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Nitric acid in the stratosphere based on Odin observations from 2001 to 2009 – Part 2: High-altitude polar enhancements

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Abstract. The wintertime abundance of nitric acid (HNO_3) in the polar upper stratosphere displays a strong inter-annual variability, and is known to be strongly influenced by energetic particle precipitation (EPP), primarily by protons during solar proton events (SPEs), but also by precipitating auroral or relativistic electrons. We analyse a multi-year record (August 2001 to April 2009) of middle atmospheric HNO_3 measurements by the Sub-Millimeter Radiometer instrument aboard the Odin satellite, with a focus on the polar upper stratosphere. SMR observations show clear evidence of two different types of polar high-altitude HNO_3 enhancements linked to EPP. In the first type, referred to as direct enhancements by analogy with the EPP/NO_x direct effect, enhanced HNO_3 mixing ratios are observed for a short period (1 week) after a SPE, upwards of a level typically in the mid-stratosphere. In a second type, referred to as indirect enhancements by analogy with the EPP/NO_x indirect effect, the descent of mesospheric air triggers a stronger and longer-lasting enhancement. Each of the three major SPEs that occurred during the Northern Hemisphere autumn or winter, in November 2001, October–November 2003 and January 2005, are observed to lead to both direct and indirect HNO_3 enhancements. On the other hand, indirect enhancements occur recurrently in winter, are stronger in the Southern Hemisphere, and are influenced by EPP at higher altitudes.

1 Introduction

Nitric acid (HNO_3) is a key minor constituent of the middle atmosphere, part of the odd nitrogen family (NO_y), and a reservoir for the active nitrogen species (NO_x), which provide a major ozone loss catalytic cycle in the middle and upper stratosphere. Stratospheric HNO_3 has been observed by means of ground-based, balloon, aircraft and satellite instrumentation. The most complete dataset to date has been provided by the Microwave Limb Sounder (MLS) instrument aboard Upper Atmosphere Research Satellite (Santee et al., 2004), albeit not in the upper stratosphere. Since July 2004, the EOS/MLS instrument also makes global observations of HNO_3 that cover the mid and upper stratosphere (Santee et al., 2007). The infrared Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) aboard the ENVISAT satellite has also been measuring HNO_3 since July 2002 (Stiller et al., 2005), but following an interruption in 2004, observations have not been continuous.

The "Odin Sub-Millimetre Radiometer" (SMR) instrument provides a continous record of global HNO_3 observations every third day since summer 2001. We present results from a multi-year record (August 2001 to April 2009) in a two-part article. Urban et al. (2009) describes the characteristics of the satellite retrievals and the climatology and variability in the lower stratosphere, neither of which are repeated here. Here, we focus on the polar upper stratosphere-lower mesosphere. Enhanced layers of HNO_3 are commonly observed in winter at high altitudes in the polar regions, as revealed by ground-based (de Zafra and Smyshlaev, 2001) or satellite (Austin et al., 1986; Kawa et al., 1995; Lopez-Puertas et al., 2005b, hereafter LP05; Orsolini et al., 2005a, hereafter OR05; Stiller et al., 2005) observations. Exceptionally strong enhancements have been shown in connection with energetic particle precipitation (EPP) events or anomalous descent of mesospheric air, both of which provide an enhanced source of stratospheric NO_x. However, they have not been previously documented over so many years by a single satellite instrument. The aim of this paper is to show the high inter-annual variability of high-altitude HNO_3 polar enhancements, especially in the aftermath of major solar proton events (SPEs).

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2 Polar HNO_3 enhancements from 2001 to 2009

In the polar upper stratosphere in winter, HNO_3 enhanced layers appeared recurrently in the Odin/SMR observation period considered here (2001–2009). In Fig. 1, the zonal-mean zonal wind at 1 hPa and latitudes of 60° N or 60° S (top), the normalised solar radio flux at 10.7 cm (F10.7 index) and the daily A-p index of geomagnetic activity (middle), are shown to document the dynamical conditions in the polar upper stratosphere and the magnitude of solar-terrestrial coupling during that period. Winds are derived from operational analyses from the European Centre for Medium-Range Forecasting (ECMWF). Normalised intensities of GOES proton flux measurements are shown as indicators of SPEs (Fig. 1, middle), with Class-X SPEs highlighted. The period corresponds to the declining phase of the solar cycle as seen in the F10.7 index, and numerous, intense SPEs were observed (see also Jackman et al., 2008). SMR observations of HNO_3 at a potential temperature level of 1600 K and at latitudes of 60° N or 60° S in Fig. 1 (bottom), or at 1400 K as a function of equivalent latitude in Fig. 2, reveal that, while recurrent, the amplitude of these winter polar enhancements varies widely from year to year in both hemispheres. The figures can be examined together with Figs. 2, 3 in Urban et al. (2009) which show the descent of the anomalies from the upper stratosphere. The largest enhancements in the NH follow the very strong SPEs in November 2001, in October–November 2003 (Halloween storms), and in January 2005. The SH also shows inter-annual variations, with the largest enhancement occurring in austral winter 2003.

Individual winters in each hemisphere from 2001 to 2007 are presented in Figs. 3 and 4, as time versus potential temperature cross-sections of HNO_3 mixing ratio as well as mixing ratio anomalies from the winter mean, averaged over equivalent latitudes poleward of 70°. More recent winters have not been characterised by strong enhancements and are not shown. Occurrences of major Class-X SPEs are shown by pink circles with vertical lines. Enhancements appear recurrently in winter as air descending from the mesosphere always provides some amount of NO_x and HNO_3 conversion (de Zafra and Smyshlaev, 2001). Anomalously elevated stratospheric NO_x abundances can hence arise from this downward transport, if EPP phenomena such as relativistic electron precipitation, or lower energy electron precipitation from auroral activity are strong or persistent, or SPEs occur. This is refered to as the EPP/NO_x indirect effect (e.g., Randall et al., 2006). The efficiency of the mesosphere-to-stratosphere transport depends upon meteorological conditions, which are quite variable, especially in the Northern Hemisphere. Stratospheric NO_x abundances can also be amplified in-situ by most energetic EPP events, such as SPEs. This is refered to as the EPP/NO_x direct effect (e.g., Randall et al., 2006). EPP events have occasionally led to upper-stratospheric NO_2 abundances over a hundred ppb (Callis and Lambeth, 1998; OR05; LP05).

3 Direct and indirect HNO_3 enhancements following major SPEs

SMR observations show clear evidence of two different types of HNO_3 enhancements following the 3 major SPEs that occurred during the Northern Hemisphere autumn or winter over the Odin/SMR observation period considered here: November 2001, October–November 2003, and mid-January 2005. In a first type, enhancements are observed for a short period (1 week) upwards of a level typically in the mid-stratosphere, and are analogous to the EPP/NO_x direct effect. In a second type, the descent of mesospheric air triggers a stronger and longer-lasting enhancement, analogous to the EPP/NO_x indirect effect. In both cases though, HNO_3 is mostly created in the stratosphere (unlike the NO_x).

A first example showing both direct and indirect enhancements follows the powerful November 2001 SPE: a short-lived (one week) layer of enhanced HNO_3 appears above 1200 K, extending upward into the upper stratosphere-lower mesosphere (at least 2000 K). It is followed in December by a much stronger and longer-lasting descending enhancement, in fact the strongest anomaly in the SMR record for the NH. Probably the best studied SPEs were associated with the violent "Halloween" solar storms of autumn 2003. Both direct and indirect HNO_3 enhancements were observed for the first time by MIPAS (LP05; OR05). Immediately following the SPE, a short-lived (about 1 week) stratospheric layer of enhanced HNO_3, peaking at 2–2.5 ppb, was observed above 35 km by LP05. Newly reprocessed MIPAS retrievals indicate that the HNO_3 enhancements extend into the upper stratosphere and lower mesosphere (Jackman et al., 2008). In the indirect enhancement, several weeks after the SPE, an anomalous HNO_3-rich layer was first observed at about 45 km (OR05), and intensified considerably while descending confined in vortex air. By mid-January, it had reached 30 km and vortex-averaged HNO_3 abundances were as high as 13–15 ppb, leading to double-peaked high-latitude HNO_3 profiles. Vortex-averaged SMR measurements are in good agreement with those of MIPAS cited above. In individual SMR profiles, mixing ratios were as high as 7 ppb at 40–45 km in early November 2004. A weaker direct HNO_3 enhancement was also observed by MIPAS in the Southern Hemisphere following the SPEs (LP05). The corresponding enhancement is SMR is very weak, in the range of 1–2 ppb at 40 km in individual profiles, not conclusively above the background levels.

Multiple HNO_3 enhancements also appear during the winter 2004/2005. The indirect enhancement is starting early in December, while a direct enhancement coincides with the strong SPE of mid-January 2005, which ranked as number 11 of the last 4 decades (Jackman et al., 2007). However, the high fluxes for the most energetic protons (>100 Mev), supported by calculation of ionisation rates (Verronen et al., 2005; Seppälä et al., 2008) indicate that the SPE penetrated as deep into the stratosphere as the Halloween 2003 event.

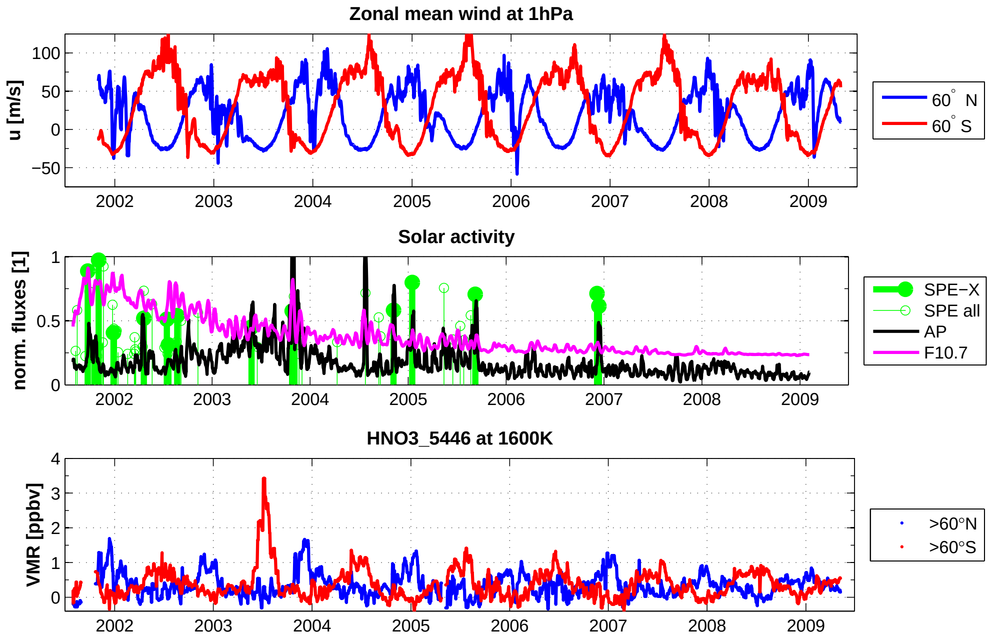

Fig. 1. (top) Zonal-mean zonal winds at 1 hPa and at latitudes of 60° N or 60° S, derived from ECMWF operational analyses. (middle) Normalised solar radio flux at 10.7 cm, A-p index of geomagnetic activity, and normalised proton flux for both major (Class-X) as well as minor SPEs. (bottom) HNO_3 mixing ratio (ppb) at a potential temperature level of 1600 K and at equivalent latitudes poleward of 60° N or 60° S. X-axis is time, from August 2001 to April 2009.

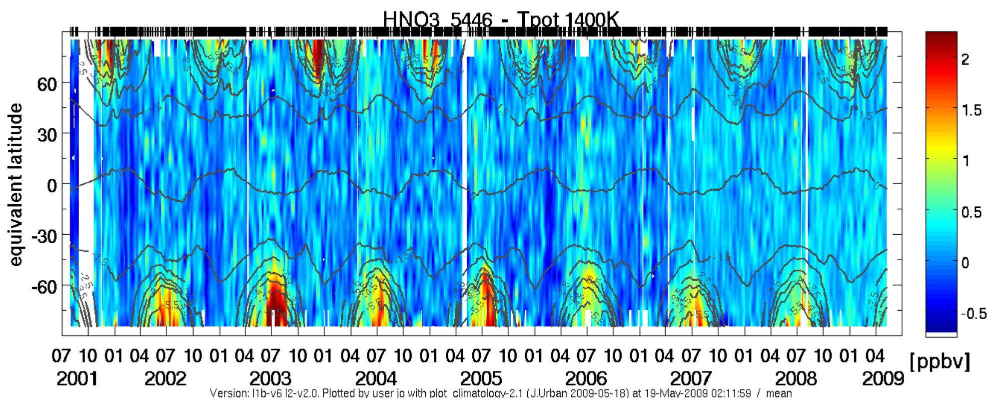

Fig. 2. Time vs. equivalent latitude HNO_3 mixing (ppb) from August 2001 to April 2009, at a potential temperature of 1400 K (near 40 km). Tick marks indicate the beginning of months.

The SMR observations indeed indicate HNO_3 enhancements reaching lower levels, about 1000 K, than during the Halloween 2003 event, and being in the 10–15 ppb range at 30 km in early January. High carbon monoxide (CO) abundance can also be used to infer the descent of lower mesospheric air into the stratosphere, and vortex-averaged MLS observations already show a separated peak in CO at the 1400 K by mid-December (Manney et al., 2007). Hence the SPE, and the direct enhancement, occurred after the winter descent, and the indirect enhancement, had already started.

The above-mentioned three cases are the clearest cases in the Northern Hemisphere, when both direct and indirect HNO_3 enhancements are observed. In the autumn 2004, a much weaker, short-lived enhancement can also be seen after a SPE in November, extending upward of 1500 K. The two SPEs of December 2006 gave rise to a direct enhancement with a maximum near 1300 K–1500 K, but it was weak and did not extend upwards as in the three clearly defined cases mentioned above. The indirect enhancement maximised in mid-January 2007.

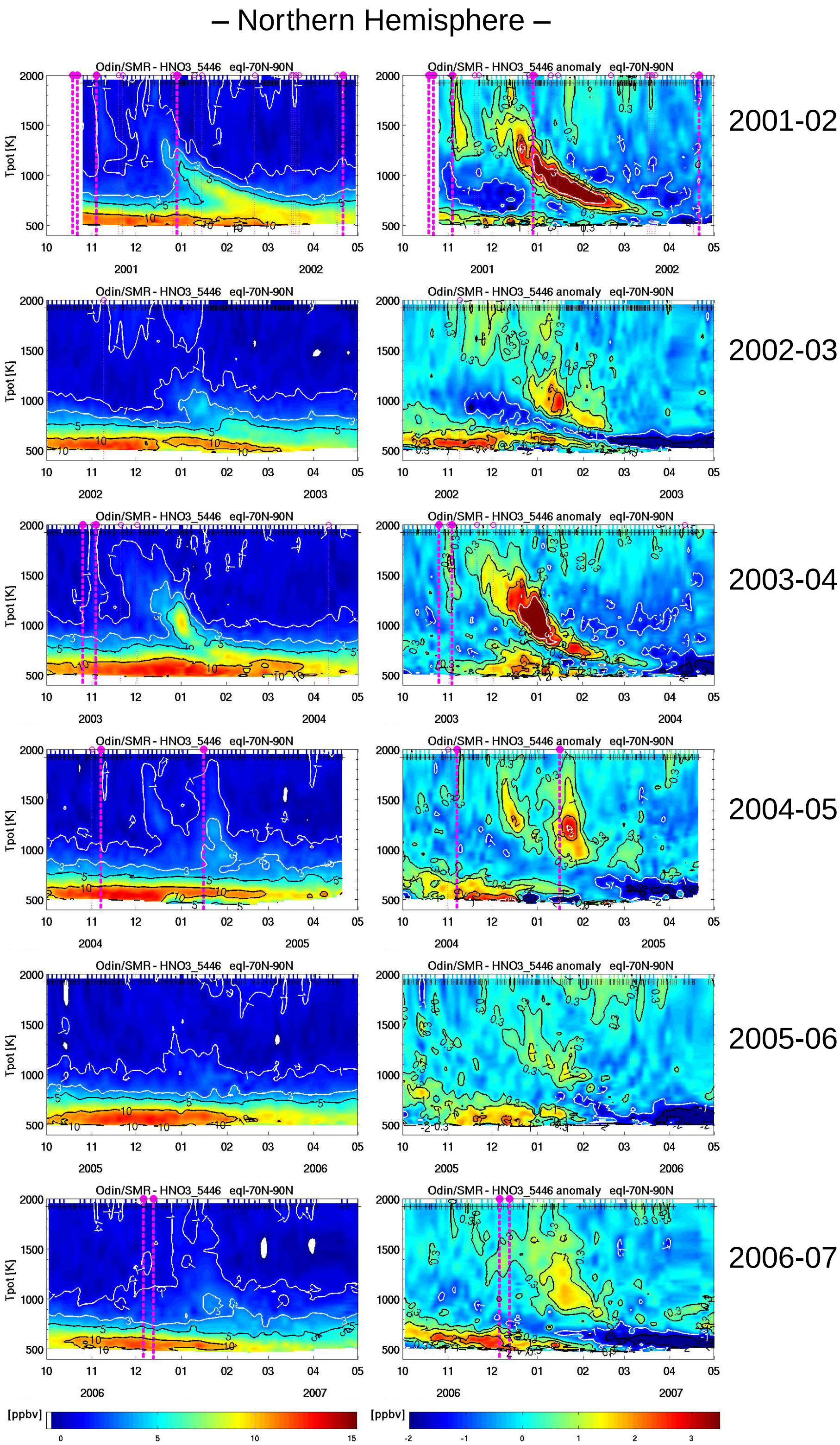

Fig. 3. Time vs. potential temperature evolution of vortex-averaged (equivalent latitudes 70° N–90° N) HNO_3 mixing ratio (ppb) (left column) and deviations from the winter mean (i.e. anomalies, right column), during NH winters 2001/2002 through 2006/2007. X-axis is labelled with months. Major, class X SPEs are indicated by pink circles with vertical lines.

4 Discussion

The time development of the HNO_3 anomalies involves the interplay of middle atmospheric dynamics and chemistry, and solar-terrestrial coupling. Two chemical pathways have been suggested in LP05 to explain the direct HNO_3 enhancements: either gas phase reactions of NO_2 with OH, both enhanced by the SPE, or else ion chemistry (Solomon et al., 1981). A recent ion chemistry model study by Verronen et al. (2008) indicates that abundances of HNO_3 observed by MIPAS following the Halloween 2003 SPEs are largely consistent with production above 35km resulting from ion-ion recombination between ion clusters. They showed that the latter mechanism dominated both the gas phase formation and the heterogeneous conversions of N_2O_5 into HNO_3 upon hydrated ion clusters. Whether the short duration of the enhancement can be solely explained by chemistry or by a combination of transport and chemistry, requires further modelling studies.

Following earlier suggestions (Bohringer et al., 1993; Kawa et al., 1995), de Zafra and Smyshlaev (2001) estimated that such N_2O_5 heterogeneous conversions upon hydrated ion clusters were responsible for the enhancements they observed in ground-based microwave measurements at the South Pole. This is the likely mechanism for the indirect enhancements, although this conclusion awaits quantitative model results. Sulphate aerosols were also suggested to play a role below 35 km (Bekki et al., 1995). The indirect enhancements require a large downward flux of NO_2 to generate N_2O_5, a process favoured by vortex confinement, hence explaining the stronger effect in the SH. The background abundance of hydrated ion clusters is thought to be generated by galactic cosmic rays. How EPP influences that background abundance is an open question.

While the HNO_3 enhancements share some characteristics of the NO_x enhancements, such as high-altitude origin, polar confinement and descent, the latter are not always followed by HNO_3 enhancements. An enhanced descent from the mesosphere into the stratosphere well-confined by a strong vortex would be leading to a large EPP/NO_x indirect effect. Three such strong descent events have been observed to occur during the vortex recovery from the mid-winter stratospheric sudden warmings in 2004, 2006 and 2009 (Randall et al., 2005, 2006, and reference therein; Manney et al., 2009), or as revealed by inspection of zonal-mean zonal winds at 1 hPa in Fig. 1. NO_x enhancements have been shown during the 2004 and 2006 events (see above references). The EPP/NO_x indirect effect did not give rise to large HNO_3 enhancements. This would be consistent with the ion cluster chemistry requiring darkness to build up HNO_3, conditions which are not provided in late winter and spring. In fact, there is only a signature of a weak enhancement during the NO_x descent in March–April 2006 at the highest levels measured by SMR. These NO_x descents in 2004 and 2006 were nevertheless important for the upper stratospheric ozone budget, leading for example to nearly 60% ozone destruction at 45 km in spring 2004 (Natarajan et al., 2004; Randall et al., 2005). Hence, while the short-lived, direct HNO_3 enhancements could be triggered by SPEs in any season, SPEs contribute to indirect enhancements only from late autumn to winter. Hence, the seasonal timing of EPP events largely constrain their impact on HNO_3 enhancements. Another effect of stratospheric warmings is that they could dampen the HNO_3 anomalies by bringing polar air to sunlit regions.

Polar HNO_3 enhancements in the SH also display a strong inter-annual variability (Fig. 4). But, in the SH, the vortex is less variable than in the NH, and the variations in EPP is playing a key role in year-to-year variability of polar NO_x (Randall et al., 2007) or HNO_3 enhancements. The strongest enhancement is seen to occur in austral winter 2003, and was studied by Stiller et al. (2005) using MIPAS data. The vortex-averaged magnitude of about 7 ppbv at 1400 K is in good agreement with MIPAS measurements. Stiller et al. (2005) concluded that the enhancement originated from strong descent of mesospheric air enriched in NO_x by enhanced auroral activity. Indeed, Tanskanen et al. (2005) indicated a high occurrence of magnetic substorms and auroral activity in 2003. It can be noticed in Fig. 1 that the SH vortex was not the strongest in austral winter 2003.

In SMR observations over the SH, we do not observe any clear-cut case showing both direct and indirect enhancements following SPEs. There is however a series of weak short-lived enhancements throughout the austral winter 2002, including a pronounced one following the July 2002 SPE, extending upward of about 1200 K. As shown by the zonal-mean zonal wind in Fig. 1 (top), this winter was characterised by a large dynamical variability that led to the sudden stratospheric warming of late September 2002 (Allen et al., 2003; Orsolini et al., 2005b). As for the January 2005 event in the NH, the direct July 2002 enhancement would have occurred after the winter descent had started. It is not possible to clearly disentangle the effects of dynamical variability and SPEs, without a detailed model study. In particular, it is not clear why only the second SPE in July 2002 led to an upward-extended structure, that is characteristic of direct enhancements.

Inspection of Figs. 3 and 4 (as well as Figs. 2, 3 in Urban et al., 2009) reveals that, in the Northern Hemisphere, the enhancements descending from the upper stratosphere merge with the main layer when the abundance is still high, as clearly seen during the two strongest episodes in 2002 and 2004. In the SH, on the contrary, the descending layer normally reaches the lower stratosphere when the main layer abundance has already decreased considerably due to polar stratospheric cloud formation, during which there is uptake of HNO_3 from the gas phase. A further point to note is that, as the high-altitude layer descends, the mixing ratios increase down to a certain level, which is quite variable, indicating continued production.

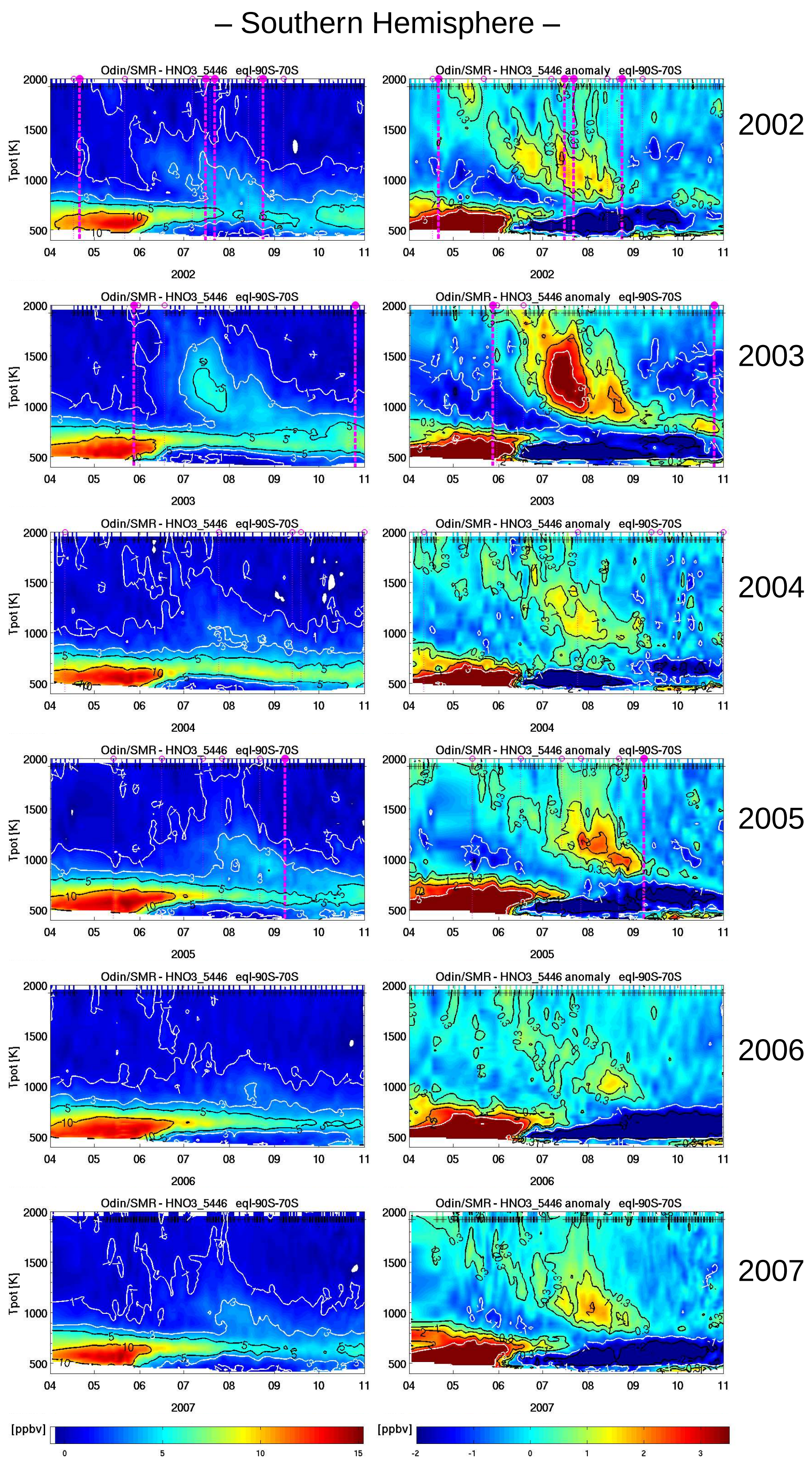

Fig. 4. Same as Fig. 2, for SH (equivalent latitudes 70° S–90° S) winters 2002 through 2007.

5 Summary

The Odin/SMR observations provide for the first time a multi-year record of polar HNO_3 enhancements at high altitudes, and their downward propagation inside the winter polar vortex. Outstanding enhancements are seen during major SPEs or strong mesospheric descent events. Direct enhancements take the form of a short-lived (about 1 week) layer of enriched HNO_3, extending from typically 35 km upwards into the upper stratosphere-lower mesosphere. The indirect enhancements are characterised by slowly-descending, larger anomalies, and are generally stronger in the Southern Hemisphere. Not all SPEs contribute to the indirect enhancements. Their effects depend not only of their intensity and penetration depth, but also on seasonal timing and meteorological conditions. On the other hand, indirect enhancements can occur without the occurrences of SPEs, and are influenced by EPP at higher altitudes. The occurrence of both direct and indirect HNO_3 enhancements following a SPE was first observed by MIPAS (LP05; OR05) following the Halloween solar storms of autumn 2003, and is here confirmed by the SMR data in at least 2 additional cases.

The descending low or high HNO_3 anomalies appear somewhat analogous to the tropical "tape-recorder" effect (Mote et al., 1996), that describes how low-latitude tracer anomalies imprinted at the tropopause level ascend over years, keeping a memory of their initial composition, and giving rise to layered anomalies in the tropical stratosphere. In this case, it is rather acting at high latitudes, and in reverse (propagating downwards) fashion, and on a faster (seasonal) scale: HNO_3 anomalies are imprinted in the upper stratosphere, descending to the lower stratosphere over the course of the winter, and giving rise to a double-layered profile.

Further modelling studies are needed as a step toward implementing appropriate schemes to represent these processes affecting the stratospheric NO_y budget into global chemical transport models.

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