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Antarctic ozone loss in 1989–2010: evidence for ozone recovery?

J. Kuttippurath[1], F. Lefèvre[1], J.-P. Pommereau[1], H. K. Roscoe[2], F. Goutail[1], A. Pazmiño[1], and J. D. Shanklin[2]

[1]Université Versailles-Saint-Quentin, LATMOS/CNRS, UMR8190, Guyancourt, France
[2]British Antarctic Survey, Cambridge, UK

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Correspondence to: J. Kuttippurath (jayanarayanan.kuttippurath@latmos.ipsl.fr)

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Abstract

We present a detailed estimation of chemical ozone loss in the Antarctic polar vortex from 1989 to 2010. The analyses include ozone loss estimates for 12 Antarctic ground-based (GB) stations. All GB observations show minimum ozone in the late September–early October period. Among the stations, the lowest minimum ozone values are observed at South Pole and the highest at Dumont d'Urville. The ozone loss starts by mid-June at the vortex edge and then progresses towards the vortex core with time. The loss intensifies in August–September, peaks by the end of September–early October, and recovers thereafter. The average ozone loss in the Antarctic is revealed to be about 33–50 % in 1989–1992 in agreement with the increase in halogens during this period, and then stayed at around 48 % due to saturation of the loss. The ozone loss in the warmer winters (e.g. 2002, and 2004) is lower (37–46 %) and in the colder winters (e.g. 2003, and 2006) is higher (52–55 %). Because of small inter-annual variability, the correlation between ozone loss and the volume of polar stratospheric clouds yields ~0.51. The GB ozone and ozone loss values are in good agreement with those found from the space-based observations of the Total Ozone Mapping Spectrometer/Ozone Monitoring Instrument (TOMS/OMI), the Global Ozone Monitoring Experiment (GOME), the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY), and the Aura Microwave Limb Sounder (MLS), where the differences are within ±5 % and are mostly within the error bars of the measurements. The piece-wise linear trends computed from the September–November vortex average GB and TOMS/OMI ozone show about −4 to −5.6 DU (Dobson Unit) yr^{-1} in 1989–1996 and about +1 DU yr^{-1} in 1997–2010. The trend during the former period is significant at 95 % confidence intervals, but the trend in 1997–2010 is significant only at 85 % confidence intervals. Our analyses suggest a period of about 9–10 yr to get the first detectable ozone recovery signal at the 95 % confidence intervals with the current ozone trends in the Antarctic. Thus, this study reveals that the recovery of the Antarctic ozone is well on course.

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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). To monitor the changes in ozone levels, a number of instruments has been deployed in and around the continent, under the Global Atmosphere Watch (GAW) and Network for the Detection of Atmospheric Composition Change (NDACC) consortia. Among which, the System d'Analyse par Observation Zénithale (SAOZ) technique (Pommereau and Goutail, 1988) is able to measure year-round, even inside the polar circle at high solar zenith angles (SZAs), as compared with the Brewer, Dobson, and other UV space-borne total column observations. This is because the SAOZ ozone measurements are taken in the visible bands. Studies have already shown a significant reduction in the stratospheric halogen amounts (e.g. Jones et al., 2011). Therefore, a subsequent decrease in ozone loss and thus, the recovery of ozone is expected in a few decades (Austin et al., 2010a,b). This further illustrates the necessity of the constant surveillance of ozone in the polar stratosphere to detect any possible changes that may affect the prediction of its future evolution.

Although there are some ozone loss estimations available for the Antarctic, they are either modelled or incomplete due to limitations of the analysed observations (Austin and Wilson, 2006; Huck et al., 2007; Lemmen et al., 2006; Tilmes et al., 2006). For instance, the ozone loss estimations by Huck et al. (2007) present a time series from 1992 to 2004, but are largely limited to a parameterised tracer from a regression model and reanalysis data. The analysis of Tilmes et al. (2006) uses measurements either from the Halogen Occultation Experiment (HALOE) or from the Improved Limb Atmospheric Spectrometer (ILAS). Both instruments are spatially restricted as far as their observation of the Antarctic vortex is concerned (up to 80° S for HALOE and up to 88° S for ILAS, depending on season). In addition, the number of measurements is very few and they do not always cover the peak ozone loss period of late September/early October. Although there are some studies using the Polar Ozone and Aerosol Measurement

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(POAM) data, a continuous long-term ozone loss analysis is still not available using these data (Bevilacqua et al., 1997; Hoppel et al., 2005). The Chemistry-Climate Model (CCM) based studies are mostly exploited for the projection of ozone recovery (Austin et al., 2010a,b). Therefore, an analysis of an ozone loss time series that we have for the Arctic (e.g. Harris et al., 2010; Kuttippurath et al., 2010b; Tilmes et al., 2006; Goutail et al., 2005; Rex et al., 2004) is obviously missing for the Antarctic. We fill this gap with a comprehensive ozone loss analysis using GB and satellite measurements. 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. (2010a). In this study, we extend the same method to the measurements from 1989 to 2010 in order to construct an ozone loss time series to further elucidate the evolution of ozone loss in the southern polar vortex.

Some 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 the Antarctic ozone using measurements from GB and satellite instruments by applying a temperature/saturation correction to the data. They showed a negative trend of about 4–5 DU (Dobson Unit) yr^{-1} during the period 1978–1996 and an insignificant positive trend or a levelling off thereafter. Similar trends were also estimated by Wohltmann et al. (2007), who applied a process-oriented multi-variate regression model to the high latitude total ozone measurements for their estimates. A study by Hassler et al. (2011) showed the stabilisation of ozone loss rates at South Pole during the period 1991–2009. However, a recent work by Salby et al. (2011) showed a significant positive trend in the September–November Total Ozone Mapping Spectrometer/Ozone Monitoring Instrument (TOMS/OMI) ozone during 1997–2009. Therefore, in addition to the long-term ozone loss assessment for the Antarctic, we also compute ozone trends to discuss the aforementioned results from a perspective of ozone recovery.

This study is organised in the following way: the introduction follows the data used for the analyses and the method applied to the ozone loss derivation in Sect. 2. Since we have already presented details of the method in a previous article (Kuttippurath et al., 2010b), only a short description will be given to follow the study. In Results, Sect. 3.1 deals with the minimum ozone measurements at each station, which give an idea about the minimum ozone observation period and the changes in minimum values in the wake of the decreasing halogen content in the stratosphere. The ozone loss estimates for the Antarctic stations and for the whole region are presented in Sect. 3.2. In Discussion, Sect. 4.1 assesses the derived ozone loss and their inter-annual variability. The measured loss is compared with results from other studies in Sect. 4.3 and ozone trends in the Antarctic are analysed in Sect. 4.4. Section 5 concludes the study with our main findings.

2 Materials and methods

2.1 Ground-based ozone column

We use measurements from 12 GB stations deployed in and around the continent, such that they cover the entire region to provide a representative analysis for Antarctica. As the Antarctic vortex is very stable and inter-annual variations in the meteorology are very small, the selection of the stations is less dependent on estimated ozone loss (Kuttippurath et al., 2010a). Nevertheless, the analysis for each year contains data from at least eight stations and hence, assures a robust diagnosis. Among the various instruments, the SAOZ observations from Dumont d'Urville, Concordia (Dome C), Faraday-Vernadsky (after 1996) and Rothera are used. These total column measurements in the visible region have a precision of 4.7 % and an accuracy of 5.9 % (Hendrick et al., 2011) and different SAOZ slant column measurements are consistent within ±3 % (Roscoe et al., 1999). The Neumayer measurements are performed with a similar UV-Visible instrument of the Differential Optical Absorption Spectroscopy (DOAS) and the precision of these measurements is about 2 % (Frieß et al., 2005). In addition, the Dobson measurements performed (Dobson, 1957) at Arrival Heights, Faraday-Vernadsky (before 1996), Halley, Marambio, South Pole (Amundsen-Scott) and Syowa, and the Brewer measurements (Brewer, 1973) from Belgrano and Zhongshan are also considered. The precision of these observations was estimated as 0.5 % or 1 DU (Basher, 1982), but is subject to the accuracy of absorption cross-sections and a known significant temperature dependence in the UV and stray light at large SZAs (Hendrick et al., 2011), which were not taken into account for the retrievals used here.

2.2 Satellite ozone column

To compare with the GB ozone loss estimations, total ozone version (v)8.5 from TOMS aboard 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 the TOMS ozone aboard different 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 SZA <84° (Levelt et al., 2006). Therefore, a continuous series comparable to that of GB is available from TOMS and OMI from 1989 to 2010, with the exception of 1994 and 1995. The ozone column data from the Global Ozone Monitoring Experiment (GOME) aboard the European Remote Sensing (ERS)-2 satellite (Burrows et al., 1999) and GOME-2 on the Meteorological Operational satellite, and the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) on board the Environmental Satellite (Bovensman et al., 1999) are also considered. The GOME and SCIAMACHY data used here are retrieved with similar techniques i.e. TOSOMI/TOGOMI (Total Ozone retrieval scheme for SCIAMACHY/GOME based on the OMI DOAS algorithm) (Eskes et al., 2005; Valks et al., 2004). Note that a similar algorithm was also applied for the retrievals of total ozone from TOMS and OMI. GOME observations from 1996 to 2003, GOME-2 from 2006 to 2009, and SCIAMACHY from 2002 to 2009 are considered, as they were available during this study period. The uncertainty of the SCIAMACHY and GOME 1/2 data is 2–3.3 % (Eskes et al., 2005; Loyola et al., 2011). Our comparisons between GB and satellite measurements at all GB stations show a reasonably good agreement with the TOMS/OMI data, but a clear high bias of about 2–5 % with data from GOME and SCIA-

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MACHY. This offset has been corrected for the analyses presented here (except for the minimum ozone). However, as shown by Hendrick et al. (2011) there is still some bias of the order of 2 % between TOMS/OMI and SAOZ observations in the Antarctic with a strong seasonal dependence. As this bias was not systematic, it was not possible to correct it here.

In order to compare the GB ozone loss in early winter, we have calculated ozone columns from the Aura Microwave Limb Sounder (MLS) ozone v3.3 profiles, which are averaged within $2^\circ \times 2^\circ$ around each station. The uncertainty of the MLS ozone profiles is about 5–10 % in the stratosphere (Livesey et al., 2011; Froidevaux et al., 2008). The South Pole overpass data are produced by averaging the four nearest longitude points (0, 90, 180, and 270° E) at 82° S, the southernmost latitude of the MLS observations. The ozone columns from MLS ozone mixing ratio profiles are calculated between 10 and 60 km using the MLS pressure and temperature data. Our primary comparisons show a good agreement between the MLS overpass column with that of GB at all stations. However, since the MLS column is computed from 10 to 60 km, the column contributions from below and above the altitude range have to be accounted for and are about 7–15 DU or 2–5 %, as computed from the comparison with total ozone observations from 12 Antarctic GB stations. This deficit has been taken into account for the ozone loss analyses.

2.3 Tracer simulations from REPROBUS

We use the REPROBUS chemical transport model (CTM) (Lefèvre et al., 1994) discussed by Kuttippurath et al. (2010a) to simulate the passive tracer from 1989 to 2010 for the chemical loss computations. Our new simulations, however, use the European Centre for Medium-Range Weather Forecasts (ECMWF) Reanalysis (ERA) – interrim meteorological data to force the model runs (Dee et al., 2011). The version used in this work has a horizontal resolution of $2^\circ \times 2^\circ$ on 60 vertical levels from the surface to 0.1 hPa. We use the passive tracer simulated by the model intialised in 1989, but we reinitialise the tracer fields on every June to match the Antarctic winter period. The

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ECMWF ozone data were used for the initialisation of the model runs. The passive tracer columns used here are the averages within 100 km of each station.

2.4 Ozone loss derivation 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 (Goutail et al., 1999) to the selected observations. In this method, the ozone chemical loss is computed as the difference between the passive tracer initialised identically to ozone at the beginning of the CTM run and the measured ozone (ground-based ozone - model tracer). For instance, Fig. 1 illustrates the ozone loss estimated inside the vortex from all GB measurements for the Antarctic winter 2006. Generally, each station shows different timings for the onset, progress and maximum in the ozone loss, depending on the exposure to sunlight and distribution of temperature. Note that the transport of ozone depleted air masses over the station can also affect the onset period (Hassler et al., 2011; Kuttippurath et al., 2010a).

There are some variations in ozone distribution inside the vortex with 2 separate air masses - the edge region with a breadth of about 15° around the brim 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 mostly in the edge region, with occasional excursions between the edge and core of the vortex. On the other hand, Dumont d'Urville is frequently in the edge region, but occasionally inside the vortex core and the stations at 70–90° S are mostly inside the vortex core. A detailed discussion of the station positions and related observational features can be found in Kuttippurath et al. (2010b). On average (Fig. 1 solid line), the ozone loss in the region starts by mid-June at the edge of the vortex 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. The loss stays around the peak loss, or reduces thereafter with respect to the meteorological

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conditions and vortex persistence. The estimated ozone depletion has an uncertainty of about 3–5 % (Kuttippurath et al., 2010a).

3 Results

3.1 Minimum ozone measurements

The minimum ozone distribution usually indicates the amount of maximum ozone loss in the region. In addition, any change in the minimum values and day when the minimum is observed can also indicate the change in ozone trends. Therefore, Fig. 2 presents the minimum ozone observed by various instruments at each site during 1989–2010. Among the stations, South Pole shows the lowest values of about 100 DU, while Dumont d'Urville shows the highest of about 180 DU. These exclude the early years 1989–1992 and the warm winters (2002, 2004, 2007, and 2010), where the vortex dissipated earlier than in other winters due to enhanced wave activity (WMO, 2011; Tully et al., 2011). In 1989–1992, most stations observe a similar minimum of about 140–150 DU, except at Dumont d'Urville, where it is around 200 DU. Similarly, the warm winters of 2002 and 2004 show a comparatively higher minimum at all stations; ∼140 DU at South Pole and ∼175 DU at other stations, excluding Dumont d'Urville. The other stations register the minimum ozone in between these two extremes. For instance: Faraday-Vernadsky, Marambio, Syowa and Zhongshan show values near to 140 DU, but Rothera and Concordia show slightly lower values of about 130 DU. The minimum ozone observed at Belgrano, Halley, and Neumayer is comparable with those at the South Pole and is about 110 DU. This suggests that the centre of the vortex is not always at the South Pole, as also mentioned by Hassler et al. (2011). Among the winters the lowest minimum is observed in 2006 at most stations. However, the minimum observed by the Brewer at Zhongshan was about 85–90 DU in 2001 and is the lowest among the stations/winters. The day when the minimum was measured is

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between day 260 and 270 at all stations, which falls in the end of September and early October period, indicating the period of maximum ozone loss in the Antarctic.

The satellite measurements are generally in good agreement with the GB observations and their best agreement is found at Halley, Arrival Heights and Dumont d'Urville. Nevertheless, the GB instruments measure a comparatively lower minimum at Rothera and a higher minimum at Faraday-Vernadsky and Syowa in 1989–2002. There is a known low bias (∼10 %) in the Rothera data related to the uncertainties in the air mass factor used in the retrievals (Roscoe et al., 1999; Hendrick et al., 2011), which could be a reason for the offset found at Rothera. The GOME and SCIAMACHY data always show a high bias relative to the GB and TOMS/OMI data, as they are not bias corrected, because we wanted to analyse the minimum ozone they observe. The GB and satellite differences are mostly within ±30 DU, depending on station and winter. Note that these values are extracted from the available measurements by each instrument, because some instruments lack continuous measurements during the August–October period. Therefore, the observational characteristics and measurement gaps could also contribute to the observed differences in the minimum ozone values.

3.2 Ozone loss above the stations

Figure 3 depicts the ozone (DU) and chemical ozone loss (DU and %) inside the vortex at different Antarctic stations as computed from various data sets in mid-September/mid-October, the minimum ozone period. The average ozone distributions show equivalent features that are discussed for each station/winter in Sect. 3.1 with corresponding changes in ozone values. Therefore, we now discuss the ozone loss estimated from these ozone measurements for each station. For instance, at Arrival Heights, the GB ozone is shown to be about 32 % or 100 DU in 1989 and it rapidly increased to 160 DU or 50 % by 1993. The loss gradually reduced to 130 DU or 42 % during the next three years and then increased again to 175 DU or 53 % by 2001. During the warm winter of 2002, the loss reduced considerably to 135 DU or 34 %. Since 2002, there have been two very cold and four relatively warm winters that show extreme

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values in ozone loss. The largest loss of about 175 DU or 53–56 % was in the colder winters of 2003 and 2006 and the lowest loss of around 145 DU or 42 % was in the warmer winters of 2004, 2005, 2007 and 2010.

The evolution of ozone loss at other GB stations is similar, but with slight differences in the ozone loss values. On average, the loss at Syowa and Zhongshan is similar to that of Arrival Heights, about 45 %, whilst the loss at Marambio and Faraday-Vernadsky is slightly smaller, about 43 %. Halley and Rothera exhibit larger ozone loss of about 50 % and the stations installed inside the vortex core, Belgrano, Neumayer and South Pole, show the highest loss of about 55–60 %. In contrast, the lowest ozone loss of about 38 % is estimated at the edge region station Dumont d'Urville. All stations show the lowest ozone loss in 2002 (about 110–140 DU or 35–40 %) and the highest in 2006, except for Marambio, where it shows the lowest loss in 1997. This discrepancy could be due to the lack of continuous measurements in the late September–early October period in 2006 at Marambio. The edge stations (Marambio, Faraday-Vernadsky and Dumont d'Urville) show comparatively lower ozone loss than the stations inside the vortex core (South Pole, Halley and Belgrano) due to relatively warmer vortex conditions. The satellite observations return identical ozone loss values to those of GB at all stations and the agreement is exceptionally good at Halley, Neumayer, Syowa and Zhongshan, within ±1 %. However, the differences are slightly larger at Dumont d'Urville in 1998–2004, about 2–3 % or 5–15 DU.

3.3 Ozone loss averaged over all stations

Figure 4 shows the vortex average chemical ozone loss estimated in the Antarctic from the GB and satellite data during 1989–2010. In general, the loss starts in mid-June/early July, in agreement with PSCs and chlorine activation on sunlit parts of the vortex, except in the early years of ozone loss (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. The warm winter of 2002 and the early years 1989–1990 show late onset of ozone loss. All years exhibit a higher loss rate during August–September and the peak loss during the end of September and early October period. On average, the maximum loss until mid-October is around 120 DU or 40 % 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, where the colder winters (2000, 2003 and 2006) show a slightly greater reduction of about 170 DU or 55 %. As anticipated, the warmer winters show the opposite pattern of lower ozone loss, as demonstrated by the 40 % loss in 2002.

All satellite observations show a remarkably similar evolution of ozone loss in all winters and the differences among various data sets are within ±5 % or ±15 DU. The excellent agreement amongst the data, including SCIAMACHY which has no South Pole data in its average, affirms that the addition or removal of a station hardly makes any significant change to the average ozone loss. The evolution and maximum ozone loss in the ozone column computed from MLS ozone profiles exhibit an exceptionally good agreement with the GB estimations. The MLS data are available from the beginning of each winter for all stations and hence, their average represents data from all stations, in which many of them have different scales of ozone loss in the June–August period. However, the GB average contains measurements only from the edge stations (e.g. Dumont d'Urville and Rothera), which gives rise to the difference in the ozone loss values between GB and MLS in June–August. This comparison corroborates the potential and strength of the Aura MLS measurements to be used for ozone total column loss analyses in the polar regions, where polar night measurements are not possible by UV-visible instruments.

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4 Discussion

4.1 Inter-annual variability of ozone loss

Figure 5 shows the inter-annual variations in vortex average chemical ozone loss deduced from different data sets in two different periods. In the Antarctic, the maximum reduction in ozone in each winter is observed by the end of September to early October period and hence, a temporal average of this period is compared to the conventional October average. In the former period, the GB estimations show a steep increase of ozone loss from 90 to 155 DU or from 33 to 44 % in 1989–1994. There was then a sharp decrease to 125 DU or 42 % by 1996, although the loss again slipped gradually to the decadal maximum of 181 DU or 53 % in 2001. The warming in 2002 reduced the loss to 151 DU or 40 %, but it remained around 160 DU or 48 % thereafter, with the highest ozone loss in the coldest winter of 2006, about 53 %. These ozone loss estimations mostly follow the amount of halogens in the stratosphere during the respective periods, as they slowly increased from 1989–1994, peaked during 1996–2000 and then started to decrease thereafter (e.g. Newman et al., 2007; Jones et al., 2011; WMO, 2011). The October average also shows a similar ozone loss evolution, but the highest loss is still observed during the mid-September/mid-October period. Therefore, we have derived a ten-day ozone loss average during the peak loss period: 26 September to 5 October to find the maximum loss during each winter in 1989–2010 for a reference and these are given in Table 1. The ozone loss rates between day 225 and 275, during which most GB instruments have measurements, are also deduced. The ozone loss in this time window is analogous to that discussed for the other seasons, but as expected, the values are about 2–4 % higher. The loss rates also mark a comparable temporal evolution, with the largest loss rates (0.71 % day^{-1}) in the coldest winter of 2006 and the lowest (0.39 % day^{-1}) in the warmest winter of 2002.

In agreement with the ozone loss analyses, the average ozone values in mid-September/mid-October (Fig. 5 left) show a reciprocal evolution in each winter. The ozone evolution during the period is consistent with that discussed in Sect. 3.2, but

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with slight differences in ozone values. The October average shows identical behaviour in ozone distributions, but slightly higher than the aforesaid ones. This implies that the conventional October average used in other studies of the Antarctic ozone (WMO, 2011 and references therein) does not correspond to the maximum ozone loss period.

The gross features of the October average ozone (Fig. 5 right) from GB and satellite are in excellent agreement with the HALOE and Stratospheric Aerosol and Gas Experiment (SAGE) October average ozone shown by Yang et al. (2008). The ozone and ozone loss values are also in good accordance with the distribution of average temperature and heat flux during each winter (e.g. Yang et al., 2008; Salby et al., 2011), where the highest temperatures were observed in 2002 and the lowest in 2006. As noted for ozone, the difference in the vortex average temperature in mid-September/mid-October 2002 to that of other winters is ~10 K, indicating the severity of the warming in this particular winter.

4.2 Correlation between ozone loss and V_{psc}

A compact relationship between ozone loss (in DU) and volume of PSCs (V_{psc}) has been found in the Arctic (Rex et al., 2004; Harris et al., 2010). We now check how this relation comes to effect in the Antarctic. Figure 6 delineates the correlation between the GB ozone loss computed between 26 September and 5 October (Table 1) and the average V_{psc} from May through to November over 350–675 K in 1989–2010. We have excluded the anomalous winter 2002 from the correlation analysis. The correlation between the ozone loss and V_{psc} shows about 0.51–0.59 for ozone loss in both relative (%) and absolute (DU) units, smaller than that observed in the Arctic, where the correlation yields ~0.9 (Rex et al., 2004; Harris et al., 2010). We have already seen the weak inter-annual variability of ozone loss and temperature in the Antarctic. The study by Cacciani et al. (1997) has also shown that the occurrence of PSCs in the Antarctic vortex core has reached the saturation level. Alternatively, every other winter is warm and early final warmings are frequent and hence, significant occurrence of PSCs is limited to relatively cold winters in the Arctic (Manney et al., 2005; Kuttippurath et al., 2010b). Therefore, ozone loss has shown correspondingly large year to year variability (WMO, 2011; Kuttippurath et al., 2010b; Harris et al., 2010; Goutail et al., 2005). Such extreme warm winters are apparently absent in the Antarctic and thus, there is no positive one-to-one correlation between ozone loss and V_{psc}.

4.3 Ozone loss: comparison with other studies

There are no comprehensive long-term ozone loss analyses for the Antarctic region to compare with our analyses. As discussed in Sect. 1, the available estimations from Tilmes et al. (2006) use 15 HALOE ozone profiles inside the vortex in each winter (1991–2004), which is further limited to the sampling pattern of the satellite. Additionally, temporal resolution of these measurements is confined to spring. Conversely, the tracer correlation method applied to the ILAS measurements are available mostly in the winter months of each year since 1992, while the maximum loss in the Antarctic is observed in spring and therefore, these analyses are not complete. On the other hand, Huck et al. (2007) use a parameterised tracer derived from a regression model and a reanalysis ozone total column data for their ozone total column loss estimation. As the derived loss increasingly depends on the tracer values, the relative loss ([tracer-ozone]/tracer in %) with respect to the tracer that can largely alleviate the effect of tracer on the computed loss is also not available. Therefore, both studies are not directly comparable to our estimations. However, the total ozone column loss deduced by Huck et al. (2007) shows a peak loss of around 120 DU in most years, with the largest of about 130 DU in 2001 and the lowest of about 88 DU in 2002. The partial column loss estimated over 380–550 K from the satellite ozone profiles by Tilmes et al. (2006) show about 120–145 DU. The peak loss deduced from the available measurements (maximum three ozone profiles) is about 155 DU in 2003 and the lowest in 1996–1997, about 115 DU (Tilmes et al., 2006). Both analyses show lower ozone loss than in our estimations. The offset with Huck et al. (2007) could be due to the difference between the tracers and the ozone data used in both analyses. The length of ozone column

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(over 380–550 K) and limitations of the data used for the loss analysis could be the reasons for the offset with the loss derived by Tilmes et al. (2006).

4.4 Multi-variate regression of vortex average ozone

4.4.1 Data, method and model

A process oriented multi-variate regression model is constructed and applied to determine the ozone trends. We use the September–November GB and TOMS/OMI vortex average ozone for this study. The 1994–1995 data gap in TOMS/OMI is filled with corresponding overpass analyses using the multi-sensor reanalyses (MSR) data, which primarily consists of the Solar Backscatter Ultraviolet (SBUV) ozone observations during the period. Previous studies have successfully used this data cluster in various scientific and trend studies (e.g. de Laat and van Weele, 2011). A recent study by Salby et al. (2011) showed that there is a significant positive trend in the September–November TOMS/OMI ozone in 1997–2009 in the Antarctic. So we also consider the September–November GB and TOMS/OMI ozone average in 1989–2010. More importantly, we use the vortex averaged data from GB and TOMS/OMI to further elucidate the significance of the trends.

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

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$$
\begin{aligned}
Y(t) = {} & K && \text{(Constant)}\\
& + C^1\mathrm{LT}(t) && \text{(Linear trend)}\\
& + C^2\mathrm{CT}(t) && \text{(Change in trend)}\\
& + C^3(\mathrm{SF} \times \mathrm{QBO})(t) && \text{(Solar flux} \times \text{QBO)}\\
& + C^4\mathrm{Aer}(t) && \text{(Aerosol)}\\
& + C^5\mathrm{HF}(t) && \text{(Heat flux)}\\
& + C^6\mathrm{AAO}(t) && \text{(AAO)}
\end{aligned}
\qquad (1)
$$

where t is time, K is a constant and C^1 to C^6 are the regression coefficients of the respective proxies. To describe the total ozone variability, we take the planetary wave drive proxy (heat flux – calculated from the ECMWF operational analyses at 70 hPa/40–90° S in August–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/) at 10.7 cm wavelength, the Quasi-Biennial Oscillation (QBO) at 40 hPa (http://www.geo.fu-berlin.de/met/ag/strat/produkte/qbo/), and the Aerosol optical thickness (http://data.giss.nasa.gov/modelforce/strataer/) to account for the Mount Pinatubo volcanic aerosol injection. In order to better explain the variability of ozone, we use SF × QBO instead of individual solar flux and QBO terms. The feasibility and advantages of this method have been explained in detail 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 is 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 use both piece-wise linear trend (PWLT) (Reinsel et al., 2002, 2005) and EESC functions (PWLT terms replaced with EESC) (Brunner et al., 2006; Vyushin et al., 2010) to derive ozone trends and to check the consistency and significance of the derived results.

4.4.2 Results: contribution of proxies

Figure 7 shows the contribution of explanatory variables used and resulting diagnosis using the PWLT regression, which excludes ozone observations of the anomalous winter 2002. The PWLT or EESC regression model explains about 80 % of the ozone variability as deduced from the R^2 value (e.g. Roscoe and Haigh, 2007), which indicates a very good correlation between the measurements and regressed data. It shows that the SF × QBO contributes about 7 DU in 1989 and 2001 in agreement with the solar maximum during these years and −4 DU in 1996 and 2007. The variability in ozone columns by the changes in solar activity is about 2–3 % (Soukharev et al., 2006) and our results are within the predicted lines. Presumably, aerosols significantly contribute to the ozone reduction in 1991–1992, about 10–22 DU, reiterating the influence of the Pinatubo volcanic aerosols on the ozone change (e.g. Hofmann et al., 1992). The heat flux and AAO 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 −6 DU (2001) and +6 DU (2000). The enhanced wave activity (heat flux) contributes about 5–7 DU in 1992–1993 and 2004. Similarly, suppressed planetary wave activity makes strong vortices and hence, higher ozone reduction in the very cold winters of 1994 (−5 DU), 1998 (−11 DU), and 2006 (−7 DU).

The linear trend describes the contribution of gas phase chemical ozone loss from the increase in halogen loading that peaked during 1996–2000. Therefore, the linear trend shows a maximum contribution of about −44 DU in 1996 and a gradual decrease thereafter. A matching statistic is obtained from the EESC regression (not shown). That

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is, the ozone reduction in the Antarctic dominates the halogen/chlorine loading and hence, the contributions of PWLT or EESC outweigh those of others such as heat flux or solar flux × QBO. The resulting ozone trends computed from the ozone anomalies after removing the contribution of the explanatory variables using the PWLT and EESC regression analyses are listed for various scenarios in Table 2.

4.4.3 Results: vortex average ozone trends

As expected, the regression functions applied to the GB and TOMS/OMI data show a clear negative trend of about 5.4–5.8 DU yr^{-1} from the PWLT model and 2.6–2.8 DU yr^{-1} from EESC regressions in 1989–1996. These trends are significant at 95 % confidence intervals. The lower values of EESC regression hint that the trend during the period cannot completely be explained by the reduction in ozone depleting substances, as discussed elsewhere (e.g. WMO, 2011; Vyushin et al., 2010). The PWLTs deduced from this study are in good agreement with those found in Yang et al. (2008), who estimate a corresponding value (−4.5 to −5 DU yr^{-1} in 1978–1996) from the high latitude GB and satellite data. A very similar trend of about −4 DU yr^{-1} was also deduced from an assimilated ozone data set for the high latitudes by Brunner et al. (2006). The slight differences in the trend values of these studies are within error bars but could in any case be due to the difference in data, regression model, and method used for the trend analyses.

In 1997–2010, the regression on GB and TOMS/OMI ozone yields a trend of about +1 DU yr^{-1} from the PWLT and +0.7 DU yr^{-1} from EESC functions and are significant at 85 % confidence intervals, but not significant at 95 % confidence intervals. This indicates a slow recovery of the Antarctic stratospheric ozone, even though the 95 % confidence interval is still elusive. These results are consistent with those derived from the CCM/CTM simulations in 1997–2009, which exhibit a trend of about +1 DU yr^{-1} (Austin et al., 2010a,b; Kiesewetter et al., 2010). Our results thus contradict the significant positive trends claimed by Salby et al. (2011) using the TOMS/OMI observations during 1997–2009. This could be due to two reasons: (i) their study did not consider

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the exclusive vortex measurements, but the latitudinal average over 70–90° S and (ii) their study includes the observations from the exceptional winter 2002. Because, our diagnosis (i) yield a positive trend at 95 % confidence intervals for EESC based regression in both cases, including or excluding the winter 2002 data (Table 2). Also, (ii) if 2002 ozone data are included in the regression procedure, the trend increases by about ∼15 % and significance increases by 20 %. However, (iii) our diagnosis with GB measurements inside the vortex core reduces the trends and their significance by about 20 %, indicating the sensitivity of the vortex data. Furthermore, (iv) the regression analyses with the TOMS/OMI data always give higher trend values than those of GB in all our sensitivity studies. Therefore, our results affirm the necessity of a robust data selection procedure for trend studies in the polar regions.

It could be argued that heat flux should compensate for the anomaly of 2002, but the fact that inserting 2002 greatly increases the recovery trend and our certainty that the events of 2002 have nothing to do with any recovery in chlorine chemistry, suggests that heat flux does not compensate for the 2002 anomaly. Therefore, ozone observations from the anomalous winter 2002 must be discarded from the trend studies.

In order to test the strength of the positive trends and to understand the effect of Antarctic stratospheric meteorology in the derived results in 1997–2010, we computed the trends without heat flux in the regressions. The resulting ozone trends in 1997–2010 are not very different from our previous analysis, as both GB and TOMS/OMI data show an insignificant (at 95 % confidence intervals) trend of about +0.7 DU yr^{-1} from the EESC and +1 DU yr^{-1} from the PWLT regressions. In all cases studied here, the trends in 1997–2010 are positive (∼+1 DU yr^{-1}) and significant at 85 % confidence intervals. It gives a clear message that the Antarctic ozone is recovering and a tangible ozone recovery signal should appear in a few years.

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Therefore, we calculated the number of years needed to achieve a significant positive signal in the 95 % confidence intervals using the method of Reinsel et al. (2005), assuming the trend and variance continue as they have from 1997–2010. That is

$$n_{\text{year}} = \left[\frac{(1+z_\beta)\sigma_N}{|\omega_0|} \sqrt{\frac{1+\phi}{1-\phi}} \right]^{2/3} \quad (2)$$

where n_{year} is the number of years to be found, σ_N is the standard deviation of the residuals, z_β is the value corresponding to the 95 % confidence intervals from the standard normal table, $|\omega_0|$ is +1 DU yr^{-1} and ϕ is the autocorrelation of the residuals with one year lag. The experiment suggests it will be 8.8 yr for GB and 9.3 yr for TOMS/OMI to obtain the first detectable signal at the 95 % confidence intervals for both GB and TOMS/OMI ozone. When the inter-annual temperature changes are ignored (i.e. without heat flux), a significant signal can be traced in 10 yr. The EESC based regression shows a relatively longer period for the GB and TOMS/OMI data, 10 and 11 yr, respectively. Apparently, these computations are in very good agreement with that estimated from the ozonesonde measurements by Hassler et al. (2011), who report that 2017–2021 would be the first period of ozone recovery detectable at the South Pole.

5 Conclusions

A comprehensive analysis of chemical ozone loss in the Antarctic vortex from 1989 to 2010 is presented using ground-based Brewer, DOAS, Dobson, and SAOZ, and space-borne TOMS/OMI, GOME, SCIAMACHY, and Aura MLS observations. The passive method is applied to find the ozone loss at each station, and then averaged to find the mean loss in the Antarctic. On average, the ozone loss at Arrival heights, Belgrano, Concordia, Dumont d'Urville, Faraday-Vernadsky, Halley, Marambio, Neumayer,

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Rothera, South Pole, Syowa and Zhongshan, shows about 160–180 DU or 48 %, except for the early years 1989–1991 and the extreme winters of 2002 and 2006. The loss in 1989–1991 and 2002 is about 110–140 DU or 33–40 %, and in 1992–2010 is around 160 DU or 48 %. The vortex edge region stations (e.g. Dumont d'Urville) show a lower loss than that of the stations inside the vortex core (e.g. South Pole). 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 recovers thereafter. The estimated ozone loss time series is consistent with the EESC and temperature distribution in each winter. Probably because of saturation of the vortex core with PSCs and small inter-annual variability of ozone loss, the V_{psc}-ozone loss correlation yields about 0.5, which stands in stark contrast with the Arctic scenario where it is about 0.9. The TOMS/OMI, Aura MLS, GOME and SCIAMACHY observations also exhibit a proportional progress of ozone and ozone loss as for the ground-based measurements throughout the period (1989–2010). The differences among these data are within ±5 %, and are mostly within the error limits of the ground-based and satellite observations.

The piece-wise linear trends estimated from September–November vortex average ground-based stratospheric ozone column in the Antarctic show a clear negative trend of about −5 to −5.6 DU yr^{-1} in 1989–1996 and a positive trend of about +1 DU yr^{-1} in 1997–2010. The ground-based analyses are well supported by the TOMS/OMI observations. The trends based on piece-wise and equivalent effective stratospheric chlorine functions are significant at the 95 % confidence levels in 1989–1996 but are significant only at the 85 % confidence levels thereafter, in 1997–2010. Our forecast suggests that it will take another 8–10 yr to be able to detect a 95 % confidence levels trend in the Antarctic ozone, if the current +1 DU yr^{-1} is continued. So the ozone trends indicate that the ozone in the Antarctic is recovering. However, the Antarctic ozone loss/hole will prevail in much of this century with the given rate of the estimated positive trend and will take another 50 yr to regain the 1980 levels of ozone. Our study confirms that the recovery of the ozone hole is currently camouflaged by natural variability in ozone,

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as exemplified by the response to planetary waves (heat flux), and solar and QBO terms.

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Table 1. The maximum ozone loss (26 September/5 October average) in DU (Dobson Unit) and % and the ozone loss rates estimated between day 225 and 275 from ground-based (GB) measurements in the Antarctic. The selection of this time-line depends on the measurement capability of most GB instruments, where most of them have measurements so that the analyses fairly represent the average of the Antarctic. The loss rates are given in DU/day and % day^{-1} in 50 Days. The uncertainty of the estimated ozone loss is about 3–5 %.

Year	Loss DU	Loss %	Loss Rate DU/day	Loss Rate % day^{-1}
1989	111	37	1.96	0.65
1990	112	37	1.17	0.50
1991	132	43	1.56	0.51
1992	143	48	2.55	0.80
1993	158	49	2.55	0.39
1994	161	51	0.78	0.55
1995	160	51	2.75	0.53
1996	122	41	1.37	0.41
1997	156	49	1.96	0.59
1998	150	50	1.96	0.67
1999	152	48	1.96	0.58
2000	167	51	1.96	0.53
2001	182	53	1.96	0.53
2002	167	40	1.56	0.39
2003	168	54	1.96	0.61
2004	155	45	1.76	0.45
2005	154	50	1.67	0.53
2006	175	55	2.05	0.71
2007	159	50	1.67	0.52
2008	168	53	1.96	0.64
2009	147	49	1.67	0.53
2010	154	46	1.67	0.51

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Table 2. Antarctic ozone trends in DU yr^{-1} estimated from the September–November vortex average ground-based (GB) and TOMS/OMI observations using the PWLT (piece-wise linear trend) and EESC (equivalent effective stratospheric chlorine) regressions. The error bars represent their significance at 95 % confidence intervals.

Instrument	Period	All measurements		All meas./No heat flux		No 2002		No 2002, No heat flux	
		EESC	PWLT	EESC	PWLT	EESC	PWLT	EESC	PWLT
GB	1989–1996	−2.30±1.68	−5.35±2.91	−1.97±2.11	−4.91±3.86	−2.65±1.43	−5.48±2.44	−2.56± 1.55	−5.28±2.68
	1997–2010	0.65±0.47	1.08±1.28	0.55±0.59	1.14±1.70	0.75±0.40	0.94±1.07	0.72±-0.44	0.93±1.18
TOMS/OMI	1989–1996	−2.40±1.85	−5.64±2.84	−2.11± 2.15	−5.25±3.64	−2.76±1.58	−5.79±2.37	−2.69±1.69	−5.62±2.55
	1997–2010	0.67 ±0.52	1.15±1.25	0.59±-0.61	1.21±1.60	0.78±0.44	1.01±1.04	0.75±0.47	0.99±1.12

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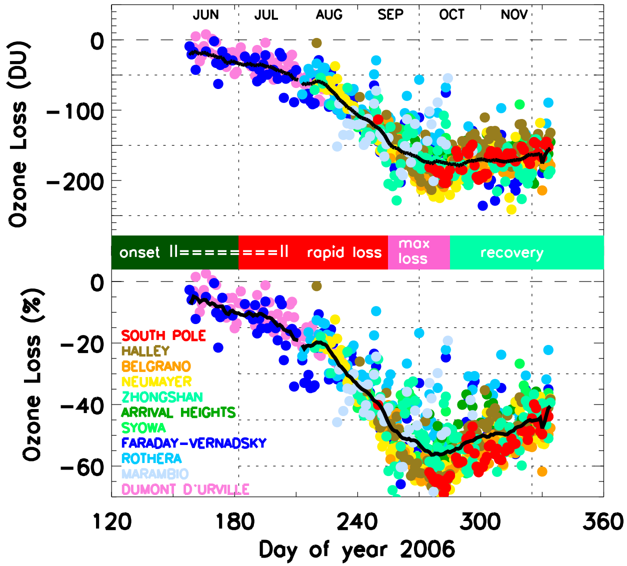

Fig. 1. Chemical ozone loss estimated from 11 ground-based station observations inside the vortex in the Antarctic winter 2006. The ozone loss is estimated as the measured ozone minus the modelled passive tracer (ozone-passive tracer), which is initialised on first of June. The average loss estimated from the observations is shown in solid line. The dotted lines represent 50, 150, and 250 DU or 15, 30, 45 and 60 % of ozone loss. The vertical lines represent day 180, 270 and 325.

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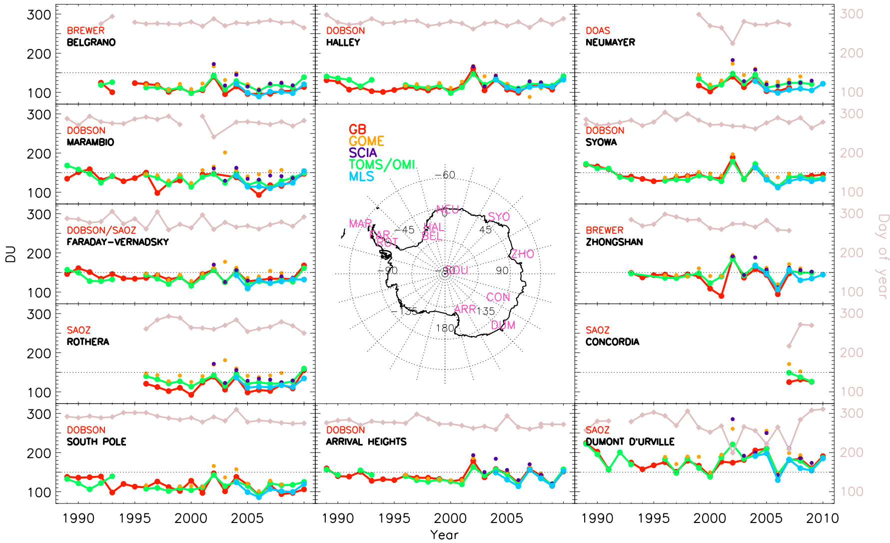

Fig. 2. Distribution of minimum ozone at 12 Antarctic ground-based (GB) stations (red). The corresponding satellite overpass data from TOMS/OMI (green), MLS (pale blue), GOME (yellow), and SCIAMACHY (SCIA – violet) are also shown. The SCIMACHY and GOME measurements are shown by dots for clarity reasons. Station names on the map are demarcated with the first three letters of the stations. The type of GB stations (e.g. SAOZ at Concordia) are also marked on the plots. The SAOZ record at Faraday/Vernadsky starts in 1996. The pale grey line indicates the day when the minimum is observed by the GB sensor (axis on the right). The horizontal dotted lines represent 150 DU of ozone.

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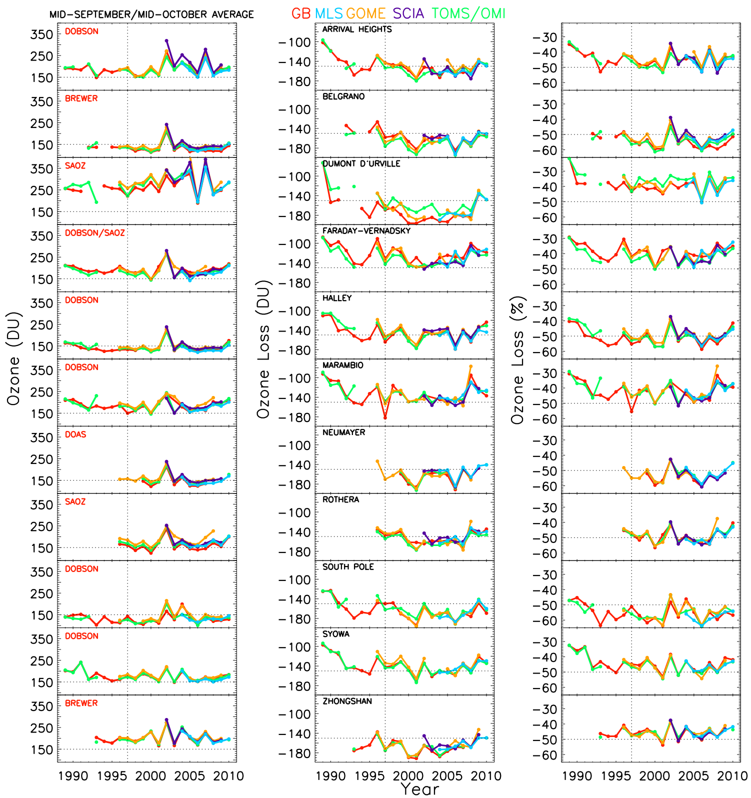

Fig. 3. The average ozone (left) and chemical ozone loss in DU (middle) and % (right) estimated inside the vortex from long-term ground-based (GB) ozone measurements at various Antarctic stations during the mid-September to mid-October period of 1989–2010. The corresponding satellite data from TOMS/OMI (green), MLS (pale blue), GOME (yellow), and SCIAMACHY (SCIA – violet) are also shown. The horizontal dotted lines represent 150 DU of ozone or 150 DU/50 % of ozone loss. The vertical dotted lines represent year 1997.

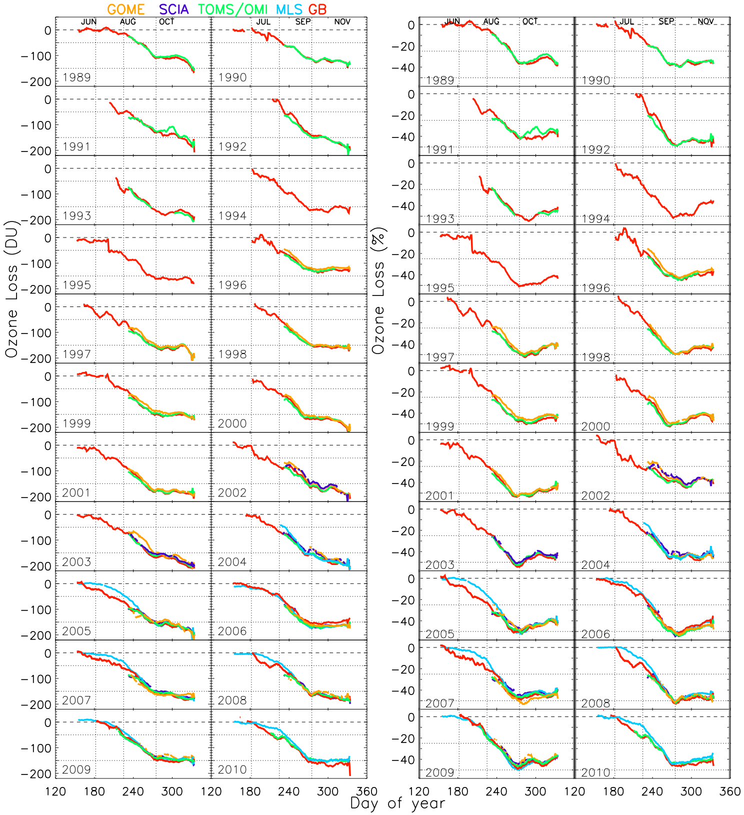

Fig. 4. Temporal evolution of the chemical ozone loss estimated from ground-based (GB) observations inside the vortex are compared to those of TOMS/OMI (green), MLS (pale blue), GOME (yellow), and SCIAMACHY (SCIA – violet) in DU (left) and in % (right) in 1989–2010. The MLS has data for all stations from June onwards, while the GB average is consisted of edge stations in the early winter period. The horizontal dotted lines represent 50 and 150 DU or 25 and 50 % of ozone loss, while the vertical lines represent day 181, 225 and 275.

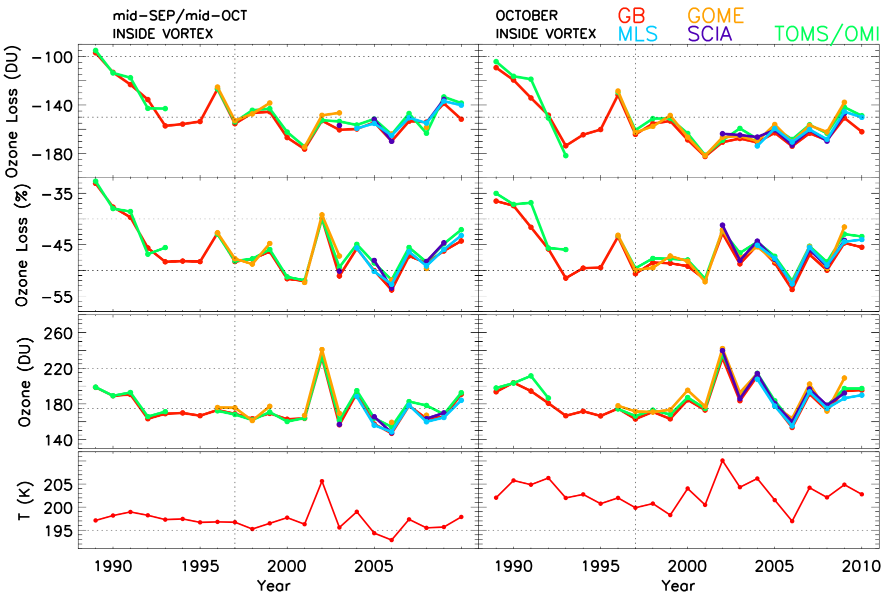

Fig. 5. The ground-based (GB) ozone (in Dobson Unit – DU) and chemical ozone loss (in DU and %) inside the vortex in mid-September/mid-October and October during the Antarctic winters 1989–2010 compared to those of TOMS/OMI (green), MLS (pale blue), GOME (yellow), and SCIAMACHY (SCIA – violet). The corresponding vortex average temperature (in Kelvin – K) from the ECMWF operational analyses is also shown. The horizontal dotted lines represent 100 and 150 DU or 40 and 50 % ozone loss, 175 and 220 DU ozone and 195 K temperature in the respective plots. The vertical lines represent year 1997.

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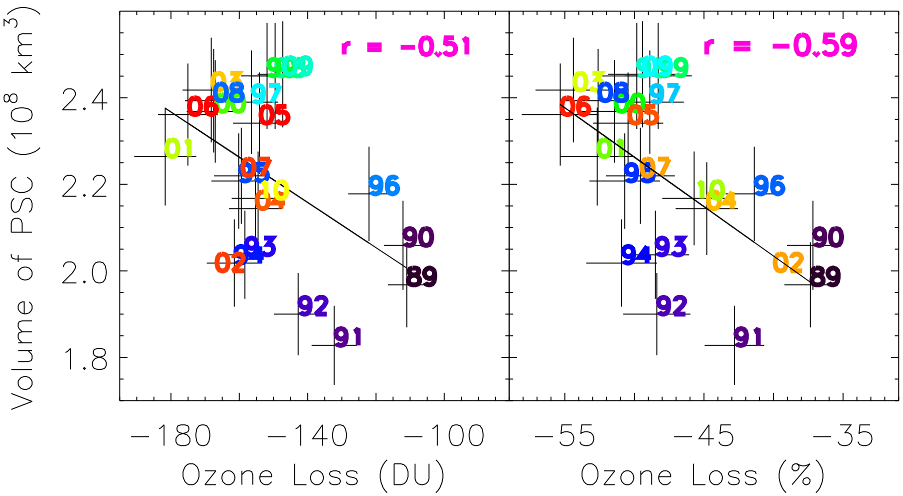

Fig. 6. The correlation between ozone loss from ground-based measurements (taken from Table 1) and the volume of polar stratospheric clouds (PSCs) calculated from the ECMWF operational analyses in 1989–2010. The correlations (r) exclude the anomalous winter 2002.

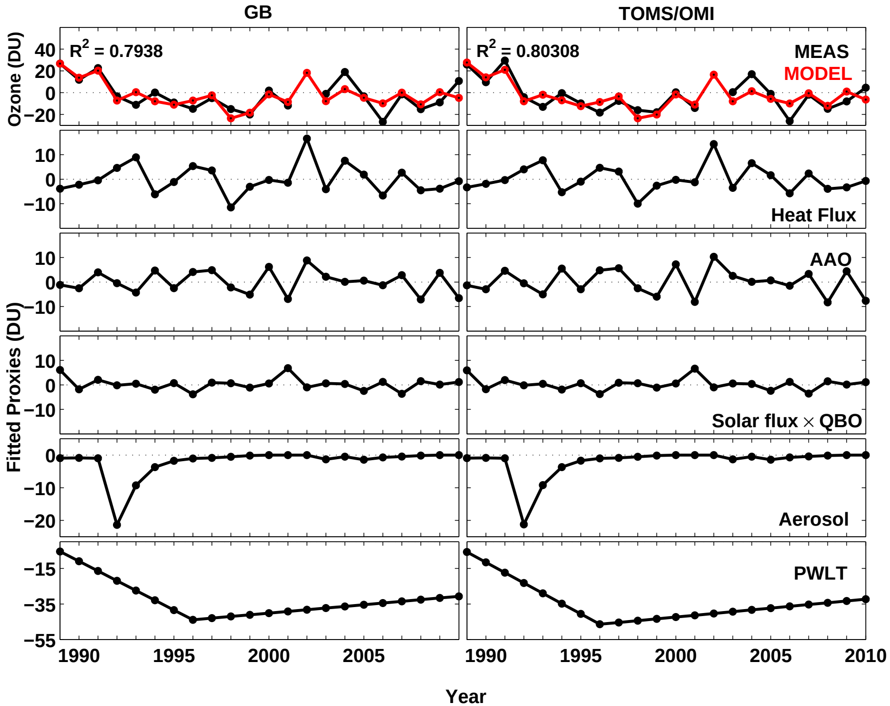

Fig. 7. The vortex average September–November average ozone trends estimated using a multi-variate regression model for the ground-based and TOMS/OMI observations in 1989–2010. The 1994–1995 data gap in TOMS/OMI is filled with the Multi-sensor reanalysis total column values, which mainly come from SBUV space-borne ozone measurements during the period. Top to bottom: deseasonalised ozone (MEAS) and the regression model (top panel), and the contribution of heat flux (second panel), Antarctic Oscillation – AAO (third panel), solar flux multiplied by quasi-biennial oscillation at 40 hPa (fourth panel), Aerosol (fifth panel), and the piece-wise linear trend (bottom panel).

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