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# Antarctic ozone loss in 1979–2010: first sign of ozone recovery

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**Abstract.** A long-term ozone loss time series is necessary to understand the evolution of ozone in Antarctica. Therefore, we construct the time series using ground-based, satellite and bias-corrected multi-sensor reanalysis (MSR) data sets for the period 1989–2010. The trends in ozone over 1979–2010 are also estimated to further elucidate its evolution in the wake of decreasing halogen levels in the stratosphere. Our analysis with ground-based observations shows that the average ozone loss in the Antarctic is about  $-33$  to  $-50\%$  ( $-90$  to  $-155$  DU (Dobson Unit)) in 1989–1992, and then stayed at around  $-48\%$  ( $-160$  DU). The ozone loss in the warmer winters (e.g. 2002 and 2004) is lower ( $-37$  to  $-46\%$ ), and in the very cold winters (e.g. 2003 and 2006) it is higher ( $-52$  to  $-55\%$ ). These loss estimates are in good agreement with those estimated from satellite observations, where the differences are less than  $\pm 3\%$ . The ozone trends based on the equivalent effective Antarctic stratospheric chlorine (EEASC) and piecewise linear trend (PWLT) functions for the vortex averaged ground-based, Total Ozone Mapping Spectrometer/Ozone Monitoring Instrument (TOMS/OMI), and MSR data averaged over September–November exhibit about  $-4.6$  DU yr<sup>-1</sup> over 1979–1999, corroborating the role of halogens in the ozone decrease during the period. The ozone trends computed for the 2000–2010 period are about  $+1$  DU yr<sup>-1</sup> for EEASC and  $+2.6$  DU yr<sup>-1</sup> for the PWLT functions. The larger positive PWLT trends for the 2000–2010 period indicate the influence of dynamics and other basis functions on the increase of ozone. The trends in both periods are significant at 95% confidence intervals for all analyses. Therefore, our study suggests that Antarctic ozone shows a significant positive trend toward its recovery, and hence, leaves a clear signature of the successful implementation of the Montreal Protocol.

## 1 Introduction

Ozone loss in the Antarctic stratosphere has been an issue of intense research since its discovery in the 1980s (Farman et al., 1985). Several estimates of ozone loss are available for Antarctica since then. However, most of them deal with the ozone loss analysis for individual winters, modelled or incomplete due to limitations of the analysed observations (e.g. Austin et al., 2010; Lemmen et al., 2006; Tilmes et al., 2006; Hoppel et al., 2005), and thus this makes the inter-annual comparison very difficult. For instance, the chemistry–climate model (CCM)-based studies are mostly exploited for the projection of ozone recovery (e.g. Austin et al., 2010). Although there are many studies using satellite data, a continuous long-term ozone loss analysis is still not available using these data (Bevilacqua et al., 1997; Hoppel et al., 2005; Tilmes et al., 2006). Therefore, we present a comprehensive ozone loss analysis in the Antarctic using ground-based and satellite measurements for the 1989–2010 period, similar to that in the Arctic (Goutail et al., 2005). In this we use the same model, measurements, and method to construct the whole time series, which makes a continuous, coherent and comparable long-term analysis. This analysis can also be regarded as an extension of the study of Huck et al. (2007), who presented an ozone loss analysis using the total column for the 1992–2004 period. The passive technique is used to derive the ozone loss from observations, in which the contribution from transport is separated from the photochemical ozone loss. A detailed description of this approach (e.g. Goutail et al., 1999) and its application to the Antarctic winters 2005–2009 can be found in Kuttippurath et al. (2010).

Many studies have already discussed the trends of ozone in the Antarctic stratosphere. For instance, a study by Yang et al. (2008) discussed the trends in Antarctic ozone using ground-based and satellite measurements and showed a trend of around  $-4.5$  DU (Dobson Unit)  $\text{yr}^{-1}$  during 1978–1996 and an insignificant positive trend thereafter. Similar trends were also estimated by Wohltmann et al. (2007), who applied a multi-variate regression model to the total ozone measurements. A study by Hassler et al. (2011) showed the stabilisation of ozone loss rates at South Pole over 1991–2009. A recent work by Salby et al. (2011) reported a significant positive trend in the September–November Total Ozone Mapping Spectrometer/Ozone Monitoring Instrument (TOMS/OMI) ozone during 1997–2009. However, we here use two different regression methods to estimate the trends in ozone during 1979–2010. Furthermore, we consider the measurements inside the vortex and use two additional satellite data sets to check the robustness of the derived trends. These data and approach have hitherto not been used for trend studies for this region, which is the significance of this diagnosis.

The plan of this paper is as follows: the data used for the analyses and the method applied to the ozone loss derivation are given in Sect. 2. The ozone loss estimates for the Antarctic are presented in Sect. 3.1. Section 4.1 assesses the derived ozone loss and its inter-annual variability. The ozone trends in the Antarctic are analysed in Sect. 4.3. Section 5 concludes the study with the main findings.

## 2 Data and methods

### 2.1 Ground-based observations

We use measurements from 12 ground-based stations deployed in and around the continent, such that they cover the entire region to provide a representative analysis for Antarctica. Figure 1 shows the positions of the stations and Table 1 shows further details about them. As the Antarctic vortex is stable and inter-annual variations in the meteorology are small compared to those of the Arctic, the estimated ozone loss is less dependent on the selection of the stations, as demonstrated in Kuttippurath et al. (2010). The analysis for each year contains data from at least eight stations. The SAOZ data used are the zenith sky sunrise and sunset measurements between  $86$  and  $91^\circ$  solar zenith angle (SZA) (Pommereau and Goutail, 1988) from Dumont d'Urville, Concordia (Dome C), Faraday/Vernadsky (after 1996) and Rothera. These measurements in the visible region have the advantage of early winter observations even inside the polar circle, which is not possible by most satellite or other ground-based instruments. The measurements have a random error of  $4.7\%$  and systematic error of  $3.6\%$  (Hendrick et al., 2011) and different SAOZ slant column measurements are consistent within  $\pm 3\%$  (Roscoe et al., 1999). The ozone observations at Neumayer station are performed by the DOAS, and

the random error of these measurements is about  $2\%$  (Frieß et al., 2005). In addition, the Dobson measurements (Dobson, 1957) performed at Arrival Heights, Faraday/Vernadsky (prior to 1996), Halley, Marambio, South Pole (Amundsen-Scott) and Syowa, and the Brewer measurements (Brewer, 1973) from Belgrano (MK IV) and Zhongshan (MK IV) are also considered. The random error of these observations is estimated as  $1\%$  or  $1$  DU and the total error as  $3\%$  (Basher, 1982), but these are subjected to the accuracy of absorption cross sections and a known significant temperature dependence in the ultraviolet and stray light at high SZAs (Hendrick et al., 2011), which were not taken into account for the retrievals used here. The recent studies indicate that the random errors of well-maintained Brewer observations are of the order of  $0.15$ – $0.25\%$  and the total error of about  $2.5\%$  (Scarnato et al., 2010; Kerr and McElroy, 1995).

### 2.2 Satellite observations

To compare with the ozone loss estimates from the ground-based observations, version 8.5 total column ozone measurements from TOMS onboard Nimbus-7, Meteor-3, and Earth Probe are used (Bhartia and Wellemeyer, 2002). The uncertainty of the TOMS ozone column data is  $3.3\%$ , and the bias among ozone products from the different TOMS platforms is  $1$ – $2\%$  (Kroon et al., 2008). Since 2005, the OMI data are used as the continuation of the TOMS series and the uncertainty of the OMI ozone column is  $2$ – $5\%$  for  $\text{SZA} < 84^\circ$  (Levelt et al., 2006). Therefore, a continuous series comparable to the ground-based series is available from TOMS and OMI from 1979 to 2010, with the exception of 1994 and 1995. However, as shown by Hendrick et al. (2011) there is still some bias of the order of  $2\%$  between TOMS/OMI and SAOZ observations at Dumont d'Urville, Antarctica, with a strong seasonal dependence. As this bias was random, it was not possible to correct here for the TOMS/OMI data. In addition, we have used a bias-corrected reanalysed ozone data set, the Multi-Sensor Reanalysis (MSR), compiled from various satellite observations during the period 1979–2008. The Global Ozone Monitoring Experiment 2 (GOME-2) total column data (Eskes et al., 2005) are used to extend the MSR data to 2010 for the trend analyses. These data sets have a bias of about  $1 \pm 2\%$  compared to various satellite observations (van der A et al., 2010). Previous studies have successfully used these data in various scientific studies (e.g. de Laat and van Weele, 2011).

### 2.3 Determining ozone loss inside the vortex

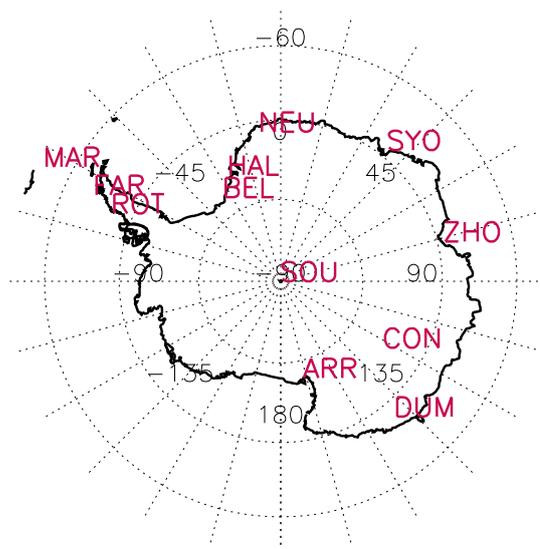
To find the amount of chemical ozone loss inside the vortex, we select the measurements using the vortex edge criterion of Nash et al. (1996) and apply the passive method (e.g. Kuttippurath et al., 2010; Goutail et al., 1999) to the selected observations. In this method, the chemical ozone loss is computed as the difference between the measured ozone

**Table 1.** Antarctic stations and their latitude (Lat.), longitude (Long.), type of instrument (Inst.), and observation period (Period) considered in this study.

Station	Lat.	Long.	Inst.	Period
South Pole	89.9° S	24.8° W	Dobson	1979–2010
Belgrano	77.9° S	34.6° W	Brewer	1992–2010
Arrival Heights	77.8° S	166.7° W	Dobson	1988–2010
Halley	75.6° S	26.8° W	Dobson	1979–2010
Concordia	75.1° S	123.4° E	SAOZ	2007–2010
Neumayer	70.7° S	8.3° W	DOAS	1999–2007
Zhongshan	69.4° S	76.4° E	Brewer	1993–2007
Syowa	69.0° S	39.6° E	Dobson	1979–2010
Rothera	67.6° S	68.1° W	SAOZ	1996–2010
Dumont d'Urville	66.7° S	140.0° E	SAOZ	1988–2010
Faraday/Vernadsky	65.3° S	64.3° W	Dobson	1979–2010
Marambio	64.2° S	56.7° W	Dobson	1987–2010

and the passive ozone tracer initialised identically to ozone at the beginning of each run (i.e. ozone–passive ozone tracer in DU and  $[100 \times (\text{ozone} - \text{passive ozone tracer}) / \text{passive ozone tracer}]$  in %). We use the REPROBUS chemical transport model (CTM) (Lefèvre et al., 1994) to simulate the passive ozone tracer from 1989 to 2010 for the loss computations. The model version used in this work has a horizontal resolution of  $2 \times 2^\circ$  on 60 vertical levels from the surface to 0.1 hPa. Our simulations use the ERA-interim meteorological data to force the model runs (Dee et al., 2011). The long-term simulations start in 1989, but we reinitialise the passive ozone tracer field on every first of June by setting the passive ozone tracer equal to the actual ozone. Note that, although the satellite measurements are available since 1979, the passive ozone tracer simulations start in 1989 due to the then availability of ERA data for our model runs and hence, our ozone loss analyses start in 1989. The ERA-interim ozone data were used for the initialisation of the model runs for each year. The passive ozone tracer columns used here are the averages within 100 km of each station.

Figure 2 illustrates the ozone loss inside the vortex from all ground-based measurements for the winter 2006. Generally, each station shows different timings for the onset, progress and maximum in the ozone loss, depending on the history of the exposure of the air parcels observed to contact with polar stratospheric clouds (PSCs) at sunlit parts of the vortex. The transport of ozone depleted air masses over the stations can also affect the onset period (Hassler et al., 2011; Kuttippurath et al., 2010). There are some variations in ozone distribution inside the vortex with two separate air masses – the edge region, with a latitudinal extent of about  $15^\circ$  around the perimeter of the vortex as identified by Roscoe et al. (2012), and the vortex core. The behaviour at any one station depends on which air mass is above it, and many stations do not have the same air mass above them throughout the ozone hole period. Faraday/Vernadsky and Rothera are most often in the edge region, with occasional excursions between the edge



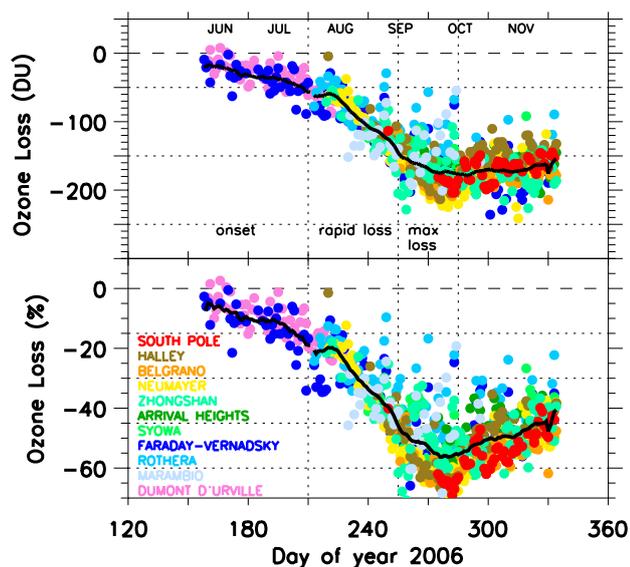
**Fig. 1.** The stations in the Antarctic from which the measurements are taken for this study. The stations are represented by their first three letters, and details are given in Table 1.

and core of the vortex. On the other hand, Dumont d'Urville is frequently outside the vortex, but occasionally inside the vortex core, and the stations at  $70\text{--}90^\circ$  S are most often inside the vortex core. A detailed discussion of the station positions and related observational features can be found in Kuttippurath et al. (2010). On average (Fig. 2 solid line), the ozone loss in the region starts by mid-June and rapidly increases to  $-160$  DU or  $-52\%$  by the end of September. The maximum ozone loss of  $-185$  DU or  $-56\%$  was observed at the end of September/early October 2006 in agreement with the minimum ozone period. The loss reduces thereafter with respect to the meteorological conditions and vortex persistence. The estimated ozone loss has an uncertainty of about 3–5% (Kuttippurath et al., 2010).

## 3 Results

### 3.1 Antarctic ozone loss

Figure 3 shows the average chemical ozone loss estimated inside the vortex from the ground-based, TOMS/OMI and MSR data during 1989–2010. This is the average ozone loss found inside the vortex from all stations, as shown by the black solid line in Fig. 2. No special scaling is performed to account for the differences in the position of the stations in the vortex, as we find the average ozone loss inside the whole vortex. Furthermore, our analysis shows insignificant differences between the loss estimated using various vortex edge criteria (inside vortex, vortex core and over the equivalent latitudes (EqLs)  $65\text{--}90^\circ$  S). The ozone loss over the whole season appears to be about the same in the edge region as in



**Fig. 2.** Chemical  $O_3$  loss estimated from observations at 11 ground-based stations inside the vortex in the winter 2006. The  $O_3$  loss is estimated as the measured  $O_3$  minus the modelled passive  $O_3$  tracer, which is initialised on first of June. The average loss estimated from the observations is shown as a solid line. The dotted lines represent  $-50$ ,  $-150$ , and  $-250$  DU and  $-15$ ,  $-30$ ,  $-45$  and  $-60\%$  of  $O_3$  loss. The vertical lines represent days 210, 255 and 285.

the core, but it is more rapid in the core and starts later. In general, the loss starts in mid-June/early July, in agreement with the appearance of PSCs and heterogeneous chlorine activation on the sunlit parts of the vortex (e.g. Solomon, 1999; Solomon et al., 1986), except during 1989–1990 where it begins in early August. On the same note, the ozone loss onset in the very cold winters such as in 2003 and 2006 is about a month earlier, in early June. Similarly, the warm winters 2002 and 1989–1990 show late onset of ozone loss. All years exhibit a higher loss rate during August–September (about  $-1.7$  DU  $\text{day}^{-1}$  or  $-0.5\%$   $\text{day}^{-1}$ ), and peak loss during the end of September and early October period. On average, the maximum loss until mid-October is around  $-120$  DU or  $-48\%$  in 1989–1990, in agreement with the lower abundances of stratospheric halogens during this period (WMO, 2011), and around  $-160$  DU or  $-48\%$  thereafter due to saturation of ozone loss (e.g. Solomon, 1999; Solomon et al., 2005; WMO, 2011), where the very cold winters (2000, 2003 and 2006) show a slightly greater reduction of about  $-170$  DU or  $-55\%$ . As anticipated, the warmer/shorter winters show the opposite pattern of ozone loss, as the  $-40\%$  loss in 2002. The TOMS/OMI and MSR analyses also show a similar evolution of ozone loss in all winters and the differences are mostly within  $\pm 3\%$ . However, the advantage of ground-based observations is that they facilitate early winter (June–August) ozone loss analyses, which cannot be captured by most satellite observations. Therefore, we have not

shown the MSR data before August, which are made from various satellite observations.

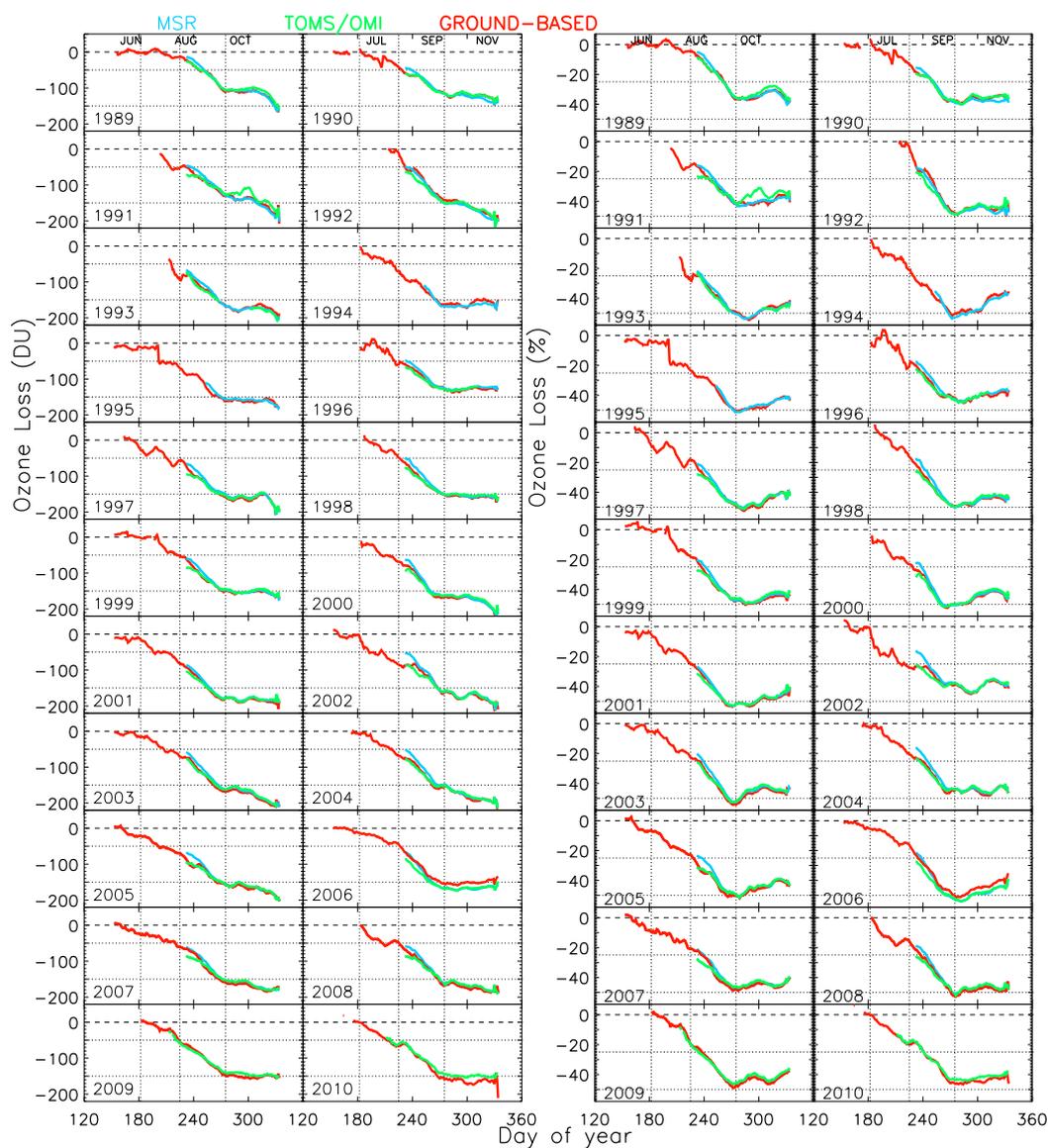
## 4 Discussion

### 4.1 Inter-annual variability of ozone loss

Figure 4 shows the inter-annual variations in the vortex averaged cumulative ozone loss deduced from the ground-based and satellite data averaged from mid-September to mid-October, and Table 2 shows the cumulative ozone loss averaged during the peak loss period: 26 September to 5 October. The ground-based estimates show a steep increase of ozone loss from  $-90$  to  $-155$  DU or from  $-33$  to  $-48\%$  between 1989 and 1994 in agreement with the increasing stratospheric chlorine during the period, but it remained around  $-160$  DU or  $-48\%$  thereafter, with the largest loss in 2006, about  $-53\%$ , and lowest loss of about  $-40\%$  in 2002. The instantaneous ozone loss rates between 13 August and 2 October, during which most ground-based instruments have measurements, are also given in the table. The loss rates show a similar time evolution, with the largest loss rates ( $-0.63$  to  $-0.67\%$   $\text{day}^{-1}$ ) in the very cold winters of 1998 and 2006. The lower ozone loss rate estimated for 2002 is due to the early vortex split during the winter, and the exceptional loss rate in 1992 is due to the additional and faster heterogeneous ozone loss on the Pinatubo aerosols (e.g. Hofmann et al., 1992). In agreement with the ozone loss analyses, the ozone values averaged over the mid-September to mid-October period show a reciprocal evolution in each winter. In sum, the evolution of ozone loss time series is in tune with the increasing stratospheric chlorine levels during the 1989–1999 period and then correlates with the inter-annual variations in temperature thereafter, as also reported in previous studies (e.g. Yang et al., 2008; Salby et al., 2011).

### 4.2 Comparison with other studies

There are two long-term ozone loss studies that can be compared to our analyses: the total column ozone loss analyses by Huck et al. (2007) and the partial column ozone loss analyses by Tilmes et al. (2006). The study by Huck et al. (2007) uses a parameterised passive ozone tracer for the calculation of the ozone loss, and they found a peak loss of around  $-120$  DU in most years, with the largest loss of about  $-130$  DU in 2001 and the lowest loss of about  $-88$  DU in 2002. These estimates are smaller than the loss estimated in our study, which could be due to the differences in passive ozone tracers used in the respective calculations. Note that when we use a passive ozone tracer computed using their formula to calculate the ozone loss from ground-based measurements, we also find relatively lower ozone loss values (of up to  $-50$  DU for the maximum loss, depending on year) than our original loss estimates using the REPROBUS passive ozone tracer.



**Fig. 3.** Temporal evolution of the chemical  $O_3$  loss estimated from ground-based observations (red) inside the vortex is compared to that from TOMS/OMI (green) and MSR (blue) in DU (left panel) and in % (right panel) for 1989–2010. The horizontal dotted lines represent  $-50$  and  $-150$  DU of  $O_3$  loss (left panel) and  $-25$  and  $-50$  % of  $O_3$  loss (right panel), while the vertical lines represent day 181, 225 and 275.

Tilmes et al. (2006) have estimated the partial column ozone loss over 380–550 K using the tracer correlation method from the satellite ozone profiles for the period 1992–2004. The peak loss deduced from the available measurements is about  $-155$  DU in 2003, and the lowest is about  $-115$  DU during 1996–1997. These values are also smaller than our ozone loss estimates, as they consider a partial column, which consists of only two-thirds of the total column ozone loss in the Antarctic (e.g. Kuttippurath et al., 2012).

### 4.3 Multi-variate regression of vortex averaged ozone

Since it is difficult to accurately estimate trends directly from ozone observations, we have constructed and applied a multi-variate regression model to the ground-based, TOMS/OMI and MSR total column data. Note that the 1994–1995 data gap in TOMS/OMI is filled with corresponding overpass analyses using the MSR data. We consider the deseasonalised September–November ozone average over 1979–2010. Apart from using various data sets for the trend analysis, we have also used various vortex definitions to group each data set, i.e. the data averaged inside the vortex, vortex

**Table 2.** The cumulative ozone ( $O_3$ ) loss estimated from 1 June averaged between 26 September and 5 October. The loss is computed as the ground-based  $O_3$  – passive  $O_3$  tracer in DU and  $100 \times (\text{ground-based } O_3 - \text{passive } O_3 \text{ tracer})/\text{passive } O_3 \text{ tracer}$  in %. The instantaneous ozone loss rates estimated between 13 August and 2 October (dates in the year covered by all measurement stations) from ground-based measurements in the Antarctic are also shown. The loss rates are given in  $\text{DU day}^{-1}$  and  $\% \text{ day}^{-1}$ . The uncertainty of the estimated  $O_3$  loss is about 3–5 %.

Year	13 Aug–2 Oct		26 Sep–5 Oct	
	Loss rate $\text{DU day}^{-1}$	Loss rate $\% \text{ day}^{-1}$	Loss DU	Loss %
1989	−1.96	−0.61	−111	−37
1990	−1.57	−0.51	−112	−37
1991	−1.57	−0.43	−132	−43
1992	−2.35	−0.80	−143	−48
1993	−1.57	−0.45	−158	−49
1994	−1.67	−0.55	−161	−51
1995	−1.76	−0.53	−160	−51
1996	−1.47	−0.45	−122	−41
1997	−1.96	−0.59	−156	−49
1998	−1.96	−0.67	−150	−50
1999	−1.96	−0.61	−152	−48
2000	−1.86	−0.51	−167	−51
2001	−1.86	−0.53	−182	−53
2002	−1.47	−0.26	−167	−40
2003	−1.86	−0.57	−168	−54
2004	−1.67	−0.47	−155	−45
2005	−1.76	−0.51	−154	−50
2006	−2.05	−0.63	−175	−55
2007	−1.96	−0.55	−159	−50
2008	−2.15	−0.55	−168	−53
2009	−1.76	−0.61	−147	−49
2010	−1.96	−0.57	−154	−46

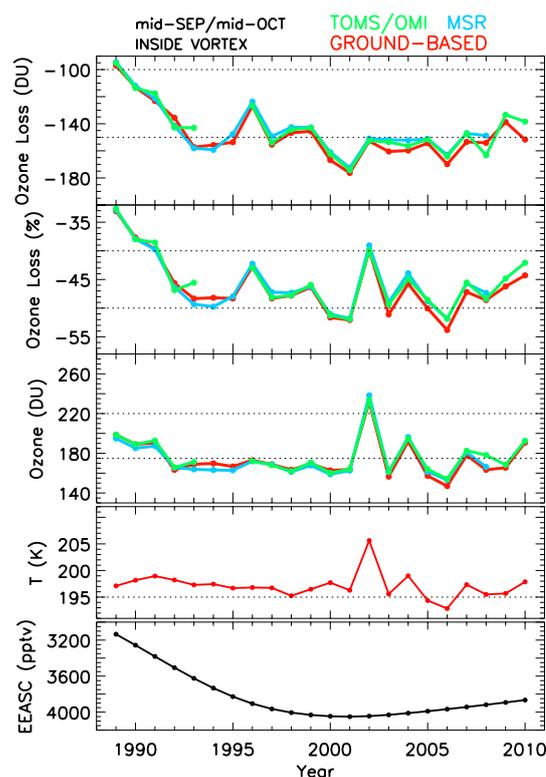
core and over 65–90° S EqL to diagnose the robustness of the derived trends.

### 4.3.1 Method and model

This model is very similar to that of Wohltmann et al. (2007) and Steinbrecht et al. (2004), where ozone ( $Y$ ) variability is expressed as:

$$\begin{aligned}
 Y(t) = & K && (\text{constant}) \\
 & +C_1t_1 && (\text{linear trend}) \\
 & +C_2t_2 && (\text{change in trend}) \\
 & +C_3(\text{SF} \times \text{QBO})(t) && (\text{solar flux} \times \text{QBO}) \\
 & +C_4\text{Aer}(t) && (\text{aerosol}) \\
 & +C_5\text{HF}(t) && (\text{heat flux}) \\
 & +C_6\text{AAO}(t) && (\text{AAO}) \\
 & +\epsilon(t) && (\text{residual})
 \end{aligned}$$

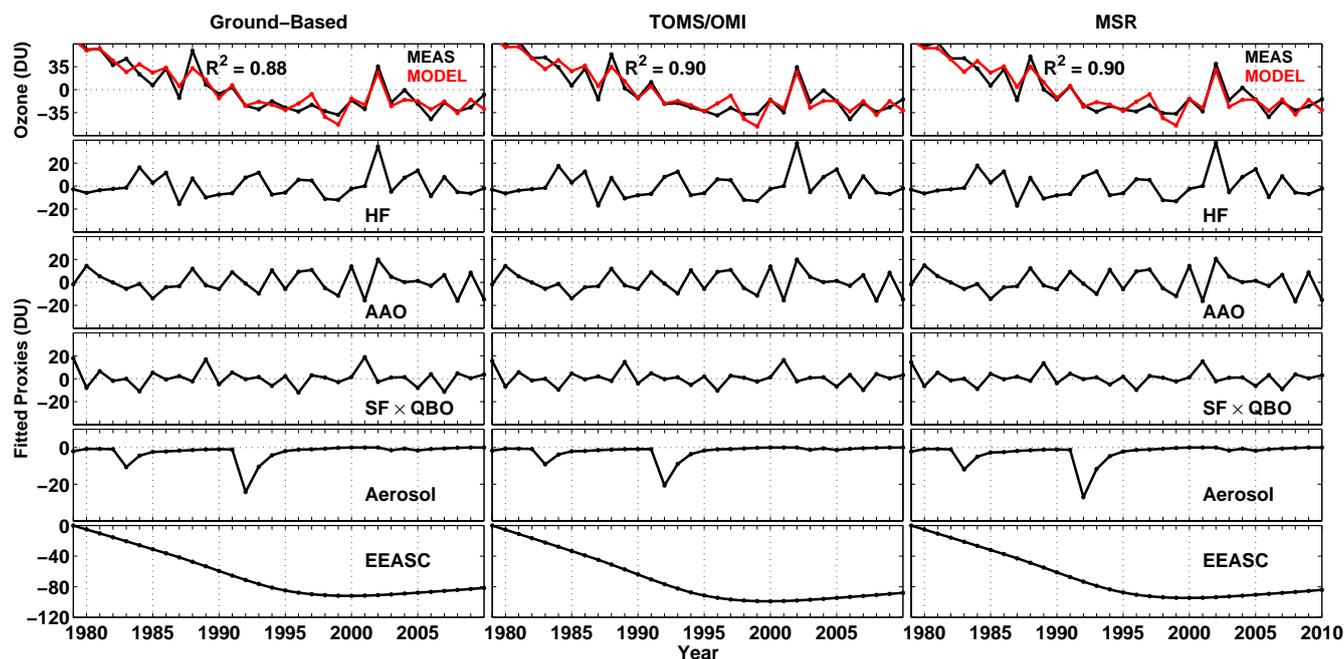
where  $t$  is time period from 1979 to 2010,  $t_1$  is the number of years from 1979 to 2010,  $t_2$  is the number of years



**Fig. 4.** The ground-based  $O_3$  (in DU) and cumulative chemical  $O_3$  loss (in DU and %) inside the vortex averaged between mid-September and mid-October during the Antarctic winters 1989–2010 compared to those of TOMS/OMI (green) and MSR (blue). The corresponding vortex averaged temperature (K) at a potential temperature of 475 K from the ECMWF (European Centre for Medium-Range Weather Forecasts) operational analyses is also shown. The equivalent effective Antarctic stratospheric chlorine (EEASC) data are shown in the inverted scale in the bottom panel. The horizontal dotted lines represent −100 and −150 DU of  $O_3$  loss (top), −40 and −50 %  $O_3$  loss (second panel from top), 175 and 220 DU of  $O_3$  (third panel from top) and 195 K temperature (fourth panel from top) in the respective plots.

from 2000 to 2010,  $K$  is a constant and  $C_1$  to  $C_6$  are the regression coefficients of the respective proxies. This piecewise linear trend (PWLTL) describes a linear change in ozone after removing all dynamical proxies and aerosol influence during the period (e.g. Reinsel et al., 2002, 2005).

To describe the total ozone variability, we use the planetary wave drive proxy (i.e. heat flux calculated from the ERA interim analysis at 70 hPa/40–90° S, averaged over August and September as described by Kuttippurath and Nikulin, 2012), the Antarctic Oscillation (AAO) (<ftp://ftp.cpc.ncep.noaa.gov/cwlinks/>), solar flux (SF) ([ftp://ftp.ngdc.noaa.gov/STP/SOLAR\\_DATA/SOLAR\\_RADIO/FLUX/Penticton.Adjusted/monthly/](ftp://ftp.ngdc.noaa.gov/STP/SOLAR_DATA/SOLAR_RADIO/FLUX/Penticton.Adjusted/monthly/)) at 10.7 cm wavelength, the quasi-biennial oscillation (QBO) at 40 hPa (<http://www.geo.fu-berlin.de/met/ag/strat/produkte/qbo/>),



**Fig. 5.** The vortex averaged September–November  $O_3$  trends estimated using a multi-variate regression model for the ground-based, TOMS/OMI, and MSR data in 1979–2010. Top to bottom: deseasonalised  $O_3$  (MEAS) and the regression model (top panel), the contribution of heat flux – HF (second panel), Antarctic Oscillation – AAO (third panel), solar flux (SF) multiplied by quasi-biennial oscillation (QBO) at 40 hPa (fourth panel), aerosol (fifth panel), and the equivalent effective Antarctic stratospheric chlorine – EEASC (bottom panel).

and the aerosol optical thickness (<http://data.giss.nasa.gov/modelforce/strataer/>) to account for the El Chichon (1982) and Mount Pinatubo (1991) volcanic aerosol injections. In order to better explain the variability of ozone, we use  $SF \times QBO$  instead of individual solar flux and QBO terms, as explained by Roscoe and Haigh (2007). All proxies, except heat flux, are averaged over the September–November period to match the mean ozone taken during the same period. The southern hemispheric aerosol average data, which are shifted by +6 months to account for the transport of aerosols to the Antarctic, are considered. The selection of a 6 month shift of the aerosol data was based on a sensitivity test using various options (0, 3, 6, and 9 months of shift), for which a 6 month shift gave the best correlation between the regression model and ozone.

We have repeated the regression analysis after replacing the PWLT term ( $C_1 t_1 + C_2 t_2$ ) by  $C_1 EEASC(t)$ , EEASC being the equivalent effective Antarctic stratospheric chlorine (e.g. Brunner et al., 2006; Vyushin et al., 2010). Our EEASC uses the WMO A1-2010 scenario with a mean age of air of 5.5 yr for the polar stratosphere, age of air spectrum width of 2.75 yr (i.e. the half width of the age of air) and the bromine scaling factor of 60 to account for the greater ozone depletion potential of bromine compared to that of chlorine on a per atom basis. Further details about the EEASC formulation can be found in Newman et al. (2007). It should be noted that the trend in EEASC is in  $pptv\ yr^{-1}$  and the regression coefficient of EEASC from the model is in  $DU\ pptv^{-1}$

and hence the trend in ozone is expressed in  $DU\ yr^{-1}$  (i.e.  $DU\ pptv^{-1} \times pptv\ yr^{-1}$ ) (e.g. Stolarski et al., 2006). Since the trends derived from the PWLT model is in  $DU\ yr^{-1}$  itself, the estimated trends from both models can be compared directly.

#### 4.3.2 Contribution of proxies

Figure 5 shows the contribution of explanatory variables and resulting diagnosis using the EEASC regression for ground-based, TOMS/OMI and MSR data. Note that we have taken the turning point (TP) or break point for the PWLT regression as 2000, to coincide with the peak in EEASC. The regression analyses on all three data sets show similar evolution of ozone, regressed data, and the explanatory parameters. The PWLT and EEASC regression models explain about 88–90% of the ozone variability as deduced from the  $R^2$  estimate (e.g. Roscoe and Haigh, 2007). The  $SF \times QBO$  contributes about +20 DU in 1979, 1989 and 2001 and –10 DU in 1984, 1996 and 2007. The variability in ozone columns resulting from the changes in solar activity is about 2–3%, (Soukharev and Hood, 2006) and our results are within the expected range. The aerosol loading due to the eruption of El Chichon and Pinatubo significantly contribute to the ozone reduction of about –10 DU in 1983 and –26 DU in 1992, respectively, which reiterates the key role of aerosols on the heterogeneous ozone loss processes (e.g. Hofmann et al., 1992). The heat flux and AAO

**Table 3.** Antarctic ozone trends in  $\text{DU yr}^{-1}$  estimated from the deseasonalised September–November vortex averaged ground-based, TOMS/OMI, and MSR data using the PWLT (piecewise linear trend) and EEASC (equivalent effective Antarctic stratospheric chlorine) regressions. The regression results without considering heat flux are also shown in the bottom panel. The error values represent 95 % confidence intervals. The results are shown for various vortex averaged calculations: inside vortex, over the equivalent latitudes (EqLs) 65–90° S and inside vortex core.

Data	Period	Inside vortex		EqL: 65–90° S		Vortex core	
		PWLT	EEASC	PWLT	EEASC	PWLT	EEASC
Ground-based	1979–1999	$-4.68 \pm 0.88$	$-4.18 \pm 0.65$	$-4.62 \pm 0.87$	$-4.14 \pm 0.66$	$-5.02 \pm 0.89$	$-4.51 \pm 0.65$
	2000–2010	$+2.58 \pm 2.16$	$+1.03 \pm 0.16$	$+2.33 \pm 2.13$	$+1.02 \pm 0.16$	$+2.59 \pm 2.19$	$+1.11 \pm 0.16$
TOMS/OMI	1979–1999	$-5.03 \pm 1.12$	$-4.50 \pm 0.63$	$-4.89 \pm 1.00$	$-4.38 \pm 0.62$	$-5.24 \pm 1.12$	$-4.70 \pm 0.66$
	2000–2010	$+2.87 \pm 2.74$	$+1.11 \pm 0.16$	$+2.67 \pm 2.47$	$+1.08 \pm 0.15$	$+2.84 \pm 2.74$	$+1.16 \pm 0.16$
MSR	1979–1999	$-4.81 \pm 1.11$	$-4.31 \pm 0.62$	$-4.68 \pm 0.99$	$-4.19 \pm 0.62$	$-5.02 \pm 1.11$	$-4.50 \pm 0.65$
	2000–2010	$+2.91 \pm 2.73$	$+1.06 \pm 0.15$	$+2.68 \pm 2.43$	$+1.03 \pm 0.15$	$+2.91 \pm 2.73$	$+1.11 \pm 0.16$
Without heat flux							
Ground-based	1979–1999	$-4.53 \pm 1.08$	$-4.07 \pm 0.80$	$-4.45 \pm 1.13$	$-4.01 \pm 0.85$	$-4.87 \pm 1.10$	$-4.40 \pm 0.80$
	2000–2010	$+2.77 \pm 2.66$	$+1.00 \pm 0.20$	$+2.55 \pm 2.78$	$+0.99 \pm 0.21$	$+2.79 \pm 2.70$	$+1.08 \pm 0.20$
TOMS/OMI	1979–1999	$-4.87 \pm 1.15$	$-4.38 \pm 0.80$	$-4.72 \pm 1.14$	$-4.25 \pm 0.84$	$-5.09 \pm 1.12$	$-4.58 \pm 0.81$
	2000–2010	$+3.08 \pm 2.82$	$+1.08 \pm 0.20$	$+2.90 \pm 2.81$	$+1.05 \pm 0.21$	$+3.04 \pm 2.75$	$+1.13 \pm 0.20$
MSR	1979–1999	$-4.66 \pm 1.22$	$-4.18 \pm 0.80$	$-4.51 \pm 1.15$	$-4.06 \pm 0.84$	$-4.87 \pm 1.21$	$-4.38 \pm 0.81$
	2000–2010	$+3.12 \pm 3.00$	$+1.03 \pm 0.20$	$+2.91 \pm 2.83$	$+1.00 \pm 0.21$	$+3.11 \pm 2.98$	$+1.08 \pm 0.20$

contributions mostly follow the dynamics of each winter, as both explain wave forcing and meteorology of the winters (Sexton, 2001; Randel et al., 2002). The contribution of AAO is between  $-18 \text{ DU}$  (e.g. 1985, 2001, 2008 and 2010) and  $+20 \text{ DU}$  (e.g. 1980, 2000 and 2002). The enhanced wave activity (heat flux) contributes about  $+18 \text{ DU}$  in 1984, 1993, and 2005, with the largest contribution of about  $+38 \text{ DU}$  in 2002. Similarly, suppressed planetary wave activity makes strong vortices, and hence, higher ozone reduction in the very cold winters of 1987 ( $-18 \text{ DU}$ ), 1998 ( $-12 \text{ DU}$ ) and 2006 ( $-10 \text{ DU}$ ). The analyses with PWLT regression also yield a very similar contribution of the proxies, which are exempted from this discussion to avoid repetitions. The regression analysis shows that the halogen loading (EEASC) dominates the ozone reduction. The resulting ozone trends computed from the deseasonalised ozone anomalies after removing the contribution of the explanatory variables are listed in Table 3.

### 4.3.3 Vortex averaged ozone trends during 1979–1999

The regression functions applied to the ground-based measurements show a trend of about  $-4.1$  to  $-5.2 \text{ DU yr}^{-1}$  from both regressions over 1979–1999 and are significant at 95 % confidence intervals. As presented in Table 3 (top panel), these results did not change significantly when the data were analysed with respect to different vortex criteria (i.e. inside vortex, vortex core, and over the EqLs 65–90° S). The trends derived from ground-based observations are in very good agreement with those found from TOMS/OMI and MSR data. The similar trends deduced from both EEASC

and PWLT regressions imply that the ozone decrease over 1979–1999 is dominated by the increase in halogens during the period, consistent with the results of previous studies (e.g. WMO, 2011, and references therein). These trends are in good agreement with those found by Yang et al. (2008), who estimated a corresponding value ( $-4.5$  to  $-5 \text{ DU yr}^{-1}$  in 1978–1996) from the ground-based and satellite data using the cumulative sum method. A very similar trend of around  $-4 \text{ DU yr}^{-1}$  was also deduced from an assimilated ozone data set by Brunner et al. (2006). The slight differences in the trend values of these studies are within error bars.

### 4.3.4 Vortex averaged ozone trends during 2000–2010

The results for the 2000–2010 period show a trend of about  $+1 \text{ DU yr}^{-1}$  from the EEASC and  $+2.3$  to  $+2.9 \text{ DU yr}^{-1}$  from PWLT functions and are significant at 95 % confidence intervals for the ground-based data averaged with respect to various vortex criteria. These trends are also in very good agreement with those estimated from TOMS/OMI and MSR data. The EEASC-based results are consistent with those derived from the CCM/CTM simulations for the 1997–2009 period, which exhibit an EEASC-based ozone trend of around  $+1 \text{ DU yr}^{-1}$  (Austin et al., 2010; Kieseewetter et al., 2010). The trends derived from EEASC regression are smaller than those obtained from PWLT. This implies that the ozone increase during 2000–2010 cannot be explained by the reduction in ozone depleting substances alone, but there are strong influences from the dynamics and other parameters. Note also that similar differences between the PWLT and EEASC-

based trend values are also reported in previous studies for mid-latitude (up to 60° N/S) ozone (WMO, 2011; Vyushin et al., 2010). The significant positive trend during the period reinforces the notion that the Antarctic ozone is recovering, as reported by Salby et al. (2011).

#### 4.3.5 Influence of inter-annual variation in meteorology

It is well known that the temperature controls the PSC formation, chlorine and bromine activation, and hence, the spring-time ozone depletion. Therefore, to test the strength of the positive trends and to understand the impact of inter-annual variability of Antarctic meteorology on the derived results during 2000–2010, we computed the trends without heat flux in the regression models, which are shown in Table 3 (lower panel). The resulting estimates show similar values for EEASC-based regression, around +1 DU yr<sup>-1</sup> using all data sets, and are significant at 95 % confidence intervals. The PWLT regressions show slightly higher values, +2.5 to +3.1 DU yr<sup>-1</sup>, and are also significant at 95 % confidence intervals, except for the ground-based measurements averaged over 65–90° EqLs. These results, however, show a clear ozone recovery signal even without subtracting the added variability induced by dynamics.

## 5 Conclusions

A comprehensive analysis of chemical ozone loss in the Antarctic vortex from 1989 to 2010 is presented using total ozone observations from ground-based Brewer, DOAS, Dobson, and SAOZ, and space-borne TOMS/OMI and MSR data. The passive technique is applied to find the ozone loss at each station, and then averaged to find the mean loss. The loss in 1989–1991 and 2002 is about –110 to –140 DU or –33 to –40 %, and during 1992–2010 (except 2002) is around –160 DU or –48 %. In general, the ozone loss in the Antarctic starts by mid-June and intensifies in August–September, peaks by the end of September/early October, and ozone recovers thereafter, consistent with the results of previous studies. The ozone loss estimated from TOMS/OMI and MSR data also exhibits a proportional progress of ozone and ozone loss as for the ground-based measurements throughout the period (1989–2010), where the differences with the ground-based estimates are less than ±3 %.

The piecewise and EEASC-based trends estimated from the September–November vortex averaged ground-based ozone column show a trend of about –4.1 to –5.2 DU yr<sup>-1</sup> for the period 1979–1999 and a trend of around +1 DU yr<sup>-1</sup> with EEASC and +2.3 to +2.9 DU yr<sup>-1</sup> with PWLT functions during 2000–2010. These trend analyses are significant at the 95 % confidence levels in both periods for all vortex averaged data clusters. The ground-based analyses are well supported by those of the TOMS/OMI and MSR data. In 1979–1999, both piecewise and EEASC-based ozone trends show very similar values, corroborating the dominance of stratospheric halogens on the ozone decrease in that period.

However, the larger values derived from the PWLT regression for the 2000–2010 period suggest the greater influence of dynamics plus other regression indices not considered here on the increase of ozone during the period. These results thus show the first sign of ozone recovery. However, the Antarctic ozone loss/hole will prevail in much of this century with the given rate of the estimated positive trend and due to the still high levels of stratospheric chlorine. It will take another fifty years to regain the 1980 level of ozone.

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