



HAL
open science

Analysis of the spatial distribution of the unusual NO₂ enhancements in the Arctic polar upper stratosphere and mesosphere observed by GOMOS-Envisat on January-March 2004

Jean-Baptiste Renard, Slimane Bekki, Pierre-Louis Blelly, Quentin Bourgeois, Gwenaël Berthet, Alain Hauchecorne

► **To cite this version:**

Jean-Baptiste Renard, Slimane Bekki, Pierre-Louis Blelly, Quentin Bourgeois, Gwenaël Berthet, et al.. Analysis of the spatial distribution of the unusual NO₂ enhancements in the Arctic polar upper stratosphere and mesosphere observed by GOMOS-Envisat on January-March 2004. *Journal of Geophysical Research Space Physics*, 2009, 114, pp.A12323. 10.1029/2009JA014174 . hal-00414776

HAL Id: hal-00414776

<https://hal.science/hal-00414776>

Submitted on 17 Mar 2015

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Analysis of the spatial distribution of the unusual NO₂ enhancements in the Arctic polar upper stratosphere and mesosphere observed by GOMOS-Envisat in January–March 2004

Jean-Baptiste Renard,¹ Slimane Bekki,² Pierre-Louis Blelly,³ Quentin Bourgeois,^{1,4} Gwenaël Berthet,¹ and Alain Hauchecorne²

Received 18 February 2009; revised 3 July 2009; accepted 1 September 2009; published 31 December 2009.

[1] Anomalously enhanced NO₂ concentrations are sometimes observed in the polar winter upper atmosphere. The enhancements over Antarctica from May to August 2003 were probably due to auroral electron precipitation, producing high amounts of NO in the upper mesosphere that were converted to NO₂ during downward transport to the stratosphere. Another enhancement was detected in the Arctic middle stratosphere in October–November 2003, due this time to energetic solar proton precipitation. This enhancement was quickly followed by a new Arctic NO₂ enhancement produced by auroral electrons in November 2003. Finally, a last enhancement was detected in the lower mesosphere from January to April 2004. Although it was proposed that this enhancement could also be due to auroral electrons, uncertainties remained concerning the absolute value of the NO₂ enhancement and its spatial coverage. We propose here a new analysis of the Global Ozone Monitoring by Occultation of Stars (GOMOS) nighttime measurements of the NO₂ enhancements. Instead of using daily zonally averaged data as done previously, we consider only the profiles containing the maximum values of the NO₂ enhancement. Unlike all the previous enhancements, the NO₂ content of the January 2004 appears to be longitudinally and latitudinally dependent inside the polar circle. The enhancement starts on 17 January 2004, with mixing ratios of up to a ppmv at altitudes above 60 km on 21 January. The enhancement looks like a “hot spot” above the polar cap. Then the enhancement spreads while it propagates downward into the stratosphere. It is accompanied by perfectly coincident strong ozone depletion; in particular, ozone is almost totally destroyed in mid-February at about 50 km. The vertical extent and horizontal spread of this NO₂ enhancement strongly differ from the November 2003 enhancement attributed to auroral electron precipitation. The possible origins of this unusual pattern are discussed.

Citation: Renard, J.-B., S. Bekki, P.-L. Blelly, Q. Bourgeois, G. Berthet, and A. Hauchecorne (2009), Analysis of the spatial distribution of the unusual NO₂ enhancements in the Arctic polar upper stratosphere and mesosphere observed by GOMOS-Envisat in January–March 2004, *J. Geophys. Res.*, 114, A12323, doi:10.1029/2009JA014174.

1. Introduction

[2] Two types of NO₂ enhancements were recently observed using numerous satellite instruments. A NO₂ enhancement associated with a mesospheric electron precipitation was detected above Antarctica from May to August 2003; it was well documented by *Funke et al.*

[2005] using the mid infrared limb emission Fourier transform spectrometer Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) onboard Envisat [*Fischer and Oelhaf*, 1996]. A strong NO₂ enhancement associated with a solar proton precipitation was detected by several satellite instruments in the northern polar stratosphere in October–November 2003 and was compared to modeling computation [e.g., *Semeniuk et al.*, 2005]. Maximum concentration of NO₂ were observed around 45 km at night using the Global Ozone Monitoring by Occultation of Stars (GOMOS) [*Seppälä et al.*, 2004] and MIPAS instruments onboard the Envisat satellite [*Orsolini et al.*, 2005; *López-Puertas et al.*, 2005]. A more extensive study over the 2003–2004 polar winter was also conducted by *Seppälä et al.* [2007] using the GOMOS data, showing various NO₂ enhancements during this period. Various sources (solar

¹Université d'Orléans, LPC2E, CNRS, Orléans, France.

²LATMOS, Université Pierre et Marie Curie (Paris 6), Institut Pierre-Simon Laplace, Paris, France.

³CESR, Toulouse, France.

⁴Now at Institute for Atmospheric and Climate Science, ETH Zurich, Zurich, Switzerland.

proton events, energetic electron precipitation and auroral energy electron precipitation) were proposed for the enhancements mentioned in the papers referenced here.

[3] High levels of NO₂ were also observed in the middle stratosphere at sunset/sunrise in Polar Ozone and Aerosol Measurements (POAM) III, Stratospheric Aerosol and Gas Experiment (SAGE) III [Randall *et al.*, 2005], Atmospheric Chemistry Experiment (ACE) [Rinsland *et al.*, 2005], and Halogen Occultation Experiment (HALOE) data [Natarajan *et al.*, 2004] from February to April 2004. Using preliminary MIPAS data and some GOMOS data, Renard *et al.* [2006] have shown that this last NO₂ enhancement started at about 60 km on 22 January, and was not linked to a solar proton event. At the same time, a geomagnetic storm occurred after two solar coronal mass ejections. Renard *et al.* [2006] speculated that the enhancement could be due to precipitations of electrons with energy of a few hundred keV, and a peculiar nighttime chemistry involving negative ion in the D layer of the ionosphere. On the other hand, López-Puertas *et al.* [2007] and Funke *et al.* [2007], using a more recent version of the MIPAS data, have concluded that the detection of the enhancement started around 15 January and can be due to a conventional downward transport of upper atmosphere NO_x produced throughout the 2003–2004 polar winter by auroral and precipitating electrons. Other studies have also concluded that the January 2004 event has a significant dynamical origin [Randall *et al.*, 2006; Siskind *et al.*, 2007]. In particular, Hauchecorne *et al.* [2007] and Manney *et al.* [2005] have shown that a strong mesospheric air descent indeed occurred during this period because there was an intense mesospheric warming in late December and a enhanced gravity wave breaking in the upper mesosphere. Then, the enhancements in NO₂ appeared when NO was converted to NO₂ in the lower mesosphere, while NO_x (= NO+NO₂) carried on being transported to the middle stratosphere until April 2004. The hypothesis of the auroral production is further supported by observations of clear change in the radio wave diurnal propagation in January 2004 as a consequence of the descent of NO_x into the mesosphere [Clilverd *et al.*, 2006, 2007]. All these studies have also shown that the enhancements started a few days before the date proposed by Renard *et al.* [2006].

[4] Although all these late results suggest that the origin and the time evolution of the January 2004 enhancement are well identified and understood, there remain very large uncertainties concerning the total amount of the NO₂ enhancement detected by the various satellite instruments. Funke *et al.* [2007] estimated it to about 100 ppbv or more of NO_x in the mesosphere, using MIPAS and HALOE data. Renard *et al.* [2006] estimated it to more than 200 ppbv using GOMOS and MIPAS data. Finally, Hauchecorne *et al.* [2007] found zonal averaged mixing ratios of more than 600 ppbv in NO₂, although they claimed that values exceeding 1 ppmv could be found in some individual profiles. Also, the vertical extent of the enhancement and its evolution during the downward transport in the work of Hauchecorne *et al.* [2007] differ very significantly from the results of Clilverd *et al.* [2007], although both studies used data coming from the same instrument (GOMOS) but not using the same latitude coverage.

[5] There are differences in the estimation of the absolute values of the enhancement and of its vertical extent, raising questions of using satellite data from different instruments and having different latitude criteria for the data analysis. Also, some problems can appear considering the GOMOS single vertical profiles, which exhibit different signal-to-noise ratios from one profile to another. The various authors have proposed different selection criteria for excluding the noisy GOMOS measurements, thus giving different analysis of the intensity of the enhancement.

[6] Meanwhile, an intensive validation work of GOMOS ozone and NO₂ profiles have been conducted using balloon-borne instruments [Renard *et al.*, 2008]. Therefore, we propose here to reanalyze the polar winter GOMOS data above Antarctica during 2003 and the Arctic during 2003–2004 using the results of the validation work in order to better characterize the NO₂ enhancements, their evolutions and the impact on ozone levels. In addition, the estimation of the amount of the NO₂ enhancement and of its impact on ozone will be conducted here using the individual profiles instead of zonal and daily averaged data used in previous studies. Such analysis could help to better document the spatial distribution and the temporal evolution of the January–March 2004 enhancement when compared to the other enhancements.

2. GOMOS Data

[7] GOMOS onboard Envisat is an instrument for the retrieval of stratospheric and mesospheric species that exhibit absorption lines in the UV-visible and near-infrared domain [Bertaux *et al.*, 2004; Kyrölä *et al.*, 2004]. GOMOS performs measurements using the stellar occultation method during the setting of stars, allowing the retrieval of the vertical profiles of ozone from about 110 km to the troposphere, and of NO₂ from about 75 km to the lower stratosphere. Several hundred occultations are conducted per day, leading to a global latitude and longitude coverage. Nevertheless, the signal-to-noise ratio varies from one individual profile to another, since stars with different magnitudes and temperatures are observed. Some profiles can be noisier than others because of problems in the pointing system or the presence of polar stratospheric clouds, which can produce artificial oscillations [Renard *et al.*, 2008]. Also, at given latitude and longitude ranges, the number of observed stars changes from one day to another, depending on their conditions of visibility. Finally, the spatial sampling is not regular because of the nonuniform distribution of luminous stars in the sky. As a result, accuracy and hence the meaning of the results can vary from one day to another when producing maps of the vertical distribution of the species using zonal and daily averaged data, especially when there are gaps in the latitude and longitude coverage because of the absence of observable stars. These spatial conditions of measurements differ strongly from those performed when using the Sun as light source (like SAGE) or limb spectrometers. Therefore, the treatment of GOMOS data for scientific use must differ from the one performed routinely on most data that have regular sampling and almost constant signal-to-noise ratio.

[8] GOMOS was mainly dedicated to evaluate accurately the trend of the species at all latitudes, in the stratosphere

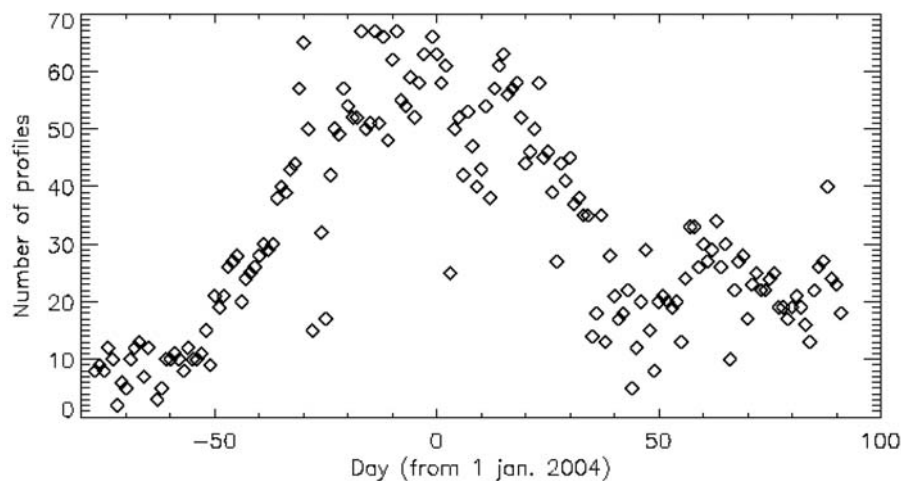


Figure 1. Total number of profiles kept each day for the 2003–2004 winter above the Arctic.

and the mesosphere using averaged data. Nonetheless, one can expect that specific case studies can be carried out using individual profiles if they are accurate enough. Fortunately, because of the large number of occultations performed each day, even an extensive data filtering could leave tens of star occultations per day inside the polar circle. The problem is the quality of the data selection criteria.

[9] In the present analysis, the data used are the version V5.0, provided by the European Space Agency. During the validation exercise with balloon vertical profiles available in the lower and middle stratosphere, *Renard et al.* [2008] estimated an accuracy of about 3×10^8 molecule cm^{-3} for nighttime GOMOS NO₂ vertical profiles obtained for stars having magnitude from -1.4 to 2.7 . No bias in the estimation of the altitude has been found. These results were obtained using GOMOS data from 25 km to the balloon float altitude, just below 40 km. Thanks to the inversion method used, least squares fit and regularization, it seems reasonable to adopt, as a first guess, the same accuracy in term of concentration all along the profile, at least in the middle and upper stratosphere. This can be assumed because the values of the transmission spectra are similar (i.e., the star flux is only slightly attenuated by the atmosphere) in the middle and upper atmosphere. On the other hand, the profiles are less accurate in the lower stratosphere when the flux of the star is close to the detection limit because of longer path lengths for the lines of sight. Obviously, the uncertainty increases with increasing altitude when the amounts are expressed in mixing ratios.

[10] In the following, we will consider profiles from geographic latitudes higher than 60° . In practice, because of the GOMOS spatial sampling, the cutoff is above 63° . This is close to the polar cap, which starts at about 66.5° .

[11] We have calculated the standard deviation of all the winter 2003–2004 GOMOS nighttime individual profiles available for measurements in the 80–110 km altitude range, after applying on each profile a vertical sliding smoothing over 3 consecutive data points in order to reduce the noise. This altitude range (that contains about 20 data points) has been chosen because no NO₂ enhancements are expected in this range, even in the case of electron precip-

itations, since all previous studies have shown that the enhancements occur below 65 km. The flux of the transmission spectra at this altitude range is similar; thus, this calculation gives values that are directly relevant to the instrumental noise and to the star brightness. Also, it can indicate if pointing problems occurred during the occultation and if the retrieval is biased by local mesospheric clouds.

[12] We have performed a statistical analysis of the GOMOS profiles and have found that the NO₂ enhancement in the middle and upper stratosphere cannot be accurately detected, both for the altitude of the maximum concentration and absolute value of the enhancement, when the standard deviation of the upper part of the profile is greater than at least 3×10^8 molecule cm^{-3} . This value is similar to the one determined during the GOMOS validation exercise with balloon instruments. This further supports the adoption of this value of accuracy as a criterion for selecting the GOMOS data expressed in concentrations versus altitude. Then, we propose to reject all the profiles that have a standard deviation greater than 3×10^8 molecule cm^{-3} in the 80–110 km altitude range, and, obviously, profiles obtained during twilight and daytime. With such filtering, only enhancements of NO₂ greater than 3×10^8 molecule cm^{-3} can be detected. In practice, adopting at least a 2-sigma criterion, enhancements greater than 6×10^8 molecule cm^{-3} can be assumed to be real.

[13] After applying this procedure, globally about 20% of the profiles are kept for the Antarctic 2003 and Arctic 2003–2004 enhancements. Depending on the stars availability, between 10 and 80 profiles are available daily. Figure 1 presents the total number of profiles kept each day for the 2003–2004 winter. As expected, the number of profiles is at maximum around the end of the year, when the nights are the longest. It can be noticed that a drop of number of profiles occurs around mid-February, due to a lack of bright stars observed at this period. Figure 1 shows the problem of irregular GOMOS sampling that can produce some signal to noise variations over the year when the profiles are zonally and temporally averaged. Also, such variations can occur when considering only individual profiles.

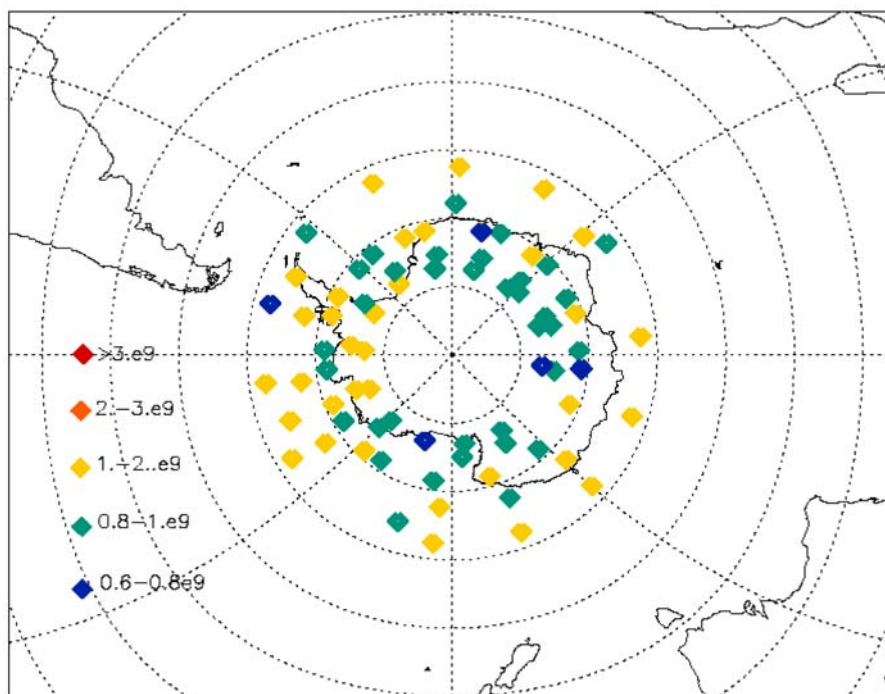


Figure 2. Spatial coverage of the Antarctic NO₂ enhancement in the upper stratosphere above 40 km on 25 July 2003. The solid diamonds represent the maximum concentrations for each profile in molecule cm⁻³. The enhancements occur in a similar altitude range.

[14] All the profiles obtained using the brightest stars having temperature above 5000 K are kept with this procedure. On the other hand, some profiles obtained with stars having a magnitude down to 2.5 and a temperature above 5000 K are kept, and some others are rejected. This seems to be related to the individual star spectrum (width and temporal variability of the Fraunhofer lines) but also to the GOMOS pointing quality. Thus, no simple rules can be established for our error criterion that is relevant to the magnitude and temperature of low-magnitude stars.

[15] It is worth stressing that this filtering procedure uses only a posteriori criterion based on an analysis of the individual profiles. It relies only on the signal-to-noise ratio of the profiles without a priori hypothesis concerning the star magnitude and temperature. This method is a new approach of analysis of GOMOS measurements in the middle and upper stratosphere. The quality and consistency of the results obtained with this approach are discussed in the following sections.

3. Spatial Distributions of the Enhancements

[16] The daily spatial sampling of GOMOS makes it possible to estimate the latitude and longitude coverage of the various strong NO₂ enhancements at any altitude above 40 km (in order to be above the usual polar NO₂ amounts), whatever their origin is. Figures 2–5 present the spatial coverage of the NO₂ enhancement and its maximum concentration value in the upper stratosphere and mesosphere above the Antarctic on 25 July 2003, and above the Arctic on 20 November 2003, on 21 January 2004, and 3 March 2004, respectively. Only enhancements greater than 6×10^8 molecule cm⁻³ above 40 km present over three con-

secutive vertical points and over at least two time-consecutive vertical profiles are taken into account in order to distinguish unambiguously between real features and noise. It must be noticed that the enhancements occur in a similar altitude range. In Figure 2, the enhancement covers totally the polar cap, and has almost constant values. Similar conclusions can be derived from Figure 3. On the other hand, only a small part of the polar cap is covered by the enhancement in Figure 4. In Figure 5, the enhancement covers totally the polar cap, but with a strong variability for the NO₂ amounts. Thus, it appears that the spatial distribution of January 2004 enhancement differs strongly from the others, covering only a small part of the polar cap.

[17] Figure 6 shows the percentage of GOMOS data above polar cap where an NO₂ enhancement at altitude above 40 km is present for the Arctic 2003–2004 polar winter. It must be noticed that the curve is globally continuous, showing that the profile selection method proposed here does not lead to scattered results. For the proton events of November 2003, the NO₂ enhancement starts at the beginning of November and covers on average 90% of the polar cap throughout November. A similar pattern is found for the Antarctic 2003 NO₂ enhancement associated with electron events. On the other hand, the January 2004 enhancement covers at its beginning only about 20% of the polar cap. The coverage increases rapidly during the two last weeks of January, reaching about 60% at the beginning of February. Then the increase slows down and reaches a maximum coverage at the beginning of March 2004 when the enhancement, due to the mesospheric descent, has totally penetrated the stratosphere through the stratopause (50 km). This increase could be probably due to the spreading of the NO_x cloud in various directions during

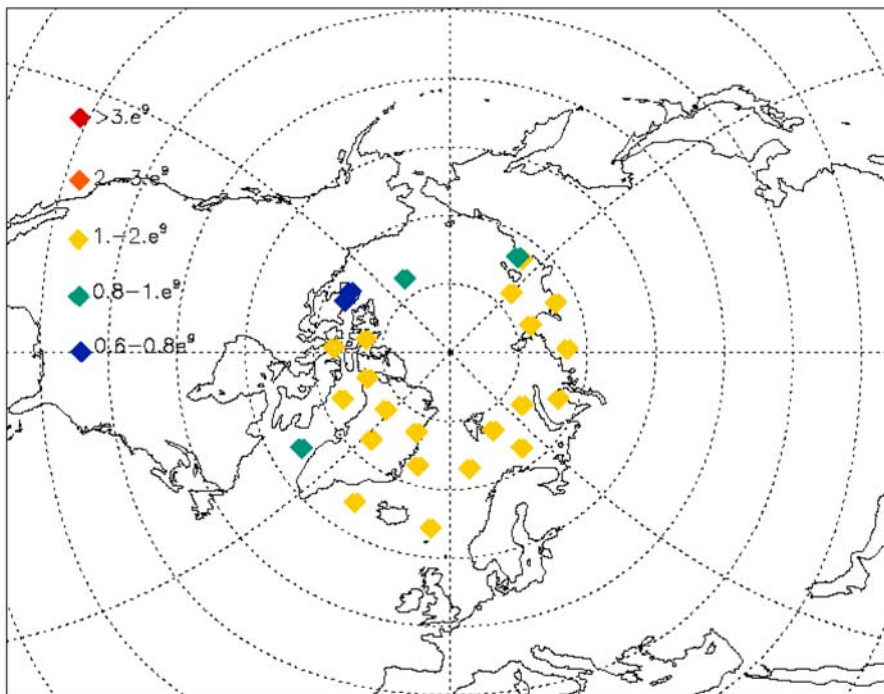


Figure 3. Spatial coverage of the Arctic NO₂ enhancement above 40 km on 20 November 2003. The solid diamonds represent the maximum concentrations for each profile in molecule cm⁻³. The enhancements occur in a similar altitude range.

the downward transport process. Even when the coverage reaches 95%, a careful analysis of the measurements shows that the horizontal distribution of the enhancement is not entirely homogenous.

[18] This large spatial variability in the concentration fields can certainly bias the analysis when the profiles are zonally averaged. In all the previous published works (including ours) using GOMOS and MIPAS data, the profiles

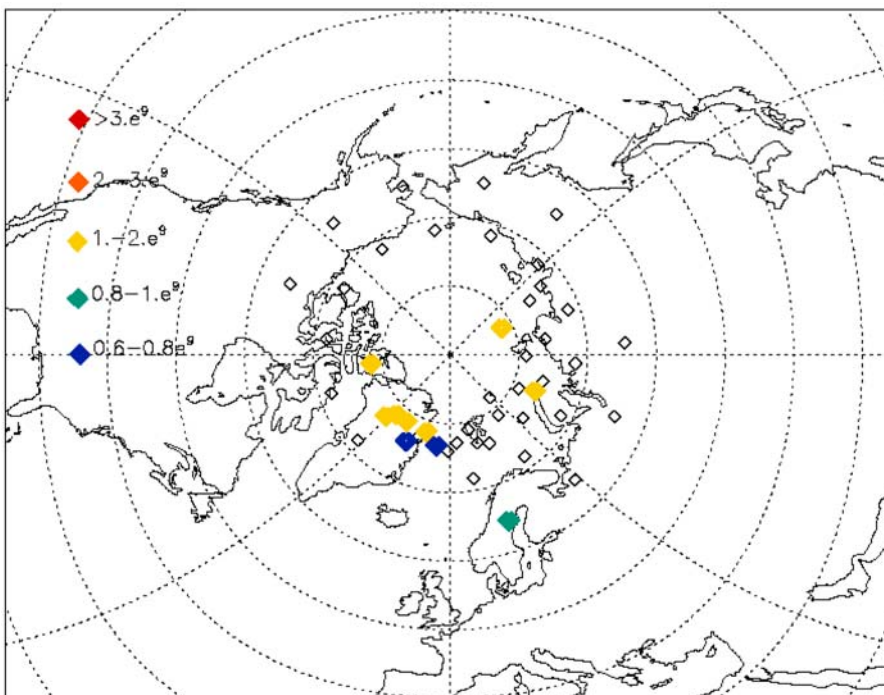


Figure 4. Spatial coverage of the Arctic NO₂ enhancement above 40 km on 21 January 2004. The solid diamonds represent the maximum concentrations for each profile in molecule cm⁻³, and the open diamonds represent the GOMOS profiles where no enhancement is present. The enhancements occur in a similar altitude range.

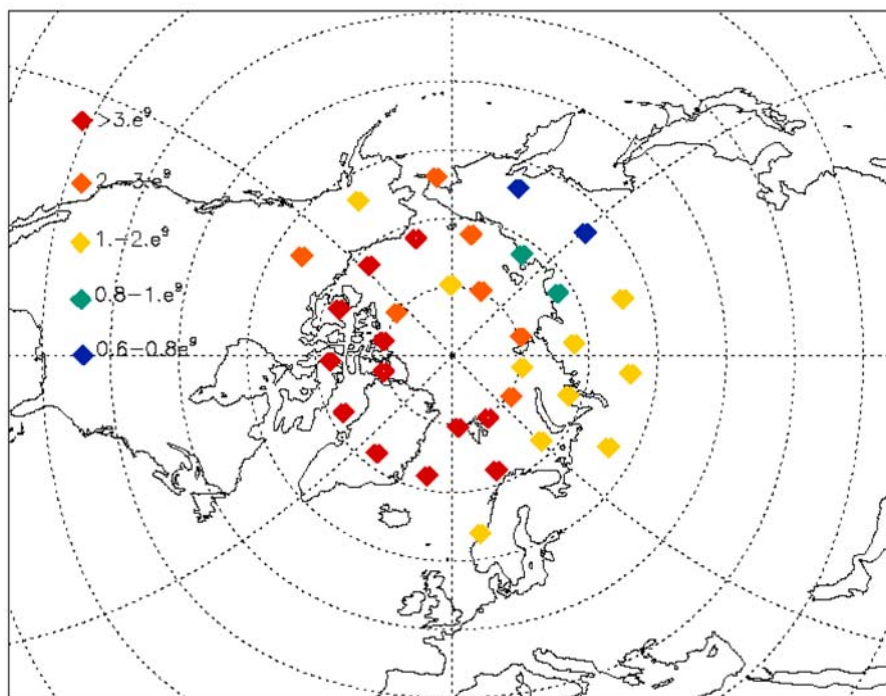


Figure 5. Spatial coverage of the Arctic NO₂ enhancement above 40 km on 3 March 2004. The solid diamonds represent the maximum concentrations in molecule cm⁻³. The enhancements occur in a similar altitude range.

were averaged resulting in strongly underestimated values for the NO₂ maximum for the January–April 2004 enhancement, especially during the first phase. It is therefore necessary to consider only the individual GOMOS profiles exhibiting anomalously large NO₂ concentrations in order to evaluate better both the positions and the absolute values of the enhancements.

4. January 2004 Enhancement

[19] Because the enhancement has a small spatial extent, like a “hot spot” within the polar cap but not necessary

centered over the pole (Figure 4), we propose a new method for a better analysis of the temporal evolution of the enhancement. The profile containing the strongest value of the NO₂ enhancement is the only profile taken into account per day (considering altitude range starting a few km above the altitude of the November 2003 enhancement residual). Taking into account the GOMOS validation work of Renard *et al.* [2008], the errors on the NO₂ profiles is of about ±25%, at least in the middle stratosphere. Since NO₂ has a short life time (not a chemical tracer), considering individual profiles instead of spatially and temporally averaged profiles is one of the best approaches in order to

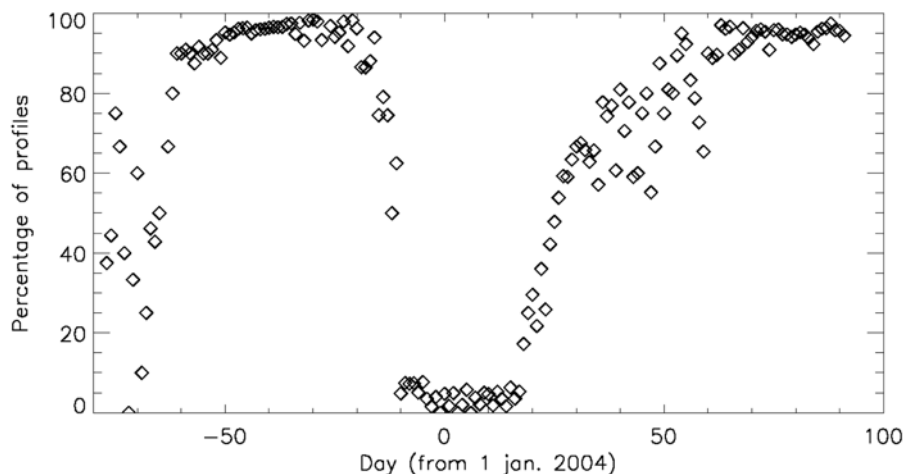


Figure 6. Percentage of GOMOS data where the NO₂ enhancement is present in the middle and upper stratosphere during the 2003–2004 polar winter.

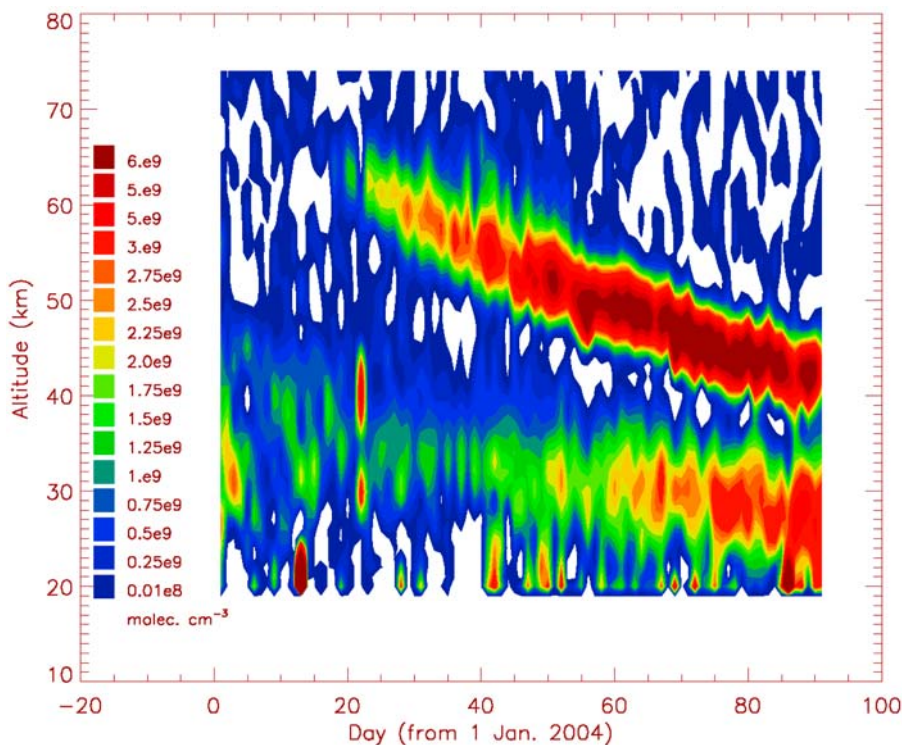


Figure 7. Temporal evolution of the Arctic NO₂ concentration profile at the beginning of 2004; for each day, only the profile with the highest NO₂ concentration is plotted.

estimate the absolute value of the enhancement when it is local, as shown here.

[20] Figures 7 and 8 present the temporal evolution obtained with such approach, for the concentration and mixing ratio profiles, respectively. Although the noise remains more or less constant when considering concen-

trations, as it can be seen in Figure 7, the mixing ratio noise increases with increasing altitude. Presenting results only in mixing ratios can produce “false detections” like the blobs in Figure 8 at altitudes above 0.1 hPa at the beginning of January. Thus, the altitude and absolute values of the enhancement are searched for considering first data

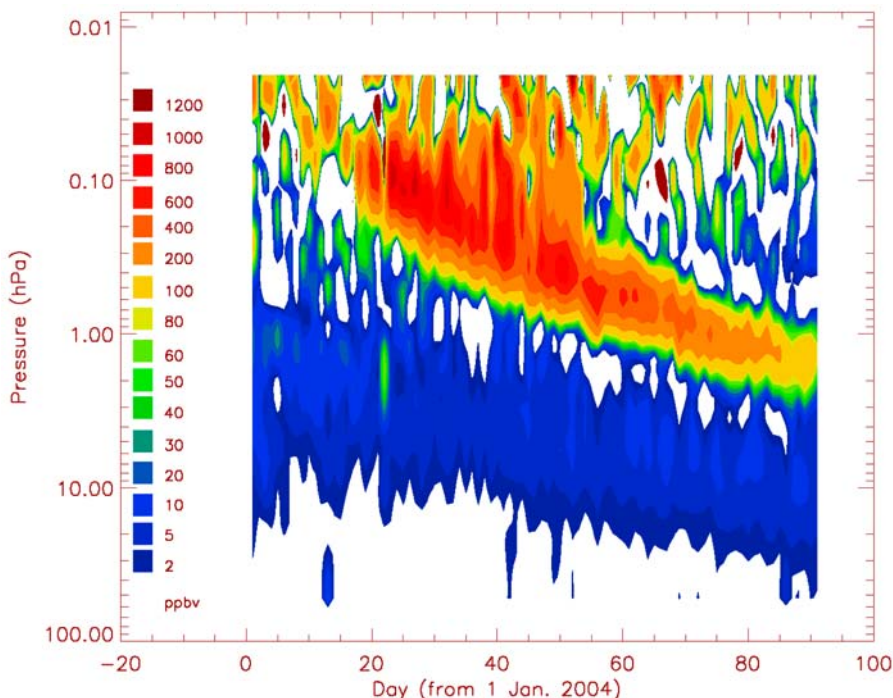


Figure 8. Same as Figure 7, but expressed in mixing ratio versus pressure.

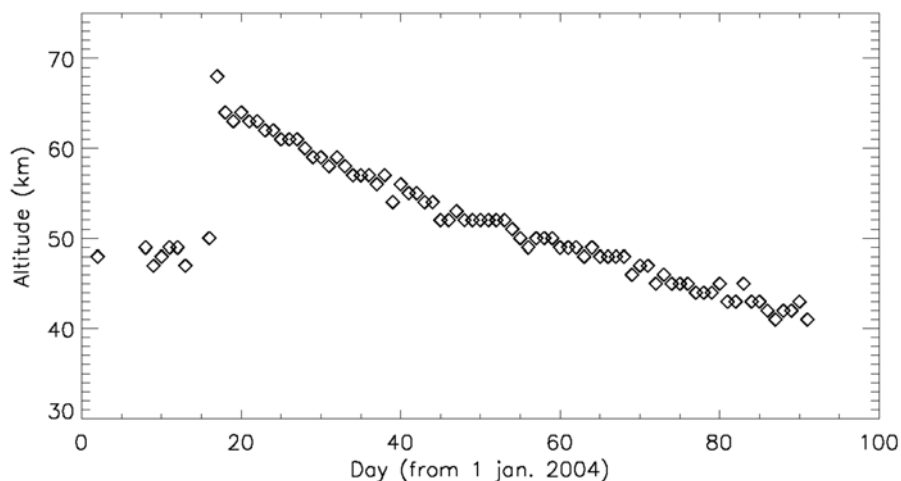


Figure 9. Temporal evolution of the altitude of the January–April 2004 enhancement expressed in mixing ratio. No obvious enhancement can be detected during the first days of January 2004 (the altitudes of the maxima around 50 km at this period correspond to “artificial” enhancements produced by the conversion of concentrations to mixing ratios).

expressed in concentrations. Then these enhancement values can be converted to mixing ratios.

[21] The results presented here are noisier than the ones obtained when averaging tens of profiles. Nonetheless, the vertical and temporal features of the NO₂ enhancement can still be analyzed accurately. The vertical shape of the enhancement and its temporal evolution are in agreement with the *Hauchecorne et al.* [2007] map, but not with the *Clilverd et al.* [2007] map on which the vertical position of the enhancement in mid February is strongly scattered. This difference could be due to the fact that, in the former study, the profile selection was limited to latitudes greater than 80°, whereas, in the latter study, profiles over a wider latitude range were considered, making the results more sensitive to the bias associated with the inhomogeneous concentration fields and the irregular spatial sampling of GOMOS. By averaging profiles, both studies should have underestimated the values of the enhancement, which is around 1 ppmv.

[22] In our case, the altitude of the initial enhancement expressed in concentration and in mixing ratio is about 65 km, occurring on the 17 January 2004 (Figure 9). This starting date is slightly later than the date (around 15 January) derived from the MIPAS data by *Funke et al.* [2007]. The maximum mixing ratio is observed three days later, on 21 January (this date was assigned to the beginning of the enhancement by *Renard et al.* [2006] using the preliminary MIPAS data). Then, the altitude of the enhancement decreases due to the downward transport. It must be noticed that the scatter on the altitude of the enhancement is low, of the order of 1 km, and in perfect agreement with results obtained by the other studies using averaged data. Thus, we can conclude that the GOMOS profiles are indeed accurate enough to be used individually.

[23] Figure 10 shows the evolution of the GOMOS NO₂ mixing ratio (corresponding to the concentration peak) with time. There is a bit of scatter, with uncertainties of a few hundred of ppbv around mid-February because of the lack

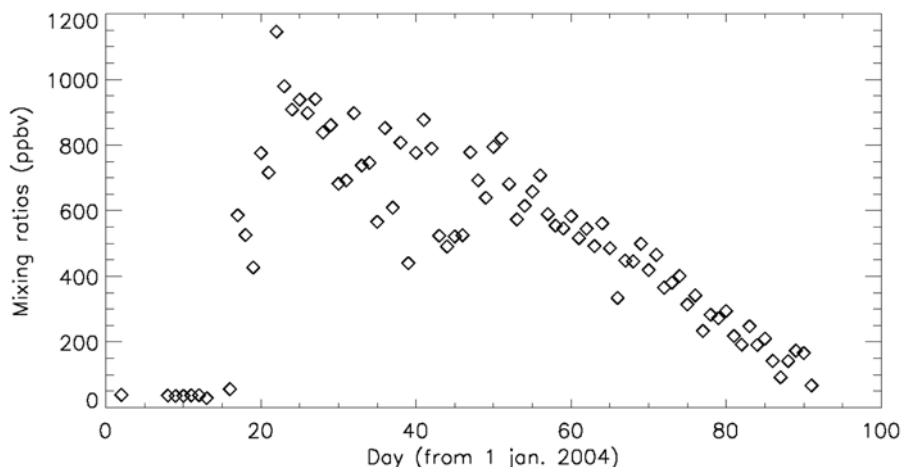


Figure 10. Temporal evolution of the January–April 2004 maximum NO₂ mixing ratio (corresponding to the concentration peak).

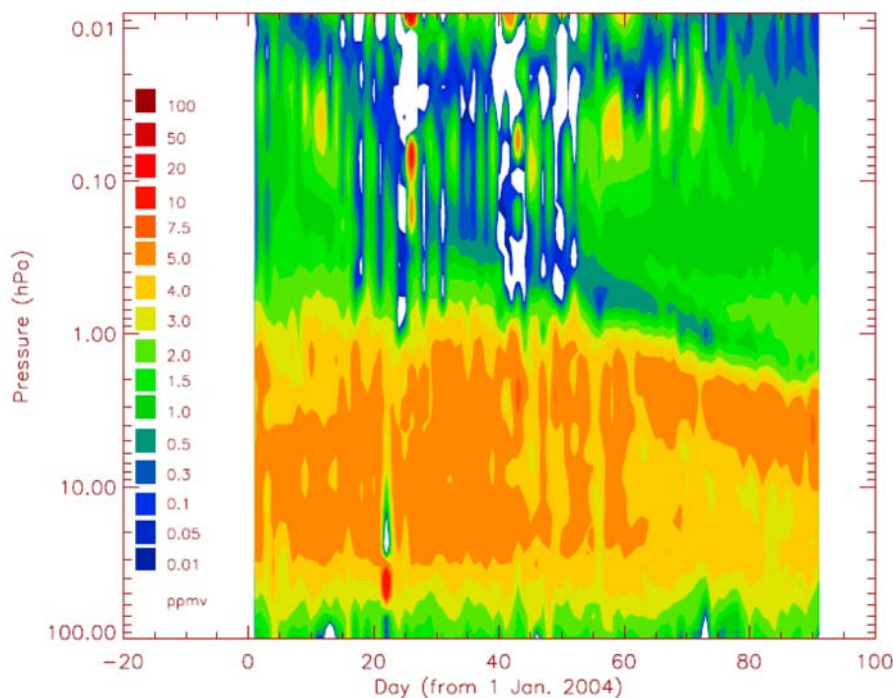


Figure 11. Temporal evolution of the Arctic ozone profiles in mixing ratio at the beginning of 2004; only the ozone profiles corresponding to the same NO₂ profiles shown in Figures 7 and 8 are plotted.

of bright stars that usually give more accurate results. Nevertheless, the decay of NO₂ mixing ratio with decreasing altitude is clearly observed, showing once again that our methodology appears to work well. Figure 10 confirms that the enhancements appear suddenly, with a value of about 0.5 ppmv on 17 January, and reaches a maximum, above 1 ppmv, on 21 January. Then the values slowly decrease almost linearly with time.

[24] It must be noticed that some inconsistencies seem to appear between Figures 7–10 for the altitude of the NO₂ maximum concentrations and the maximum mixing ratios before 15 January. This is a perfect illustration of the bias that can produce the conversion from concentrations to mixing ratios when the possible enhancements are smaller than the noise.

[25] Figure 11 presents the evolution of ozone mixing ratio profile with time using the same occultations as for NO₂ (i.e., one profile per day). We must use the same profiles for the two species in order to be able to evaluate the effect of the NO₂ enhancement on ozone. Once again, the profiles are accurate enough, but this time up to an altitude of 110 km, so the three altitude regions of ozone maxima appear distinctly (not shown here). A strong local decrease of ozone is clearly visible, from around 0.1 hPa at the end of January 2004 to 1 hPa until April 2004. The temporal evolution of the altitude of this local ozone depletion is the same as the evolution of the altitude of the NO₂ enhancement (Figure 12), indicating that the two phenomena are intimately linked. It must be noticed that, because of the weak brightness of the stars observed at the end of January, the ozone values are close to noise level, preventing an accurate estimation of the ozone amount at the top altitudes of the NO₂ enhancement (on the other

hand, the NO₂ enhancement is so strong that it can be detected even using stars having weak brightness). The perfect correlation found between the NO₂ enhancements and ozone (0.98 after 25 January) is an additional validation of the data selection procedure that result in consistent and meaningful results.

[26] Figure 13 presents the evolution of the mixing ratios of the ozone depletion with time. Each point is retrieved at the altitude of the NO₂ maximum enhancement. It is most certainly in situ chemical ozone destruction. If it was a tongue of ozone-depleted descending air, the ozone mixing ratio would be minimal at the beginning of the descent, between day 20 and 40, and not between day 45 and 60 as observed. Indeed, from the end of January to mid-February, the ozone content is divided by about a factor of 2. Then, when the NO₂ enhancement enters the stratosphere after mid-February, almost all the ozone is destroyed. This is the consequence of the large amounts of NO₂ still present in the core of the NO_x perturbation; the peak of NO₂ concentration is still of order of half of a ppmv, which is still almost 2 orders of magnitude above the background levels. This results in a massive increase in the ozone destruction by the NO₂ catalytic cycle. It is one of the dominant ozone-destroying cycles in the stratosphere and its efficiency is directly proportional to NO₂ except at high-latitude winter during the polar night (the rate-limiting step is the reaction between NO₂ and atomic oxygen O). As the NO₂ mixing ratio decays (see Figure 8), the perturbation to the ozone chemical budget diminishes as indicated by the increase in ozone amount. The ozone concentration responds quickly (on diurnal time scales) to NO₂ changes because, unlike the lower stratosphere, photochemistry is the dominant process in the ozone budget in the upper stratosphere. As a result,

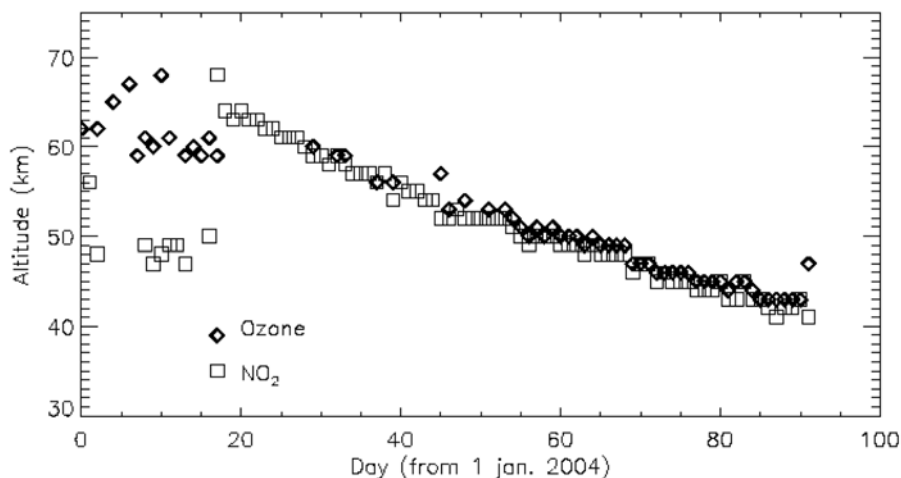


Figure 12. Temporal evolution of the altitude of the ozone depletion and of the altitude of the maximum NO₂ enhancement. The excellent match between both altitudes confirms that the ozone destruction is certainly linked to the NO₂ enhancement.

there is an excellent correlation between NO₂ changes and ozone changes. By April, ozone has more or less recovered its climatological value of the order of a couple ppmv at such altitude.

5. Particularities of the January 2004 NO₂ Enhancement

[27] As said before, the January 2004 NO₂ enhancement strongly differs from the other enhancements recorded before, both in term of the amplitude of the enhancement and of its vertical distribution. The January enhancement manifests itself with about 1 ppmv of NO₂ that is first observed at an altitude of 65 km. Its vertical extent is close to 10 km, and the NO₂ mixing ratio decreases above the altitude of the maximum (see Figure 8). These features can be contrasted with those of other NO₂ enhancements that have been recently documented, where the NO₂ mixing ratio is more or less constant with increasing altitude, at least up to 0.02 hPa.

[28] We have reanalyzed the GOMOS data in order to produce maps of the temporal evolution of the other enhancements that can be compared to the map of the January 2004 enhancement. As done previously (Figures 7 and 8), the profile containing the strongest value of the NO₂ enhancement is the only profile taken into account per day. Figure 14 presents the temporal evolution of the NO₂ mixing ratios for the Antarctic enhancement on May–August 2003 that was produced by electron precipitation [Funke *et al.*, 2005]. The Arctic NO₂ enhancement that started late October–early November 2003 was produced by a strong solar proton event [López-Puertas *et al.*, 2005]. Our analysis of GOMOS data confirms the existence of another Arctic enhancement that occurred a month later in the mesosphere [López-Puertas *et al.*, 2007; Seppälä *et al.*, 2007]. Figure 15 shows the temporal evolution of the NO₂ enhancement in the upper stratosphere and lower mesosphere for the November–December 2003 period. The last November enhancement appears in Figure 15 as a permanent blue zone up to an altitude of 70 km.

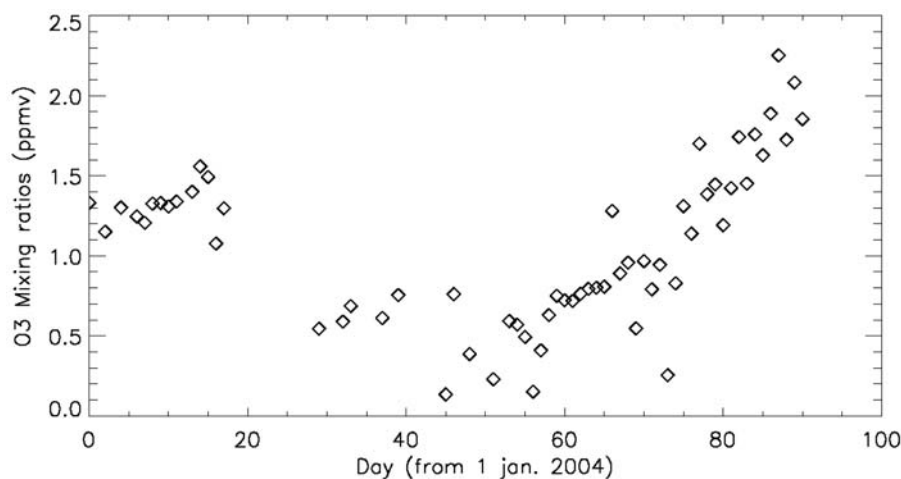


Figure 13. Temporal evolution of the ozone mixing ratios corresponding to the NO₂ concentration peak. The values of about a couple of ppmv at the end of the period are representative of climatology values.

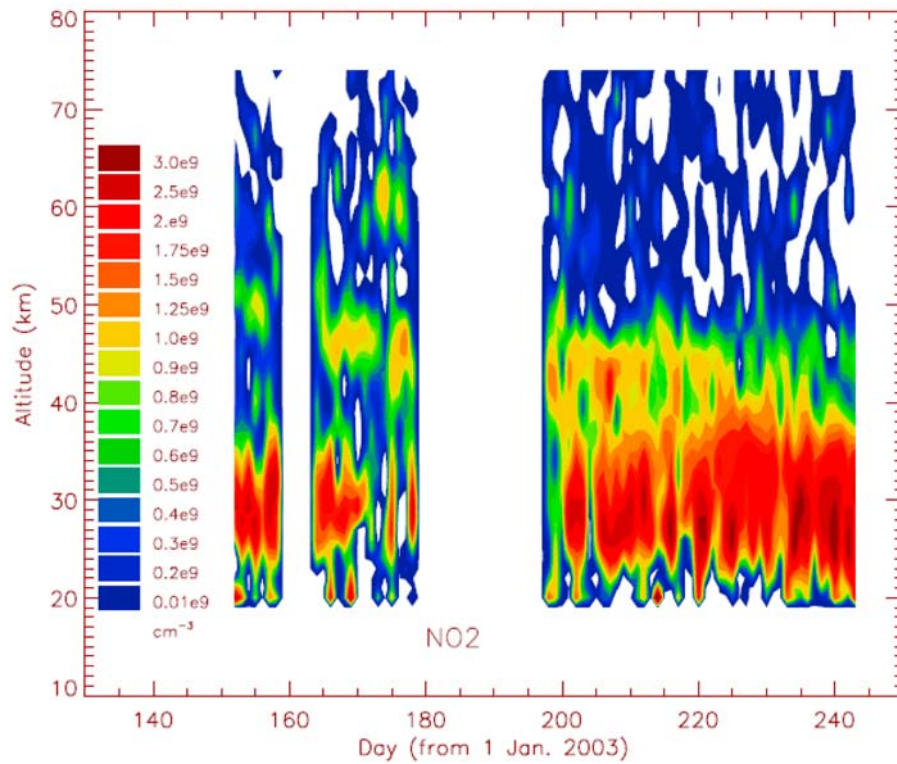


Figure 14. Temporal evolution of Antarctic NO₂ mixing ratios in mid-2003; for each day, only the profile with the highest NO₂ concentration is plotted.

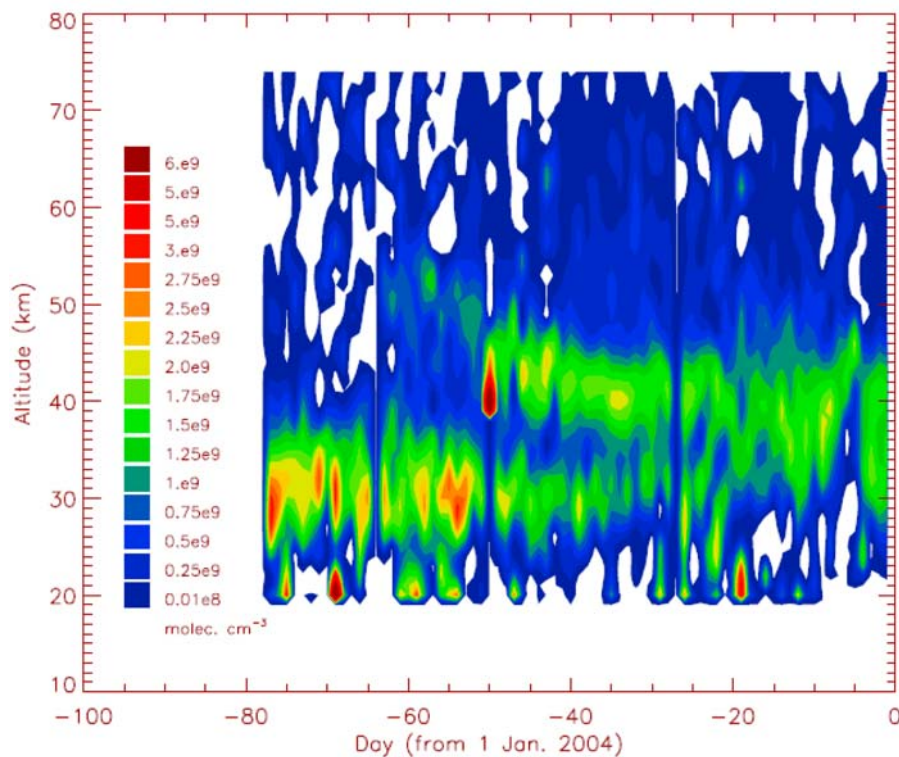


Figure 15. Temporal evolution of Arctic NO₂ mixing ratios at the end of 2003; for each day, only the profile with the highest NO₂ concentration is plotted.

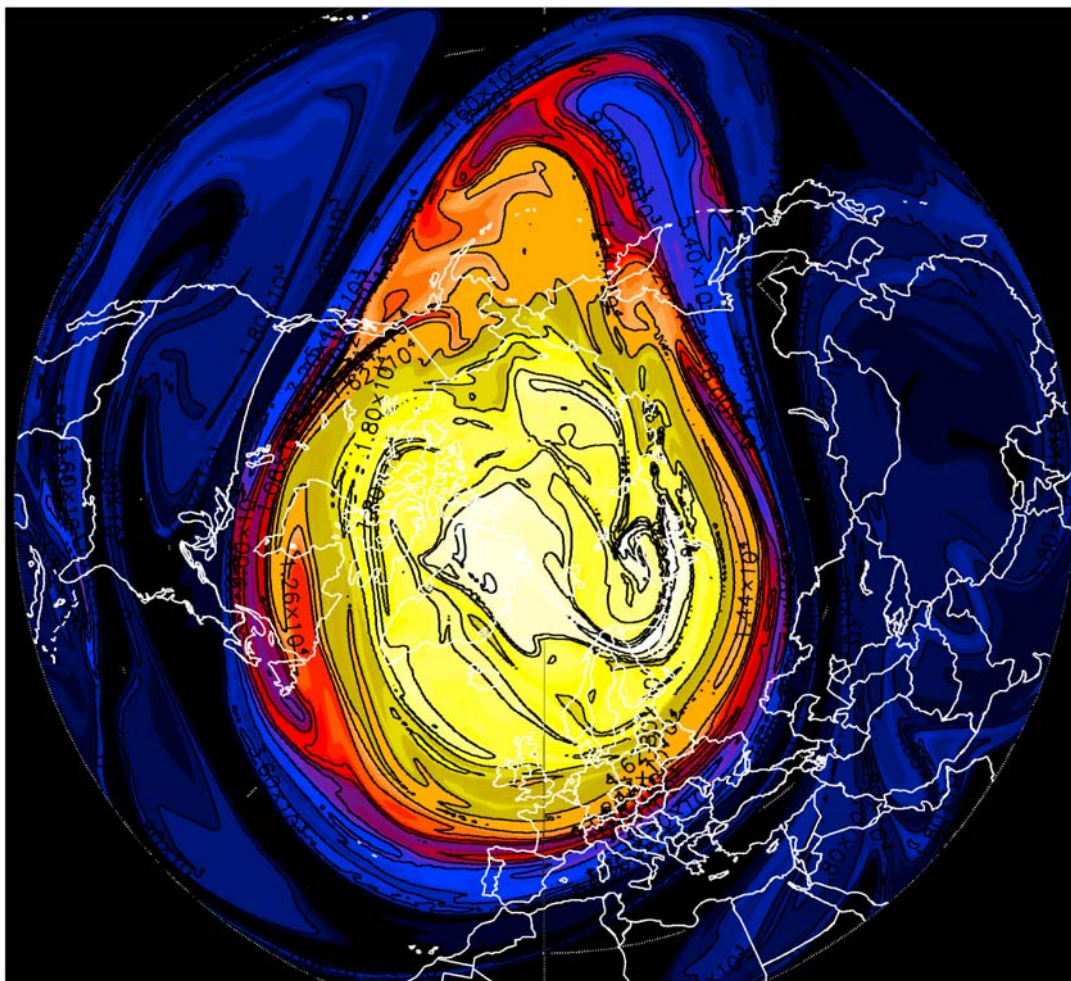


Figure 16. Output of the MIMOSA advection model showing at 1900 K (above 40 km) some correlation between maximum values of potential vorticity stretched like a “tongue” and the localization of the NO₂ enhancements on 21 January 2004.

[29] The May–July 2003 and November–December 2003 enhancements were probably produced by electron events. The amplitude of the enhancements is of the order of several hundreds of ppbv which is lower than the enhancement observed in January 2004. Also, their vertical extents are more diffuse, with NO₂ mixing ratios remaining more or less constant in the lower mesosphere, at least over a vertical extent of about 15 km (up to the highest altitudes where the GOMOS data can be analyzed). As a result, it is very difficult to identify a maximum in the profiles or a well-defined altitude for these two enhancements. All these differences between the two electron events and the January 2004 event suggest that this last event could have a different origin, or could have occurred during different geophysical conditions in the mesosphere and the stratosphere. No strong proton events have been detected at the start January 2004, so the proton origin must be rejected for this NO₂ enhancement.

[30] If the January 2004 enhancement has an auroral electron origin, the dynamical context must differ very significantly compared to the other enhancements. Only a small “tongue” of mesospheric air must have been trans-

ported downward and only over a short period of time, perhaps a few days, which is in contrast with the global coverage of the polar cap by the enhancement during the others mesospheric descents. This could be the result of the anomalous warming at the end of 2003 linked with gravity wave breaking [Hauchecorne *et al.*, 2007]. This hypothesis may be supported by the outputs of the MIMOSA advection model [Hauchecorne *et al.*, 2002] somehow showing at 1900 K (above 40 km) some correlation between maximum values of potential vorticity stretched like a “tongue” and the localization of the NO₂ enhancements on 21 January 2004 (Figure 16). Nevertheless, it must be noticed that no analysis data are available in the mesosphere and the structure of the polar vortex at 1900 K was most likely different from what it was 20 km higher. More thorough dynamical studies must be conducted in the future in order to demonstrate whether a mesospheric descent over such a small spatial extent could really have occurred during that period.

[31] To examine all the possible explanations for the nature of the enhancement, without being in favor of one explanation compared to the others, it might be tempting to

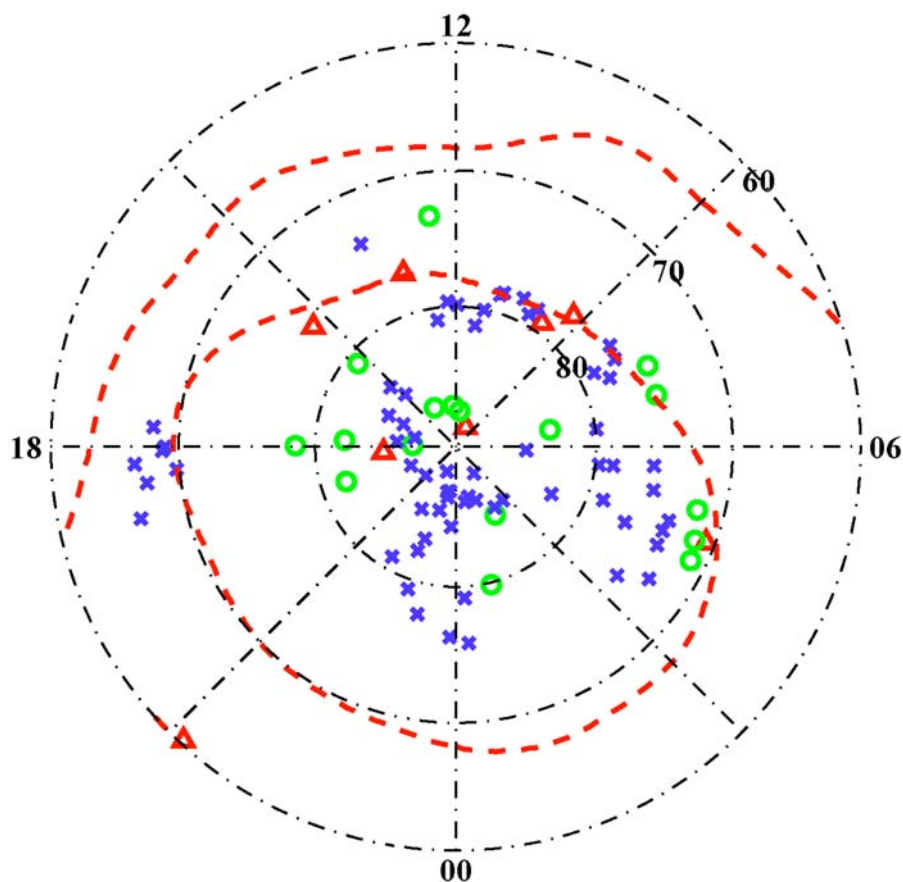


Figure 17. Locations of the NO₂ enhancements between 17 January and 31 January 2004 in polar magnetic representation. The triangles are the positions of the maxima observed between 17 and 19 January, the small circles are those observed between 20 and 22 January, and the crosses are those observed between 23 and 31 January. The magnetic north pole is at the center, the dash-dotted wide circles represent the magnetic latitudes above 60°, and the position in longitude is represented using the magnetic local time. The dashed lines represent a mean location of the boundaries of the auroral oval, derived from the *Hardy et al.* [1987] statistical study on electron precipitation.

look for another origin related to particle precipitations. But, no significant particle precipitations were recorded at this time. The fact that the enhancement began on 17 January 2004 seems to rule out the magnetic storms of 21–22 January as a cause, though it is intriguing that the maximum of the NO₂ enhancement was detected just these days (where the enhancement could had reached a critical altitude level for the NO to NO₂ conversion). We have shown above that, even with its non regular horizontal sampling, GOMOS allows an estimation of the locations of the enhancement. A statistical examination of these locations shows that they are not always centered above the geographical pole or inside the polar circle. So the geographical coordinates may not be the most relevant coordinates for the enhancement. Actually, when studying phenomena in the Earth's magnetosphere and ionosphere, data are often analyzed in a coordinate system based on the magnetic field. Especially when working at high latitudes and when comparing satellite with ground based measurements, one requires a magnetic coordinate system that is smooth at the poles and accurate at different heights. One system which is well suited is the Altitude Adjusted

Corrected Geomagnetic (AACGM) coordinate system [Baker and Wing, 1989; Bhavnani and Hein, 1994].

[32] To better estimate the spatial extension of the “NO₂ tongue,” we have taken into account all the GOMOS profiles having a NO₂ enhancement greater than $0.7 \times \text{Max}(\text{NO}_2)$, where $\text{Max}(\text{NO}_2)$ is the highest value detected each day. No more than a few profiles are kept each day for the analysis, without significantly decreasing the amplitude of the maximum NO₂ mixing ratios presented above. Figure 17 displays the location of the NO₂ maxima observed with GOMOS in a magnetic polar representation. The magnetic north pole is located at the center of the plot and the latitude decreases as we move away from this point; the parameter measuring the angular position is the magnetic local time (MLT).

[33] MLT at a given location is determined by the angle subtended at the geomagnetic axis between the geomagnetic midnight meridian and the meridian that passes through the location. The geomagnetic meridian containing the subsolar point defines the geomagnetic local noon while the opposite meridian defines the geomagnetic midnight. In Figure 17, noon (12 MLT) is on the top, midnight (00 MLT) is at the

bottom, dawn (06 MLT) is on the right-hand side and dusk (18 MLT) is on the left-hand side. The dash-dotted circles represent the magnetic latitudes 60°, 70°, and 80°. Such a representation is well suited for studies on the magnetosphere because of the fixed location of the Sun. The dashed lines represent a mean location of the boundaries of the auroral oval, derived from the Hardy *et al.* [1987] statistical study on auroral electron precipitation.

[34] On this plot, the triangles are the position of the enhancements observed between 17 and 19 January, the circles are those observed between 20 and 22 January and the crosses those observed between 23 and 31 January. We have distinguished these three periods, because of the two coronal mass ejections that occurred on the 20 and 21 January, that then hit the Earth magnetosphere on 22 January, which could more or less bias the analysis.

[35] It appears that 5 of the 8 NO₂ maxima between 17 and 19 January are observed on the borders of the auroral oval, mainly on the dayside. The maxima between 20 and 22 present some consistency with the previous enhancements (11 points over 17 on the day side), as if the polar vortex was starting to transport the enhancement over the polar region, independently of the coronal mass ejection event. The enhancements after 22 January are widely dispersed, possibly as a result of the dynamics of the vortex. Nevertheless, because of the low number of data points considered, one can conclude that this localization effect is just fortuitous.

[36] NO₂ measurements by most satellite instruments are not available during this period of the year in this region; the other ones may not be accurate enough above 50 km for this type of longitude-resolved study that requires individual profiles because their data have always been averaged and/or assimilated in order to detect the enhancement. As a result, information concerning the true location and altitude of the enhancement could be lost or diluted. Thanks to the good accuracy of the GOMOS measurements in the upper stratosphere and lower mesosphere, it is possible to determine the altitude and the location of the NO₂ enhancement, and to follow its rapid evolution. Unambiguously, the GOMOS observations show that the January 2004 enhancement started with small vertical and spatial extent, in contrast to the May–August 2003 enhancement.

[37] It is highly probable that the correlation of the locations of the initial maxima with the auroral oval is fortuitous. On the other hand, if it is not the case, this could indicate that the phenomena responsible for this particular NO_x enhancement could be associated to an unusual particle precipitation event in the auroral and polar region. This highly speculative process could occur on the dayside magnetosphere or at the magnetopause.

6. Conclusion

[38] Four episodes of NO₂ enhancements were observed in 2003–2004 above the Antarctic and Arctic regions. Two were due to auroral electrons process (May–August 2003, November–December 2003) and one to solar proton process (October–November 2003). Thanks to the high accuracy of the GOMOS measurements, the individual profiles can be used to characterize better the spatial distribution of the January 2004 enhancement. We can conclude that it has

a much smaller initial spatial extent than the other events. About 1 ppmv of NO₂ and its associated effect on ozone were detected at the altitude of the enhancement that was then transported downward into the stratosphere. In particular, ozone was almost totally destroyed in mid-February at about 50 km. The enhancements tend to be found on the day side of the auroral event.

[39] The auroral origin and particular dynamical events are a more common explanation of the enhancement, although, in our case, the extent of the enhancement is unusually well localized at the beginning. At this stage, no other convincing and definitive mechanisms can be proposed for this enhancement. Satellite data of the fluxes of energetic electrons and protons should be tentatively analyzed in order to see whether some unusual phenomenon occurred in the magnetosphere and the ionosphere in January 2004. Also, since some particular dynamical conditions must be invoked in order to produce only a small descending tongue of NO₂-rich mesospheric air, some modeling work should be conducted in the future to better document the occurrence and the spatial and temporal evolution of such enhancement.

[40] **Acknowledgments.** The authors want to thank all their colleagues involved in GOMOS validation and in the data analysis for their contributions to the estimation of the accuracy of the GOMOS measurements, and Yvan Orsolini (NILU, Norway) for fruitful discussions.

[41] Zuyin Pu thanks the reviewers for their assistance in evaluating this paper.

References

- Baker, K. B., and S. Wing (1989), A new magnetic coordinate studies at high latitudes, *J. Geophys. Res.*, *94*, 9139–9143, doi:10.1029/JA094iA07p09139.
- Bertaux, J.-L., et al. (2004), First results on GOMOS/ENVISAT, *Adv. Space Res.*, *33*, 1029–1035, doi:10.1016/j.asr.2003.09.037.
- Bhavnani, K. H., and C. A. Hein (1994), An improved algorithm for computing altitude dependent corrected geomagnetic coordinates, *Sci. Rep. PL-TR-94-2310*, Phillips Lab., Bedford, Mass.
- 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.
- Clilverd, M. A., A. Seppälä, C. J. Rodger, N. R. Thomson, and J. Lichtenberger (2007), Temporal variability of the descent of high-altitude NO_x inferred from ionospheric data, *J. Geophys. Res.*, *112*, A09307, doi:10.1029/2006JA012085.
- Fischer, H., and H. Oelhaf (1996), Remote sensing of vertical profiles of atmospheric trace constituents with MIPAS limb-emission spectrometers, *Appl. Opt.*, *35*(16), 2787–2796, doi:10.1364/AO.35.002787.
- Funke, B., M. López-Puertas, S. Gil-López, T. von Clarmann, G. P. Stiller, H. Fischer, and S. Kellmann (2005), Downward transport of upper atmospheric NO_x 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.
- Funke, B., M. López-Puertas, H. Fischer, G. P. Stiller, T. von Clarmann, G. Wetzel, B. Carli, and C. Belotti (2007), Comment on “Origin of the January–April 2004 increase in stratospheric NO₂ observed in northern polar latitudes” by Jean-Baptiste Renard et al., *Geophys. Res. Lett.*, *34*, L07813, doi:10.1029/2006GL027518.
- Hardy, D. A., M. S. Gussenhoven, R. Raistrick, and W. J. McNeil (1987), Statistical and functional representations of the pattern of auroral energy flux, number flux, and conductivity, *J. Geophys. Res.*, *92*, 12,275–12,294, doi:10.1029/JA092iA11p12275.
- Hauchecorne, A., S. Godin, M. Marchand, B. Heese, and C. Souprayen (2002), Quantification of the transport of chemical constituents from the polar vortex to midlatitudes in the lower stratosphere using the high-resolution advection model MIMOSA and effective diffusivity, *J. Geophys. Res.*, *107*(D20), 8289, doi:10.1029/2001JD000491.
- Hauchecorne, A., J.-L. Bertaux, F. Dalaudier, J. M. Russell III, M. G. Mlynarczyk, E. Kyrölä, and D. Fussen (2007), Large increase of NO₂ in the north polar mesosphere in January–February 2004: Evidences 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, doi:10.1016/S0273-1177(03)00590-8.
- López-Puertas, M., B. Funke, S. Gil-López, T. von Clarmann, G. P. Stiller, M. Höpfner, S. Kellmann, H. Fischer, and C. H. Jackman (2005), Observation of NO_x enhancement and ozone depletion in the Northern and Southern Hemispheres after the October–November 2003 solar proton events, *J. Geophys. Res.*, *110*, A09S43, doi:10.1029/2005JA011050.
- López-Puertas, M., B. Funke, T. von Clarmann, H. Fischer, and G. P. Stiller (2007), The stratospheric and mesospheric NO_y in the 2002–2004 polar winters as measured by MIPAS/Envisat, *Space Sci. Rev.*, *125*, 403–416, doi:10.1007/s11214-006-9073-2.
- Manney, G. L., K. Krueger, J. L. Sabutis, S. A. Sena, and S. Pawson (2005), The remarkable 2003–2004 winter and other recent warm winters in the Arctic stratosphere since the late 1990s, *J. Geophys. Res.*, *110*, D04107, doi:10.1029/2004JD005367.
- Natarajan, M., E. E. Remsberg, L. E. Deaver, and J. M. Russell III (2004), Anomalously high levels of NO_x in the polar upper stratosphere during April, 2004: Photochemical consistency of HALOE observation, *Geophys. Res. Lett.*, *31*, L15113, doi:10.1029/2004GL020566.
- Orsolini, Y. J., C. E. Randall, G. L. Manney, and M. Santee (2005), An upper stratospheric layer of enhanced HNO₃ following exceptional solar storms, *Geophys. Res. Lett.*, *32*, L12S01, doi:10.1029/2004GL021588.
- 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 strong upper stratospheric Arctic vortex, *Geophys. Res. Lett.*, *33*, L18811, doi:10.1029/2006GL027160.
- Renard, J.-B., P.-L. Blelly, Q. Bourgeois, M. Chartier, F. Goutail, and Y. J. Orsolini (2006), Origin of the January–April 2004 increase in the stratospheric NO₂ observed in the northern polar latitudes, *Geophys. Res. Lett.*, *33*, L11801, doi:10.1029/2005GL025450.
- Renard, J.-B., et al. (2008), Validation of GOMOS-Envisat vertical profiles of O₃, NO₂, NO₃, and aerosol extinction using balloon-borne instruments, *J. Geophys. Res.*, *113*, A02302, doi:10.1029/2007JA012345.
- Rinsland, C. P., C. Boone, R. Nassar, K. Walker, P. Bernath, J. C. McConnell, and L. Chiou (2005), Atmospheric Chemistry Experiment (ACE) Arctic stratospheric measurements of NO_x during February and March 2004: Impact of intense solar flares, *Geophys. Res. Lett.*, *32*, L16S05, doi:10.1029/2005GL022425.
- Semeniuk, K., J. C. McConnell, and C. H. Jackman (2005), Simulation of the October–November 2003 solar proton events in the CMAM GCM: Comparison with observations, *Geophys. Res. Lett.*, *32*, L15S02, doi:10.1029/2005GL022392.
- 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.
- Seppälä, A., M. A. Cliver, and C. J. Rodger (2007), NO_x enhancements in the middle atmosphere during 2003–2004 polar winter: Relative significance of solar proton events and the aurora as a source, *J. Geophys. Res.*, *112*, D23303, doi:10.1029/2006JD008326.
- 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.
- S. Bekki and A. Hauchecorne, LATMOS, Université Pierre et Marie Curie (Paris 6), Institut Pierre-Simon Laplace, 4 Pl. Jussieu, Boite 102, F-75005 Paris, France.
- G. Berthet and J.-B. Renard, Université d'Orléans, LPC2E, CNRS, 3A Ave. de la Recherche Scientifique, F-45071 Orléans CEDEX 2, France. (jbreward@cns-orleans.fr)
- P.-L. Blelly, CESR, 9 Ave. du Colonel Roche, F-33028 Toulouse CEDEX 4, France.
- Q. Bourgeois, Institute for Atmospheric and Climate Science, ETH Zurich, Universitätstr. 16, CH-8092 Zurich, Switzerland.