CHEMICAL OZONE LOSS IN THE ARCTIC AND ANTARCTIC STRATOSPHERE BETWEEN 1992 AND 2005

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Abstract. Ozone depletion in the lower polar stratosphere is strongly dependent on the the amount of chlorine activation that is controlled by polar stratospheric cloud (PSC) occurrence and thus by temperature. We define a new measure, the PSC formation potential (PFP), suitable for assessing ozone depletion in both polar regions. In contrast to the previously defined volume of possible PSC existence (V_{PSC}), this measure is normalized by the vortex volume in accordance with loss in column ozone averaged over the polar vortex and includes the lifetime of the vortices. Chemical ozone loss, derived between 1991 and 2005 for Arctic and Antarctic winters, correlates well with the PFP in the Arctic. In the Antarctic, chemical ozone loss is saturated. The year-to-year variation of the PFP for the Arctic has risen over the past thirty years with a maximum value in winter 2005. Accordingly, maximum chemical ozone loss has reached Antarctic loss values.

1. Introduction

Ozone-depleting substances are slowly decreasing in the atmosphere as a consequence of the Montreal Protocol so that a recovery of the ozone layer is expected over the coming decades [*WMO*, 2003]. The observed cooling of the polar stratosphere in winter on an annual- and global-mean basis in the last two decades [*WMO*, 2003] enhances the potential for both the existence of polar stratospheric clouds (PSC). A compact relation between chemical ozone loss and the volume of air below the temperature threshold for PSC existence ($\mathrm{V_{PSC}}$) was derived for Arctic winters [*Rex et al.*, 2004; *Tilmes et al.*, 2004]. $\mathrm{V_{PSC}}$ is used as a measure of the potential for halogen activation in the polar vortex. However, $\mathrm{V_{PSC}}$ is dependent on the vortex volume, whereas the chemical loss in column ozone is independent of the volume of the vortex because it is an average value over the entire vortex. Here, we extend this measure to create a measure of halogen activation that is normalized with the vortex volume and therefore suitable for comparison with chemical loss in column ozone, the PSC formation potential (PFP). Using PFP instead of $\mathrm{V_{PSC}}$ is especially important if the volume of the different vortices considered differs greatly, for example, in comparing Arctic and Antarctic conditions. This allows us to discuss how climate change, i.e. changing temperatures and dynamics in the stratosphere, may impact the potential for ozone destruction in the polar vortex. Changing vortex dynamics may indeed have a significant impact on the ozone hole polar winter and spring in the Antarctic [*Huck et al.*, 2005][1]. Moreover, we will discuss the ozone column that would occur in the absence of chemical changes, thus for assumed chemically unperturbed conditions (proxy ozone column [*Tilmes et al.*, 2006]) during winter and spring. Decreasing proxy ozone columns for the

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[1] *Huck et al.* [2005] discuss the evolution of the ozone mass deficit calculated between July and the end of November in the Antarctic. The ozone mass deficit is not directly comparable with the chemical loss in column ozone averaged over the polar vortex as derived here. Here, only the chemical signal is considered.

Arctic indicates dynamical changes in the stratosphere.

2. Differences between Arctic and Antarctic Winter Meteorology

The calculation of V_{PSC} (Fig. 1, top panel) requires a given PSC threshold temperature (T_{PSC}). T_{PSC} is defined here as the threshold temperature for the existence of nitric acid trihydrate (NAT) [*Hanson and Mauersberger*, 1988]. It is calculated assuming seasonally dependent stratospheric profiles of HNO_3 and H_2O derived from ILAS-II observations for the Arctic and Antarctic, including the effect of denitrification and dehydration. The UK Met Office (MetO) and ECMWF reanalysis (ERA 40) [*Randel et al.*, 2004, and refs. therein] are used to calculate V_{PSC} between 400-550 K potential temperature. The ERA40 reanalysis is unreliable in the Antarctic before the satellite era in 1979 [*Randel et al.*, 2004; *Simmons et al.*, 2004] and will not be investigated. Further, Antarctic temperatures after 1979 in ERA40 show a cold bias and an unrealistic vertical structure [*Manney et al.*, 2005a, b; *Randel et al.*, 2004]. Therefore, MetO V_{PSC} values are slightly lower than the ERA40 data. Additionally, V_{PSC} was derived using ECMWF operational analyses and data from the Free University of Berlin (FU-Berlin) [*Rex et al.*, 2004], averaged between 360–550 K (Fig 1, top panel, red and green symbols). The impact of averages of different altitude intervals for the calculation of V_{PSC} is small (see electr. suppl.). For the Arctic, the different data sets agree rather well between 1966 and 1998 [*Manney et al.*, 2005a]. Before 1966, radiosonde instrumentation was limited in the Arctic stratosphere and temperatures are very uncertain. In summary, between 1966 and 1998, the conclusions from all long-term meteorological data sets about the general evolution of V_{PSC} are consistent [*Manney et al.*, 2005a].

Figure 1.

Antarctic temperatures are in general below the PSC threshold for most of the winter between mid-May and the end of September. The variation of V_{PSC} between different winters using MetO data is small. ERA40 and MetO data indicate a slight increase of V_{PSC} between 1991 and 2005 as well as a stronger variability during the last four years (Fig. 1, top panel, colored triangles). V_{PSC} in the Arctic is much smaller than in the Antarctic and shows a strong variation between warm and cold winters (between 0.1×10^7 km^3 in 2001–2002 and 4.6×10^7 km^3 in 2005, based on MetO analysis). An increase of large V_{PSC} values, especially in the last decade, is obvious as described by *Rex et al.* [2004].

The volume of the vortex (V_{Vortex}) (Fig. 1, middle panel) – which is the volume enclosed by the vortex edge, defined as the location of the maximum gradient of potential vorticity (PV) [*Nash et al.*, 1996] – is significantly larger for the Antarctic than for the Arctic. The evolution of MetO and ERA40 values is in agreement, with differences of less than 10% in the overlapping time period. Although V_{PSC} depends on V_{Vortex}, the two values are not correlated (Fig. 1, top and middle panel). The Arctic and Antarctic V_{Vortex} values indicate a decrease since 1979, which is most significant since 1998 for the Antarctic, whereas V_{PSC} have increased slightly since 1990.

The vortex volume should not have an impact on the loss in column ozone averaged over the vortex. To normalize V_{PSC} the ratio between V_{Vortex} is derived as a measure of the possible fraction of the vortex volume exposed by PSCs. A large value of this ratio is expected to correspond to large ozone loss *rates* independent of the lifetime of the vortex. To obtain a measure of the ozone loss potential in each winter, which is also determined by the lifetime of the vortex, we integrated the value V_{PSC} / V_{Vortex} over all days when the vortex existed (using the Nash criterion for 3 potential temperature levels: 475 K, 550K and 650 K) between mid-June and September for Antarctic winters and between mid-December and the end of March for Arctic winters and then divided this sum by the number of days in the period considered. This quantity will be referred to as “PSC formation potential of the polar vortex” (PFP) and is suitable for correlation with the accumulated loss in column ozone in both hemispheres, because it is normalized with regard to V_{Vortex}.

Antarctic PFP values for the MetO and ERA40 data are in agreement within

10% during the overlapping time period. Between 1991 and 2005, PFP indicates an increase of ≈ 25% for Antarctic and cold Arctic winters (Fig. 1, bottom panel and Figure 2). Further, the variation of the Arctic PFP has increased during the last 30 years with the largest value occurring in winter 2005 and the smallest value occurring in 2004. PFP for the coldest Arctic winter is ≈ 27% smaller compared to the smaller PFP value derived for the Antarctic. In 1995 and 1998 radiosonde instrumentation changed at many stations with a systematic bias between the two instrument types. For Alaska stations, an apparent warming of up to 2 K during the nighttime and a cooling during daytime at 50-100 hPa is reported [*Elliott et al.*, 2002]. Underestimated nighttime temperatures – most of the PSC are likely to occur during the night – lead to an overestimation of V_{PSC} before 1995/1998. Any conclusion of increasing V_{PSC} and the resulting increasing PFP since 1995/1998 can therefore not be an artifact caused by changes in radiosonde instrumentation.

3. Calculation of Chemical Loss in Column Ozone

Chemical ozone loss for the Antarctic is derived from HALOE satellite measurements [*Russell et al.*, 1993] using the tracer-tracer correlation method [e.g., *Proffitt et al.*, 1990; *Tilmes et al.*, 2004; *Müller et al.*, 2005]. A detailed description of the technique and a discussion about uncertainties due to mixing processes is given in *Tilmes et al.* [2004]; *Engel et al.* [2006]; *Müller et al.* [2005]. It is shown that this technique is a reliable tool for calculating chemical ozone loss. Antarctic ozone loss is derived here using tracer-tracer correlations in a similar manner to that used for the Arctic. Using this technique, the early winter reference function has to be derived carefully to calculate chemical ozone loss [*Tilmes et al.*, 2004; *Müller et al.*, 2005]. For the Antarctic, no HALOE observations are available in the early vortex. Therefore, ILAS and ILAS-II O_3/N_2O relations for winters 1997 and 2003 are converted to an O_3/CH_4 relation for application to HALOE measurements. For this purpose, we use a CH_4/N_2O relation derived (see electr. suppl.) using 11 flights from whole air sampler measurements between 1995 and 2002, at different seasons, and measured at high northern latitudes [*Engel et al.*, 2002]. ATMOS measurements [*Michelsen et al.*, 1998] show that very similar relations between CH_4/N_2O in the Arctic and Antarctic (Plate 2 of that paper), thus, we assume the derived relation to be also valid for Antarctic conditions. Further, we use CH_4/HF relations derived from HALOE measurements to calculate the O_3/HF reference relation as described in detail by *Tilmes et al.* [2004].

Antarctic chemical ozone loss in column between 350–550 K was derived for all HALOE measurements available inside the vortex core – poleward of the poleward edge of the vortex [*Nash et al.*, 1996] – using HF as the long-lived tracer (Fig.2, electr. suppl.). Owing to the orbit of the HALOE instrument and the location of the polar vortex, in some winters there are no measurements available in the second part of September/October (5 out of 13) for the Antarctic and in March/April (3 out of 14) for the Arctic and therefore no ozone loss values are available for the analysis in Section 4, in Fig. 2.

Arctic chemical ozone loss was derived in a previous study [*Tilmes et al.*, 2004] for winters between 1991–92 and 2002–03 between 380–550 K and for winter 2005 by *von Hobe et al.* [2006] using tracer-tracer correlations. As shown by *Tilmes et al.* [2004], no significant ozone loss was observed below 380 K before winter 2005. Further, the amount of ozone loss differences between different winters above 550 K [*Hoppel et al.*, 2005] is insignificant if loss in column ozone is considered. Chemical ozone loss values derived from ozone soundings are available between 1992 and 2004 [*Rex et al.*, 2004] and for winter 2005 [*Rex*, 2006].

4. Ozone Loss and PSC Formation Potential

Chemical ozone loss, as described in Section 3, is shown in relation to PFP in Figure 2. Here, PFP is averaged between 350-550 K based on the MetO analysis. The relation between ozone loss and PFP in the Arctic is more compact compared to the previously deduced relation between ozone loss and V_{PSC} [*Tilmes et al.*, 2004; *Rex et al.*, 2004]. In winter 2005, chemical loss in column ozone reached

Figure 2.

Antarctic values, whereas the PFP value is significantly smaller than Antarctic values (Fig. 2), as explained below. The study by *Manney et al.* [2006] showed that Arctic local chemical ozone loss in 2005 did not reach the very large values observed in winter 2000. However, in considering chemical loss in column ozone, winter 2005 reached maximum values. This winter is characterized by significant ozone destruction at very low altitudes below 460 K [*von Hobe et al.*, 2006; *Rex*, 2006]. For the Antarctic, the column ozone loss does not change with changing PFP within the uncertainty of the results. This is because the column of proxy ozone in the Antarctic does not differ much from values in column ozone loss. Therefore, chemical ozone loss is almost saturated in this region and although the potential for further ozone loss exists, no ozone is available at lower altitudes (as shown by *Tilmes et al.* [e.g., 2006]). The chemical signal of Antarctic ozone depletion averaged over the polar vortex does not show any evidence of a chemical recovery due to decreasing chlorine content [*Engel et al.*, 2002], but rather indicates a slight increase calculated between 350 and 550 K potential temperature (see Figure 2). The recovery due to decreasing stratospheric chlorine content will become visible if the saturation (that already occurred in the nineties) no longer occurs, which cannot be expected before ≈ 2015 [*Newman et al.*, 2004].

For the Arctic, proxy ozone is substantially larger in comparison to austral spring (Fig. 2, open triangles). Slightly smaller values of proxy ozone are obvious for colder and less disturbed Arctic winters. For cold Arctic winters, less than half the amount of the proxy ozone is destroyed at the altitudes considered. Therefore, the potential for larger amounts of chemical ozone loss is present for the Arctic winters in the near future until the halogen loading of the stratosphere has significantly decreased. The effect of Pinatubo on chemical ozone loss based on HALOE observations in Arctic winters 1992 and 1993 is significantly larger than that derived from ozone soundings and other studies [e.g., *Lucic et al.*, 1999; *Manney et al.*, 1994].

5. Discussion and Conclusion

We use 47 years of meteorological analyses and 14 years of satellite data to analyze the sensitivity of chemical ozone loss in both the Arctic and Antarctic to temperature changes and, extending the work of *Rex et al.* [2004], to dynamical changes in the polar stratosphere. The meteorological conditions are different in the two hemispheres and both the column ozone at the beginning of the winter and the proxy ozone during the winter are much larger in the Arctic. For the cold Arctic and Antarctic winters, PFP has been increasing over the last 30 years because of decreasing temperatures and decreasing vortex volume. Correspondingly, chemical ozone loss values in the Arctic reached Antarctic values in 2005, whereas the entire ozone column is much larger in the Arctic compared to the Antarctic. The linear relation between column ozone loss and PFP indicates a potential for further increasing ozone loss values in the near future, because Arctic ozone loss is currently not saturated. Antarctic chemical ozone loss is almost saturated and therefore, increasing PFP cannot change ozone loss values. Increasing chemical ozone loss might occur if the recent tendency for greater variability of Arctic winter conditions is driven by increasing greenhouse gases. Further, stronger planetary wave activity may enhance the proxy ozone in early winter. This may change the volume of the vortex for both the Arctic and Antarctic and therefore impact the PFP and thus chemical ozone loss.

Three-dimensional coupled chemistry climate models (CCMs), the main tool for predicting the future of polar ozone, show deficiencies in particular with regard to the prediction of temperatures and dynamics of the polar stratosphere [e.g., *WMO*, 2003]. It has been suggested that the relation between PSC formation potential and ozone loss could be used to evaluate the temperature sensitivity of accumulated polar chemical ozone loss in CCMs [*Eyring et al.*, 2005]. Different vortex volumes simulated by different climate models may have a significant impact on the simulated ozone loss. Using the PFP instead of V_{PSC} to describe the climate sensitivity of chemical ozone loss in CCMs, the impact of different vortex dynamics will be separated from the chemical signal. In this way, the chemical signal of ozone changes can be analyzed although models might miscalculate the volume of the vortex. Therefore, using the PFP is also recommended for assessing the performance of CCMs.

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Figure Captions

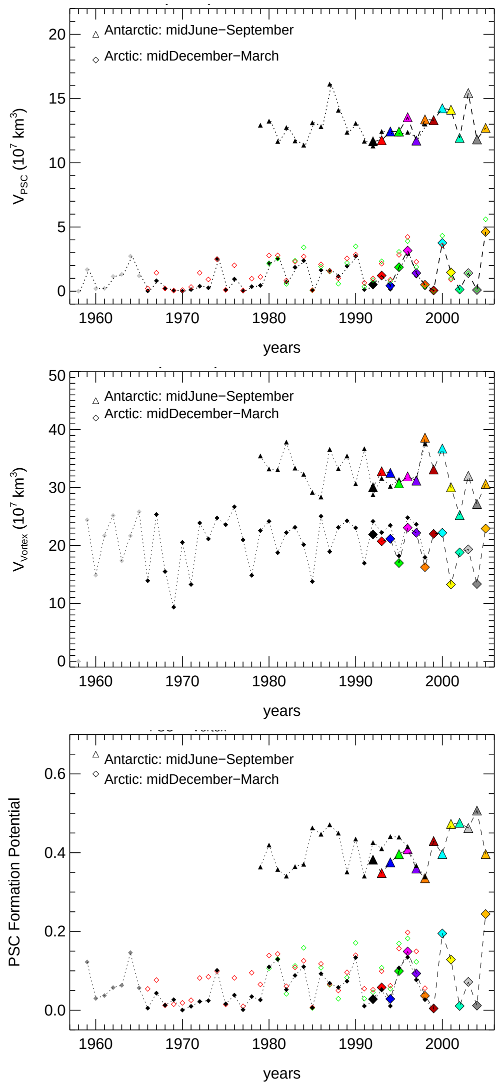

Figure 1. V_{PSC} (top panel), V_{Vortex} (middle panel) and PFP (bottom panel) (see text) averaged between 400–550 K and between mid-June and the end of September for Antarctic winters (triangles) and between mid-December and the end of March for Arctic winters (diamonds) between 1991 and 2005 derived using MetO analyses (colored symbols, connected by a dashed line) and between 1958 (1979 for the Antarctic) and 1999 using ECMWF reanalysis (ERA 40) (gray/black solid symbols, connected by a dotted line). Arctic V_{PSC} based on FU-Berlin meteorological data (red diamonds) and ECMWF ERA-15 data extended by operational ECMWF analysis (green diamonds), averaged between 360 and 550 K are shown.

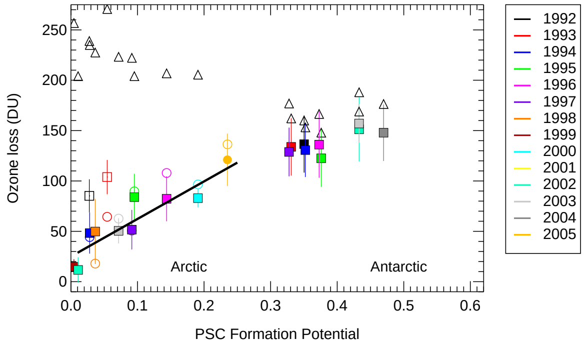

Figure 2. Relation between the column ozone loss (DU) in March (NH) and second part of September and October (SH) and PFP for the years 1992 to 2005 between 350–550 K using the tracer-tracer method (colored solid squares). Correspondingly, the column in proxy ozone is shown, open symbols. The linear relation for the Arctic (calculated excluding winters 1992 and 1993 that are strongly impacted by the eruption of Mt. Pinatubo, open squares) is shown as a black line. Moreover, ozone loss values calculated from ozone sounding [*Rex et al.*, 2004] are shown (open circles) and from the O_3 measurements onboard M55 Geophysica (solid circle) [*von Hobe et al.*, 2006].