

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

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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 O₃ 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 O₃ vmr variations between maximum and minimum conditions of a number of solar and
20 geomagnetic indices (F10.7 cm solar radio flux, Ap index, ≥ 2 MeV electron flux). The indices are
21 26-day averages centred at 1 April, 1 May, and 1 June while O₃ 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 O₃ signal associated to the
24 geomagnetic activity (Ap index) around 1 April, on average reaching amplitudes between -5% and
25 -10% of the respective O₃ background. The O₃ 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 O₃ vmr pattern simulated with a three-
28 dimensional chemistry-transport model, which includes particle impact ionisation.

29

1 **1 Introduction**

2 Energetic particles (\sim keV - \sim 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 (\sim 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 (\sim seconds – hours) and therefore more important near its
16 source region in the mesosphere, while NO_x has a relatively long life time (\sim 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 (\sim 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 –

1 2012 reported by Funke et al. (2014) reveal that particle induced NO_x is indeed transported
2 downwards to the middle stratosphere at polar latitudes, while further model studies suggest that the
3 subsiding of NO_x leads to strongly reduced stratospheric O_3 concentrations ($\sim 30\%$) 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 O_3 in actual data sets, because the downwards propagating signal of the EPP
6 indirect effect on stratospheric and mesospheric O_3 throughout the polar winter has not been
7 explicitly observed so far. Note that, NO_x can be only transported downwards inside a stable large-
8 scale dynamical structure, which provides sufficient subsidence and prevents 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 O_3
12 volume mixing ratios (vmr), superposing the EPP indirect effect.

13 Therefore our study is focused on 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 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 ≥ 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 ≥ 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 μm - 14.6 μm) emission
26 spectrometer, allowing simultaneous observations of several atmospheric trace gases, including O₃.
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 O₃ 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 O₃ 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 O₃ data between the two
17 periods, which was estimated using a multi-linear parametric trend model (Eckert et al., 2014). To
18 accept an O₃ data point, the recommended filter criteria for MIPAS O₃ 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 NO₂
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. O₃) 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 O₃ 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 O₃ 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 O₃ 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 O₃ 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. Additionally, measurements in the relevant mode are only performed every day, consequently
33 only covering about one day out of three before April 2007 and every other day afterwards.

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

6 **2.3 Solar data and geomagnetic indices**

7 The data of the indices were obtained from two different websites provided by the National
8 Geophysical Data Center. In detail the flux of the 10.7 cm radio emission from the sun (F10.7) and
9 the geomagnetic Ap index (Ap), commonly used proxies for solar variation and geomagnetic
10 activity, respectively, were downloaded from <http://spidr.ngdc.noaa.gov/spidr/>. The ≥ 2 MeV
11 electron flux (2 MeV), including the flux of all electrons with energy levels above 2 MeV, was
12 measured by the Geostationary Operational Environmental Satellites (GOES) and the corresponding
13 time series were downloaded from
14 ftp://ftp.ngdc.noaa.gov/STP/SOLAR_DATA/SATELLITE_ENVIRONMENT/Daily_Fluences/.

15 Note that the 2MeV data set also considers contamination effects on the electron detectors on the
16 spacecrafts due to protons >32 MeV. Furthermore the 2MeV data is obtained from geostationary
17 satellites which perform in-situ measurements in the radiation belts and consequently do not
18 directly provide observations of precipitating particles. However, it is very likely that there is at
19 least a positive relation between 2MeV and precipitating relativistic radiation belt particles. Thus,
20 the 2MeV is not used as a proxy of precipitating particles but as an indicator of the influence from
21 the magnetosphere. Precipitating particle integral fluxes in polar regions are observed by sun-
22 synchronous Polar orbiting Operational Environmental Satellite (POES) detectors and the
23 corresponding data correlates better with geomagnetic indices than the GOES electron fluxes
24 (Sinnhuber et al., 2011). However, the respective measurements of the POES instruments tend to
25 underestimate the fluxes from ground-based observations during weak geomagnetic activity
26 (Rodger et al., 2013). Since this study focus on 2002 – 2011 and an essential part of this time
27 interval overlaps with low geomagnetic activity, GOES data and Ap are used instead of POES
28 measurements. The time series of all data sets are based on daily values, which were arithmetically
29 averaged to 26-day means centred at 1 April, 1 May, and 1 June. The means were separately
30 calculated for each index for the individual years from 2002 – 2011, however, 2MeV data are only
31 available until 2010.

1 **2.4 Numerical modelling**

2 The three-dimensional chemistry and transport model (3dCTM; Sinnhuber et al., 2012, appendix 1)
3 used here is based on the Bremen 3dCTM (e.g. Wissing et al., 2010), extending on 47 pressure
4 levels from the tropopause up to the lower thermosphere (~10 – 140 km) with a latitude/longitude
5 resolution of $2.5^\circ \times 3.75^\circ$. The model was recently updated with a variable H₂ and O₂ distribution,
6 leading to proper HO_x and consequently night time O₃ values at altitudes >60 km (see Sect. 1). The
7 3dCTM is driven by meteorological data obtained from simulations of the three-dimensional
8 dynamical model LIMA (Berger 2008) and the advection is calculated by applying the second-order
9 moments scheme reported by Prather (1986). In the stratosphere, a family approach for the chemical
10 families: O_x (O + O(¹D) + O₃), NO_x (N + NO + NO₂), HO_x (H + OH + HO₂), BrO_x (Br + BrO),
11 ClO_x (Cl + ClO + 2Cl₂O₂), and CHO_x (CH₃ + CH₃O₂ + CH₃OOH + CH₃O + HCO) is used, but was
12 not used for O_x, HO_x, and NO_x in the mesosphere/lower thermosphere region.

13 In this study the 3dCTM was used to investigate the impact of precipitating particles on O₃ inside
14 the Antarctic polar vortex at altitudes from 20 - 70 km. After a multi-year two-dimensional model
15 spin-up, two simulations from 2003 – 2009 were performed. The first run (base run) does not
16 consider any energetic particles, while the second run (EP run) includes ionisation effects by both
17 protons and electrons, using the ionisation rates provided by the Atmospheric Ionisation Module
18 Osnabrück (AIMOS; Wissing and Kallenrode 2009). The resulting NO_x production per created ion
19 pair includes various ionic and neutral reactions depending on the atmospheric background state
20 (Nieder et al., 2014). Simple parameterisations are used for the production of HO_x (Solomon et al.,
21 1981) and O (Porter et al., 1976). Note that heterogeneous chemistry was not included, which only
22 becomes important during spring in the lower stratosphere. Both model runs considered constant
23 solar minimum conditions ($F_{10.7} = 70 \cdot 10^{-22} \text{ W m}^{-2} \text{ Hz}^{-1}$) to exclude O₃ variations due to solar
24 activity. The obtained O₃ model results of both runs were separately selected according to the
25 vertical MIPAS retrieval grid for direct comparisons to the observations, repeating the described
26 algorithm to calculate the 26-day running means. Finally, in order to derive the O₃ vmr variations
27 solely originating from precipitating particles, the obtained averages of the base run were subtracted
28 from the corresponding O₃ values of the EP run. The results were divided by the arithmetic mean of
29 both runs and eventually multiplied by 100%.

30

31 **3 Results and discussion**

1 **3.1 Satellite observations**

2 **3.1.1 O₃ response from 2002 - 2011**

3 The 26-day O₃ vmr averages from 2002 - 2011 of each altitude-time interval (1 April – 1 November,
4 20 - 70 km) were individually grouped into years of high and low index activity. For this purpose
5 the index median of the corresponding time series of the 26-day average of an index (F10.7, Ap,
6 2MeV) centred around 1 April was calculated, only including years of actually available O₃
7 observations. Therefore the median of an index time series works as a threshold, dividing the entire
8 time interval from 2002-2011 in years of high (above the median) and low (below the median)
9 index activity. Note that the classification of the years does not only depend on the chosen index,
10 but due to data gaps also on the considered height-time interval as well as the instrument used.
11 Afterwards the arithmetic O₃ mean of the years of low index activity was subtracted from the O₃
12 mean of the years of high index activity, eventually dividing this absolute O₃ difference by the
13 arithmetic O₃ average of the entire observation period and multiplying the results by 100% for more
14 handy values. Thus the calculated relative O₃ difference (referred to as O₃ amplitude here)
15 represents the impact of the respective index on the O₃ background. To reduce the measurement
16 noise of the individual instruments, the results of all three instruments were merged by simply
17 calculating the arithmetic average but only if the corresponding O₃ amplitude of all three
18 instruments was available. Note that due to the major sudden stratospheric warming centred around
19 27 September (Azeem et al., 2010) the O₃ observations from 1 September - 1 November 2002 were
20 excluded. In contrast, the solar proton event in the end of October 2003 (Jackman et al., 2005) was
21 neglected due to its late occurrence. The performed analyses with O₃ observations, considering the
22 indices from 1 May and 1 June (not shown here), revealed no essential differences compared to 1
23 April or the structures became less obvious. Comparisons with earlier periods are not reasonable
24 because the vortex first builds up in April. Therefore the focus is set on the O₃ response to indices
25 centred around 1 April. The O₃ amplitude was calculated for all three regions (CORE, EDGE, and
26 OUTSDIE) which were introduced in Sect. 2.1. The corresponding results reveal that the pattern
27 found inside the EDGE region are fairly similar and less noisy compared to the features observed in
28 the CORE area (not shown here). In contrast the O₃ amplitudes outside the Antarctic polar vortex
29 are fundamentally different. An example for the O₃ response associated to 1 April Ap in the EDGE
30 and the OUTSIDE region derived from MIPAS measurements is presented in Fig. 2, showing
31 considerably disagreeing structures and essentially weaker amplitudes, especially below 50 km.
32 Thus comparison between the individual regions of the Southern Hemisphere ensures, that, the

1 pattern found in the EDGE region are actually originating from the Antarctic polar vortex.
2 Figure 3 displays the corresponding results of the O₃ amplitude from 2002 - 2011, but only for
3 values above the significance level of 95% while shaded areas show regions between the
4 significance level of 95% and 99%. The significance was calculated according to a Student's t-test,
5 based on the error of the mean of the 26-day running O₃ means and assuming the worst case
6 scenario of absolute error propagation. The MIPAS O₃ measurements (left column) reveal a high
7 negative response to Ap (upper row) in early Antarctic winter >60 km, on average ranging around
8 -10%. Further striking negative O₃ amplitudes occur in July between 30 and 40 km as well as
9 around 1 October at ~30 km, at least weakly indicating the downward transport of the Ap signal in
10 stratospheric O₃ due to NO_x predicted by model studies (e.g. Reddman et al., 2010). In contrast, a
11 positive O₃ amplitude is found at the beginning of the winter between ~25 km and ~55 km (~10 –
12 20%), as well as at altitudes <30 km throughout the winter (up to ~20% in October at ~20 km) and
13 above the indicated subsiding layer of negative amplitudes. But considering that most of these
14 features drop below the significance level of 95% by combining the data of all three instruments
15 (right column), a more detailed investigation of these patterns is not reasonable. However, the
16 results of the merged data set show a well pronounced subsiding negative Ap signal from ~50 km in
17 June down to ~25 km in October, which is disrupted in August, while the generally positive
18 structures below 30 km are also still present.

19 The O₃ response to 2 MeV (middle row) derived from MIPAS observations also indicates a
20 downwelling of negative O₃ amplitudes, descending from ~60 km in June down to ~30 km in late
21 August. Additionally, the MIPAS O₃ response to 2MeV in early winter is reversed compared to the
22 corresponding influence from Ap on O₃, which does not originate from missing 2MeV data from
23 2011. Strong positive O₃ amplitudes are generally observed throughout the winter below 30 km,
24 exceeding values of ~20% in April and October, as well as during October between 30 km and 50
25 km where the maximum amplitude is lower (~10%). The positive features can be validated with the
26 composite results even if they are damped in the region below 30 km. However, this is not the case
27 for the negative response, except for a small area in June in the lower mesosphere. Considering that
28 the Ap responds to lower particle energy levels compared to 2MeV and that the behaviour of both
29 indices is essentially different from 2002 - 2010 (see Fig. 4), the different O₃ amplitudes associated
30 to Ap and 2MeV are still reasonable.

31 The O₃ response to F10.7 (lowermost row) is fairly similar between MIPAS and the merged
32 measurements, and both also agree with the respective pattern observed for Ap, including the
33 indicated downwelling of negative O₃ amplitudes during midwinter from 50 to 25 km. The

1 composite O₃ shows strong positive amplitudes in May >55 km which originate from SMR
2 measurements and are most likely due to the low vertical resolution of the SMR instrument at these
3 altitudes (see Sect. 2.2.3). The high agreement between the results of Ap/O₃ and F10.7/O₃ might
4 originate from the coupling of both indices during solar maximum years (Gray et al., 2010, their
5 Fig. 1). In order to investigate a possible cross-correlation between solar radiation and geomagnetic
6 disturbances, the analysis was repeated for years of moderate solar activity, only including 2005 –
7 2010 (Fig. 4). Similar analyses to extract a more distinct solar signal during times of approximately
8 constant geomagnetic activity were not reasonable, because the respective years of nearly constant
9 Ap values (2002, 2005, 2006, 2008, 2010) do not provide a sufficient amount of data in MIPAS and
10 SMR measurements.

11 **3.1.2 O₃ behaviour during solar minimum activity (2005 - 2010)**

12 Figure 5 displays the obtained O₃ amplitudes for solar quiet times (2005 – 2010) associated to 1
13 April Ap, again only showing values above 95% significance level and shading the area of regions
14 between 95% and 99%. The MIPAS O₃ response to Ap indicates a subsiding negative signal (~-10
15 to -15%), starting in late June slightly below 50 km and propagating downwards to ~25 km
16 throughout the winter. However, the middle part of the downwelling between late July and late
17 September is below the significance level of 95% and therefore not shown here. Furthermore, the
18 hinted subsidence is closely surrounded by well pronounced positive O₃ amplitudes, especially
19 below ~30 km which maximise in September (>20%). There is also a negative structure centred at 1
20 June at ~60 km, which cannot be caused by NO_x but most likely results from HO_x formation (see
21 Sect. 1). Considering the composite results, the downwelling Ap signal in O₃ becomes apparent and
22 robust but slightly weaker (~-10%) while the positive features are also damped but still present. The
23 mesospheric response is generally weak and the high positive O₃ amplitudes in May are again
24 caused by the SMR measurements.

25 The 2MeV impact on MIPAS O₃ shows generally agreeing features with the influence of the
26 geomagnetic activity and is also of similar magnitude, however, the downwelling negative signal is
27 hinted to already start in late May at ~55 km. In contrast to the O₃ response to Ap, the downwards
28 propagating 2MeV signal is less robust and can be only guessed in the composite O₃ amplitude,
29 while the positive structures (~10 - 15%) in August below 30 km and in September between 30 km
30 and 50 km are still present. In general, the 2MeV features are less obvious in the O₃ composite,
31 except for the positive O₃ amplitudes above the hinted downward transport. Nevertheless, the
32 agreement between Ap and 2MeV pattern is quite strong, in MIPAS observations in particular,

1 although both parameters are only indirectly related to O₃. However, the O₃ structure associated to
2 both indices is far too similar and additionally found in all three instruments to be a coincidence,
3 even if the descending O₃ response to 2 MeV is weaker. Since Ap represents lower particle energy
4 levels compared to the 2MeV and both indices are only moderately correlated (see Fig. 4), the
5 similar results strongly indicate a related source mechanism, suggesting solar wind variability.

6 Considering the entire process, that energetic particles produce NO_x which eventually destroys
7 stratospheric O₃, the Ap impact observed in O₃ (see Fig. 5) is expected to be reversed in NO_x, at
8 least in the stratosphere. In order to investigate this in more detail, the analysis was repeated for 1
9 April Ap and NO_x. Here NO_x is represented only by NO₂ from MIPAS observations, because the
10 respective NO measurements are quite noisy compared to NO₂, especially below 30 km. This is still
11 reasonable because NO is converted to NO₂ during night and therefore NO₂ is the major fraction of
12 NO_x inside the Antarctic polar vortex. The corresponding results include the years 2005 – 2010 and
13 are displayed in Fig. 6, supporting that the stratospheric O₃ depletion can be indeed associated to the
14 catalytic NO_x/O₃ cycle. The Ap signal in NO₂ is stronger by the factor of 2 - 5, compared to the
15 respective O₃ amplitudes. The sharp gradient in mid July originates from 2005 NO₂ data, which are
16 not available afterwards. However, the general structure of the subsiding Ap signal in NO₂ is still
17 similar with and without 2005 observations. Note that the essentially smaller NO₂ amplitudes in
18 October below the significance level of 95% are not in conflict with the respective well pronounced
19 negative O₃ response, because the latter one results from an accumulation effect from the NO₂
20 above. Furthermore, large negative NO₂ amplitudes throughout the entire winter below ~30 km are
21 observed, matching the high positive O₃ response to Ap. A possible reason for this behaviour might
22 be that NO₂ is stored in reservoir species, like ClONO₂, HNO₃, and N₂O₅, due to reactions with ClO,
23 OH, and NO₃, respectively. However, N₂O₅ is converted to HNO₃ via water ion cluster chemistry
24 (López-Puertas et al., 2005, their reactions 1 and 8 – 12), which was also investigated with respect
25 to EPP for conditions without solar proton events by Stiller et al. (2005). These reactions
26 eventually lead to lower NO_x concentrations, consequently slowing down the catalytic O₃ depletion.
27 Based on the corresponding MIPAS climatologies (not shown here), HNO₃ is more important until
28 mid July, while ClONO₂ is dominating afterwards and its influence becomes essentially crucial in
29 spring due to heterogeneous chemistry which has taken place before. This suggested NO₂-ClONO₂
30 mechanism is supported by Whole Atmosphere Community Climate Model results reported by
31 Jackman et al. (2009, their Fig. 6 and 7), who simulated the impact of the SPE in July 2000 on
32 stratospheric O₃ and NO_y (= NO_x + NO₃ + N₂O₅ + HNO₃ + HO₂NO₂ + ClONO₂ + BrONO₂).

33 Furthermore, the positive O₃ amplitudes below ~30 km could be also partly explained by the self

1 healing effect of O₃ (Jackman and McPeters, 1985). Altitude regions of reduced O₃ will lead to
2 increased solar UV radiation in the layers directly below. This is accompanied by a higher
3 production of atomic oxygen and would consequently increase the formation of O₃. However, this
4 proposed mechanism would only have an additional effect, contributing to the formation of O₃ in
5 the atmospheric layer right below the subsidence, but cannot account for the entire region. Note that
6 this layer is also present throughout the entire winter, and thus an influence from the vortex above is
7 unlikely but any further investigations are beyond the scope of this study.

8 Additionally, the area of high positive Ap/O₃ structure between 35 km and 50 km from August -
9 September cannot be completely explained by the NO_x/O₃ cycle. In detail, the respective Ap
10 influence of NO₂ is close to 0 and consequently well below the 95%, while the respective MIPAS
11 ClONO₂ amplitude (not shown here) reveals positive values, which are also mostly below the 95%
12 significance level. These results are at least not in conflict with a higher O₃ amplitude. Furthermore,
13 this positive Ap impact on O₃ is essentially less visible in the composite results than in MIPAS data,
14 and a corresponding composite analysis for Ap/NO₂ is necessary for a more detailed investigation.
15 But this is not possible due to non-existing NO₂ measurements from SABER and SMR. Thus no
16 definite explanation can be given at this state and this feature is a subject of a future work.
17 However, it should be pointed out that this structure does not harm the underlying mechanism
18 proposed to explain the identified negative O₃ amplitude and subsequent downward transport.

19 **3.2 Comparison with 3dCTM**

20 The simulated O₃ amplitude between the EP run and base run, representing high and low
21 geomagnetic activity, respectively, is displayed in Fig. 7. Note that the modelled O₃ amplitude is
22 also referred to as O₃ amplitude here, which is justified because “observed” and “modelled” O₃
23 amplitude still hold the same physical meaning, even if the calculation algorithm is slightly
24 different. It is reasonable to investigate the complete simulated time interval from 2003 – 2009,
25 because the model runs represent solar minimum conditions similar to the years 2005 - 2010. The
26 results reveal apparent negative O₃ amplitudes propagating downwards throughout the winter with
27 maximum negative values during midwinter between 45 km and 60 km. The subsidence shows
28 larger negative O₃ amplitudes compared to the measurements and is also much broader, which
29 might be due to the constant F10.7 and the prescribed dynamics, both reducing the inter-annual
30 variability of O₃. Furthermore, we performed an on/off experiment, while in reality the EEP indirect
31 effect is a persistent feature. Below 30 km the observed high positive O₃ amplitudes associated to
32 Ap are only indicated in the model results by essentially weaker and additionally negative

1 amplitudes. However, the model amplitudes are at least less negative compared to the values above.
2 The second positive region above the downwelling is completely missing. Further note that the
3 strong positive response during late winter/early spring below 30 km might not be reproduced by
4 the model due to missing heterogeneous chemistry. The proposed self healing effect of O₃ (see Sect
5 3.1.2) was also tested, using O¹D as a proxy for the O₃ photolysis rate in the Lyman-alpha band and
6 calculating the O¹D amplitude (not shown here). However, the expected positive response directly
7 below the downwelling is only partly visible and even below the 67% significance level.

8 The qualitative agreement between model results and observations in the stratosphere suggests that
9 the subsiding Ap signal found in O₃ is actually originating from particle precipitation. However, the
10 simulated downwelling starts at altitudes >60 km while observations reveal no obvious structures in
11 the mesosphere, possibly caused by satellite sampling. As already stated in Sect 3.1.2, the
12 mesospheric behaviour cannot be caused by NO_x, because the NO_x/O₃ cycle is not efficiently
13 working at these altitudes. Thus the O₃ depletion >50 km could be accounted to OH production,
14 which is most likely overestimated in the model and consequently leads to an increased O₃
15 depletion not observed by the satellite instruments.

16

17 **4 Conclusions**

18 We have investigated the O₃ behaviour inside the Antarctic polar vortex from 2002 - 2011, observed
19 by three independent satellite based instruments ENVISAT/MIPAS, Odin/SMR, and
20 TIMED/SABER. These O₃ vmr measurements, based on 26-day running means from 1 April – 1
21 November covering altitudes from 20 – 70 km, were individually grouped into high and low index
22 activity according to the 26-day averages centred around 1 April, 1 May, and 1 June of different
23 solar and geomagnetic indices (F10.7, Ap, 2MeV). After minimising the direct influence of the solar
24 radiation by only considering the period of solar minimum activity from 2005 – 2010 we found a
25 negative O₃ response caused by geomagnetic activity (Ap) from 1 April in all three instruments,
26 ranging from -5% to -10% and propagating downwards throughout the Antarctic winter from ~50
27 km down to ~25 km. This subsiding negative signal in O₃ is above the significance level of 95%
28 and overlaps with the corresponding positive NO₂ response to 1 April Ap, supporting that NO_x is
29 indeed the cause of the O₃ depletion. We could also show that the high positive O₃ response below
30 30 km, which is present during the entire winter, is in agreement with respective negative NO₂
31 structures. The cause of the NO₂ behaviour is possibly related to the formation of the reservoir
32 species ClONO₂ and HNO₃, slowing down the catalytic destruction of O₃ by Cl. The O₃ pattern
33 induced by the magnetosphere (2MeV) from 1 April are similar but weaker, compared to the

1 respective geomagnetic activity, still suggesting a related source mechanism between 2MeV and Ap
2 like solar wind variability. The composite observations of all three instruments are in good
3 qualitative agreement with 3dCTM simulation, revealing similar O₃ pattern induced by the
4 geomagnetic activity from 1 April while the simulated O₃ response is larger but still in the same
5 order of magnitude.

6 However, we have to point out that the validity of the subsiding O₃ depletion associated to
7 geomagnetic activity and NO_x is not ensured due to the short time series of only 6 years at most.
8 Thus, we conclude that precipitating particles are strongly indicated as a factor contributing to
9 stratospheric O₃ during Antarctic winter, but we cannot prove the link unambiguously.

10

11 **Authors' contribution**

12 T. F. analysed the satellite and indices data and wrote the final script. G. S., J. U.+K. P., and M. M.
13 provided the O₃ data from ENVISAT/MIPAS, Odin/SMR, and TIMED/SABER, respectively, and
14 all of them contributed to interpretation. H. N. performed the 3dCTM simulations. M. S. initiated
15 the study and contributed to interpretation.

16

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26

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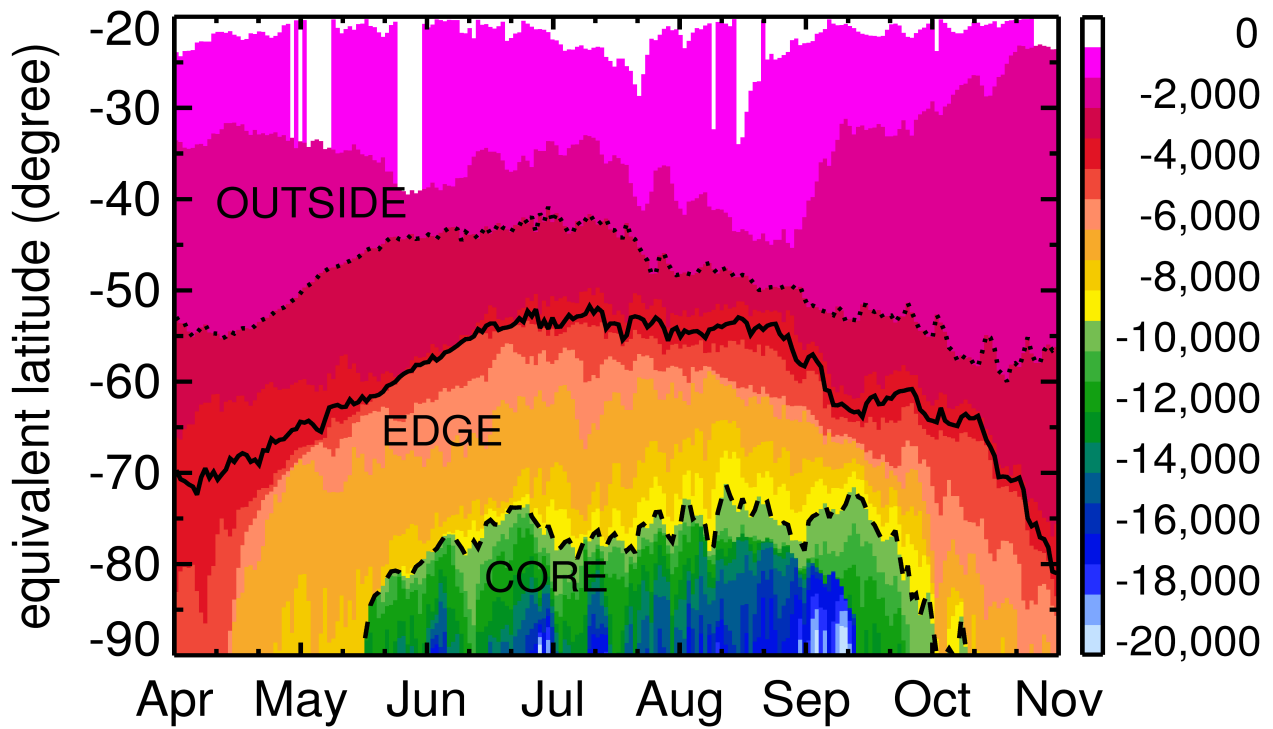
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1 Table 1. Limits, derived from the potential vorticity ($10^{-6} \text{ K m}^2 \text{ s}^{-1} \text{ kg}^{-1}$), of the southern hemispheric
 2 regions CORE, EDGE, and OUTSIDE at the individual heights. The altitudes are adapted from the
 3 MIPAS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

nominal height (km)	CORE	EDGE	OUTSIDE	nominal height (km)	CORE	EDGE	OUTSIDE
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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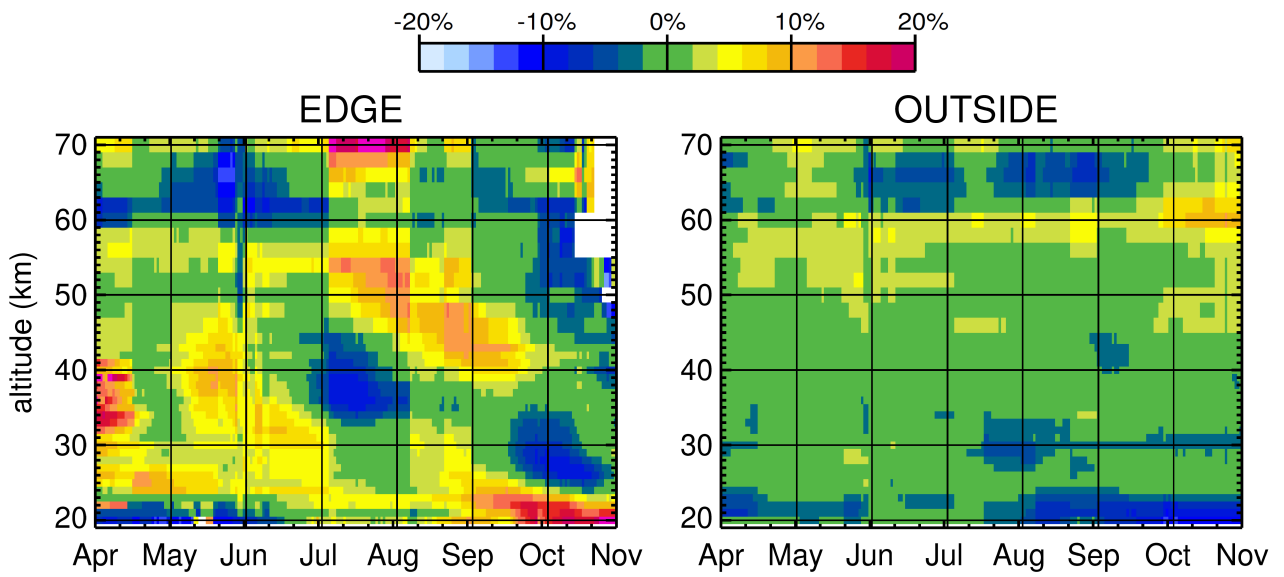
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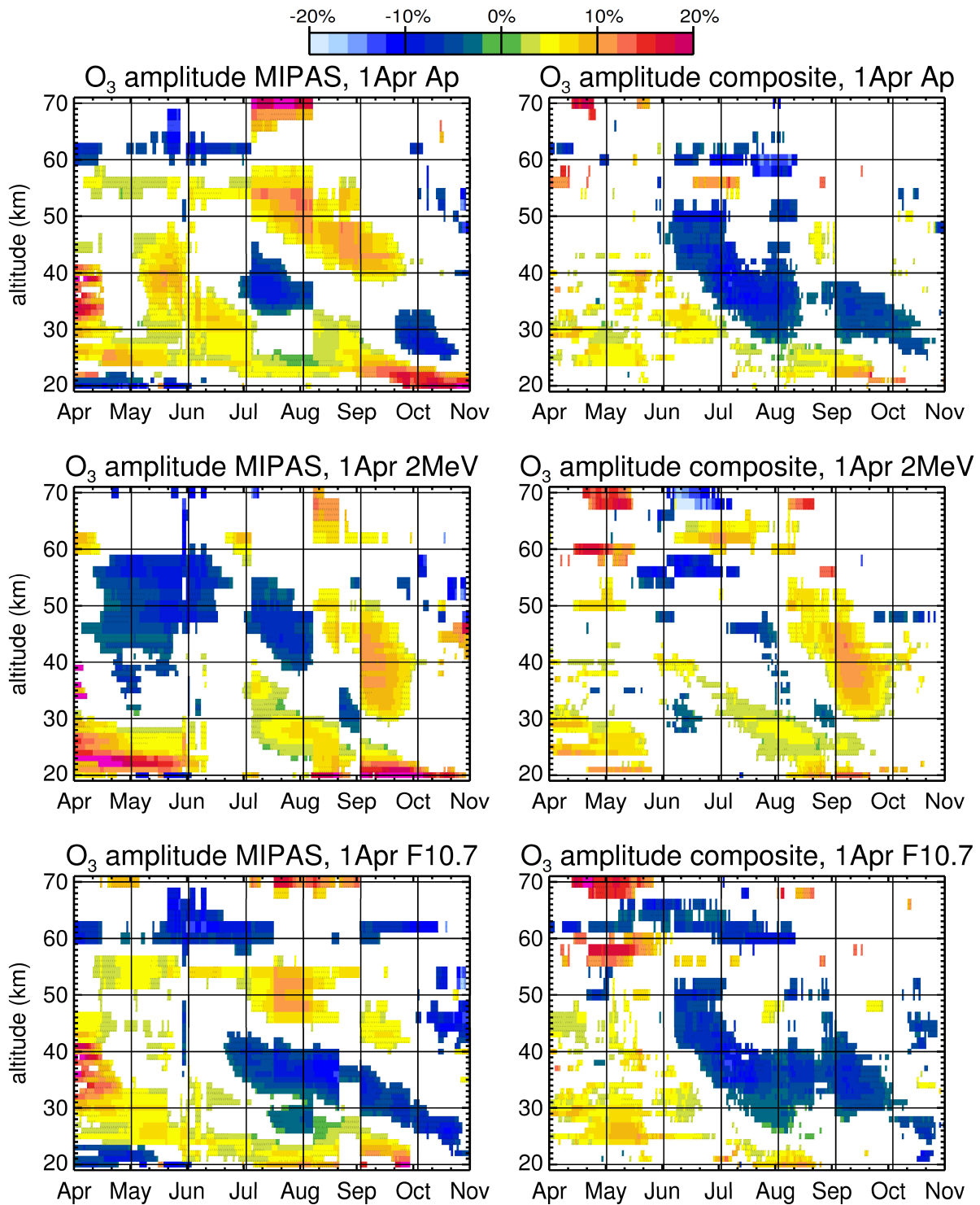
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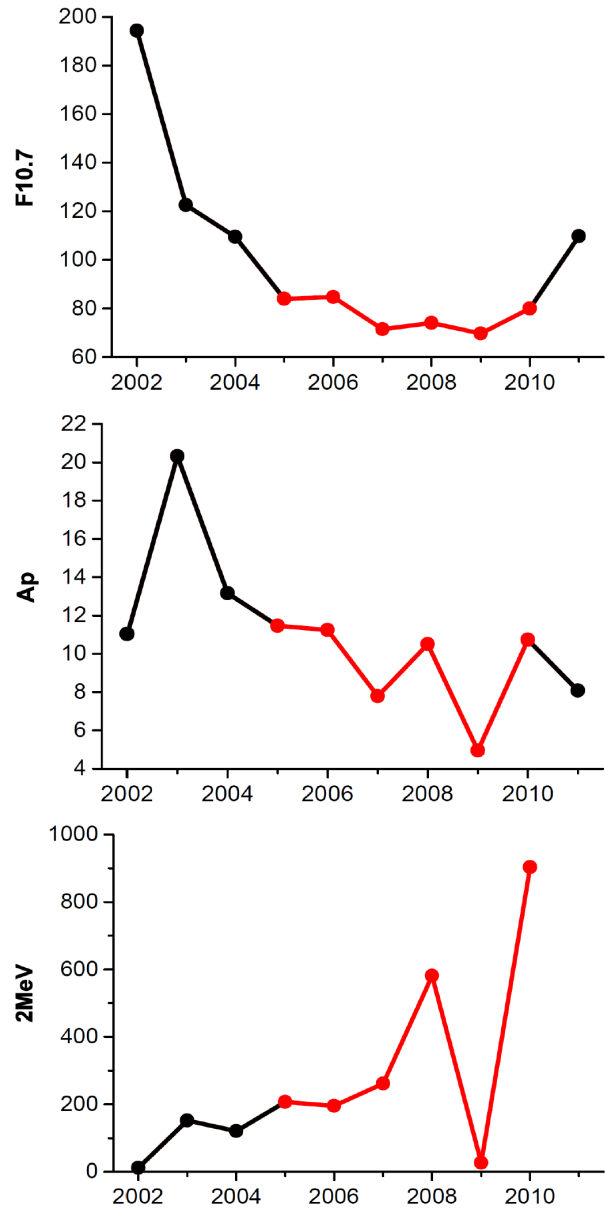


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 2 Figure 2. Example of the O₃ amplitude (see Sect. 3.1.1 for definition) observed by MIPAS from
 3 2002 – 2011 between years of high Ap index and years of low Ap index centred around 1 April, for
 4 the regions EDGE (left) and OUTSIDE (right).



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2 Figure 3. O₃ amplitude (see Sect. 3.1.1 for definition) inside the Antarctic polar vortex between
 3 years of high index values and years of low index values, namely Ap index (upper row), ≥ 2 MeV
 4 electron flux (middle) as well as F10.7 cm solar radio flux (lowermost row) centred around 1 April,
 5 derived from MIPAS (left column) and composite (MIPAS+SMR+SABER, right column)
 6 observations from 2002 – 2011. Shown are only values above the significance level of 95%.
 7 Additionally, regions between the significance level of 95% and 99% are shaded, according to a
 8 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 ≥ 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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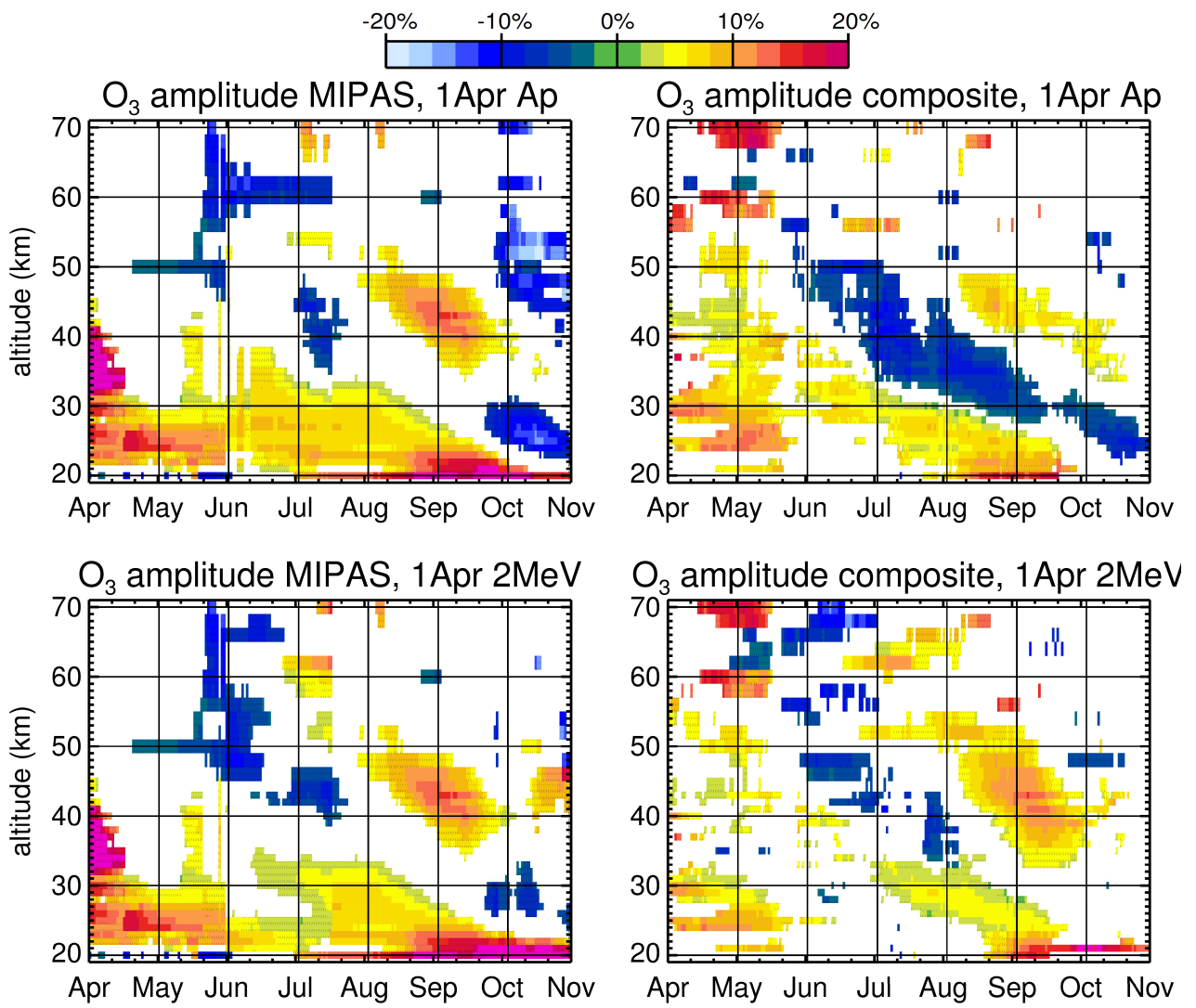
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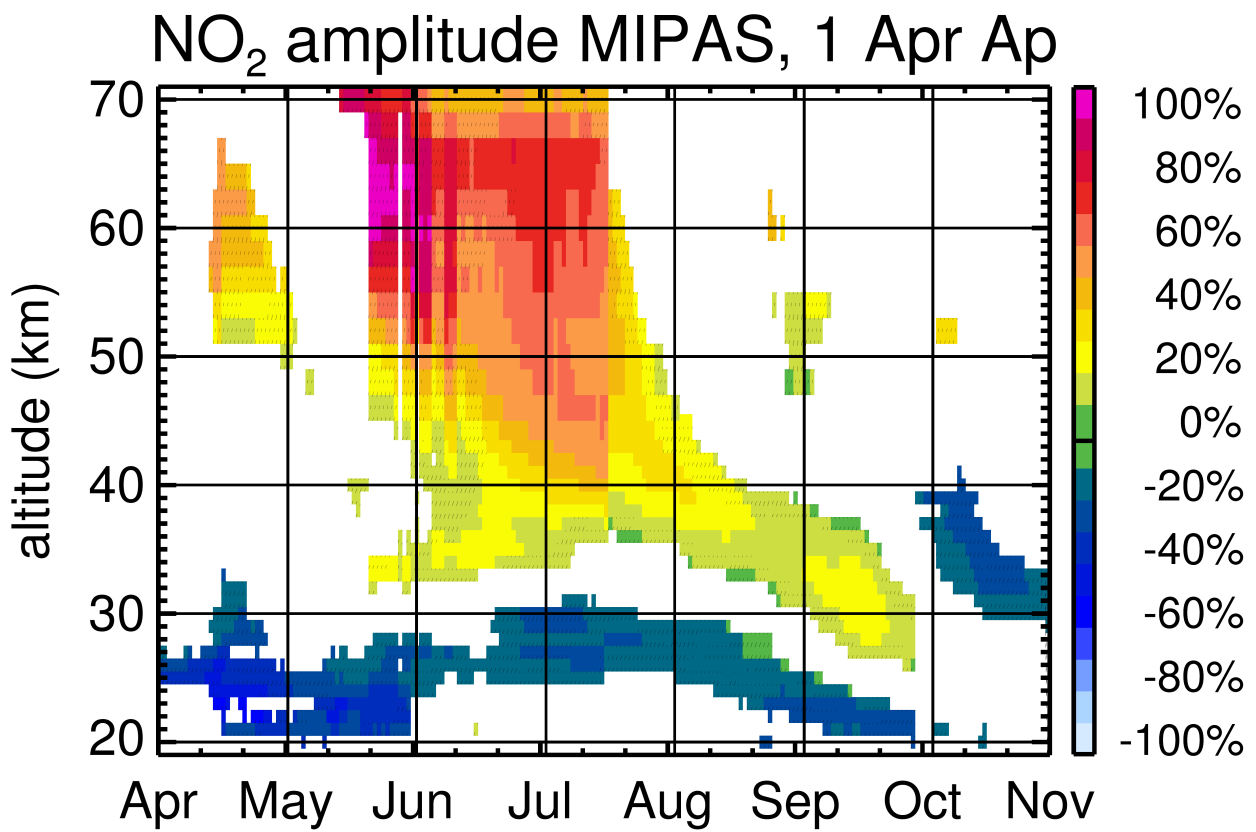
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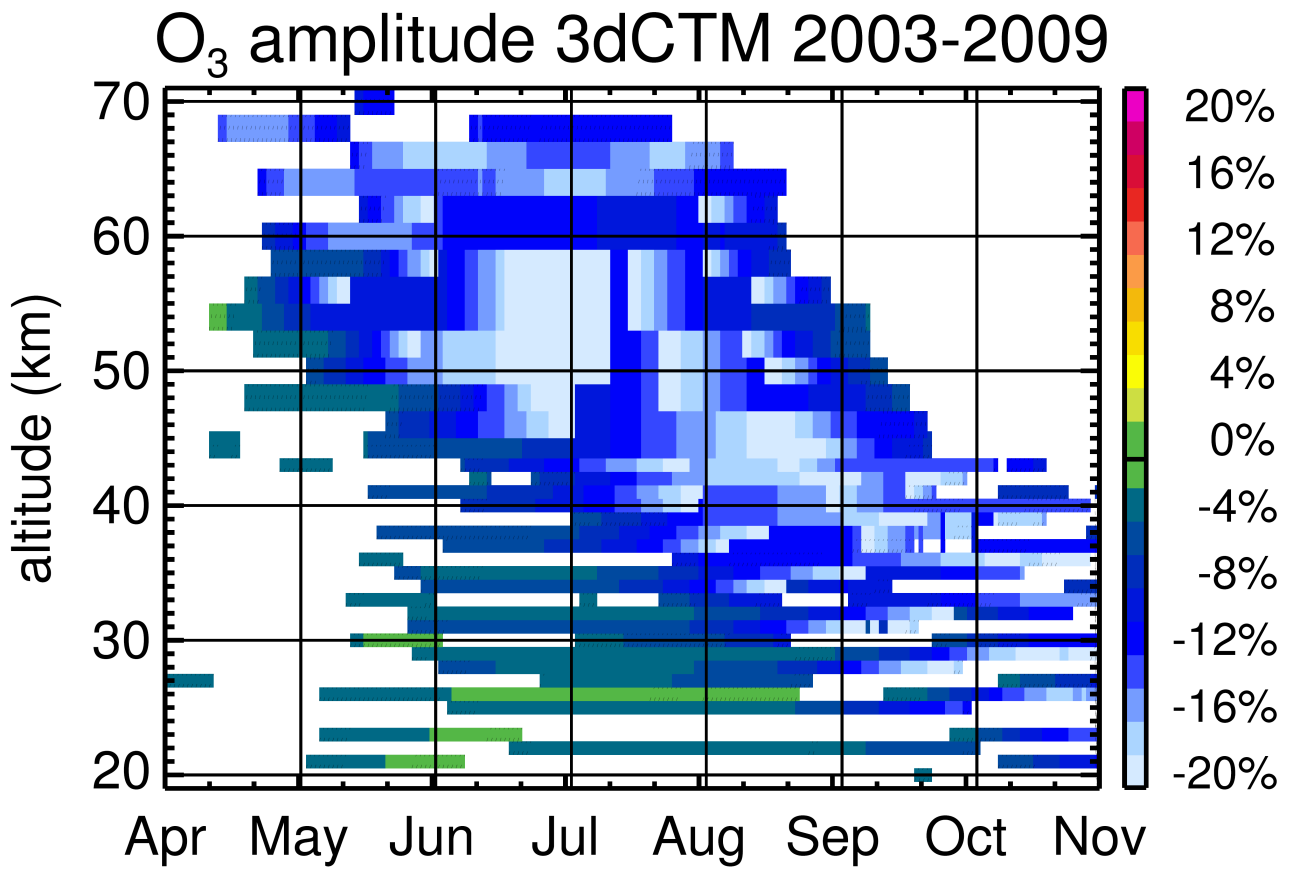


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 2 Figure 5. Same as Figure 3 but only for Ap index (upper row) and ≥ 2 MeV electron flux (lower
 3 row) from 2005 – 2010.



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Figure 6. Same as Figure 3, but only for the NO₂ amplitude associated to 1 April Ap index. The NO₂ was derived from MIPAS observations from 2005 – 2010.



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- 2 Figure 7. O₃ 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.