14.0 (compared to a cycle 23 mean of 12.9). The SH winter 2003 and NH winters 2003–2004 and 2004–2005 have prolonged periods when the average A_p is above the mean. During the NH winters the increased geomagnetic activity correlates well with SPEs seen in the proton flux measurements. The SH winters 2004 and 2006 and NH winter 2005–2006 have low average A_p, except for a short period in Jul 2004 when a moderate SPE occurred. As in the work by *Siskind* [2000] we calculated a 4-month average A_p index (hereafter $\hat{A}_p$), which can be used to describe geomagnetic activity effects on high-altitude NO_x production. The lowest panel in Figure 1 presents $\hat{A}_p$ for the polar winter months (NH: Oct–Jan, SH: May–Aug). During most winters $\hat{A}_p$ is above the mean, suggesting an increase in thermospheric NO_x production by auroral energy particles.

4. Results

[10] Figure 2 shows the NH polar nighttime NO_2 and O_3 volume mixing ratio (VMR) below 70 km from GOMOS. Large amounts of downwelling NO_2 are observed in winters 2003–2004 and 2004–2005. The first NO_2 enhancement period in Oct–Dec 2003 originates from the SPEs in Oct–Dec [*Seppälä et al.*, 2004]. In December 2003 a sudden stratospheric warming occurs, disrupting the polar vortex. This causes the SPE produced NO_x to mix with the smaller amounts of NO_x from lower latitudes and results in a dilution and reduction of the high polar NO_x values. In January 2004 the vortex is re-formed and high amounts of NO_2 are again observed. According to GOMOS results the NO_x descent starts in early January [*Hauchecorne et al.*, 2007] and is seen until the end of February when the observation period for that winter ends. Measurements from other instruments made after January have shown high NO_x amounts lasting at least through July 2004 [*Natarajan et al.*, 2004; *Randall et al.*, 2005]. GOMOS observations now show that these enhancements are first seen at mesospheric altitudes in January. According to *Clilverd et al.* [2006] the January 2004 enhancements are of thermospheric origin and not produced in the middle atmosphere *in situ* by particle precipitation (Figure 1: high $\hat{A}_p$, no SPE).

[11] In the following winter high amounts of descending NO_2 are again observed simultaneously with increased geomagnetic activity and small SPEs (Nov 2004). In the last winter, 2005–2006, prior to February, no anomalous NO_2 amounts are observed, which is in good agreement with the lack of EPP (no SPEs, low $\hat{A}_p$). In early February the NO_2 observations show a sudden NO_x enhancement of >30 ppbv between 60 and 70 km, with highest values on the order of 200ppbv at 60 km. However, as indicated by Figure 1 EPP was low throughout the NH winter 2005–2006, thus providing no significant stratospheric/mesospheric source of NO_x. *Randall et al.* [2006] have reported results from the Atmospheric Chemistry Experiment, which observed high NO_x values in the NH polar region in Feb–Mar 2006, not seen here as no GOMOS data is available after February, concluding that the NO_x enhancements were a result of exceptional meteorology leading to enhanced, confined descent in Feb 2006. *Siskind et al.* [2007] suggest that the strong descent results from anomalous gravity wave filtering.

[12] In the NH the O_3 VMR in the upper stratosphere shows significant variation from year to year. Distinct

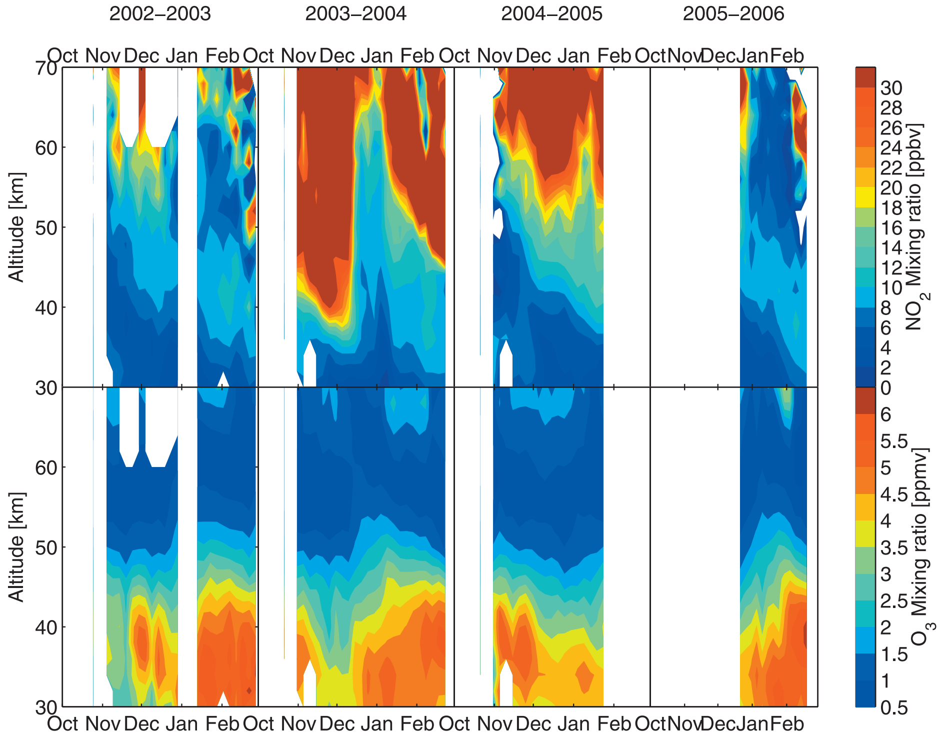

Figure 2. (top) GOMOS Northern hemisphere polar winter NO_2 mixing ratio [ppbv]. Five-day average of latitudes poleward of 60°N (months Oct–Feb). Years from left 2002–2003, 2003–2004, 2004–2005, 2005–2006. Altitudes 30–70 km. (bottom) Same as top but for O_3 [ppmv]. White areas indicate that no measurements were available or the measurements were flagged. The volume mixing ratios were calculated using ECMWF neutral densities below, and MSIS-model densities above 50 km.

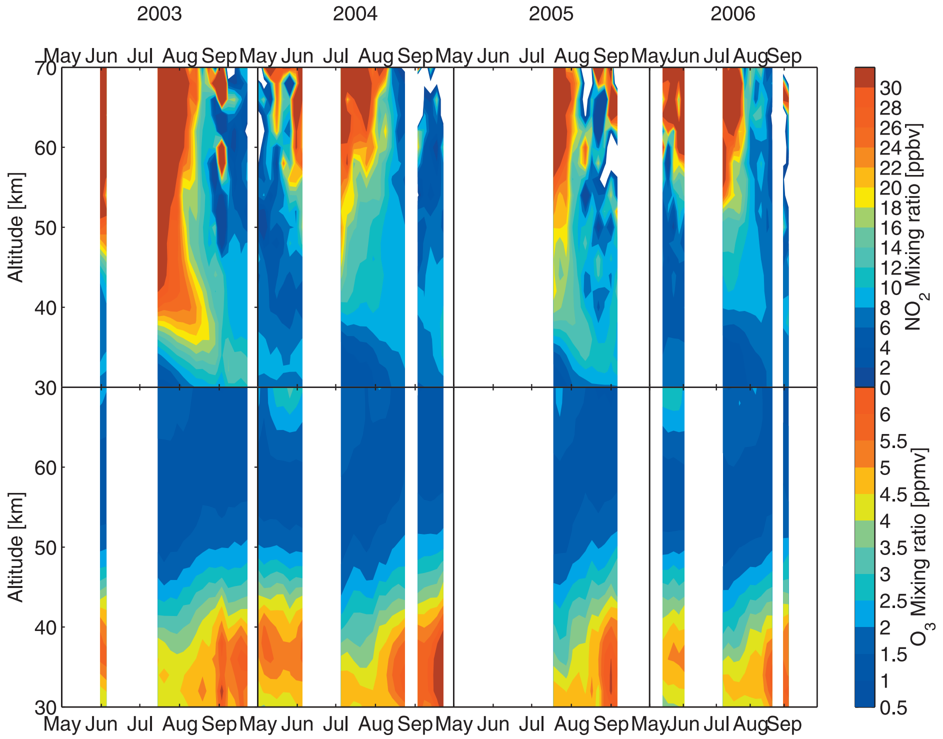

Figure 3. GOMOS Southern hemisphere polar winter (top) NO_2 and (bottom) O_3 as in Figure 2 (months May–Sept). Years from left 2003, 2004, 2005, 2006.

Table 1. SPE Occurrence, the 4-Month Average A_p Index, and Polar NO_2 Column Density[a]

	Season	SPEs	$\hat{A}_p$	NO_2
NH	2002–03	No	16.7	13.6
	2003–04	Yes	25.2	27.3
	2004–05	Yes	16.8	18.7
	2005–06	No	7.8	14.0
SH	2003	Yes	23.1	23.6
	2004	Yes	12.2	12.0
	2005	Yes	14.8	15.4
	2006	No	7.5	9.8

[a] The 4-month average A_p index is $\hat{A}_p$; see text. Polar column density is 46–56 km (NH Oct–Jan, SH:May–Aug) average over time periods shown in Figures 2 and 3 [$10^{13}cm^{-2}$]. For the NO_2 column value all available measurements (poleward of 60°N/S) are first used to calculate an average polar NO_2 profile, from which the column value is calculated.

changes in the O_3 VMR appear simultaneously with the largest NO_2 increases. At 50 km the lowest, unaveraged, values observed are <1 ppmv (Dec 2003 and Jan 2005).

[13] In the SH, in Figure 3, high amounts NO_2 are observed above the stratopause throughout every winter of our study period. During the first winter, 2003, GOMOS measurements show daily averages as high as 160 ppbv around 60 km. The NO_2 enhancements starting in late May–early June 2004 do not correspond to increases in precipitating protons or geomagnetic activity: an SPE occurs on July 25 but enhanced values are observed clearly before that. In 2005 no observations are available in the polar region before July and hence no definite start time for the increase of NO_x can be drawn.

[14] In 2004 the SH NO_x enhancements are first seen in the mesosphere in May and later in early July at 40 km. However, prior to the moderate SPE on July 25 the average A_p was only 9.2, which would not be expected to result in significant production of high-altitude NO_x [*Siskind*, 2000]. In 2006, when the average A_p was also low, though daily averages show more variation than in 2004, average NO_2 VMRs >30 ppbv are observed above 50 km, but in the stratosphere the average VMRs are <18 ppbv. This indicates that, either even low particle precipitation (as indicated by the A_p index and GOES measurements) combined with the SH polar vortex can result in high NO_x VMR or an additional NO_x source would be required.

[15] In the SH, large year to year variations are observed in the O_3 VMR. At 50 km altitude values around 1ppmv are observed every year from 2003 to 2005, with the smallest quantities in the month of July.

[16] The largest year to year variations in NO_2 values are observed in the NH. In February 2004 peak individual values of ~800 ppbv were observed near the stratopause. In the SH the largest NO_2 values seen by GOMOS are from winter 2003, with the individual profiles in the upper stratosphere-lower mesosphere showing peak values of 100–200ppbv. Similar peak NO_x values from the SH have been reported from MIPAS observations [*Funke et al.*, 2005].

[17] Finally we calculate the average 46–56 km NO_2 column density for each polar winter period and compare it with $\hat{A}_p$ and SPE occurrence. The results are summarized in Table 1. The values presented in the table show a nearly linear relationship between $\hat{A}_p$ and NO_2 for most winters. This linearity is shown clearly in Figure 4 which presents the NO_2 column vs. $\hat{A}_p$ for both hemispheres. Figure 4 shows that there is more variability in the $NO_x/\hat{A}_p$ correlation in the NH than in the SH, most likely due to the more stable SH polar vortex and more variable dynamical conditions in the NH.

5. Discussion

[18] GOMOS O_3 and NO_2 measurements during four recent winter seasons in the Arctic and Antarctic were used to study effects of EPP on the winter polar middle atmosphere. Significant increases in NO_x VMRs were observed nearly every year. In the Antarctic, downwelling NO_2, confined by the stable vortex, reaches altitudes as low as 30 km by September 2003 and 2005.

[19] A correlation between stratospheric NO_x increases and geomagnetic activity (A_p index) has previously been observed in the SH [e.g., *Siskind*, 2000; *Randall et al.*, 2007]. In our analysis we detected a similar correlation in the NH, despite the lack of stable polar vortex conditions known to exist in the SH; a linearity, valid for both hemispheres, was found between the average winter A_p index and upper stratospheric NO_2 column. The NH NO_x increases in 2004–2005, when particle precipitation as significant as that during the Halloween Storms in 2003 was not present, were as large as expected from SH correlations. This is a new result that confirms the need for polar night observations in quantifying interannual variability in EPP effects.

[20] The exceptional meteorological conditions in the NH in 2004 and 2006, discussed by *Randall et al.* [2007], which led to effective downward transport of thermospheric NO_x resulted in the highest ever observed NO_x in the stratosphere in the spring and early summer. However, in both cases, stratospheric altitudes were not affected until late January–February and thus the GOMOS October–

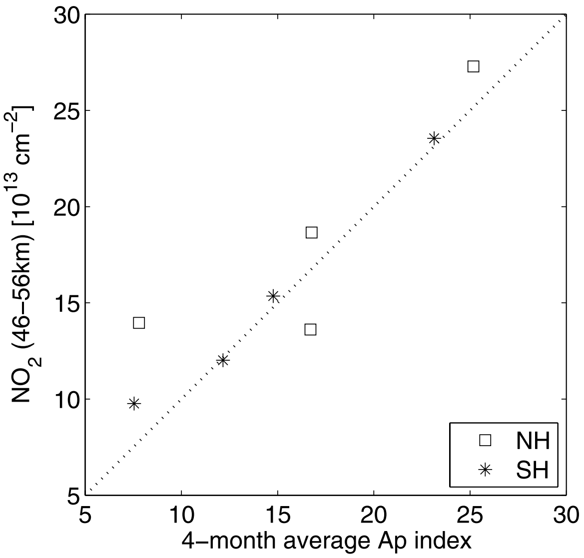

Figure 4. The 4-month average A_p index ($\hat{A}_p$) and the NO_2 column (46–56 km) from Table 1. Squares: NH, Stars: SH. The dashed line represents linear fit.

January observations used to calculate the NO_2 columns were made during more typical NH dynamical conditions. Thus the presented NO_2 columns and the found linearity between the NO_2 and $\hat{A}_p$ are not significantly affected by the exceptional meteorological conditions, but represent the more typical NH polar winter, including stratospheric warming periods in 2003–2004 and 2005–2006. If NO_x observations from the NH spring were available (GOMOS observations end in February) and included in the analysis, the linearity would, as a result of the exceptional conditions, be expected to be broken.

[21] The GOMOS O_3 observations show wintertime variation of up to 50% in upper stratospheric O_3. Determining the role of the EPP produced NO_x in the O_3 variation requires further modelling and is the subject of future work.

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References

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_x, *Geophys. Res. Lett.*, *33*, L19811, doi:10.1029/2006GL026727.

Funke, B., M. López-Puertas, S. Gil-Lopez, T. von Clarmann, G. P. Stiller, H. Fischer, and S. Kellmann (2005), Downward transport of upper atmospheric NOx 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.

Hauchecorne, A., et al. (2005), First simultaneous global measurements of nighttime stratospheric NO_2 and NO_3 observed by Global Ozone Monitoring by Occultation of Stars (GOMOS)/Envisat in 2003, *J. Geophys. Res.*, *110*, D18301, doi:10.1029/2004JD005711.

Hauchecorne, A., J.-L. Bertaux, F. Dalaudier, J. M. Russell, M. G. Mlynczak, E. Kyrölä, and D. Fussen (2007), Large increase of NO_2 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.

Kyrölä, E., et al. (2004), GOMOS on Envisat: An overview, *Adv. Space Res.*, *33*, 1020–1028.

Kyrölä, E., et al. (2006), Nighttime ozone profiles in the stratosphere and mesosphere by the Global Ozone Monitoring by Occultation of Stars on Envisat, *J. Geophys. Res.*, *111*, D24306, doi:10.1029/2006JD007193.

Natarajan, M., E. E. Remsberg, L. E. Deaver, and J. M. Russell (2004), Anomalously high levels of NO_x in the polar upper stratosphere during April, 2004: Photochemical consistency of HALOE observations, *Geophys. Res. Lett.*, *31*, L15113, doi:10.1029/2004GL020566.

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_x in 2006 linked to 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.

Seppälä, A., P. T. Verronen, E. Kyrölä, S. Hassinen, L. Backman, A. Hauchecorne, 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*(19), L19107, doi:10.1029/2004GL021042.

Siskind, D. E. (2000), On the coupling between the middle and upper atmospheric odd nitrogen, in *Atmospheric Science Across the Stratopause*, *Geophys. Monogr. Ser.*, vol. 123, edited by D. E. Siskind, S. D. Eckermann, and M. E. Summers, pp. 101–116, AGU, Washington, D. C.

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.

Vennerstrøm, S., and E. Friis-Christensen (1996), Long-term and solar cycle variation of the ring current, *J. Geophys. Res.*, *101*, 24,727–24,736.

L. Backman, E. Kyrölä, A. Seppälä, V. Sofieva, J. Tamminen, and P. T. Verronen, Earth Observation, Finnish Meteorological Institute, P.O. Box 503, FI-00101 Helsinki, Finland. (annika.seppala@fmi.fi)

M. A. Clilverd, Physical Sciences Division, British Antarctic Survey, High Cross, Madingley Road, Cambridge CB3 0ET, UK.

C. E. Randall, Laboratory for Atmospheric and Space Physics, University of Colorado, Boulder, CO 80309-0392, USA.