

## The solar proton events in 2012 as observed by MIPAS

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[1] During the solar proton events (SPE) on 23–30 January and 7–15 March 2012, the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) on Envisat monitored atmospheric temperature and composition with global coverage. In the Northern Hemisphere, the January SPE started at the end of a polar stratospheric warming period. The SPE effect is superimposed by large-scale subsidence of mesospheric NO<sub>x</sub>-rich air, which partly masks direct chemical SPE effects. SPE-induced NO<sub>x</sub> increases by 5, 20, 50, and 100 ppbv at altitudes of 50, 57, 60, and 70 km, respectively, are observed during the January SPE and those by 2, 5, 10, 20, 30, and 35 ppbv at altitudes of 47, 50, 53, 60, 63, and 66 km, respectively, during the March SPE. SPE-related ozone loss is clearly observed in the mesosphere, particularly in the tertiary ozone maximum. A sudden short-term HNO<sub>4</sub> increase immediately after the January SPE hints at SPE-triggered HO<sub>x</sub> chemistry. In the Southern Hemisphere, a large NO<sub>x</sub> response is observed (increases by 2, 5, 10, 20, and 30 ppbv at 52, 56, 59, 63, and 70 km in January and by 2, 5, 10, 20, 30, and 35 ppbv at 47, 50, 53, 60, 63, and 66 km in March), while the effect on other species seems much less pronounced than in the Northern Hemisphere. SPE-related destruction of mesospheric ozone in the Southern Hemisphere was much more pronounced after the March SPE than the January SPE but in both cases, ozone recovered within about a day.

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### 1. Introduction

[2] During coronal mass ejections of the sun, energetic particles are emitted which can reach the Earth and affect the chemistry of the atmosphere. Since the Earth's magnetic field guides the particles to the poles, where they ionize the atmosphere down to stratospheric altitudes, polar HO<sub>x</sub>, nitrogen, and chlorine chemistry can be affected, which in consequence can affect ozone concentrations [Funke *et al.*, 2010,

and references therein]. In this paper, we discuss trace gas measurements by the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) in the period from January to April 2012, in which two major solar proton events (SPE) happened.

### 2. MIPAS

[3] MIPAS is a limb emission Fourier transform spectrometer designed to globally measure temperature and trace gas profiles [Fischer *et al.* 2008]. It was operated from July 2002 to April 2012. In this paper, we use temperature and trace gas profiles provided by the KIT/IAA research data processor [von Clarmann *et al.*, 2003]. Analysis of data recorded after 2004 when the MIPAS observation mode was changed towards reduced spectral resolution (RR) is described in von Clarmann *et al.* [2009]. Retrieval of gases where non-local thermodynamic equilibrium has to be considered (namely, CO, NO, and NO<sub>2</sub>) is discussed by Funke *et al.* [2005, 2009]. Since publication of these papers, improved calibrated spectra have been provided by ESA, and several updates of the retrieval strategy have been implemented. Besides minor technical upgrades, the major improvement with respect to former data versions was achieved by jointly fitting H<sub>2</sub>O and O<sub>3</sub> mixing ratios simultaneously with the temperature and elevation pointing retrieval. This avoids mapping of inadequate climatological assumptions of H<sub>2</sub>O and O<sub>3</sub> concentrations on temperature retrievals and propagation to subsequently retrieved species. Further, the treatment of separation of background continuum emission from calibration correction has been improved for the methane retrieval. The retrieval of HNO<sub>4</sub> has been developed by Stiller *et al.* [2007] and later adapted to MIPAS RR measurements. All data used here are version V5\_\*\_221, except for methane and N<sub>2</sub>O, whose version numbers are V5\_CH4\_223 and V5\_N2O\_223, respectively, and were retrieved from MIPAS spectra measured in the nominal observation mode.

### 3. Solar Activity

[4] In the increasing phase of Solar cycle 24, sunspot AR1429 was the source of several coronal mass ejections. On 23 January 2012, protons hit the Earth's atmosphere and caused ionization down to 4 hPa, and on 27 January ionization took place even at the 10 hPa level (Figure S1). An X5 class eruption from sunspot AR1429 caused another SPE in the 7–15 March 2012 time period. The proton flux data from GOES 13 provided by the National Oceanic and Atmospheric Administration Space Weather Prediction Center (see [www.swpc.noaa.gov/ftpmenu/lists/particle.html](http://www.swpc.noaa.gov/ftpmenu/lists/particle.html)) were used to compute the hourly averaged ionization rates using the methodology discussed in Jackman *et al.* [1980].

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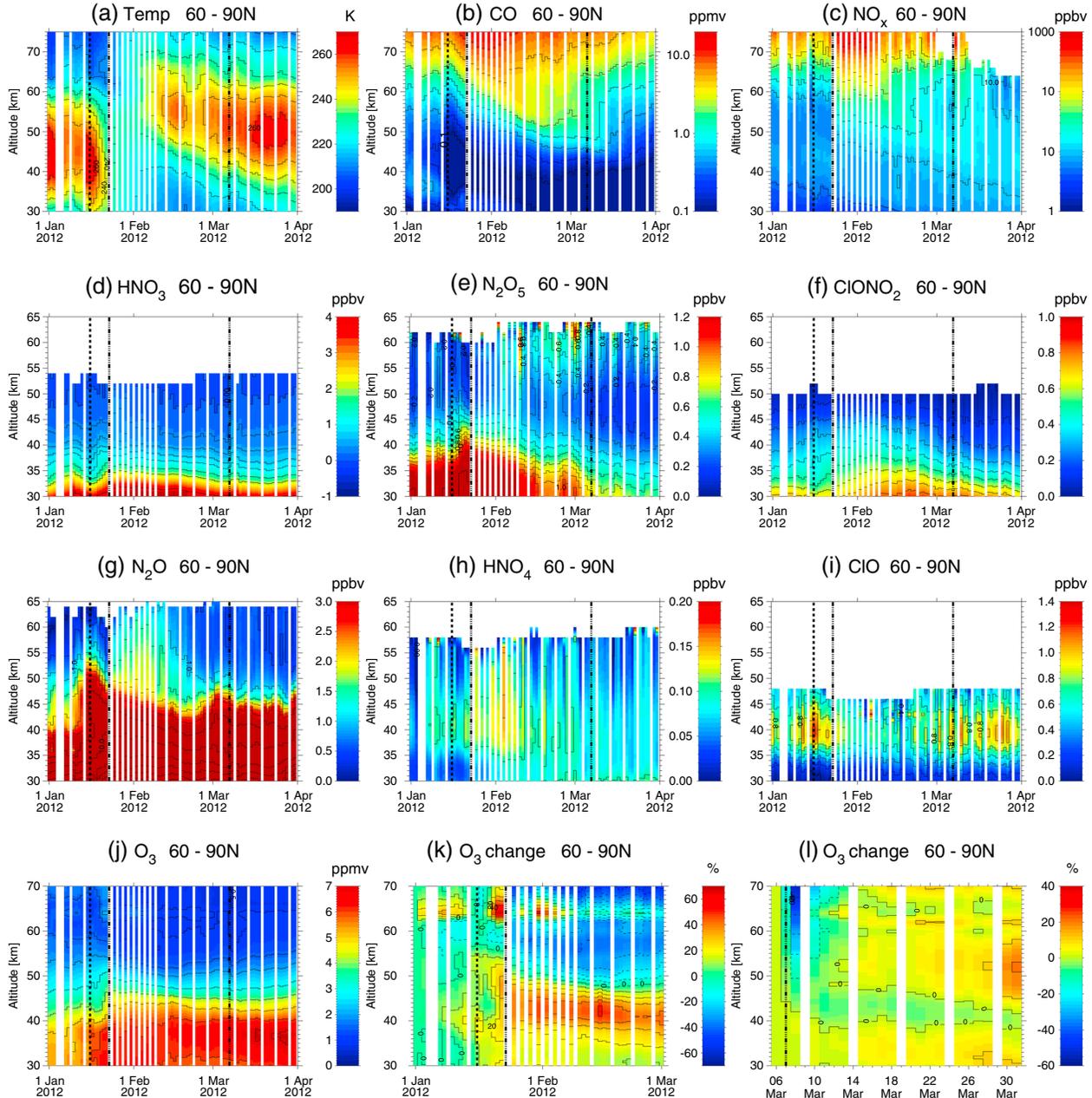
Especially from 7–9 March, the energy spectrum was fairly hard with a significant flux of high energy (>30 MeV) protons, which penetrated deep into the atmosphere, causing ionization down to 20 hPa.

#### 4. The Northern Hemisphere

##### 4.1. Meteorology

[5] Over the northern polar cap, sudden stratospheric warming (SSW) events along with mesospheric cooling occurred during January (Figure 1a). In the middle stratosphere, remnants of subsided mesospheric CO-rich air continue their

downward movement and indicate continued polar winter conditions, while in the upper stratosphere, lower CO mixing ratios hint at air of sub-polar stratospheric origin mixed towards polar latitudes during the sudden warming event (Figure 1b). From late January on, an elevated stratopause was observed, and mesospheric air again subsided into the stratosphere until the final warming in early March. The temporal co-incidence of the warming events with the SPEs makes the assignment of the atmospheric response to the SPE versus dynamical effects convoluted. In this work, we aim to separate the chemical from the dynamical effects on the composition changes.



**Figure 1.** Temporal evolution of northern polar cap mean temperature and trace gas mixing ratios as a function of altitude. Weighting by the cosine of latitude has been applied to account for the area represented by each data point. The black dotted vertical line marks the onset of the stratospheric warming, while the dashed-dotted lines are the SPEs. White lines represent data gaps due to non-availability of a sufficient number of nominal mode measurements. The changes in (k) and (l) refer to the profile of the first day plotted.

## 4.2. Nitrogen Chemistry

[6] A large abrupt increase of NO and NO<sub>2</sub> is observed around the date of the January SPE. In Figures 1c and S2a, we show [NO<sub>x</sub>] = [NO] + [NO<sub>2</sub>] rather than its components in order to avoid masking by diurnal cycle or diurnal sampling effects. Prior to the SSW, nominal descent led to an increase in NO<sub>x</sub> at 60–70 km. During the SSW, NO<sub>x</sub> at these altitudes decreased due to mixing of lower latitude air into the vortex. During the SPE, however, an abrupt additional NO<sub>x</sub> increase by 5, 20, 50, and 100 ppbv at altitudes of 50, 57, 60, and 70 km is observed which is not accompanied by a corresponding increase of CO and thus is attributed to the SPE (Figure 1b). During February and early March, NO<sub>x</sub>-rich air subsides further down into the stratosphere, similar as after the SPE in 2003 [López-Puertas *et al.*, 2005a]. After the SPE on 7–8 March, another abrupt increase of NO<sub>x</sub> by 2, 5, 10, 20, 30, and 35 ppbv at altitudes of 47, 50, 53, 60, 63, and 66 km is observed (Figure S3a).

[7] The seasonal build-up of HNO<sub>3</sub> is interrupted by a decrease due to the stratospheric warming and recovers after 17 January. On top of this increase, abrupt enhancements of HNO<sub>3</sub> mixing ratios are observed above approximately 45 km during the January SPE and above 35 km during the March SPE (Figures 1d, S2b, and 3b). Such instantaneous HNO<sub>3</sub> increase has already been observed during the SPE in 2003 [López-Puertas *et al.* 2005b] and is attributed to a recombination reaction of H<sup>+</sup> and NO<sub>3</sub><sup>-</sup> ions on water cluster ions as proposed by Verronen *et al.* [2008]. Beyond this, the conversion of N<sub>2</sub>O<sub>5</sub> to HNO<sub>3</sub> on protonated water clusters as suggested by de Zafra and Smyshlyaev [2001] may play a role, particularly at 30–35 km where N<sub>2</sub>O<sub>5</sub> is observed to slightly decrease (Figure 1e) and where no excess NO<sub>x</sub> is observed. This reaction could be accelerated by the SPE-induced excess availability of protonated water clusters.

[8] In February and early March, a small part (less than 10%) of the enhanced NO<sub>x</sub> above 50 km is transformed into N<sub>2</sub>O<sub>5</sub> (Figure 1e); while mesospheric N<sub>2</sub>O<sub>5</sub> has to our knowledge never been reported before, this feature cannot be assigned to the SPE because it is observed by MIPAS in every polar spring in both hemispheres (Stiller *et al.*, 2013, paper in preparation). The N<sub>2</sub>O<sub>5</sub> seasonal development below 45 km appears to be linked to vortex dynamics rather than the SPEs, since its spatio-temporal patterns are highly correlated to those of the temperature development.

[9] ClONO<sub>2</sub> (Figure 1f) shows a weak positive response to the January SPE in the upper stratosphere. The patterns of the long-term development of ClONO<sub>2</sub>, however, resemble those of temperature and thus seem to be driven mainly by vortex dynamics.

[10] In agreement with results by Funke *et al.* [2008a] for the SPE 2003, local SPE-related N<sub>2</sub>O formation is observed after the January SPE around 40 km altitude (Figures 1g and S4a). These changes in N<sub>2</sub>O are due to reaction of NO<sub>2</sub> with N(<sup>4</sup>S). The increase during the last days of January and for the first half of February, however, is not a direct SPE effect but seems to be due to energetic electron precipitation as suggested by Funke *et al.* [2008b].

## 4.3. HO<sub>x</sub> Chemistry

[11] A sudden HNO<sub>4</sub> increase is observed in the upper stratosphere on 25 January (Figures 1h and S4b). Since HNO<sub>4</sub> formation depends linearly both on HO<sub>2</sub> and NO<sub>2</sub>

concentrations, the increase of HNO<sub>4</sub> can be explained by the increase of any of these reactants. However, the relative increase of HNO<sub>4</sub> after the January SPE exceeds that of NO<sub>x</sub>. This indicates accelerated HO<sub>x</sub> chemistry during the proton forcing and confirms empirical conclusions by Jackman *et al.* [2001], von Clarmann *et al.* [2005], and Verronen *et al.* [2006]. A much weaker response to the March SPE was observed (Figures 1h and S5). Photolysis leads to a much smaller HNO<sub>4</sub> equilibrium concentration under sunlit conditions. The enhanced values in early February are the apex of the climatological late winter maximum which is regularly observed by MIPAS and which is attributed to excess NO<sub>x</sub> availability.

[12] Although one might expect CO removal with OH after the SPEs as reported by Funke *et al.* [2011] for the SPE in 2003, the tiny CO deficit between 50 and 60 km altitude on 25 January has dynamical origin: Nearly perfect anti-correlation with CH<sub>4</sub> (Figure S6) reveals that on this particular day, air masses have been sounded which had experienced less subsidence.

## 4.4. Chlorine Chemistry

[13] After the SPE 2003, ClO averaged over the polar cap, whose major parts were still illuminated, increased after the proton forcing [von Clarmann *et al.*, 2005], except for the dark regions poleward of 70°N, where ClO decrease was observed [Funke *et al.* 2011]. In agreement with the illumination dependence found for the SPE 2003, Damiani *et al.* [2012] found a negative response of ClO in the dark polar cap stratosphere to the January 2005 SPE. In 2012, however, no significant ClO change was observed which would exceed the observed day-to-day variability (Figure 1i). At 2 hPa, which is the region of chlorine activation, we find ionization rates about a factor of four larger during the January 2005 SPE than during that in January 2012 (both solstice). The ratio between ionization rates in October 2003 and March 2012 (both closer to equinox) is similar to that of January 2005 to January 2012. ClO responses in both January 2005 and October 2003 were in the order of 150 pptv (negative in polar night, positive in illuminated regions). Hence, assuming that the chlorine responses scale linearly with ionization rates, the expected effect would be below 50 pptv, which is below the detection limit and also lower than background variability.

[14] The ClO temporal maximum during the stratospheric warming, the broad minimum in February, and the high values in March coincide with the temporal minimum, the broad maximum, and the low concentrations of ClONO<sub>2</sub> in the respective periods (Figure 1f). These seasonal changes in the ClO/ClONO<sub>2</sub> partitioning reflect changes in temperature and illumination: Warmer temperatures accelerate ClONO<sub>2</sub> formation from ClO and NO<sub>2</sub>, while increased availability of sunlight in spring enables photolysis of ClONO<sub>2</sub>.

## 4.5. Ozone

[15] Near 65 km altitude, the formation of the tertiary ozone maximum [Marsh *et al.* 2001] is clearly seen in Figures 1j and 1k). After an interruption during the SSW, this maximum is reformed and reaches largest values on January 20, shortly after the beginning of the rapid descent [Smith *et al.* 2009]. Two days after the January SPE, about a third of the mesospheric ozone is destroyed. Destruction

of the tertiary ozone maximum has already been observed by GOMOS during/after the SPE 2005 [Seppälä *et al.* 2006]. This ozone loss coincides with the phase of excess  $\text{HNO}_4$  and is thus attributed to SPE-triggered  $\text{HO}_x$  chemistry. Mesospheric ozone concentrations, however, recover within a few days but with increasing illumination the tertiary ozone maximum fades, confirming that it is a polar night phenomenon. Immediately after the March SPE, ozone loss by up to 60% is observed in the mesosphere, and again ozone recovers within a few days (Figure 11). Around 55 km,  $\text{NO}_x$ -related ozone loss starts a couple of days after the January SPE; the pattern of ozone-depleted air coincides nicely in latitude and time with the  $\text{NO}_x$  enhancements.

## 5. The Southern Hemisphere

[16] The southern polar stratosphere and mesosphere from January to April 2012 is characterized by continuous cooling of the stratopause without any peculiarities (Figure 2a). Neither a significant response in  $\text{HO}_x$  nor in the chlorine species is observed in the Southern Hemisphere (SH, not shown). However, a pronounced immediate response of  $\text{NO}_x$  to both SPEs is observed down to the stratopause (Figure 2b). Since there is no subsidence of air over the

southern polar cap, and since the  $\text{NO}_x$  increase is observed simultaneously over a wide range of altitudes, it is attributed to SPE-induced in-situ production. Increases of 2, 5, 10, 20, and 30 ppbv at altitudes of 52, 56, 59, 63, and 70 km are observed during the January SPE, and 2, 5, 10, 20, 30, and 35 ppbv at altitudes of 47, 50, 53, 60, 63, and 66 km after the March SPE. As in the Northern Hemisphere (NH), the  $\text{NO}_x$  increase persists several weeks. In January, these increases are weaker and shorter lived than in the NH which is attributed to photochemical losses of NO in the SH upper stratosphere and above. In March, the  $\text{NO}_x$  enhancements are similar for both hemispheres.

[17]  $\text{HNO}_3$  increased immediately during both SPEs, and similar to the NH, increased values persisted only a few days (not shown). Any response of  $\text{N}_2\text{O}_5$  is masked by large background variability (not shown). A short-term (1–3 days, depending on altitude and date) negative response of ozone is noticeable after both SPEs (Figure 2c). At around 55 km altitude, the seasonal increase of ozone is overcompensated by ozone loss due to the enhanced  $\text{NO}_x$  mixing ratios.

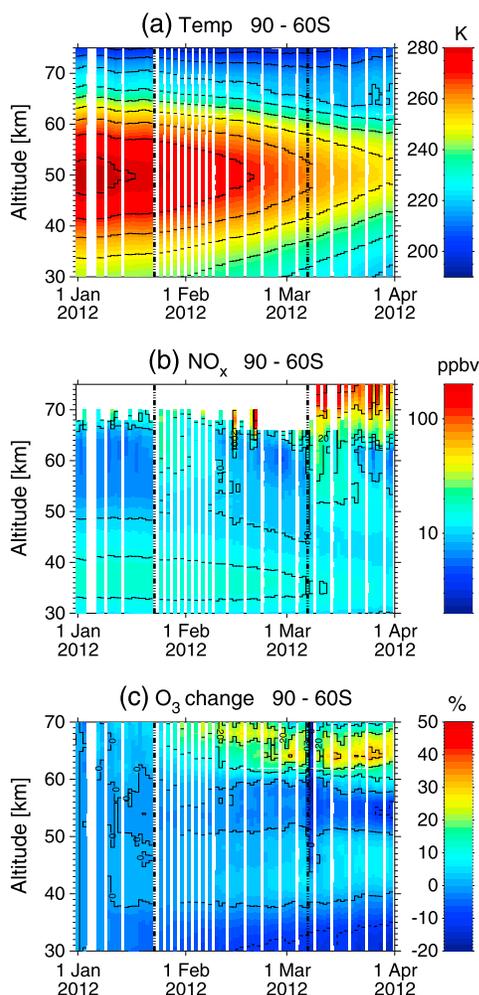
## 6. Conclusion

[18] The impact of the SPEs in January and March 2012 on the middle atmosphere is more pronounced in the NH than in the SH. The hemispheric differences are due to the Arctic vortex confinement of excess  $\text{NO}_x$  to polar night latitudes coupled with the descent of mesospheric air within the vortex. Similar to previous SPEs, the events in 2012 showed a short-term acceleration of  $\text{HO}_x$  chemistry (NH) and a strong  $\text{NO}_x$  enhancement (SH and NH). As inferred by multiplication of ionization rates by 1.25 and integration over altitude and from 60 to 90° in geomagnetic latitude in both hemispheres, the January and March SPE periods produced approximately 1.9 and 2.1 Gigamoles of odd nitrogen ( $\text{NO}_y$ ), respectively. Thus, these SPE periods were among the 12 largest in the past 50 years [see Jackman *et al.*, 2008, Table 1]. These findings corroborate the conclusions drawn from SPEs in 2003 and 2005. The major difference with respect to the 2003 “Halloween” SPE and the SPE in January 2005 is related to chlorine chemistry: While under dark/sunlit conditions ClO was observed to decrease/increase, respectively, in response to these previous SPEs, in 2012 no discernable perturbation of chlorine chemistry could be identified which would exceed sampling and noise-induced day-to-day variability. This is attributed to ionization rates lower by about a factor of four at the relevant altitude. The MIPAS data presented here are available to registered users via <http://www.imk-asf.kit.edu/english/308.php>.

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**Figure 2.** Temporal evolution of temperature and trace gases as a function of altitude for the southern polar cap. The changes in Figure 2c refer to the profile of 1 January.

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