

1 **We thank the anonymous referees for their hints to improve the paper. The referees comments**  
2 **are repeated in normal letters while our response is highlighted in bold. Note that the page**  
3 **and line numbers refer to this document.**

4  
5 **Response to Referee #1**

6 1) p. 3, line 16: Change “life time” to “lifetime”

7 **Done.**

8  
9 2) p. 7, lines 32-33: “Additionally, measurements in the relevant mode are only performed every  
10 day, consequently only covering about one day out of three before April 2007 and every other day  
11 afterwards.”

12 Comment: This sentence is puzzling to me. How can the measurements be performed every day, but  
13 yet cover about one day out of three before April 2007 and every other day afterwards? Am I  
14 missing something?

15 **Done was corrected and the sentence was changed to: “Consequently, measurements in the**  
16 **relevant mode are roughly performed one day out of three before April 2007 and every other**  
17 **day afterwards.”**

18

19 3) p. 14, line 15: Change “state” to “stage”

20 **Done.**

21

22 4) Figures 3, 5, 6, and 7: These figures have areas that are “shaded,” which indicate significance  
23 levels between 95% and 99%. I have noticed some of these “shaded” areas, but find them very  
24 difficult to see. Would it be possible to enhance the “shading,” so these areas are more prominent?

25 **Done. We increased the “shading” and also changed the “shading colour” from black to white,**  
26 **when the respective background colour is bluish. Consequently we added “in black or white”**  
27 **in caption 3 (p. 25, line 8).**

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1 **Energetic particle induced intra-seasonal variability of ozone**  
2 **inside the Antarctic polar vortex observed in satellite data**

3

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13

14 **Abstract**

15 Measurements from 2002 – 2011 by three independent satellite instruments, namely MIPAS,  
16 SABER, and SMR on board the ENVISAT, TIMED, and Odin satellites are used to investigate the  
17 intra-seasonal variability of stratospheric and mesospheric O<sub>3</sub> volume mixing ratio (vmr) inside the  
18 Antarctic polar vortex due to solar and geomagnetic activity. In this study, we individually analysed  
19 the relative O<sub>3</sub> vmr variations between maximum and minimum conditions of a number of solar and  
20 geomagnetic indices (F10.7 cm solar radio flux, Ap index,  $\geq 2$  MeV electron flux). The indices are  
21 26-day averages centred at 1 April, 1 May, and 1 June while O<sub>3</sub> is based on 26-day running means  
22 from 1 April - 1 November at altitudes from 20 - 70 km. During solar quiet time from 2005 – 2010,  
23 the composite of all three instruments reveals an apparent negative O<sub>3</sub> signal associated to the  
24 geomagnetic activity (Ap index) around 1 April, on average reaching amplitudes between -5% and -  
25 10% of the respective O<sub>3</sub> background. The O<sub>3</sub> response exceeds the significance level of 95% and  
26 propagates downwards throughout the polar winter from the stratopause down to ~25 km. These  
27 observed results are in good qualitative agreement with the O<sub>3</sub> vmr pattern simulated with a three-  
28 dimensional chemistry-transport model, which includes particle impact ionisation.

29

# 1 Introduction

2 Energetic particles (~keV - ~MeV), mainly originating from the sun but also from the Earth's  
3 magnetospheric radiation belts and the aurora region, penetrate the atmosphere down to  
4 mesospheric and stratospheric regions, depending on their energy. The particles are guided by the  
5 Earth's magnetic field lines and therefore mostly precipitate at auroral and radiation belt areas (~55°  
6 - 70° geomagnetic latitudes), depositing energy and directly influencing the chemical composition  
7 of the stratosphere and mesosphere. Due to the air compounds, precipitating particles mainly  
8 produce large abundances of  $O_2^+$  as well as  $N(^2D)$  and  $N_2^+$ .  $N(^2D)$  and  $N_2^+$  lead to increased  
9 concentrations of odd nitrogen ( $NO_x = N + NO + NO_2$ ) through a number of reactions, including  
10 dissociative recombination of  $N_2^+$  and ion-neutral chemistry with species of the oxygen family (e.g.  
11 Rusch et al., 1981). Additionally,  $O_2^+$  and water vapour initialise chain reactions associated with  
12 water cluster ion formation and accompanied recombination reactions, which eventually lead to the  
13 production of odd hydrogen ( $HO_x = H + OH + HO_2$ ; e.g. Solomon et al., 1981).

14 Both  $HO_x$  and  $NO_x$  play an important role in destroying  $O_3$  in the mesosphere and stratosphere (e.g.  
15 Lary, 1997). However,  $HO_x$  is short-lived (~seconds – hours) and therefore more important near its  
16 source region in the mesosphere, while  $NO_x$  has a relatively long lifetime (~days - months), at least  
17 during night-time conditions. Consequently,  $NO_x$  can be transported downwards inside the polar  
18 vortex (e.g. Solomon et al., 1982) from the upper mesosphere/lower thermosphere down to the  
19 stratosphere, resulting in stratospheric  $O_3$  depletion through catalytic chemical reactions in  
20 combination with solar radiation. Thus, energetic particle precipitation (EPP) indirectly affects  $O_3$   
21 during polar winter. Since  $O_3$  is the major radiative heating source in the stratosphere, variations of  
22 this gas will also influence the stratospheric temperature field and eventually lead to altered  
23 atmospheric dynamics. However, the atmospheric response to EPP is not fully understood so far.  
24 The current knowledge is discussed in more detail by Sinnhuber et al. (2012).

25 Observations of the EPP indirect effect on stratospheric polar  $O_3$  are relatively rare, at least  
26 compared to other latitudes, due to a lack of long-term  $O_3$  measurements in these regions. However,  
27 a hint for this mechanism was presented by Randall et al. (1998) which analysed the Polar Ozone  
28 and Aerosol Measurement instrument data, revealing a close anticorrelation between  $NO_2$  and  $O_3$   
29 mixing ratios in winter/spring from 1994 – 1996 in the Antarctic stratosphere (~25 – 35 km). They  
30 suggested that the relationship cannot originate from downwards transported  $O_3$ -deficient air but is  
31 due to photochemical destruction of  $O_3$  by  $NO_2$ . Further observations from several satellite  
32 instruments from 1992 – 2005 show that the stratospheric  $NO_x$  enhancement in the Southern  
33 Hemisphere is caused by EPP (Randall et al., 2007). More recent satellite observations from 2002 –

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1 2012 reported by Funke et al. (2014) reveal that particle induced  $\text{NO}_x$  is indeed transported  
2 downwards to the middle stratosphere at polar latitudes, while further model studies suggest that the  
3 subsiding of  $\text{NO}_x$  leads to strongly reduced stratospheric  $\text{O}_3$  concentrations (~30%) down to  
4 altitudes ~30 km (e.g. Reddmann et al., 2010). Thus, it appears promising to search for a link  
5 between EPP and  $\text{O}_3$  in actual data sets, because the downwards propagating signal of the EPP  
6 indirect effect on stratospheric and mesospheric  $\text{O}_3$  throughout the polar winter has not been  
7 explicitly observed so far. Note that,  $\text{NO}_x$  can be only transported downwards inside a stable large-  
8 scale dynamical structure, which provides sufficient subsidence and prevents  $\text{NO}_x$  removal/dilution  
9 by horizontal transport. These conditions are found primarily inside the Antarctic polar vortex,  
10 because the Arctic vortex is strongly disrupted by planetary waves, leading to its weakening or  
11 temporary breakdown. This large dynamical variability eventually causes high variations in  $\text{O}_3$   
12 volume mixing ratios (vmr), superposing the EPP indirect effect.

13 Therefore our study is focused on  $\text{O}_3$  vmr observations inside the Antarctic polar vortex from ~20 –  
14 70 km, derived from ENVironmental SATellite/Michelson Interferometer for Passive Atmospheric  
15 Sounding (ENVISAT/MIPAS), Thermosphere Ionosphere Mesosphere Energetics and  
16 Dynamics/Sounding of the Atmosphere using Broadband Emission Radiometry (TIMED/SABER),  
17 and Odin/Sub-Millimetre Radiometer (SMR) measurements. The intra-seasonal variability of the  $\text{O}_3$   
18 vmr values has been investigated and the relation to a number of solar and geomagnetic indices,  
19 namely the F10.7 cm solar radio flux, the Ap index, and the  $\geq 2$  MeV electron flux is analysed.

20

## 21 **2 Data analysis and numerical modelling**

### 22 **2.1 Approximation of the Antarctic polar vortex**

23 The position and the extension of the Antarctic polar vortex were estimated by using the gradient of  
24 the potential vorticity (PV) on isentropic surfaces (Nash et al., 1996). Assuming a dry atmosphere at  
25 altitudes  $\geq 20$  km, the PV was calculated from temperature, pressure, relative vorticity, and the  
26 corresponding latitude taken from ERA-Interim (<https://ecaccess.ecmwf.int/ecmwf>), the latest  
27 version of global atmospheric reanalysis data produced by the European Centre for Medium-Range  
28 Weather Forecasts (ECWMF). The reference pressure was set to 1000 hPa and the gravitational  
29 constant was considered to be dependent on latitude and height. The PV was calculated for all  
30 height intervals between 20 km and 70 km which were adapted from the MIPAS retrieval grid (see  
31 Sect. 2.2.1). Note that ERA-Interim data is primarily model-driven at mesospheric altitudes but the

1 individual PV results look reasonable at each height interval. As an example, Fig. 1 shows the PV,  
2 depending on time and equivalent latitude (EQL), during the Antarctic winter 2011 at ~40 km. The  
3 EQLs assigned to an individual PV isoline enclose the same area as the geographical latitudes of  
4 equivalent values. However, this area is located around an estimate of the vortex centre position,  
5 rather than around the geographical pole. In general, the EQL of the strongest PV gradient indicates  
6 the estimated location of the vortex edge, however, in most cases, there are at least two locations  
7 revealing gradients of similar magnitude. Therefore, Nash et al. (1996) also considered the zonal  
8 wind to locate the real vortex edge, but here we added a visual analysis instead of the zonal wind to  
9 divide the Southern Hemisphere into three non-overlapping zones: deep inside the Antarctic polar  
10 vortex (CORE) and the corresponding outermost edge (EDGE), covering all EQLs poleward the  
11 respective borders, as well as an area not influenced by the vortex (OUTSIDE), which extends from  
12 the equator to the respective OUTSIDE border. In this study we will consider the EDGE region as  
13 the Antarctic vortex area, but the CORE and OUTSIDE region are still necessary to determine  
14 whether the observed features inside the EDGE zone are actually originating from the vortex itself.  
15 The limits of the three regions of each height interval revealed no strong variation from 2002 -  
16 2011, therefore holding for every winter (Table 1). Note that the ECMWF ERA-Interim data only  
17 covers heights up to ~63 km. However, considering the behaviour of the Antarctic vortex at  
18 altitudes between 60 km and 70 km (Preusse et al., 2009, their Fig. 2a), it seems reasonable to  
19 assume that the estimated limits of the three regions at ~63 km are also valid up to 70 km.

## 20 **2.2 Ozone measurements**

### 21 **2.2.1 MIPAS**

22 MIPAS (Fischer et al., 2008) was a limb sounder on board ENVISAT, which had a sun-synchronous  
23 orbit. The main advantages of MIPAS measurements are the global coverage from 87°S – 89°N and  
24 the availability of observations during both day and night, crossing the equator at ~10:00 LT and  
25 ~22:00 LT, respectively. MIPAS was a Fourier transform infrared (4.15  $\mu\text{m}$  - 14.6  $\mu\text{m}$ ) emission  
26 spectrometer, allowing simultaneous observations of several atmospheric trace gases, including O<sub>3</sub>.  
27 MIPAS was operational from July 2002 – April 2012, but due to an instrumental failure in March  
28 2004, the entire observation period is divided into two subintervals from July 2002 – March 2004  
29 and January 2005 – April 2012 (referred to as P1 and P2 here, respectively). During P1 an almost  
30 continuous time series is available, while larger data gaps are present during P2 before October  
31 2006. Here, we use the complete data set of the most frequent observation mode (nominal mode),

1 covering the altitudes from the upper troposphere up to ~70 km at the poles which was derived from  
2 the MIPAS level-2 research processor developed by IMK/IAA. Details of the retrievals are  
3 described in von Clarmann et al. (2003), Glatthor et al. (2006), and von Clarmann et al. (2009).  
4 Note that the number of tangent heights is constant during P1 (17) and P2 (23), and that the actually  
5 available altitudes (cloud contaminated observations are disregarded) only slightly differ from day  
6 to day. The corresponding vertical resolution becomes coarser at higher altitudes (independent of  
7 the geographical location), increasing from 3.5 to 8 km (Steck et al., 2007) and from 2.5 to 5 km  
8 (Eckert et al., 2014) in P1 and P2, respectively. However, the retrieval grid in all MIPAS O<sub>3</sub> data  
9 versions used here (V3O\_O3\_9, V5R\_O3\_220, V5R\_O3\_221) is independent of the tangent  
10 heights, with a grid width of 1 km below 44 km and 2 km above. During P1/P2 O<sub>3</sub> was measured at  
11 two different wavelength intervals, ranging from 9.0 – 9.4 μm/9.6 – 9.7 μm and 12.5 – 13.5  
12 μm/12.7 – 13.2 μm in particular. However, not the full spectral ranges were used, but sub-intervals  
13 (microwindows). These were selected to minimise the computing time and to optimize the relation  
14 between the measurement-noise induced random error and other errors. These other errors originate,  
15 among further error sources, from spectral contributions of further atmospheric constituents of  
16 unknown abundances. It should also be noted that there is a bias in MIPAS O<sub>3</sub> data between the two  
17 periods, which was estimated using a multi-linear parametric trend model (Eckert et al., 2014). To  
18 accept an O<sub>3</sub> data point, the recommended filter criteria for MIPAS O<sub>3</sub> data were applied by using  
19 an averaging kernel diagonal value >0.03 as well as the visibility flag = 1 which indicates spectral  
20 available data.

21 At least 10 accepted data points inside the Antarctic polar vortex at a certain grid level were  
22 required to calculate the arithmetic average of one day, while at least 13 days were arithmetically  
23 averaged to a 26-day running mean from 1 April – 1 November, repeating this algorithm for each  
24 height interval and all years from 2002 - 2011. The time interval of 26 days was chosen to minimise  
25 a possible influence of the 27-day cycle of the sun, also ensuring that each time interval includes  
26 only one 27-day solar rotation maximum at most. The analysis was repeated for NO<sub>2</sub>  
27 (V5R\_NO2\_220, V5R\_NO2\_221) and the corresponding retrieval is described in Funke et al.  
28 (2005) and Funke et al. (2011).

## 29 **2.2.2 SABER**

30 The SABER instrument on board the TIMED Satellite has been nearly continuously operating since  
31 January 2002, measuring vertical profiles of several atmospheric parameters and minor constituents  
32 (e.g. O<sub>3</sub>) from the surface up to altitudes >100 km. The SABER measurements are governed by a

1 periodic quasi 60-day cycle, each time changing from the Southern Hemisphere mode (83°S –  
2 52°N) to the Northern Hemisphere mode (52°S – 83°N) and vice versa. Note that the “switching  
3 day” is only varying a few days from year to year. To consider both day and night O<sub>3</sub> observations,  
4 SABER Level 2A Ozone96 data v2.0 and v1.07 (<http://saber.gats-inc.com/custom.php>, Rong et al.,  
5 2009) measured at ~9.6 μm are used. However, v1.07 was only used to fill v2.0 data gaps, which  
6 seemed reasonable because the data fit quite well the results of the performed analysis during the  
7 respective periods (15 May – 31 May, 7 August – 31 August, not shown here). Consequently, the  
8 combined data set of both versions shows no larger data gap and the measurements of both versions  
9 were restricted to values <20 ppm to exclude outliers. Comparisons with the results of an increased  
10 threshold to <100 ppm revealed only minor differences (not shown here). The investigated height  
11 interval, ranging from 20 to 70 km, is divided in 38 non-overlapping subintervals and binned at the  
12 same altitudes as MIPAS data. The algorithm used to calculate the running means is also identical to  
13 the one applied for the “accepted” MIPAS data points. However, SABER needs approximately 60  
14 days to cover all local times, leading to a quasi 60-day wave like oscillation in O<sub>3</sub> if 26-day running  
15 means are used. This behaviour becomes evident at altitudes >50 km, where the averaging interval  
16 was consequently extended from 26 to 60 days. Note that the calculation of the 60-day running  
17 means required at least 30 days.

### 18 2.2.3 SMR

19 The Odin satellite mission started in February 2001 and is a joint project between Sweden, Canada,  
20 France and Finland (Murtagh et al., 2002). Odin was launched into a sun-synchronous polar orbit,  
21 carrying the SMR instrument and nominally covering the latitude range from 82.5°S – 82.5°N. The  
22 SMR makes vertical profile measurements during both day and night, while passing the equator at  
23 ~6:00/18:00 LT in the descending/ascending node. The O<sub>3</sub> data were extracted from the Odin/SMR  
24 Level 2 data product, version 2.0 (<http://odin.rss.chalmers.se/>, Urban et al., 2005), only using  
25 measurements of the frequency band centred around ~544.6 GHz, providing vertical O<sub>3</sub> profiles in  
26 the ~15-70 km altitude range. The filtering criterion used for SMR is the measurement response,  
27 which corresponds to the sum of the rows of the averaging kernel matrix. The profiles characterized  
28 by a measurement response lower than 0.9 are not reliable enough, and are therefore excluded. The  
29 algorithm to calculate the 26-day running means is identical to the one applied to MIPAS data. Note  
30 that Odin/SMR was a two-discipline satellite until April 2007, switching between atmospheric  
31 (aeronomy mode) and astronomy observations, and is entirely dedicated to aeronomy since this  
32 date. Consequently, measurements in the relevant mode are roughly performed one day out of three  
33 before April 2007 and every other day afterwards. However, the calculation of the 26-day running

**Gelöscht:** Additionally,

**Gelöscht:** only

**Gelöscht:** every day, consequently  
only covering about

1 means is still possible because the data gaps occur in a regular way, so they do not essentially  
2 worsen the 26-day averages. The vertical resolution of the data version used here is better than 3 km  
3 below 45 km, but increases to 5 – 6 km (50 – 60 km) and 7 – 10 km (60 – 70 km), leading to noisy  
4 results at altitudes >50 km compared to the other two instruments.

### 5 **2.3 Solar data and geomagnetic indices**

6 The data of the indices were obtained from two different websites provided by the National  
7 Geophysical Data Center. In detail the flux of the 10.7 cm radio emission from the sun (F10.7) and  
8 the geomagnetic Ap index (Ap), commonly used proxies for solar variation and geomagnetic  
9 activity, respectively, were downloaded from <http://spidr.ngdc.noaa.gov/spidr/>. The  $\geq 2$  MeV  
10 electron flux (2 MeV), including the flux of all electrons with energy levels above 2 MeV, was  
11 measured by the Geostationary Operational Environmental Satellites (GOES) and the corresponding  
12 time series were downloaded from  
13 [ftp://ftp.ngdc.noaa.gov/STP/SOLAR\\_DATA/SATELLITE\\_ENVIRONMENT/Daily\\_Fluences/](ftp://ftp.ngdc.noaa.gov/STP/SOLAR_DATA/SATELLITE_ENVIRONMENT/Daily_Fluences/).  
14 Note that the 2MeV data set also considers contamination effects on the electron detectors on the  
15 spacecrafts due to protons >32 MeV. Furthermore the 2MeV data is obtained from geostationary  
16 satellites which perform in-situ measurements in the radiation belts and consequently do not  
17 directly provide observations of precipitating particles. However, it is very likely that there is at  
18 least a positive relation between 2MeV and precipitating relativistic radiation belt particles. Thus,  
19 the 2MeV is not used as a proxy of precipitating particles but as an indicator of the influence from  
20 the magnetosphere. Precipitating particle integral fluxes in polar regions are observed by sun-  
21 synchronous Polar orbiting Operational Environmental Satellite (POES) detectors and the  
22 corresponding data correlates better with geomagnetic indices than the GOES electron fluxes  
23 (Sinnhuber et al., 2011). However, the respective measurements of the POES instruments tend to  
24 underestimate the fluxes from ground-based observations during weak geomagnetic activity  
25 (Rodger et al., 2013). Since this study focus on 2002 – 2011 and an essential part of this time  
26 interval overlaps with low geomagnetic activity, GOES data and Ap are used instead of POES  
27 measurements. The time series of all data sets are based on daily values, which were arithmetically  
28 averaged to 26-day means centred at 1 April, 1 May, and 1 June. The means were separately  
29 calculated for each index for the individual years from 2002 – 2011, however, 2MeV data are only  
30 available until 2010.

### 31 **2.4 Numerical modelling**



1 The three-dimensional chemistry and transport model (3dCTM; Sinnhuber et al., 2012, appendix 1)  
2 used here is based on the Bremen 3dCTM (e.g. Wissing et al., 2010), extending on 47 pressure  
3 levels from the tropopause up to the lower thermosphere (~10 – 140 km) with a latitude/longitude  
4 resolution of  $2.5^\circ \times 3.75^\circ$ . The model was recently updated with a variable H<sub>2</sub> and O<sub>2</sub> distribution,  
5 leading to proper HO<sub>x</sub> and consequently night time O<sub>3</sub> values at altitudes >60 km (see Sect. 1). The  
6 3dCTM is driven by meteorological data obtained from simulations of the three-dimensional  
7 dynamical model LIMA (Berger 2008) and the advection is calculated by applying the second-order  
8 moments scheme reported by Prather (1986). In the stratosphere, a family approach for the chemical  
9 families: O<sub>x</sub> (O + O(<sup>1</sup>D) + O<sub>3</sub>), NO<sub>x</sub> (N + NO + NO<sub>2</sub>), HO<sub>x</sub> (H + OH + HO<sub>2</sub>), BrO<sub>x</sub> (Br + BrO),  
10 ClO<sub>x</sub> (Cl + ClO + 2Cl<sub>2</sub>O<sub>2</sub>), and CHO<sub>x</sub> (CH<sub>3</sub> + CH<sub>3</sub>O<sub>2</sub> + CH<sub>3</sub>OOH + CH<sub>3</sub>O + HCO) is used, but was  
11 not used for O<sub>x</sub>, HO<sub>x</sub>, and NO<sub>x</sub> in the mesosphere/lower thermosphere region.

12 In this study the 3dCTM was used to investigate the impact of precipitating particles on O<sub>3</sub> inside  
13 the Antarctic polar vortex at altitudes from 20 - 70 km. After a multi-year two-dimensional model  
14 spin-up, two simulations from 2003 – 2009 were performed. The first run (base run) does not  
15 consider any energetic particles, while the second run (EP run) includes ionisation effects by both  
16 protons and electrons, using the ionisation rates provided by the Atmospheric Ionisation Module  
17 Osnabrück (AIMOS; Wissing and Kallenrode 2009). The resulting NO<sub>x</sub> production per created ion  
18 pair includes various ionic and neutral reactions depending on the atmospheric background state  
19 (Nieder et al., 2014). Simple parameterisations are used for the production of HO<sub>x</sub> (Solomon et al.,  
20 1981) and O (Porter et al., 1976). Note that heterogeneous chemistry was not included, which only  
21 becomes important during spring in the lower stratosphere. Both model runs considered constant  
22 solar minimum conditions ( $F_{10.7} = 70 \cdot 10^{-22} \text{ W m}^{-2} \text{ Hz}^{-1}$ ) to exclude O<sub>3</sub> variations due to solar  
23 activity. The obtained O<sub>3</sub> model results of both runs were separately selected according to the  
24 vertical MIPAS retrieval grid for direct comparisons to the observations, repeating the described  
25 algorithm to calculate the 26-day running means. Finally, in order to derive the O<sub>3</sub> vmr variations  
26 solely originating from precipitating particles, the obtained averages of the base run were subtracted  
27 from the corresponding O<sub>3</sub> values of the EP run. The results were divided by the arithmetic mean of  
28 both runs and eventually multiplied by 100%.

29

### 30 **3 Results and discussion**

#### 31 **3.1 Satellite observations**

### 1 3.1.1 O<sub>3</sub> response from 2002 - 2011

2 The 26-day O<sub>3</sub> vmr averages from 2002 - 2011 of each altitude-time interval (1 April – 1 November,  
3 20 - 70 km) were individually grouped into years of high and low index activity. For this purpose  
4 the index median of the corresponding time series of the 26-day average of an index (F10.7, Ap,  
5 2MeV) centred around 1 April was calculated, only including years of actually available O<sub>3</sub>  
6 observations. Therefore the median of an index time series works as a threshold, dividing the entire  
7 time interval from 2002-2011 in years of high (above the median) and low (below the median)  
8 index activity. Note that the classification of the years does not only depend on the chosen index,  
9 but due to data gaps also on the considered height-time interval as well as the instrument used.  
10 Afterwards the arithmetic O<sub>3</sub> mean of the years of low index activity was subtracted from the O<sub>3</sub>  
11 mean of the years of high index activity, eventually dividing this absolute O<sub>3</sub> difference by the  
12 arithmetic O<sub>3</sub> average of the entire observation period and multiplying the results by 100% for more  
13 handy values. Thus the calculated relative O<sub>3</sub> difference (referred to as O<sub>3</sub> amplitude here)  
14 represents the impact of the respective index on the O<sub>3</sub> background. To reduce the measurement  
15 noise of the individual instruments, the results of all three instruments were merged by simply  
16 calculating the arithmetic average but only if the corresponding O<sub>3</sub> amplitude of all three  
17 instruments was available. Note that due to the major sudden stratospheric warming centred around  
18 27 September (Azeem et al., 2010) the O<sub>3</sub> observations from 1 September - 1 November 2002 were  
19 excluded. In contrast, the solar proton event in the end of October 2003 (Jackman et al., 2005) was  
20 neglected due to its late occurrence. The performed analyses with O<sub>3</sub> observations, considering the  
21 indices from 1 May and 1 June (not shown here), revealed no essential differences compared to 1  
22 April or the structures became less obvious. Comparisons with earlier periods are not reasonable  
23 because the vortex first builds up in April. Therefore the focus is set on the O<sub>3</sub> response to indices  
24 centred around 1 April. The O<sub>3</sub> amplitude was calculated for all three regions (CORE, EDGE, and  
25 OUTSDIE) which were introduced in Sect. 2.1. The corresponding results reveal that the pattern  
26 found inside the EDGE region are fairly similar and less noisy compared to the features observed in  
27 the CORE area (not shown here). In contrast the O<sub>3</sub> amplitudes outside the Antarctic polar vortex  
28 are fundamentally different. An example for the O<sub>3</sub> response associated to 1 April Ap in the EDGE  
29 and the OUTSIDE region derived from MIPAS measurements is presented in Fig. 2, showing  
30 considerably disagreeing structures and essentially weaker amplitudes, especially below 50 km.  
31 Thus comparison between the individual regions of the Southern Hemisphere ensures, that, the  
32 pattern found in the EDGE region are actually originating from the Antarctic polar vortex.  
33 Figure 3 displays the corresponding results of the O<sub>3</sub> amplitude from 2002 - 2011, but only for

1 values above the significance level of 95% while shaded areas show regions between the  
2 significance level of 95% and 99%. The significance was calculated according to a Student's t-test,  
3 based on the error of the mean of the 26-day running O<sub>3</sub> means and assuming the worst case  
4 scenario of absolute error propagation. The MIPAS O<sub>3</sub> measurements (left column) reveal a high  
5 negative response to Ap (upper row) in early Antarctic winter >60 km, on average ranging around -  
6 10%. Further striking negative O<sub>3</sub> amplitudes occur in July between 30 and 40 km as well as around  
7 1 October at ~30 km, at least weakly indicating the downward transport of the Ap signal in  
8 stratospheric O<sub>3</sub> due to NO<sub>x</sub> predicted by model studies (e.g. Reddmann et al., 2010). In contrast, a  
9 positive O<sub>3</sub> amplitude is found at the beginning of the winter between ~25 km and ~55 km (~10 –  
10 20%), as well as at altitudes <30 km throughout the winter (up to ~20% in October at ~20 km) and  
11 above the indicated subsiding layer of negative amplitudes. But considering that most of these  
12 features drop below the significance level of 95% by combining the data of all three instruments  
13 (right column), a more detailed investigation of these patterns is not reasonable. However, the  
14 results of the merged data set show a well pronounced subsiding negative Ap signal from ~50 km in  
15 June down to ~25 km in October, which is disrupted in August, while the generally positive  
16 structures below 30 km are also still present.

17 The O<sub>3</sub> response to 2 MeV (middle row) derived from MIPAS observations also indicates a  
18 downwelling of negative O<sub>3</sub> amplitudes, descending from ~60 km in June down to ~30 km in late  
19 August. Additionally, the MIPAS O<sub>3</sub> response to 2MeV in early winter is reversed compared to the  
20 corresponding influence from Ap on O<sub>3</sub>, which does not originate from missing 2MeV data from  
21 2011. Strong positive O<sub>3</sub> amplitudes are generally observed throughout the winter below 30 km,  
22 exceeding values of ~20% in April and October, as well as during October between 30 km and 50  
23 km where the maximum amplitude is lower (~10%). The positive features can be validated with the  
24 composite results even if they are damped in the region below 30 km. However, this is not the case  
25 for the negative response, except for a small area in June in the lower mesosphere. Considering that  
26 the Ap responds to lower particle energy levels compared to 2MeV and that the behaviour of both  
27 indices is essentially different from 2002 - 2010 (see Fig. 4), the different O<sub>3</sub> amplitudes associated  
28 to Ap and 2MeV are still reasonable.

29 The O<sub>3</sub> response to F10.7 (lowermost row) is fairly similar between MIPAS and the merged  
30 measurements, and both also agree with the respective pattern observed for Ap, including the  
31 indicated downwelling of negative O<sub>3</sub> amplitudes during midwinter from 50 to 25 km. The  
32 composite O<sub>3</sub> shows strong positive amplitudes in May >55 km which originate from SMR  
33 measurements and are most likely due to the low vertical resolution of the SMR instrument at these

1 altitudes (see Sect. 2.2.3). The high agreement between the results of  $A_p/O_3$  and  $F10.7/O_3$  might  
2 originate from the coupling of both indices during solar maximum years (Gray et al., 2010, their  
3 Fig. 1). In order to investigate a possible cross-correlation between solar radiation and geomagnetic  
4 disturbances, the analysis was repeated for years of moderate solar activity, only including 2005 –  
5 2010 (Fig. 4). Similar analyses to extract a more distinct solar signal during times of approximately  
6 constant geomagnetic activity were not reasonable, because the respective years of nearly constant  
7  $A_p$  values (2002, 2005, 2006, 2008, 2010) do not provide a sufficient amount of data in MIPAS and  
8 SMR measurements.

### 9 **3.1.2 $O_3$ behaviour during solar minimum activity (2005 - 2010)**

10 Figure 5 displays the obtained  $O_3$  amplitudes for solar quiet times (2005 – 2010) associated to 1  
11 April  $A_p$ , again only showing values above 95% significance level and shading the area of regions  
12 between 95% and 99%. The MIPAS  $O_3$  response to  $A_p$  indicates a subsiding negative signal (~-10  
13 to -15%), starting in late June slightly below 50 km and propagating downwards to ~25 km  
14 throughout the winter. However, the middle part of the downwelling between late July and late  
15 September is below the significance level of 95% and therefore not shown here. Furthermore, the  
16 hinted subsidence is closely surrounded by well pronounced positive  $O_3$  amplitudes, especially  
17 below ~30 km which maximise in September (>20%). There is also a negative structure centred at 1  
18 June at ~60 km, which cannot be caused by  $NO_x$  but most likely results from  $HO_x$  formation (see  
19 Sect. 1). Considering the composite results, the downwelling  $A_p$  signal in  $O_3$  becomes apparent and  
20 robust but slightly weaker (~-10%) while the positive features are also damped but still present. The  
21 mesospheric response is generally weak and the high positive  $O_3$  amplitudes in May are again  
22 caused by the SMR measurements.

23 The 2MeV impact on MIPAS  $O_3$  shows generally agreeing features with the influence of the  
24 geomagnetic activity and is also of similar magnitude, however, the downwelling negative signal is  
25 hinted to already start in late May at ~55 km. In contrast to the  $O_3$  response to  $A_p$ , the downwards  
26 propagating 2MeV signal is less robust and can be only guessed in the composite  $O_3$  amplitude,  
27 while the positive structures (~10 - 15%) in August below 30 km and in September between 30 km  
28 and 50 km are still present. In general, the 2MeV features are less obvious in the  $O_3$  composite,  
29 except for the positive  $O_3$  amplitudes above the hinted downward transport. Nevertheless, the  
30 agreement between  $A_p$  and 2MeV pattern is quite strong, in MIPAS observations in particular,  
31 although both parameters are only indirectly related to  $O_3$ . However, the  $O_3$  structure associated to  
32 both indices is far too similar and additionally found in all three instruments to be a coincidence,

1 even if the descending O<sub>3</sub> response to 2 MeV is weaker. Since Ap represents lower particle energy  
2 levels compared to the 2MeV and both indices are only moderately correlated (see Fig. 4), the  
3 similar results strongly indicate a related source mechanism, suggesting solar wind variability.

4 Considering the entire process, that energetic particles produce NO<sub>x</sub> which eventually destroys  
5 stratospheric O<sub>3</sub>, the Ap impact observed in O<sub>3</sub> (see Fig. 5) is expected to be reversed in NO<sub>x</sub>, at  
6 least in the stratosphere. In order to investigate this in more detail, the analysis was repeated for 1  
7 April Ap and NO<sub>x</sub>. Here NO<sub>x</sub> is represented only by NO<sub>2</sub> from MIPAS observations, because the  
8 respective NO measurements are quite noisy compared to NO<sub>2</sub>, especially below 30 km. This is still  
9 reasonable because NO is converted to NO<sub>2</sub> during night and therefore NO<sub>2</sub> is the major fraction of  
10 NO<sub>x</sub> inside the Antarctic polar vortex. The corresponding results include the years 2005 – 2010 and  
11 are displayed in Fig. 6, supporting that the stratospheric O<sub>3</sub> depletion can be indeed associated to the  
12 catalytic NO<sub>x</sub>/O<sub>3</sub> cycle. The Ap signal in NO<sub>2</sub> is stronger by the factor of 2 - 5, compared to the  
13 respective O<sub>3</sub> amplitudes. The sharp gradient in mid July originates from 2005 NO<sub>2</sub> data, which are  
14 not available afterwards. However, the general structure of the subsiding Ap signal in NO<sub>2</sub> is still  
15 similar with and without 2005 observations. Note that the essentially smaller NO<sub>2</sub> amplitudes in  
16 October below the significance level of 95% are not in conflict with the respective well pronounced  
17 negative O<sub>3</sub> response, because the latter one results from an accumulation effect from the NO<sub>2</sub>  
18 above. Furthermore, large negative NO<sub>2</sub> amplitudes throughout the entire winter below ~30 km are  
19 observed, matching the high positive O<sub>3</sub> response to Ap. A possible reason for this behaviour might  
20 be that NO<sub>2</sub> is stored in reservoir species, like ClONO<sub>2</sub>, HNO<sub>3</sub>, and N<sub>2</sub>O<sub>5</sub>, due to reactions with  
21 ClO, OH, and NO<sub>3</sub>, respectively. However, N<sub>2</sub>O<sub>5</sub> is converted to HNO<sub>3</sub> via water ion cluster  
22 chemistry (López-Puertas et al., 2005, their reactions 1 and 8 – 12), which was also investigated  
23 with respect to EPP for conditions without solar proton events by Stiller et al. (2005). These  
24 reactions eventually lead to lower NO<sub>x</sub> concentrations, consequently slowing down the catalytic O<sub>3</sub>  
25 depletion. Based on the corresponding MIPAS climatologies (not shown here), HNO<sub>3</sub> is more  
26 important until mid July, while ClONO<sub>2</sub> is dominating afterwards and its influence becomes  
27 essentially crucial in spring due to heterogeneous chemistry which has taken place before. This  
28 suggested NO<sub>2</sub>-ClONO<sub>2</sub> mechanism is supported by Whole Atmosphere Community Climate  
29 Model results reported by Jackman et al. (2009, their Fig. 6 and 7), who simulated the impact of the  
30 SPE in July 2000 on stratospheric O<sub>3</sub> and NO<sub>y</sub> (= NO<sub>x</sub> + NO<sub>3</sub> + N<sub>2</sub>O<sub>5</sub> + HNO<sub>3</sub> + HO<sub>2</sub>NO<sub>2</sub> +  
31 ClONO<sub>2</sub> + BrONO<sub>2</sub>).

32 Furthermore, the positive O<sub>3</sub> amplitudes below ~30 km could be also partly explained by the self  
33 healing effect of O<sub>3</sub> (Jackman and McPeters, 1985). Altitude regions of reduced O<sub>3</sub> will lead to

1 increased solar UV radiation in the layers directly below. This is accompanied by a higher  
2 production of atomic oxygen and would consequently increase the formation of O<sub>3</sub>. However, this  
3 proposed mechanism would only have an additional effect, contributing to the formation of O<sub>3</sub> in  
4 the atmospheric layer right below the subsidence, but cannot account for the entire region. Note that  
5 this layer is also present throughout the entire winter, and thus an influence from the vortex above is  
6 unlikely but any further investigations are beyond the scope of this study.

7 Additionally, the area of high positive Ap/O<sub>3</sub> structure between 35 km and 50 km from August -  
8 September cannot be completely explained by the NO<sub>x</sub>/O<sub>3</sub> cycle. In detail, the respective Ap  
9 influence of NO<sub>2</sub> is close to 0 and consequently well below the 95%, while the respective MIPAS  
10 ClONO<sub>2</sub> amplitude (not shown here) reveals positive values, which are also mostly below the 95%  
11 significance level. These results are at least not in conflict with a higher O<sub>3</sub> amplitude. Furthermore,  
12 this positive Ap impact on O<sub>3</sub> is essentially less visible in the composite results than in MIPAS data,  
13 and a corresponding composite analysis for Ap/NO<sub>2</sub> is necessary for a more detailed investigation.  
14 But this is not possible due to non-existing NO<sub>2</sub> measurements from SABER and SMR. Thus no  
15 definite explanation can be given at this stage and this feature is a subject of a future work.  
16 However, it should be pointed out that this structure does not harm the underlying mechanism  
17 proposed to explain the identified negative O<sub>3</sub> amplitude and subsequent downward transport.

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### 18 3.2 Comparison with 3dCTM

19 The simulated O<sub>3</sub> amplitude between the EP run and base run, representing high and low  
20 geomagnetic activity, respectively, is displayed in Fig. 7. Note that the modelled O<sub>3</sub> amplitude is  
21 also referred to as O<sub>3</sub> amplitude here, which is justified because “observed” and “modelled” O<sub>3</sub>  
22 amplitude still hold the same physical meaning, even if the calculation algorithm is slightly  
23 different. It is reasonable to investigate the complete simulated time interval from 2003 – 2009,  
24 because the model runs represent solar minimum conditions similar to the years 2005 - 2010. The  
25 results reveal apparent negative O<sub>3</sub> amplitudes propagating downwards throughout the winter with  
26 maximum negative values during midwinter between 45 km and 60 km. The subsidence shows  
27 larger negative O<sub>3</sub> amplitudes compared to the measurements and is also much broader, which  
28 might be due to the constant F10.7 and the prescribed dynamics, both reducing the inter-annual  
29 variability of O<sub>3</sub>. Furthermore, we performed an on/off experiment, while in reality the EEP indirect  
30 effect is a persistent feature. Below 30 km the observed high positive O<sub>3</sub> amplitudes associated to  
31 Ap are only indicated in the model results by essentially weaker and additionally negative  
32 amplitudes. However, the model amplitudes are at least less negative compared to the values above.

1 The second positive region above the downwelling is completely missing. Further note that the  
2 strong positive response during late winter/early spring below 30 km might not be reproduced by  
3 the model due to missing heterogeneous chemistry. The proposed self healing effect of O<sub>3</sub> (see Sect  
4 3.1.2) was also tested, using O<sup>1</sup>D as a proxy for the O<sub>3</sub> photolysis rate in the Lyman-alpha band and  
5 calculating the O<sup>1</sup>D amplitude (not shown here). However, the expected positive response directly  
6 below the downwelling is only partly visible and even below the 67% significance level.

7 The qualitative agreement between model results and observations in the stratosphere suggests that  
8 the subsiding Ap signal found in O<sub>3</sub> is actually originating from particle precipitation. However, the  
9 simulated downwelling starts at altitudes >60 km while observations reveal no obvious structures in  
10 the mesosphere, possibly caused by satellite sampling. As already stated in Sect 3.1.2, the  
11 mesospheric behaviour cannot be caused by NO<sub>x</sub>, because the NO<sub>x</sub>/O<sub>3</sub> cycle is not efficiently  
12 working at these altitudes. Thus the O<sub>3</sub> depletion >50 km could be accounted to OH production,  
13 which is most likely overestimated in the model and consequently leads to an increased O<sub>3</sub>  
14 depletion not observed by the satellite instruments.

15

#### 16 **4 Conclusions**

17 We have investigated the O<sub>3</sub> behaviour inside the Antarctic polar vortex from 2002 - 2011, observed  
18 by three independent satellite based instruments ENVISAT/MIPAS, Odin/SMR, and  
19 TIMED/SABER. These O<sub>3</sub> vmr measurements, based on 26-day running means from 1 April – 1  
20 November covering altitudes from 20 – 70 km, were individually grouped into high and low index  
21 activity according to the 26-day averages centred around 1 April, 1 May, and 1 June of different  
22 solar and geomagnetic indices (F10.7, Ap, 2MeV). After minimising the direct influence of the solar  
23 radiation by only considering the period of solar minimum activity from 2005 – 2010 we found a  
24 negative O<sub>3</sub> response caused by geomagnetic activity (Ap) from 1 April in all three instruments,  
25 ranging from -5% to -10% and propagating downwards throughout the Antarctic winter from ~50  
26 km down to ~25 km. This subsiding negative signal in O<sub>3</sub> is above the significance level of 95%  
27 and overlaps with the corresponding positive NO<sub>2</sub> response to 1 April Ap, supporting that NO<sub>x</sub> is  
28 indeed the cause of the O<sub>3</sub> depletion. We could also show that the high positive O<sub>3</sub> response below  
29 30 km, which is present during the entire winter, is in agreement with respective negative NO<sub>2</sub>  
30 structures. The cause of the NO<sub>2</sub> behaviour is possibly related to the formation of the reservoir  
31 species ClONO<sub>2</sub> and HNO<sub>3</sub>, slowing down the catalytic destruction of O<sub>3</sub> by Cl. The O<sub>3</sub> pattern  
32 induced by the magnetosphere (2MeV) from 1 April are similar but weaker, compared to the  
33 respective geomagnetic activity, still suggesting a related source mechanism between 2MeV and Ap

1 like solar wind variability. The composite observations of all three instruments are in good  
2 qualitative agreement with 3dCTM simulation, revealing similar O<sub>3</sub> pattern induced by the  
3 geomagnetic activity from 1 April while the simulated O<sub>3</sub> response is larger but still in the same  
4 order of magnitude.

5 However, we have to point out that the validity of the subsiding O<sub>3</sub> depletion associated to  
6 geomagnetic activity and NO<sub>x</sub> is not ensured due to the short time series of only 6 years at most.  
7 Thus, we conclude that precipitating particles are strongly indicated as a factor contributing to  
8 stratospheric O<sub>3</sub> during Antarctic winter, but we cannot prove the link unambiguously.

9

## 10 **Authors' contribution**

11 T. F. analysed the satellite and indices data and wrote the final script. G. S., J. U.+K. P., and M. M.  
12 provided the O<sub>3</sub> data from ENVISAT/MIPAS, Odin/SMR, and TIMED/SABER, respectively, and  
13 all of them contributed to interpretation. H. N. performed the 3dCTM simulations. M. S. initiated  
14 the study and contributed to interpretation.

15

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25

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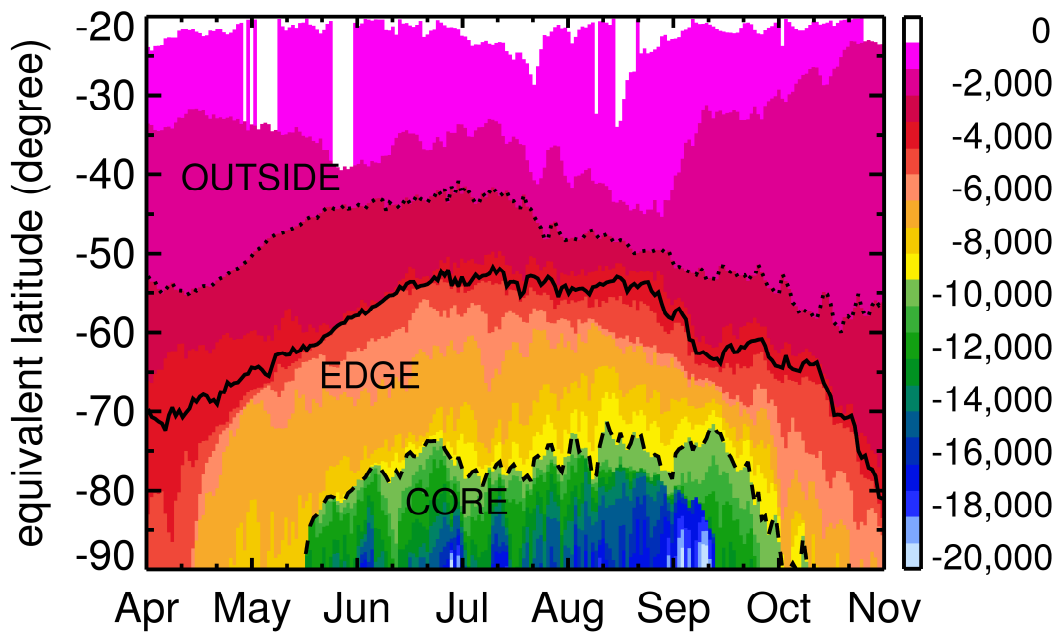
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1 Table 1. Limits, derived from the potential vorticity ( $10^{-6} \text{ K m}^2 \text{ s}^{-1} \text{ kg}^{-1}$ ), of the southern hemispheric  
 2 regions CORE, EDGE, and OUTSIDE at the individual heights. The altitudes are adapted from the  
 3 MIPAS retrieval grid and the shown potential vorticity values hold for 2002 – 2011.

<b>Nominal height (km)</b>	<b>CORE</b>	<b>EDGE</b>	<b>OUT- SIDE</b>	<b>Nominal height (km)</b>	<b>CORE</b>	<b>EDGE</b>	<b>OUT- SIDE</b>
20	-60	-50	-30	37	-3600	-1900	-1000
21	-70	-50	-30	38	-6000	-2500	-1500
22	-90	-70	-40	39	-6000	-3000	-2000
23	-150	-90	-40	40	-9000	-3500	-2000
24	-200	-100	-60	41	-9000	-4000	-2000
25	-180	-110	-60	42	-9000	-4000	-2000
26	-280	-160	-100	43	-15000	-5000	-3000
27	-360	-220	-120	44	-15000	-5500	-3000
28	-600	-250	-120	46	-22000	-8000	-4000
29	-900	-300	-150	48	-18000	-10000	-2000
30	-800	-400	-200	50	-36000	-12000	-4000
31	-1000	-400	-200	52	-32000	-16000	-4000
32	-1600	-600	-300	54	-36000	-16000	-4000
33	-2000	-800	-400	56	-60000	-30000	-5000
34	-1800	-1100	-400	58	-60000	-30000	-10000
35	-2800	-1400	-800	60	-60000	-30000	-10000
36	-4000	-1600	-1000	62 – 70	-180000	-90000	-30000

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2 Figure 1. Potential vorticity ( $10^{-6} \text{ K m}^2 \text{ s}^{-1} \text{ kg}^{-1}$ , colour scale) at ~40 km during the Antarctic winter  
 3 2011 as a function of time and equivalent latitude. The thresholds of the regions OUTSIDE (dotted  
 4 line), EDGE (solid line) and CORE (dashed line) are included. Potential vorticity was calculated  
 5 from ECMWF Era-Interim data.

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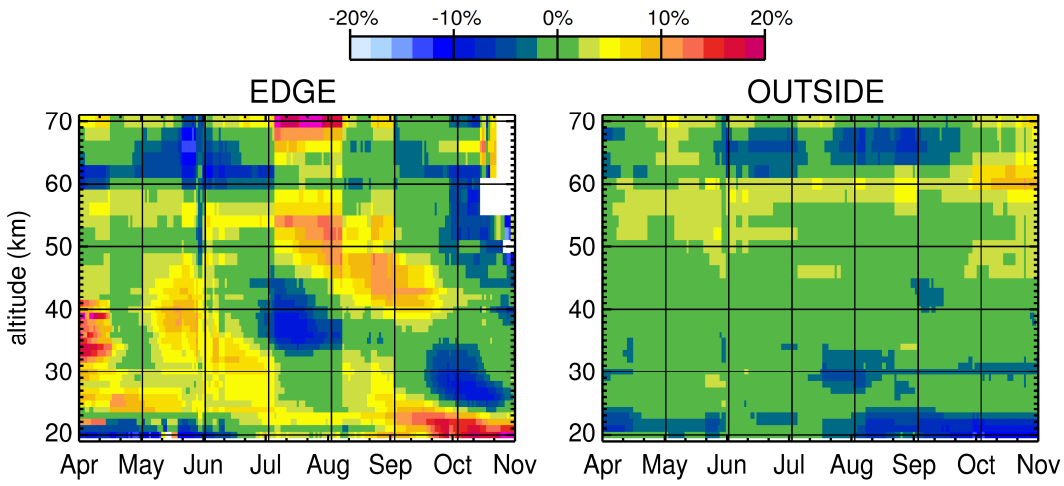
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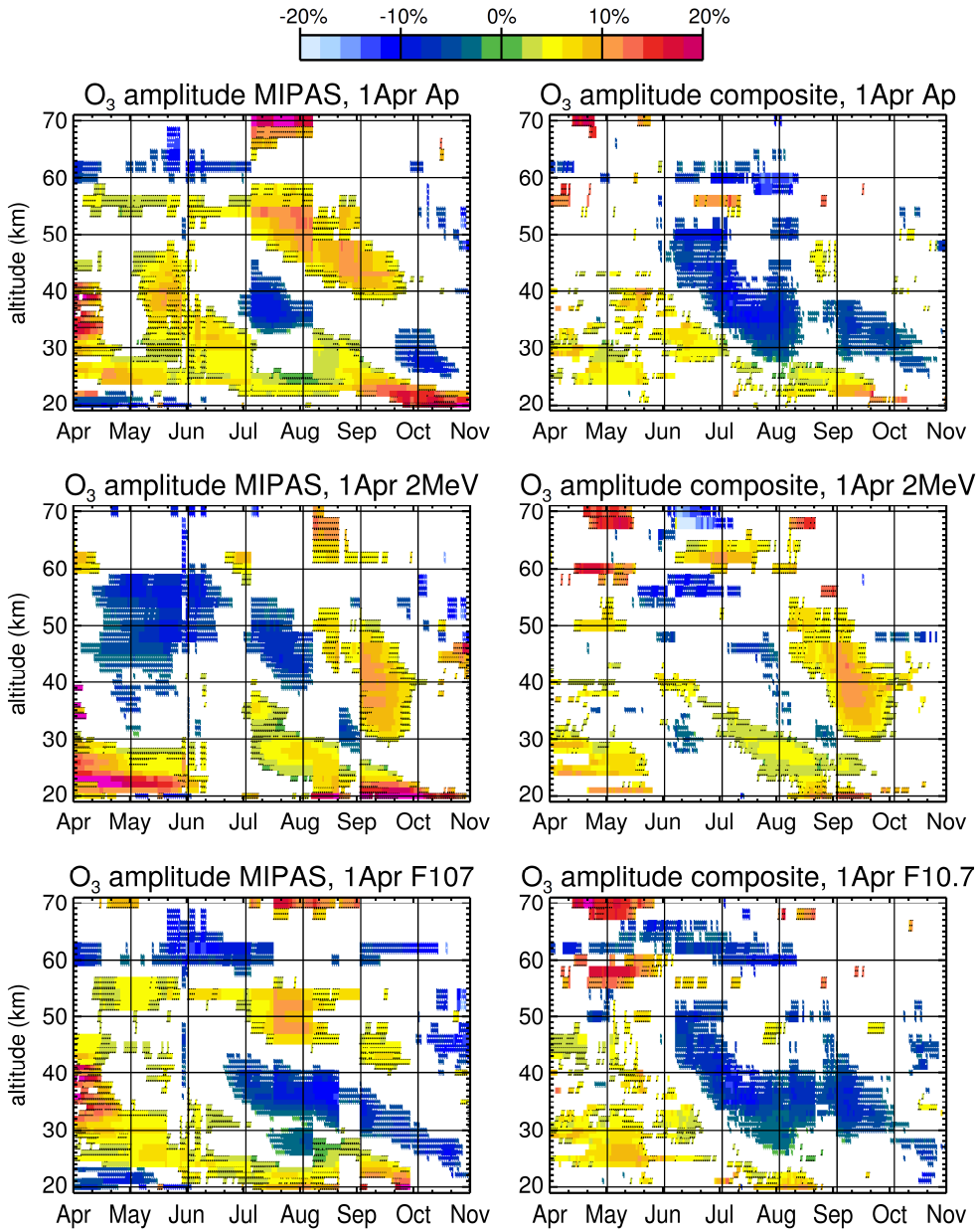


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 2 Figure 2. Example of the O<sub>3</sub> amplitude (see Sect. 3.1.1 for definition) observed by MIPAS from  
 3 2002 – 2011 between years of high Ap index and years of low Ap index centred around 1 April, for  
 4 the regions EDGE (left) and OUTSIDE (right).

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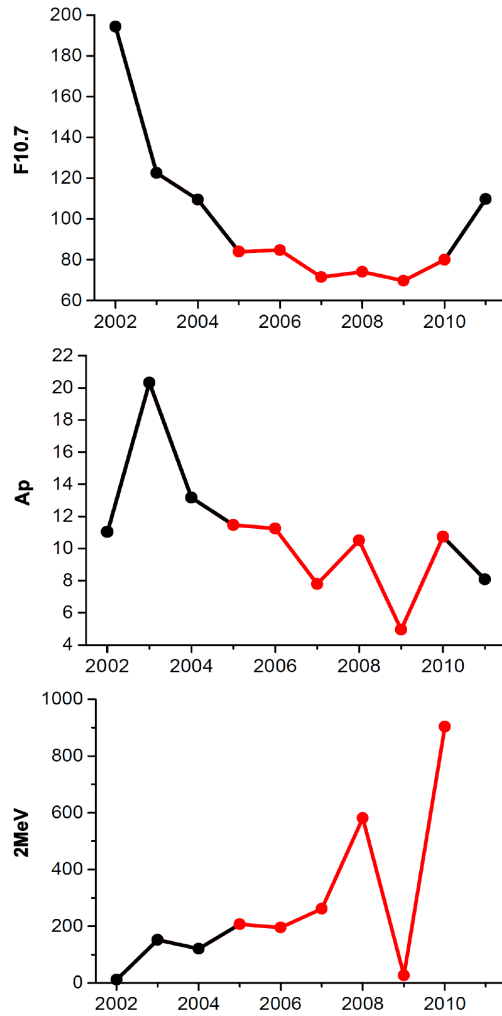


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3 Figure 3. O<sub>3</sub> amplitude (see Sect. 3.1.1 for definition) inside the Antarctic polar vortex between  
 4 years of high index values and years of low index values, namely Ap index (upper row),  $\geq 2$  MeV  
 5 electron flux (middle) as well as F10.7 cm solar radio flux (lowermost row) centred around 1 April,  
 6 derived from MIPAS (left column) and composite (MIPAS+SMR+SABER, right column)  
 7 observations from 2002 – 2011. Shown are only values above the significance level of 95%.  
 8 Additionally, regions between the significance level of 95% and 99% are shaded in black or white,  
 9 according to a Student's t-test.



2 Figure 4. Time series from 2002 – 2011 of the 26-day averages centred around 1 April of the F10.7  
 3 cm solar radio flux ( $10^{-22} \text{ W m}^{-2} \text{ Hz}^{-1}$ , top), Ap index (middle), and  $\geq 2$  MeV electron flux (electrons  
 4  $\text{cm}^{-2} \text{ day}^{-1} \text{ sr}^{-1}$ , bottom). The period of low solar activity from 2005 – 2010 is marked in red. Note  
 5 the different scaling.

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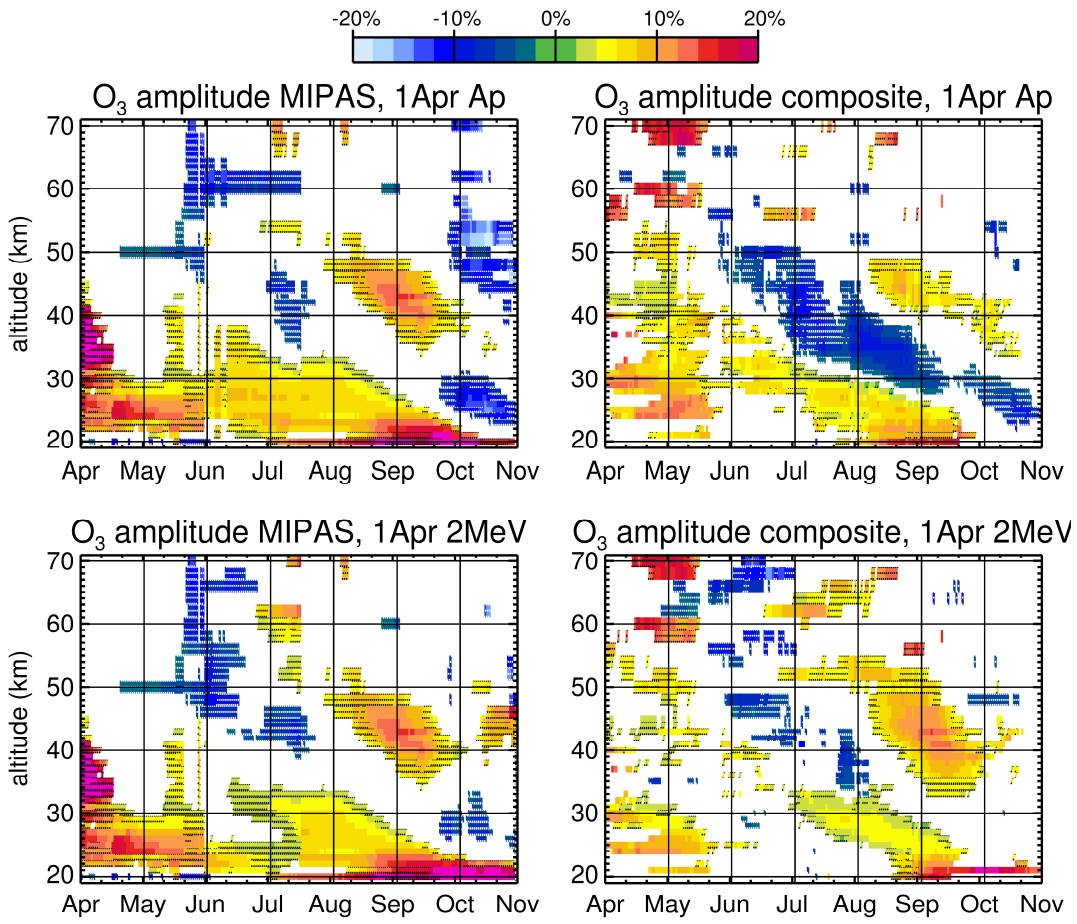
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3 Figure 5. Same as Figure 3 but only for Ap index (upper row) and  $\geq 2$  MeV electron flux (lower  
4 row) from 2005 – 2010.

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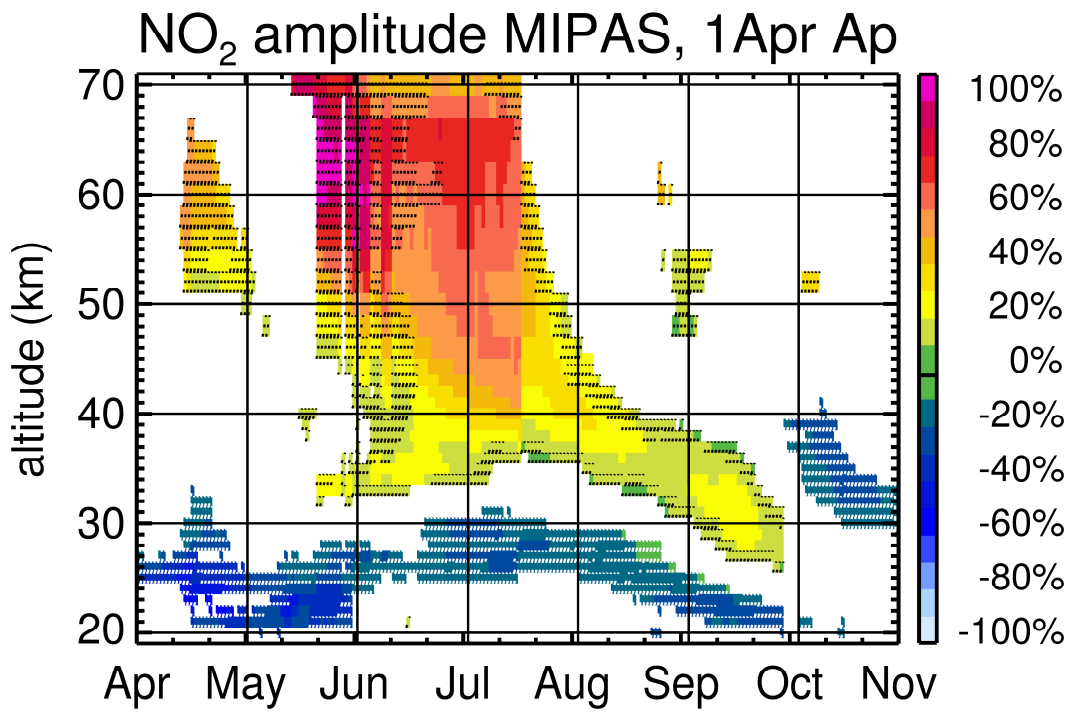
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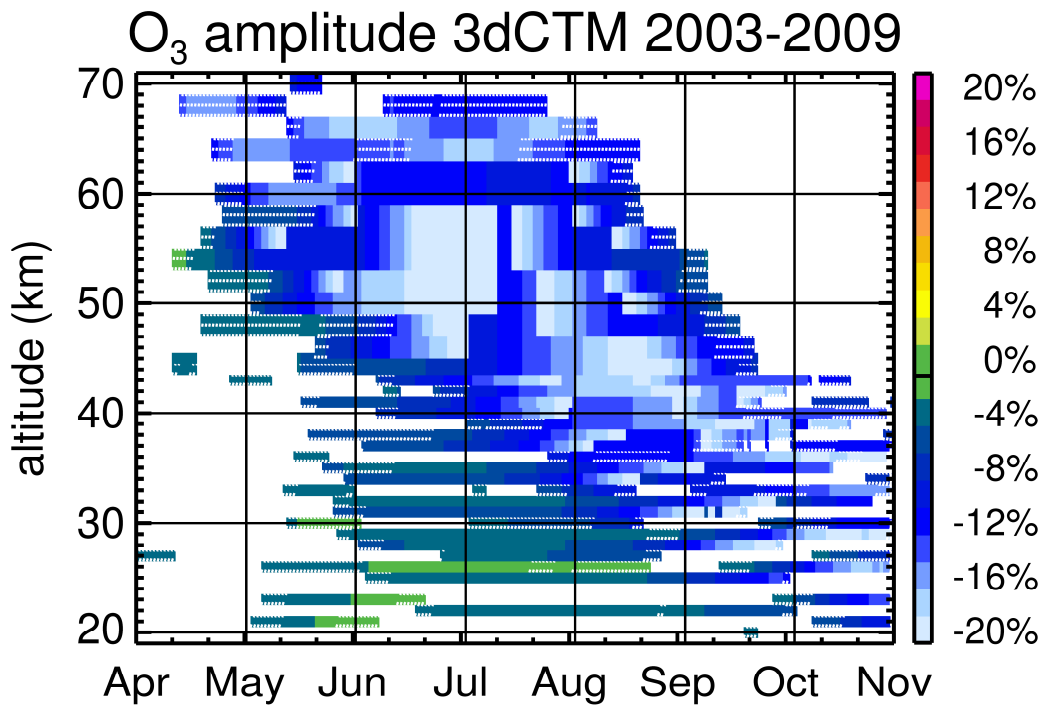
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 2 Figure 6. Same as Figure 3, but only for the NO<sub>2</sub> amplitude associated to 1 April Ap index. The NO<sub>2</sub>  
 3 was derived from MIPAS observations from 2005 – 2010.

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2 Figure 7. O<sub>3</sub> amplitude (see Sect. 2.4 for definition), simulated by the 3dCTM from 2003 – 2009.

3 Shown are values above 95% significance level, according to a Student's t-test, and areas between

4 95% and 99% significance level are shaded.