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## Solar proton events of October–November 2003: Ozone depletion in the Northern Hemisphere polar winter as seen by GOMOS/Envisat

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[1] Observations of O<sub>3</sub> and NO<sub>2</sub> made by the GOMOS instrument on board European Space Agency's Envisat satellite have been used to monitor the increase of NO<sub>2</sub> and depletion of ozone due to the solar proton events of October–November 2003. For the first time this phenomenon was measured in polar winter conditions by a satellite instrument. Results show NO<sub>2</sub> enhancement of several hundred per cent and tens of per cent ozone depletion between 36 and 60 km, an effect which lasts several months after the events. A comparison of the after-event concentrations of NO<sub>2</sub> and ozone reveals a strong negative correlation. *INDEX TERMS*: 0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334); 2455 Ionosphere: Particle precipitation; 3360 Meteorology and Atmospheric Dynamics: Remote sensing. **Citation**: 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*, L19107, doi:10.1029/2004GL021042.

### 1. Introduction

[2] Solar proton events (SPE) correspond to solar coronal mass ejections (CME) during which a large amount of protons and heavier ions are emitted, sometimes toward the Earth. Solar protons entering the Earth's magnetosphere are guided by the Earth's magnetic field and precipitate into the polar cap areas. Since the protons can have very high energy, up to tens of MeVs, they deposit their energy in the mesosphere and stratosphere. Thus they provide a direct connection between the Sun and the Earth's middle atmosphere. SPEs are sporadic although more probable during solar maxima. But when occurring they provide extreme forcing on the middle atmosphere.

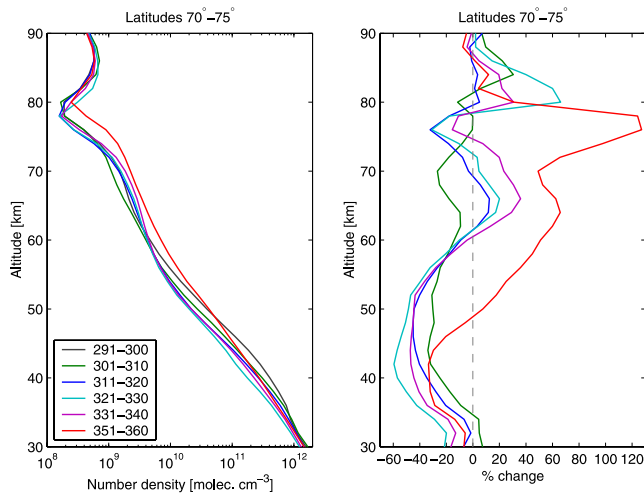
[3] The precipitating particles produce 1) odd hydrogen HO<sub>x</sub> (H + OH + HO<sub>2</sub>) through chemistry associated with ion pair production, water cluster ion formation, and subsequent neutralization, and 2) odd nitrogen NO<sub>x</sub> (N + NO + NO<sub>2</sub>) through dissociation of molecular nitrogen via charged particle impact [Crutzen *et al.*, 1975; Solomon *et al.*, 1981; Rusch *et al.*, 1981]. HO<sub>x</sub> and NO<sub>x</sub> play a key role in ozone balance of the middle atmosphere because they destroy odd oxygen through catalytic reactions [see, e.g., Brasseur and Solomon, 1986, pp. 291–299]. The produced HO<sub>x</sub> has a relatively short lifetime, but without solar radiation NO<sub>x</sub> chemical loss is inefficient. Therefore the NO<sub>x</sub> produced during polar night has a long chemical lifetime and is transported to lower altitudes and latitudes [Siskind *et al.*, 1997; Callis and Lambeth, 1998]. Significant depletions of ozone concentrations after large solar proton events have been predicted by atmospheric modelling [Rusch *et al.*, 1981; Solomon *et al.*, 1983; Reid *et al.*, 1991; Jackman *et al.*, 2000] and this phenomenon has been captured in the dayside middle atmosphere using satellite measurements [Thomas *et al.*, 1983; McPeters and Jackman, 1985; Jackman *et al.*, 2001; Randall *et al.*, 2001].

[4] The GOMOS (Global Ozone Monitoring by Occultation of Stars) instrument measures vertical profiles of several middle atmospheric gases, including O<sub>3</sub> and NO<sub>2</sub>. GOMOS makes several hundred occultations per day with good global coverage, including the polar areas. It can measure in both day and night conditions because the stellar occultation technique does not require solar radiation. Therefore, GOMOS has a unique capability of observing the effects of SPEs during polar winter. In this paper, we use GOMOS measurements from the northern hemisphere during and after the October–November 2003 SPEs to confirm the increase/persistence of NO<sub>2</sub> and depletion of O<sub>3</sub> due to the events.

[5] Late October 2003 the Sun released two powerful X-class solar flares both associated with a CME directed almost straight at the Earth. Consequently the flux of particles entering the Earth's atmosphere was greatly enhanced. During October–November 2003 there were four occasions of noticeable increase in the proton flux (NOAA NGDC–Solar Terrestrial Physics, 2004, see <http://www.ngdc.noaa.gov/stp/stp.html>). First on October 26th and second on the 28th, then again on November 2nd and 4th. The first event was caused by a solar X1-class flare and associated CME. This event lasted only few hours, but only a day later a second flare, this time even larger X17 followed by a halo-type CME led to measured maximum

### 2. Solar Storms on October–November 2003

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**Figure 1.** Left: Zonal average GOMOS ozone densities for latitudes  $70^{\circ}$ – $75^{\circ}$ . Averaging is done in 10-day periods and the measurements have been weighted with the measurement errors. Day of year 301 = 28.10.2003. Right: Relative change [%] from the zonal average profile before the SPE (291–300 in the left panel).

flux of  $3 \times 10^4$  protons  $\text{cm}^{-2}\text{s}^{-1}\text{sr}^{-1}$  ( $E > 10$  MeV). The following two events were much smaller than this although the November 4th event was preceded by a record setting X28-class flare. After the four events the quiet time level of the GOES measured proton flux was not reached until mid November.

### 3. GOMOS Observations

[6] GOMOS is a stellar occultation instrument on board the European Space Agency's Envisat satellite [Bertaux *et al.*, 1991, 2004; Kyrölä *et al.*, 2004]. Envisat, launched on March 1st, 2002, from French Guiana, is an environmental satellite carrying a total of 10 instruments including three atmospheric chemistry instruments GOMOS, MIPAS and SCIAMACHY.

[7] GOMOS measures light of a star as it appears to descend through the Earth's atmosphere. The incoming light travels through the atmosphere and is absorbed and scattered by various molecules along its path. Knowing the absorption features of the different atmospheric gases their altitude profiles can be calculated using advanced inversion methods [Kyrölä *et al.*, 1993]. The GOMOS spectral range is 250–950 nm and it measures vertical profiles of  $\text{O}_3$ ,  $\text{NO}_2$ ,  $\text{NO}_3$ ,  $\text{H}_2\text{O}$ ,  $\text{O}_2$ , neutral density, and aerosols. Also, high resolution temperature profiles are measured with two fast photometers. The altitude range of the measurements is 10–100 km for ozone and 10–50 km for the other gases. Altitude sampling frequency is better than 1.7 km. By using stars as a source of light, information about the abundance of different gases in the atmosphere can be attained from the dark side of the atmosphere as well as from the bright side.

[8] For this study we used nighttime GOMOS measurements from the northern hemisphere polar region (GOMOS processing prototype version 6.0a, geographic latitude  $\geq 45^{\circ}$ , solar zenith angle  $\geq 107^{\circ}$ , and solar zenith angle at satellite location  $> 90^{\circ}$  to avoid straylight conditions). In

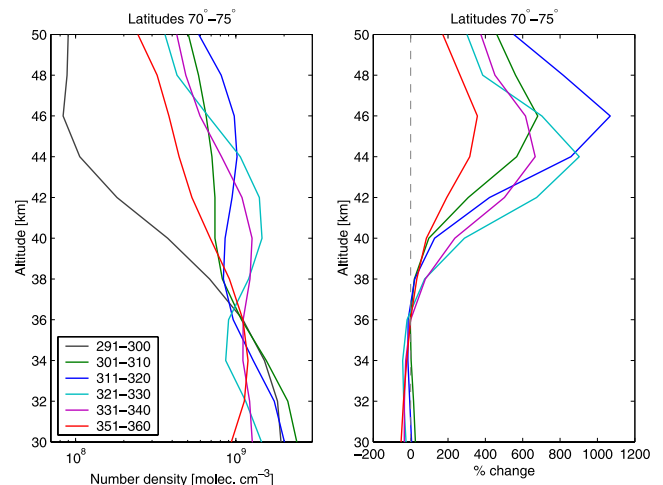
addition, to ensure good accuracy of the measurements we selected stars with temperature  $\geq 7000$  K [Bertaux *et al.*, 2004; Kyrölä *et al.*, 2004]. Finally, before analyzing the data all the selected measurement profiles were interpolated into a constant altitude grid with 2 km spacing.

### 4. Results

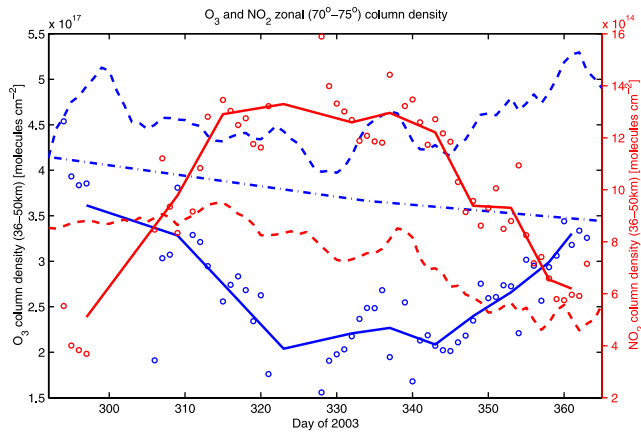
[9] Figure 1 shows zonal average vertical profiles for latitudes  $70^{\circ}$ – $75^{\circ}$  for ozone. This particular latitude band was chosen for our study because it is well covered by the measurements throughout Oct–Dec (see Figure 4 for measurement locations), and because of its vicinity to the magnetic polar cap. The profiles are ten-day means between Oct 18 and the end of the year 2003. In the first 10-day period after Oct 28, when the largest SPE took place, ozone depletion of 20–30% from the initial values (18–27 Oct) is seen at altitudes 40–58 km and near 70 km. The ozone depletion further increases at altitudes below 58 km until the end of November, when the depletion maximum near 40 km reaches 60%. At the end of December the change from the initial values is still nearly –30% between 36 and 46 km.

[10] Figure 2 shows  $\text{NO}_2$  densities from altitudes 30–50 km. Before Oct 28 the amount of  $\text{NO}_2$  is less than  $4 \times 10^8$  molecules  $\text{cm}^{-3}$  above 40 km. After the SPEs an increase of several hundred per cent above 40 km is observed. The  $\text{NO}_2$  enhancement peak in the upper stratosphere appears to be transported downward. At the end of November the increase of  $\text{NO}_2$  is still more than 100% at several altitudes.

[11] We calculated daily and 5-day mean  $\text{O}_3$  and  $\text{NO}_2$  columns for altitudes 36–50 km where the  $\text{NO}_2$  enhancement is greatest. The results are shown in Figure 3 together with ozone and  $\text{NO}_2$  columns from the FinRose-CTM model (based on the Rose model by Rose and Brasseur [1989], no SPE forcing included) and ozone climatology [Fortuin and Kelder, 1998]. Before the 28th the ozone column from GOMOS agrees with those from the model and climatology ( $\sim 4 \times 10^{17}$  molecules  $\text{cm}^{-2}$ ). After the event the ozone values decrease to almost  $1.5 \times 10^{17}$  molecules  $\text{cm}^{-2}$  at the lowest. At the end of December the ozone column values are again close to those of the climatology. The  $\text{NO}_2$  values increase by over 200% after



**Figure 2.** Like Figure 1 but for  $\text{NO}_2$  at altitudes 30–50 km.



**Figure 3.** O<sub>3</sub> (blue) and NO<sub>2</sub> (red) column densities. Daily (circle) and 5-day mean (solid line) calculated from GOMOS daily zonal average profiles for latitudes 70°–75° and altitudes 36–50 km. The dash-dot line is [Fortuin and Kelder, 1998] O<sub>3</sub> column (36–50 km) for latitude 72.5° and the dashed lines are O<sub>3</sub> and NO<sub>2</sub> columns (37–47 km) from the FinRose-CTM model. The FinRose-CTM model includes no SPE forcing.

the event and exceed the modelled values. At the end of December the values are still over 50% larger than before the October 28. The observed ozone column decrease coincides with the NO<sub>2</sub> column enhancement and the correlation coefficient  $r$  for the Oct–Dec daily O<sub>3</sub> and NO<sub>2</sub> columns is  $-0.77$  indicating a strong negative correlation.

[12] The temporal evolution of O<sub>3</sub> and NO<sub>2</sub> at 46 km (latitudes  $\geq 45^\circ$ ) is shown in Figure 4 together with the measurement locations. Before Oct 28 the ozone values are in agreement with the climatology values. After the 28th the density decreases particularly near the magnetic polar area. The ozone depletion area near the end of December is restricted between longitudes  $\sim \pm 80^\circ$ . Comparing the O<sub>3</sub> and NO<sub>2</sub> maps we notice that the areas of largest ozone depletion coincide with the areas where NO<sub>2</sub> enhancement

is most significant. To verify this we calculated correlation coefficients for the NO<sub>2</sub> and O<sub>3</sub> measurements at 46 km for 5-day periods. Figure 5 shows the measured values for one time period and variation of the correlation coefficient during Oct–Dec and the number of measurements used to calculate each correlation. Before the SPEs NO<sub>2</sub> and O<sub>3</sub> are positively correlated. However, after the SPEs they become strongly negatively correlated with  $r$  approaching a value of  $-0.8$ .

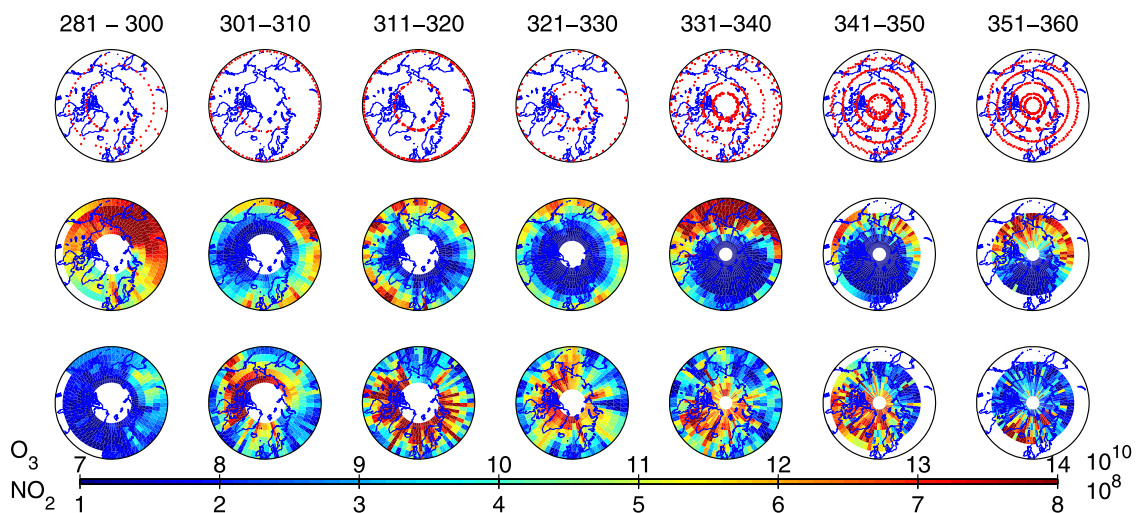
## 5. Discussion

[13] GOMOS observations show significant changes in NO<sub>2</sub> and ozone due to the October–November 2003 solar proton events. An order-of-magnitude increase in NO<sub>2</sub> results in up to 60% ozone depletion in the upper stratosphere. Even two months after the SPEs the effect can still be seen. This is the first time that SPE effects have been observed in the polar winter middle atmosphere with a good spatial and temporal coverage.

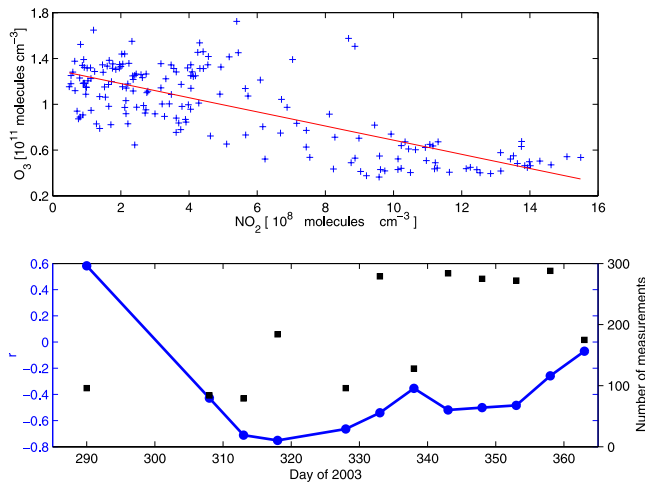
[14] NO<sub>2</sub> and NO participate in the catalytic reaction cycle which destroys ozone but neither creates nor destroys odd nitrogen. Therefore, in general it might be more beneficial to observe changes in the sum NO + NO<sub>2</sub>. However, in the nighttime stratosphere and lower mesosphere practically all NO is converted to NO<sub>2</sub>. Therefore we are confident that our nighttime NO<sub>2</sub> measurements are a good tracer for the total odd nitrogen changes.

[15] Ozone is also affected by HO<sub>x</sub> production during the events. However, this effect should be present only for a few days after the events, i.e., between Oct 26 and Nov 5 (days 299–310), because of the relatively short lifetime of HO<sub>x</sub> in the upper stratosphere. Therefore, after day 310 we can assume that the effect on ozone is solely due to the increase of NO<sub>x</sub>.

[16] The correlation between the concentrations of NO<sub>2</sub> and ozone is positive before the SPEs (Figure 5). FinRose results, as well as GOMOS measurements (January, 2003, and early October, 2003) confirm that without the SPEs the correlation is positive, possibly reflecting the same latitudi-



**Figure 4.** GOMOS measurement locations (top) and O<sub>3</sub> (middle) and NO<sub>2</sub> (bottom) densities [cm<sup>-3</sup>] at 46 km for 10-day periods. The density maps are in 5° × 5° grid showing latitudes  $\geq 45^\circ$ , filled using a Delaunay triangulation method from Qhull [Barber *et al.*, 1996].



**Figure 5.** Correlation of NO<sub>2</sub> and ozone at altitude 46 km (latitudes  $\geq 45^\circ$ ). Upper panel: Measured NO<sub>2</sub> and O<sub>3</sub> number densities (days 316–320). The red line is a first degree polynomial fit to the data points. Lower panel: Correlation coefficient *r* for 5-day periods (blue circles) and the number of used measurements (black squares).

nal dependence of NO<sub>2</sub> and O<sub>3</sub>. The correlation becomes strongly negative after the SPEs indicating a large increase in NO<sub>2</sub> and resulting ozone depletion.

[17] An interesting feature in Figure 1 is the increase of O<sub>3</sub> at 60–75 km after the events (days 311–340). The reason for this increase is presumably the development of the tertiary ozone maximum, which is known to exist in the high-latitude mesosphere [Marsh *et al.*, 2001].

[18] The MIMOSA advection model [Hauchecorne *et al.*, 2002] shows that when the SPEs occurred, the polar vortex was well formed in the upper stratosphere and almost coincident with the polar cap (not shown here). Most of the air with depleted ozone and enhanced NO<sub>2</sub> is located within the polar vortex as long as it is stable. The descent of NO<sub>2</sub> peak (Figure 2) is clearly related to the diabatic descent in the vortex which is very strong at the beginning of the winter. The rate of descent is few km/10 days, which is comparable to values reported by Eluszkiewicz *et al.* [1996] and Summers *et al.* [1997]. At the end of December, the dynamical situation changed and a sudden warming occurred in the upper stratosphere. As a result, the vortex was displaced from the pole and the latitude band  $70^\circ$ – $75^\circ$  no longer was fully inside the vortex. This explains the rapid increase of O<sub>3</sub> at this time between 40 and 60 km (Figure 1, days 351–360).

[19] It would be interesting to study the effects of these SPEs also in the southern hemisphere. There we would expect only a short-term decrease of ozone, mainly due to an increase of HO<sub>x</sub>, because in summer the increase in NO<sub>x</sub> is quickly balanced by photodissociation. When GOMOS data for 2004 becomes available, we will continue this study and follow the evolution of NO<sub>2</sub> and ozone throughout the polar night.

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