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## Large increase of NO<sub>2</sub> in the north polar mesosphere in January–February 2004: Evidence of a dynamical origin from GOMOS/ENVISAT and SABER/TIMED data

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[1] Odd nitrogen species play an important role in the stratospheric ozone balance through catalytic ozone destruction. A layer of strongly enhanced NO<sub>2</sub> was detected in the north polar mesosphere by the GOMOS/ENVISAT stellar spectrometer in mid-January 2004. Large NO<sub>2</sub> enhancements in the polar winter mesosphere have been previously reported by several authors and have been attributed to NO production by solar proton or by energetic electron precipitations. The simultaneous occurrence of an intense mesospheric warming observed by the SABER/TIMED instrument indicates that a strong air descent occurred in the polar region, transporting a large quantity of NO from the upper mesosphere-lower thermosphere to the lower mesosphere. The proposed mechanism may have a significant contribution to the budget of polar stratospheric ozone. **Citation:** Hauchecorne, A., J.-L. Bertaux, F. Dalaudier, J. M. Russell III, M. G. Mlynczak, E. Kyrölä, and D. Fussen (2007), Large increase of NO<sub>2</sub> in the north polar mesosphere in January–February 2004: Evidence of a dynamical origin from GOMOS/ENVISAT and SABER/TIMED data, *Geophys. Res. Lett.*, 34, L03810, doi:10.1029/2006GL027628.

### 1. Introduction

[2] The stratospheric ozone layer protects the life on Earth surface against harmful ultraviolet radiation from the sun. The thickness of this layer has decreased during the past 30 years due to the increase of chlorine and bromine compounds from anthropogenic origin. Following the Montreal protocol and its amendments, limiting the production of halogen species, a progressive recovery of the ozone layer is expected during the next decades but large uncertainties remain on the rate of this recovery due to the high variability of the atmosphere and the imperfect knowledge of chemical and physical processes controlling the ozone budget. Among them, the odd nitrogen family (oxidised nitrogen species) plays an important role in the ozone balance due to catalytic reactions involved in ozone

destruction. The main source of odd nitrogen in the stratosphere is the dissociation of N<sub>2</sub>O by reaction with atomic oxygen in an excited state. The presence of high level of NO in the upper mesosphere-lower thermosphere (UM-LT; ~80–100 km) is well recognized [Barth *et al.*, 1999]. It is produced by mechanisms involving solar x-rays, auroral disturbances and energetic electron precipitations. The radiatively induced downward transport of UM-LT air during the polar night is another potential source of mesospheric and stratospheric odd nitrogen as demonstrated by Russell *et al.* [1984] using NO<sub>2</sub> satellite observations. Proton precipitations occurring during solar events can increase the production of odd nitrogen in the upper stratosphere and mesosphere [Crutzen, 1975]. High concentrations of mesospheric NO observed by HALOE/UARS satellite experiment in 1992–1997 were well correlated with energetic electron precipitations [Callis *et al.*, 2002]. More recently, several space instruments measured a large increase of NO<sub>x</sub> (NO + NO<sub>2</sub>) and a simultaneous destruction of ozone in the polar upper stratosphere after the large solar proton event end of October 2003 [Degenstein *et al.*, 2005; Seppälä *et al.*, 2004; Rohen *et al.*, 2005; Jackman *et al.*, 2005; Orsolini *et al.*, 2005]. High levels of NO and NO<sub>2</sub> and low levels of O<sub>3</sub> were also observed in February–May 2004 in the north polar vortex [Natarajan *et al.*, 2004; Rinsland *et al.*, 2005; Randall *et al.*, 2005].

[3] Here, using NO<sub>2</sub> data from GOMOS/ENVISAT and temperature data from SABER/TIMED, we show observational evidences of a rapid downward transport of a large quantity of NO<sub>x</sub> from the UM-LT to the lower mesosphere during the second half of January 2004 in the north high latitudes. The transport occurred in the polar night, preventing NO to be dissociated. This period was not characterized by any noticeable energetic particle precipitation. This event is at the origin of the layer enriched in NO<sub>x</sub> observed in the polar upper stratosphere in March–May 2004.

### 2. Data

[4] GOMOS, one of ten instruments on ESA's ENVISAT mission, is a stellar occultation spectrometer dedicated to the stratospheric and mesospheric ozone monitoring at global scale [Bertaux *et al.*, 2004]. The 250–680 nm spectral range is used to determine vertical profiles of O<sub>3</sub>, NO<sub>2</sub>, NO<sub>3</sub> and aerosols. The data quality depends on star brightness and temperature and on illumination conditions. Occultations performed on night side (dark limb) are of better quality than on day side (bright limb). Hot stars have a strong UV emission and allow obtaining ozone profiles up

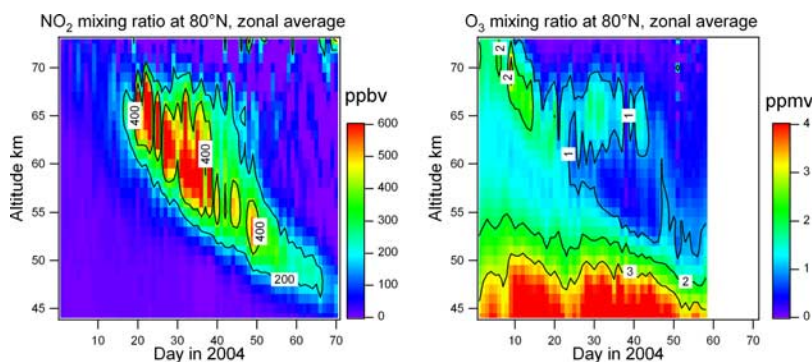
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**Figure 1.** Evolution of zonal averaged mixing ratio of (left) NO<sub>2</sub> and (right) O<sub>3</sub> at 80°N from GOMOS data between 1st January and 10 March 2004. During this period, 2 stars were available for observations around 80°N, star 8 Alpha CMi, visual magnitude  $M_{\text{vis}} = 0.4$ , brightness temperature  $T_{\text{brightness}} = 6500$  K was available from 1st January to 10 February and star 48 Alpha Hya,  $M_{\text{vis}} = 2$ ,  $B_{\text{brightness}} = 4100$  after 10th February. The brightness of star 48 is too weak in the UV to provide useful O<sub>3</sub> information after 27 February in twilight conditions.

to the lower thermosphere using UV absorption bands. Cold star ozone profiles are limited to the stratosphere using visible Chappuis band. NO<sub>2</sub> profiles extend from 20 to 50 km in typical non polar conditions and up to 70 km during polar winter when a strong NO<sub>2</sub> enhancement occurs. The typical accuracy is 5% for O<sub>3</sub> profiles and 15–20% for NO<sub>2</sub> [Hauchecorne *et al.*, 2005].

[5] SABER, one of four instruments on NASA's TIMED Mission, is dedicated to the study of the mesosphere and lower thermosphere structure and energetics, including their seasonal, latitudinal and temporal variations [Russell *et al.*, 1999]. It is a limb spectrometer operating in the near to mid-infrared. The kinetic temperature used in this study is retrieved using two wide and one narrow band CO<sub>2</sub> channels centred in the 15  $\mu\text{m}$  band.

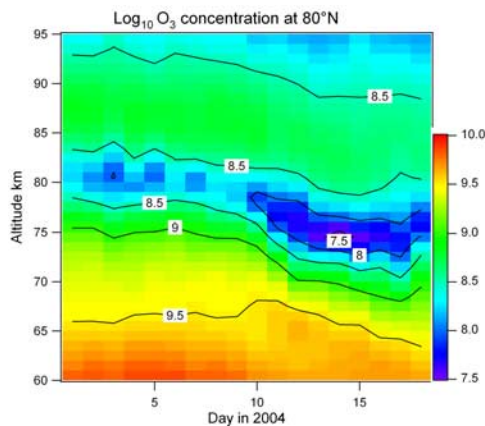
### 3. Results

[6] GOMOS performs about 400 occultations per day with a global latitude coverage depending on star availability. A given star provides 14 profiles per day (1 per orbit) at a latitude varying slowly from day to day. The present study is based on measurements from 3 stars occulted around 80°N on night side from 1st January to 10 March 2004. Until mid-February this region is in the polar night and the length of the day increases gradually in late February–early March. A very strong NO<sub>2</sub> enhancement develops between 15 and 20 January around 65 km with mixing ratios higher than 600 ppbv (Figure 1, left). Individual profiles (not shown) indicate values higher than 1000 ppbv. The descent of the bottom altitude of the enhanced NO<sub>2</sub> layer can be followed after 20 January down to 45 km on 10 March with a vertical speed decreasing gradually from 600 m/day in the late January to 200 m/day in early March. The maximum mixing ratio is almost constant until 7 February and decreases slightly until 20 February and more rapidly after this date, reaching 150 ppbv on 10 March. One part of the decrease after mid-February is due to the gradual apparition of sunlight causing a partial photodissociation of NO<sub>2</sub> in NO. The NO<sub>2</sub> concentration in the enhanced layer reaches values as high as  $4.10^9 \text{ cm}^{-3}$  in late February at 50 km, to be compared to less than  $3.10^9 \text{ cm}^{-3}$  in the climatological stratospheric layer at 30 km.

[7] The decrease in altitude of the secondary O<sub>3</sub> maximum after 10 January (Figure 2) is an indication that a rapid descent of air with high O<sub>x</sub> concentration occurred during this period in the upper mesosphere (70 to 90 km). The layer with low O<sub>3</sub> mixing ratio at 53–60 km between days 30 and 45 (Figure 1, right) coincides well with the layer enriched in NO<sub>2</sub> and its bottom altitude goes down at the same rate than the NO<sub>2</sub> layer. The ozone decrease at 55–60 km after day 30 when NO<sub>2</sub> increases is interpreted as the catalytic destruction of O<sub>3</sub> by NO<sub>x</sub>. Its decrease at all levels after day 50 is attributed to the gradual apparition of sunlight and a partial photodissociation of O<sub>3</sub> in O + O<sub>2</sub>.

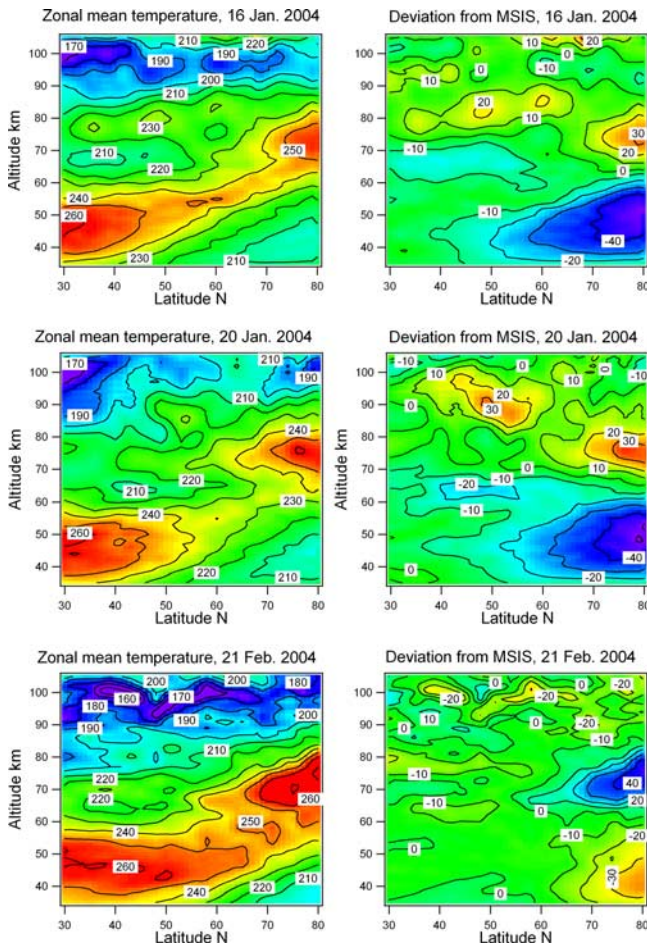
### 4. Discussion

[8] A large amount of NO<sub>x</sub> is formed in the polar middle atmosphere after large solar proton events [Jackman *et al.*, 2000]. During such events, the penetration of solar protons



**Figure 2.** Evolution of zonal averaged O<sub>3</sub> concentration around 80°N from GOMOS data between 1st and 18 January 2004 using star 7, Beta Ori, visual magnitude  $M_{\text{vis}} = 0.1$ , brightness temperature  $T_{\text{brightness}} = 14000$  K. This bright hot star provides very good O<sub>3</sub> data in the upper mesosphere due to its high UV flux but was not available after 18 January.





**Figure 3.** (left) Zonal average temperature and (right) deviation from MSIS90 climatological model (23) on (top) 16 January 2004, (middle) 20 January 2004 and (bottom) 21 February 2004 from SABER/TIMED data. Data are not available before 15 January because SABER was looking southwards.

in the atmosphere produces NO<sub>x</sub> in the whole mesosphere and even in the upper stratosphere. *Natarajan et al.* [2004] and *Rinsland et al.* [2005] attribute the NO<sub>x</sub> enhancement in February–March 2004 to the production of a large quantity of NO in the mesosphere/thermosphere during the October–November 2003 solar storm and its diabatic descent to the stratosphere during the following months. As pointed out by *Randall et al.* [2005] this hypothesis is unlikely because polar air enhanced in NO<sub>x</sub> has been diluted into mid-latitude air after the December 2003 stratospheric warming. Furthermore, there was no solar proton event in January 2004. Another proposed explanation is the production of NO<sub>x</sub> by energetic electron precipitations due to auroral phenomena in early 2004 [*Natarajan et al.*, 2004; *Randall et al.*, 2005; *Renard et al.*, 2006]. This explanation can also be rejected because there was no significant electron precipitation event before the formation of the NO<sub>x</sub> layer in mid-January. *Renard et al.* [2006] attribute the mesospheric NO<sub>2</sub> enhancement to an electron precipitation event occurring on 22 January but the layer started to develop at least one week before this date. We propose

another explanation based on the rapid downward transport in the polar night of NO from the UM-LT where it is formed by the dissociation of N<sub>2</sub> by far-UV solar radiation and energetic particle precipitations in the auroral zone [*Barth et al.*, 1999].

[9] The general mesospheric circulation is characterized by a meridional motion from the summer pole to the winter pole, a vertical ascendance (subsidence) with an adiabatic cooling (warming) at the summer (winter) pole. As consequences the mesopause is colder in summer than in winter and atmospheric species are transported downwards from the UM-LT in the winter polar region. The 2003–2004 Arctic winter was remarkable in the 50 years record of meteorological analyses [*Manney et al.*, 2005]. A strong warming occurred in the polar upper stratosphere in late December 2003 with a splitting of the polar vortex and a reversal of the zonal wind down to 10 hPa, preventing planetary waves to propagate from below and to transmit their energy and momentum flux. After this event, a strong thermal infrared cooling occurred in the polar upper stratosphere and lower mesosphere and the temperature reached very low values in mid-January. On 16–20 January 2004 SABER/TIMED observed temperature up to 40 K colder than MSIS90 climatology at 50 km [*Hedin*, 1991] and up to 30 K warmer at 75 km (Figure 3, top and middle plots). SABER/TIMED data (not shown) indicate that this situation, with cold temperatures at 40–50 km and warm temperatures at 70–75 km in the Arctic region, persisted at least until 21 February (Figure 3, bottom plots). It was favourable to the strengthening and the stability of the polar vortex in the lower mesosphere and the propagation and breaking of gravity and tidal waves in the middle and upper mesosphere [*Lindzen*, 1981]. The gravity wave propagation is particularly favoured in the region of fast westerly winds around the polar vortex. The breaking of gravity waves decelerates the westerly wind and creates a poleward meridional wind due to a lack of balance between the pressure gradient and the Coriolis force. By continuity the poleward wind induces a downward vertical wind in the polar region. A warm layer in the polar night mesosphere is a strong indication of a rapid downward motion inducing an adiabatic warming because we don't expect any significant internal heat source in the polar night in without any noticeable particle precipitation. This downward motion is expected to strongly enhance the transport of NO from the UM-LT. If NO is transported rapidly ( $\sim 10$  days) in the polar night down to  $\sim 65$  km without being photodissociated by solar UV radiation, it is transformed in NO<sub>2</sub> by the reaction with O<sub>3</sub> and can stay for months in the polar vortex. NO<sub>2</sub> is not stable above 70 km because it reacts with O to reform NO and its concentration cannot reach a detectable level. The proposed mechanism is effective only if NO remains in the polar night during its transport down to 65 km. During daytime NO is photolysed and its lifetime is shorter than 2 days above 70 km [*Brasseur and Solomon*, 1986]. Consequently, the NO<sub>2</sub> layer was no more alimented from above after day 50 as shown in Figure 2, even though dynamical conditions were still favourable to a downward transport.

[10] Our hypothesis is supported by observations of a clear change in the radio wave diurnal propagation from Svalbard VLF/LF data starting on 13 January 2004 inter-

puted by Clilverd *et al.* [2006] as a consequence of the descent of NO<sub>x</sub> into the mesosphere in January/February 2004 and the ionization of enhanced NO by Lyman- $\alpha$  radiation. The rapid descent may have even started before, around 10 January, as indicated by the rapid descent of the secondary O<sub>3</sub> maximum starting at this date. Our results are also in agreement with solar occultation observations of an enhanced NO<sub>2</sub>/NO<sub>x</sub> layer in the upper polar stratosphere in March–May 2004 [Natarajan *et al.*, 2004; Randall *et al.*, 2005; Rinsland *et al.*, 2005], descending from 45 km in early March to 38–40 km in early May. Solar occultation instruments were not able to observe NO<sub>2</sub> before March because when the NO<sub>x</sub> layer is in the mesosphere, NO<sub>2</sub> is totally photodissociated at sunset and sunrise. Night-time data from MIPAS/ENVISAT instrument reported by Randall *et al.* [2005] indicate a NO<sub>2</sub> mixing ratio larger than 100 ppbv in the upper stratosphere (1600 K isentropic level), in quantitative agreement with the 150 ppbv value from GOMOS on 10 March. The strong descent of air in the mesospheric Arctic vortex is also supported by ACE-FTS and ODIN-SMR observations of CO in early March 2004 showing a strong enhancement of CO around 50 km [Jin *et al.*, 2005].

[11] If we combine GOMOS night-time measurements from mid-January to early March with solar occultation observations from early March to early May, it is possible to follow the descent of the NO<sub>2</sub> layer in the polar vortex during almost 4 months from 65 km to 38–40 km. It is striking to note that during these 4 months the thickness of the layer does not increase. This means that the vertical turbulent mixing inside the vortex is very small even if the vertical compression of the layer when its pressure increases can compensate the vertical broadening by turbulent mixing. A very crude upper limit estimation of the vertical diffusion coefficient  $K_{zz}$  is given by  $L_z^2/T$ , where  $L_z$  is the thickness of the layer and  $T$  the length of the period. If we take  $L_z = 5$  km and  $T = 10^7$  s, we find:  $K_{zz} \leq 2.5$  m<sup>2</sup>s<sup>-1</sup>. This means also that the layer remains well confined inside the polar vortex during this 4 months.

[12] During the period when the NO<sub>x</sub> layer remains confined in the polar night (until mid-February), the O<sub>3</sub> depletion is limited to the direct conversion of NO to NO<sub>2</sub> but as soon as air masses experience sunlight hours (after mid-February) the catalytic destruction of O<sub>3</sub> by NO<sub>x</sub> becomes effective. The transport of a large quantity of NO<sub>x</sub> in the winter polar upper stratosphere by a strong dynamical event decreases significantly the O<sub>3</sub> mixing ratio in this region for at least several months. Up to 60% O<sub>3</sub> reduction has been observed by POAM III in the upper polar stratosphere during spring 2004 compared to climatology [Natarajan *et al.*, 2004]. Further studies are needed to evaluate the frequency and the intensity of such events in order to assess their contribution to the polar ozone budget on a long term basis. If it appears that these events are frequent, it will be necessary to reevaluate the contribution of stratosphere-mesosphere-lower thermosphere dynamical coupling in the polar stratospheric ozone budget.

## 5. Summary

[13] A layer of strongly enhanced NO<sub>2</sub> has been detected by GOMOS/ENVISAT starting on January 15, 2004 at

65 km in the North polar stratosphere with mixing ratios up to 1000 ppbv. The descent of the layer can be followed until March 10 down to 45 km. The simultaneous destruction of O<sub>3</sub> in the layer is clearly detected. An intense warming was observed in the north polar middle mesosphere at the time of formation of the NO<sub>2</sub> layer by SABER/TIMED. This warming is an indication of a strong air descent in the polar vortex bringing a large quantity of NO from the UM-LT. When such a descent occurs in the polar night, NO cannot be photodissociated before reaching an altitude where it is stable and it can be transformed in NO<sub>2</sub> by reaction with O<sub>3</sub> (~60–65 km).

[14] Large NO<sub>2</sub> enhancements in the polar winter upper stratosphere-lower mesosphere have been reported previously by several authors and have been attributed to the production of NO by large energetic particle precipitations (solar proton or energetic electron precipitations). These events are relatively rare. In the present case no significant particle precipitation was reported and we attribute the event to a pure atmospheric dynamical phenomenon, the breaking of atmospheric waves in the upper mesosphere inducing a strong descent of UM-LT NO into the polar vortex. If the proposed mechanism appears to be frequent, it will be necessary to reevaluate the contribution of the stratosphere-mesosphere-lower thermosphere dynamical coupling in the polar stratospheric ozone budget.

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