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Spatial, temporal, and vertical variability of polar stratospheric ozone loss in the Arctic winters 2004/05–2009/10

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Abstract

The stratospheric ozone loss during the Arctic winters 2004/05–2009/10 is investigated by using high resolution simulations from the chemical transport model Mimosa-Chim and observations from Microwave Limb Sounder (MLS) on Aura by the passive tracer technique. The winter 2004/05 was the coldest of the series with strongest chlorine activation. The ozone loss diagnosed from both model and measurements inside the polar vortex at 475 K ranges from ~1–0.7 ppmv in the warm winter 2005/06 to 1.7 ppmv in the cold winter 2004/05. Halogenated (chlorine and bromine) catalytic cycles contribute to 75–90% of the accumulated ozone loss at this level. At 675 K the lowest loss of ~0.4 ppmv is computed in 2008/09 from both simulations and observations and, the highest loss is estimated in 2006/07 by the model (1.3 ppmv) and in 2004/05 by MLS (1.5 ppmv). Most of the ozone loss (60–75%) at this level results from cycles catalysed by nitrogen oxides (NO and NO₂) rather than halogens. At both 475 and 675 K levels the simulated ozone evolution inside the polar vortex is in reasonably good agreement with the observations. The ozone total column loss deduced from the model calculations at the MLS sampling locations inside the vortex ranges between 40 DU in 2005/06 and 94 DU in 2004/05, while that derived from observations ranges between 37 DU and 111 DU in the same winters. These estimates from both Mimosa-Chim and MLS are in general good agreement with those from the ground-based UV-VIS (ultra violet–visible) ozone loss analyses for the respective winters.

1 Introduction

Unlike in the Antarctic winter stratosphere, the chemical ozone loss in the Arctic is highly variable. This variability is primarily caused by the variations in Arctic meteorology, which is disturbed by planetary wave forcing triggered by mountain orography. Wave activity and stratospheric warming often disrupt the prerequisite condition for the formation of polar stratospheric clouds (PSCs), which control the variability of ozone

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loss to a large extent. For instance, the Arctic winters 2000, 2005 and 2008 were very cold and total ozone loss reached about 20–30% in late-March (WMO, 2002, 2006; Goutail et al., 2010). In contrast, 2001, 2004, 2006 were rather warm winters with total ozone loss not exceeding ~10–15% (WMO, 2006; Manney et al., 2007).

5 In this paper, we study the ozone loss and its driving chemical cycles for the recent winters 2005–2010 and assess the variability of ozone loss in a quantitative perspective using simulations and measurements. Calculations using a high resolution chemical transport model (CTM) together with satellite observations are exploited for this purpose. While the role of halogen cycles in the polar ozone loss is relatively well 10 understood (WMO, 2006), the contribution from other cycles have less been explored. Therefore, we also investigate contribution of other ozone depleting cycles like those involving NO_x compounds in ozone loss processes in the lower and middle stratosphere (up to ~850 K isentropic level).

15 The article is organised as follows: After Introduction, the model, measurements and meteorological situation of the studied winters are presented in Sects. 2–4. The simulations of ozone loss, ozone and ClO , and their comparison with observations for the recent winters are described in Sect. 5. The simulated ozone loss and production rates, contribution of different chemical cycles to the ozone loss and the ozone column loss during the winters are analysed in Sect. 6. Section 7 briefs the conclusion of the 20 study.

2 The simulations with Mimosa-Chim CTM

25 The Mimosa-Chim CTM has been successfully used for the diagnosis of polar ozone loss in previous winters (Kuttippurath et al., 2009; Tripathi et al., 2007, 2006) and is described in detail in Tripathi et al. (2006). The model combines the Mimosa advection code (Hauchecorne et al., 2002) with the Reprobus chemistry scheme (Lefèvre et al., 1994). The model has a horizontal resolution of $1^\circ \times 1^\circ$. It has isentropic vertical coordinates on 25 levels, resolved by 5 K in the lower stratosphere. Winds and temperatures

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are taken from the European Centre for Medium Range Weather Forecasts (ECMWF) operational analyses. The diabatic transport through the isentropes is computed from the heating rate calculations by MIDRAD (Shine, 1987), driven by climatological H_2O , CO_2 and by the O_3 fields calculated by Mimosa-Chim.

5 The model includes a comprehensive description of stratospheric chemistry. Absorption cross-sections and kinetics data are based on Sander et al. (2006). The absorption cross-sections of Cl_2O_2 are taken from Burkholder et al. (1990) and are extrapolated to 450 nm. They are in very good agreement with the most recent Cl_2O_2 spectrum measurements by Papanastasiou et al. (2009). Monthly varying H_2SO_4 fields leading to the 10 formation of liquid aerosols in the CTM are computed from the outputs of a 2-D-model long-term simulation, which considers the impacts of volcanic eruptions. The heterogeneous chemistry contains reactions on binary and ternary liquid aerosols, nitric acid trihydrate (NAT), and on water-ice particles. The composition of liquid aerosols is calculated analytically (Luo et al., 1995). The ice particles are assumed to incorporate 15 HNO_3 in the form of NAT described by a bimodal size distribution (Davies et al., 2002). Cl_y and Br_y are explicitly calculated from their long-lived sources at the surface and are therefore time dependent. An additional 6 pptv of bromine in the form of CH_2Br_2 is added to Br_y to represent the contribution of brominated short lived species reaching the stratosphere (WMO, 2006).

20 For each Arctic winter considered here the model was run from 1 December to 31 March. Initialisation of ozone on 1 December was provided by the ECMWF operational analyses. Other species in Mimosa-Chim were initialised from a long-term simulation of the Reprobus CTM driven by ECMWF meteorological analyses.

3 Measurements: MLS on Aura

25 Ozone and ClO observations (version 2.2) from the Microwave Limb Sounder (MLS) aboard Aura are used to compare with the simulations. The retrieved ozone profiles have a vertical range of 215–0.02 hPa and a vertical resolution of \sim 3 km, while the

horizontal resolution of a profile is \sim 200 km. The vertical range of ClO is 100–0.1 hPa and the vertical resolution is 3–3.5 km, whereas the horizontal resolution ranges from 350 to 500 km. The estimated accuracy is 5–10% for ozone and 10–20% for ClO depending on altitude (Froidevaux et al., 2006; Santee et al., 2008).

5 4 Temperature and PSCs during the winters

Figure 1 shows the area covered by PSCs (Apsc) calculated from the ECMWF temperature and pressure data for the last six winters. PSCs are assumed here to form at the NAT frost point according to Hanson and Mauersberger (1988) and are calculated using climatological values of HNO_3 and H_2O (e.g. Tripathi et al., 2007). Winter

10 2005 shows the largest PSC area with a maximum of $1.7 \times 10^7 \text{ km}^2$ in late-January. Considerable area of PSC is also found in December–January 2008, with a maximum value of $1.4 \times 10^7 \text{ km}^2$ in mid-January. Due to a vortex split occurrence in mid-December at 475 K and a major warming in February 2010, Apsc during the winter is reduced and it shows a maximum of $1.2 \times 10^7 \text{ km}^2$ in mid-January. The warm winters
15 2006 and 2009 show much smaller PSC area, limited in late-December/early-January with a peak value of about $0.8 \times 10^7 \text{ km}^2$. In winter 2007, the largest area of PSCs, $1.0 \times 10^7 \text{ km}^2$, is observed in late-December.

5 Results

5.1 Simulated ozone loss

20 The ozone chemical loss is computed from the difference between a passive tracer initialised identically to ozone at the beginning of the simulation and the chemically integrated ozone (i.e., $\text{Tracer-Ozone}_{\text{model}}$). As an example, Fig. 2 shows the passive tracer, ozone, and the difference (chemical ozone loss) calculated at 475 K on 15 March for

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each winter. In the figure, polar vortices with high mixing ratios of around 5 ppmv corresponding to warm winters and reduced mixing ratios of around 3 ppmv corresponding to cold winters, are clearly visible. Thus, the maps tangibly illustrate the strong inter-annual variability in the meteorology and chemical ozone loss in the Arctic, with large losses diagnosed inside the polar vortex in 2005 and 2008, more limited loss in 2007 and 2010, and the absence of vortex, as of 15 March, in 2006 and 2009. The large loss simulated in a vast vortex on 15 March 2005, while other winters show a shrunk or dissipated vortex, suggest sustained and high ozone depletion during that winter. Each winter in this study is characterised by different dynamical conditions and polar processing, which lead to various magnitudes of ozone depletion. Therefore, the situation merits a thorough investigation on the chemical characterisation during these winters. We present a detailed and quantitative analyses on these ozone simulations in comparison with MLS observations in the following section.

5.2 Comparison with observation

5.2.1 Ozone loss: vertical development

Figure 3 (left panel) displays the vertical structure of the accumulated chemical ozone loss computed for the Arctic winters 2005–2010. The results are averaged inside the polar vortex defined as the area enclosed inside 65° N of equivalent latitude (EqL) (see Müller et al., 2008 for further discussions on adequate definition of polar vortex). The vortex ozone loss average computed from model grids and at the MLS sampling points show small differences. Therefore, we present the ozone loss computed at the MLS footprints inside the vortex for each winter for the comparison purpose.

Among the winters, 2005 exhibits the largest ozone loss with a maximum of 1.7 ppmv in March around 475 K. The ozone loss is spread vertically between 450 and 850 K in January–February, reaching 1.5 ppmv above 600 K in late February. In March, most of the loss is confined between 400 and 600 K. A similar level of depletion is simulated during 2010, where large reduction in ozone is estimated from mid-January to March

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at 450–800 K, and the peak ozone loss of about 1.4 ppmv is found at 600 K. Comparatively large losses are also found during the cold winters 2007 and 2008. In 2007, the ozone loss shows a double peak feature with the maximum (1.3 ppmv) at 675 K. In 2008, the ozone loss is delimited between 450 and 600 K with a peak loss of 1.4 ppmv around 475 K. Little ozone loss is computed above the 650 K level in this winter. Due to early final warming (since there was no strong or well-defined polar vortex, we take the major warming in late-January/early-February 2006 and 2009 as the final warming), the winters 2006 and 2009 show limited ozone loss, not exceeding ~0.8 ppmv. The winter 2009 presents the smallest vertical extent in diagnosed ozone loss, which is mainly controlled below 650 K until the final warming.

Figure 3 (right panel) describes the temporal evolution of the vertical distribution of the vortex averaged ozone loss derived from the observations. Ozone loss from the measurements is computed in a similar way as for the simulation (the difference between the passive ozone tracer sampled at each measurement point and observed ozone). In agreement with the simulations, comparatively large losses are estimated from the measurements in 2005, 2008 and 2010, and are reaching up to ~1.4–1.6 ppmv at the end of March depending on year. The simulations reproduce quite well the gross features of observed ozone loss in each winter, e.g. its onset in the course of the winter, the altitude of its maximum and its vertical distribution. They tend however to underestimate the measured loss, except in 2009, where the differences are mostly within 0.2 ppmv. In 2010, the computed loss from MLS is about 0.5 ppmv larger than that from Mimosa-Chim. The large loss observed during this winter seems to be due to relatively high tracer values simulated after the warming in February. In 2005, nonetheless, the model does simulate the second ozone loss maximum observed around 600 K albeit with a lesser amplitude. In both cases, the results show a large ozone loss in the middle stratosphere, as compared to other winters followed by a strong decrease in March. In addition, both model and observations find the highest loss above 500 K in 2007.

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5.2.2 Ozone: vertical distribution

Since the passive method used for the computations of ozone loss depends heavily on the tracer calculations, possible problems in tracer transport can adversely affect the evaluation. Therefore, it is fair to compare also the measured and modelled ozone fields. Figure 4 compares the vortex averaged ozone measurements to the model results sampled at the same time and location as the satellite observations. Both measurements and simulations express similar maximum and exhibit a rather good agreement, with differences within 0.4 ppmv depending on isentropic level and time. In general, the comparison yields good agreement in the lower stratosphere, below 600 K in particular, consistent with ozone loss survey. When we look into the details of ozone modelling, it reveals that the calculations are in very good agreement with the observations during the warm winters 2006 and 2009. The model captures well the ozone enhancement during the warming events, specifically at higher altitudes, in January–February 2006 and 2009. The middle stratospheric ozone simulations during these periods are higher than that of other winters in accordance with the observations. The higher ozone values due to meridional transport of ozone rich air masses from lower latitudes, associated with a major warming in February 2010, can also be seen in both data sets. Nevertheless, the simulations show some differences with observations in December–January 2005, 2008 and 2010 above 675 K, linked to differences in subsidence. Inter-annual variability in the evolution of ozone with altitude is apparent in the figure. For instance, the winter 2005 shows low ozone values in the lower stratosphere up to 600 K and the ozone maximum in the winter 2007 is comparatively lower than that of other winters. Therefore, most features found in ozone loss (Fig. 3) are analogous to the distribution of ozone in the model and MLS. Obviously, some other features such as higher ozone loss estimated with MLS data in March 2010, can be attributed to inaccuracies in the tracer simulations after the warming in February.

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5.2.3 CIO: vertical structure

Figure 5 compares the temporal evolution of MLS and Mimosa-Chim CIO for the various winters. As expected from large areas of PSCs, the observations show high chlorine activation during the winters 2005, 2008, and 2010, where enhanced CIO values are observed in the lower stratosphere up to about 600 K. In these winters, vortex averaged CIO reach 1.2–1.5 ppbv around 550 K in January. Chlorine activation usually starts in December above 475 K (in late-December during the first two winters and a little earlier in the later ones) and then extends lower down in the course of these winters. Both simulated and measured CIO occupy a larger vertical stretch and exhibit higher values in January 2010 as compared to other winters, consistent with higher ozone loss measured during the period. The simulations generally reproduce the observed CIO and its variability throughout the winters quite well, although some differences are seen. In 2005 and 2010, a stronger chlorine activation extending up to 650 K is simulated in late-December compared to the observations. In 2005, later in the winter, higher CIO values are observed in MLS extending up to mid-March. This discrepancy explains the stronger ozone loss derived from the observations around 500–600 K during this winter (Fig. 3). In 2007, Mimosa-Chim clearly underestimates the observed chlorine activation. The vortex averaged CIO in Mimosa-Chim is lower by about 0.4 ppbv, which explains the underestimation of accumulated ozone loss in the simulations in 2007. In other winters, the simulations show generally a good agreement with the observations at most altitudes. A closer examination of the ozone loss in the lower and middle stratosphere at two representative altitudes, 475 and 675 K, is presented in the following sections.

5.2.4 Comparisons: lower stratosphere

As shown by Fig. 3 the simulated loss until January is generally within 0.2 ppmv and it varies in January–March for each year at 475 K. The simulated accumulated ozone loss at the end of March is, respectively, 1.7, 0.7, 1.0, 1.3, 0.8 and 1.1 ppmv in 2005,

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2006, 2007, 2008, 2009 and 2010. These values underestimate observed values by about 0.2 ppmv, except in 2010 where the difference is larger (0.8 ppmv). For this winter, the discrepancy is due, in addition to the underestimation of chlorine activation by the model, to a sudden increase in the passive ozone tracer after the major warming in February 2010, which might induce an overestimation of the ozone loss estimated from the observations.

5 The ozone loss obtained from our study is in general good agreement with that obtained from other techniques for the winter 2005 (WMO, 2006), 2006 (Manney et al., 2007) and 2007 (Rösevall et al., 2008). Table 1 presents the comparison of ozone loss 10 derived from various measurements and model calculations for the winter 2005. The maximum loss simulated at 475 K is about 1.7 ppmv in 2005, which compares well with that of Grooß and Müller (2007), who compute around 1.6 ppmv as the peak depletion. Our ozone loss estimations are also in very good agreement with those from Singleton et al. (2007); Manney et al. (2006); Amraoui et al. (2008); Rösevall et al. (2007); 15 Jackson and Orsolini (2008) and Tsvetkova et al. (2007), as we estimate comparable values in respective periods. Nevertheless, the peak ozone loss altitude shown by the aforesaid works are generally about 25 K lower than our analysis. Such a discrepancy between the various techniques was also noted by Grooß et al. (2005a) for the winter 2003. The only diagnosis that departs considerably from all other evaluation 20 is Rösevall et al. (2007) using SMR (Submillimeter Radiometer) on Odin measurements/assimilation. This could be due to the peculiarity of their method, which is prone to more mixing and dilution in the vortex air, in addition to other reasons presented in Jackson and Orsolini (2008). It has to be recalled that a similar range of ozone loss 25 values, from 0.7 to 2.3 ppmv, were also computed for the cold Arctic winter 2000 by various methods (Newman et al., 2002).

As discussed in Sect. 5.2.3 (Fig. 5), high values of ClO in the late winters indicate a strong chlorine activation and are consistent with the area of PSCs (Fig. 1). In agreement with the measured and simulated ozone loss and Apsc, the chlorine activation is predominant in 2005, moderate in 2010, and is mild in 2006 at 475 K. The winters

5.2.5 Comparisons: middle stratosphere

5 As evident in Fig. 3, the simulated ozone loss at 675 K is around 0.2 ppmv in early-
 January in most winters. The maximum loss reaches 1.2, 0.7, 1.1, 0.8, 0.4, and
 10 1.3 ppmv in 2005, 2006, 2007, 2008, 2009, and 2010, respectively. The calculated
 losses show reasonable agreement with the observed ones at this altitude, except in
 15 2005 and 2010 where it is largely underestimated. In December 2006 and 2007 too,
 the measured losses are slightly larger than that of the calculations. The reasons for
 the offset found in 2010 are already presented in Sect. 5.2.4. The large loss simulated
 around 675 K in January–February 2005 is also confirmed by other estimations (Jin
 et al., 2006; Rex et al., 2006; Grooß and Müller, 2007; Tsvetkova et al., 2007; Jackson
 and Orsolini, 2008). The high ozone depletion simulated at 675 K are in good agree-
 20 ment with that of Grooß and Müller (2007), who estimate a similar loss at this altitude.
 However, this double peak structure is not pronounced in the analyses of Singleton
 et al. (2007) and thus, the measured and simulated results from their study are consid-
 erably lower (about 0.7 ppmv) than our results. There is only a little amount of active
 chlorine present at 675 K, as most of it is found below 600 K. Therefore, key factors
 driving ozone loss at 675 K will be discussed in the succeeding sections.

6 Discussion

6.1 Ozone loss and production rates

To gain further insights into the inter-annual variability of ozone in the Arctic vortex, we have calculated the ozone loss and production rates for the winters 2005–2010.

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Figure 6 shows the vortex averaged instantaneous ozone loss and production rates in ppbv per sunlit hour (ppbv/sh) at 475 (top panel) and 675 K (bottom panel).

6.1.1 Lower stratosphere

At 475 K, the winter 2005 shows the largest loss rate of around 5 ppbv/sh in February.

5 In 2010, loss rates of 3–5 ppbv/sh are calculated in mid-January/mid-February, while relatively lower depletion rates are found in 2008 and 2007 during these months. The warm winters 2009 and 2006 show loss rates up to 3–4 ppbv/sh in December and mid-January respectively, which are higher than those of the cold winters during the same period. There is hardly any ozone production at this isentropic level.

10 For the winters discussed here, there are no other studies with which to compare our simulated ozone loss rates. However, the ozone loss rates analysed from Match ozonesonde observations for a number of previous Arctic winters are available for the comparisons (Frieler et al., 2007). They derive ozone loss rates (seven/ten day averages) of 5–10 ppbv/sh at 490 K in 1995, 5–8 ppbv/sh at 475 K in 1996, 6–7 ppbv/sh at 500 K in 2000, 4.5–8.5 ppbv/sh at 475 K in 2001 and 4–5 ppbv/sh at 475 K in 2003 15 in January. Our results are in general good agreement with these figures. For instance, both Match observations and our simulations show higher loss rates in late-January/early-February period. Further, as shown by the measurements, the loss rates in warm winters rarely extend beyond January, but they are higher than those for most cold winters during the same period. Additionally, cold winters with sustained 20 loss show simulated loss rates of ~5 ppbv/sh in January/mid-March, consistent with the measured rates (in the Arctic winter 2000 and Antarctic winter 2003). Thus in general, the comparison provides a good agreement with earlier results and a consistent evaluation throughout the winters.

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6.1.2 Middle stratosphere

At 675 K, ozone loss and production rates tend to increase with time until February. The largest loss rates of our study are found in February–March 2008, up to 12 ppbv/sh. In 2010, elevated loss rates of 4–9 ppbv/sh are simulated in mid-February/mid-March.

5 A similar evolution of production rates is also calculated during these two winters, in which the latter shows a massive ozone production of 5–19 ppbv/sh. The large loss of 2–6 ppbv/sh is masked by enhanced production of 2–14 ppbv/sh in mid-March 2005. On the other hand, loss rates dominate over production rates in 2007 except in late-
10 February, which is consistent with the highest ozone loss found at 675 K in March. The warm winter 2006 records the largest loss and production rates in December–January in accordance with higher chlorine activation and larger ozone loss estimated during that period.

Figure 7 shows the potential vorticity (PV) maps on 15 March of each year at 675 K. Since ozone production depends solely on sunlight, the movement of vortex over ill-
15 illuminated regions causes the variations in the production. As can be seen from the figure, the displacement of vortex to the mid-latitudes explains the reasons for higher production rates in 2010, 2008 and 2005 compared to other winters at 675 K. Therefore, the major warming periods accompanied by vortex displacement, disintegration or breakdown show relatively high ozone production rates. This is clearly manifested
20 in late-January/early-February in 2006 and 2009, and late-February/early-March 2010, during which the polar vortices were severely disturbed and displaced off the pole by major warming events (Manney et al., 2006; Flury et al., 2009; Kuttippurath et al., 2010). Further, it is evident from Fig. 6 that the production rates in March increase with time, which are well anticipated as the final warming approaches. In addition,
25 the high production rates simulated in late-February 2010 exactly coincide the major warming occurred during the period. On the other hand, a pole centred vortex and hence, comparatively diminished production rates are found in 2007.

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6.2 Chemical cycles

In order to better understand the prime chemical cycles driving the ozone loss inside the vortex in the lower and middle stratosphere, we have evaluated their relative contribution to the total loss, as a function of time at both 475 and 675 K levels, for the 5 winters discussed here. This contribution is shown in Fig. 8 left panel for the lower stratosphere and right panel for the middle stratosphere. Contribution of each cycle is divided by the total contribution from all cycles and then multiplied by 100 to get the relative contribution of each cycle in percent.

6.2.1 Lower stratosphere

10 The importance of halogen cycles in ozone destroying processes in the polar lower stratosphere is well known (e.g. Vogel et al., 2003; Groß et al., 2005a; WMO, 2002) and this study too shows similar results. At 475 K, the ClO–ClO and ClO–BrO cycles represent 80–90% of the total loss (e.g. Frieler et al., 2007; Woyke et al., 1999). The ClO–O cycle contributes 10% to the loss throughout the winter at this level, consistent with a previous study at 465 K based on MLS/UARS (Upper Atmosphere Research Satellite) measurements in the Arctic and Antarctic winter of 1993 (MacKenzie et al., 1996). The ClO dimer cycle is prominent in January/mid-March (since the ozone loss before January is very small, the contribution before the period is not shown), with a maximum contribution of ~50% in mid-January/mid-February. Because of its 15 quadratic dependence on ClO, the efficiency of the ClO–ClO cycle to destroy ozone falls very rapidly when active chlorine returns to reservoir forms at the end of the winter. This is not the case for the contribution of BrO–ClO, which decreases not as rapidly in these conditions, and therefore becomes larger than that of the ClO–ClO cycle in early-March. When the ClO dimer cycle becomes less important, the contribution from 20 ClO–BrO enhances. A similar result was also observed by Butz et al. (2007) in the Arctic winter 1999 from balloon-borne measurements. From early-March onwards, the contributions of the HO_x and NO_x cycles grow quickly and become the active ozone 25

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depleting cycles in the second half of the month.

Another interesting feature to note is the contribution of the aforesaid cycles in the winter 2010. During this winter temperatures were relatively high and as emphasised previously, the vortex was subjected to a major warming and subsequent split. Therefore, in early-February the ClO–ClO contribution fell dramatically and contribution from other cycles (HO_x and BrO–ClO cycles in particular) dominated later during the winter for the reasons stated above. The contribution from HO_x dominates during warming periods, and is demonstrated by its higher contribution during the vortex dissipation (mid/late-March) or major warming periods (late-January 2006 and 2009, and mid-February 2010). This is also expected, since increase in water vapour and nitric acid during the phase of warming (e.g. Flury et al., 2009) are sources of HO_x , and hence contribution from this cycle outweighs others (e.g. Marchand et al., 2005).

The maximum contribution of the ClO–ClO cycle to the total loss varies from ~50% in cold winters to ~40% in warm winters. In contrast, contribution from ClO–BrO equals that of ClO–ClO in warm winters and decreases to ~25–30% in cold winters. The larger difference in the contribution of both cycles, during the period of sustained ozone loss, is found in the winter 2008 from January to late-February.

Our analyses on the contribution of halogens to the total loss are consistent with those derived in Friele et al. (2007). Using a photochemical box model, they show a contribution of 50% from the ClO dimer, ~27–48% from BrO–ClO and 5–10% from the ClO–O to the total loss in the Arctic winters 1995, 1996, 2000, 2001 and in the Antarctic winter 2003 in the lower stratosphere. They also find that the efficiency of the BrO–ClO cycle increases with faster photolysis rate of ClO dimer. Studies using MLS/UARS measurements for the Antarctic winters 1992–1994 also point out that these two cycles account nearly for 90% of the total loss in the lower stratosphere (Wu and Dessler, 2001). Therefore, our study affirms the fact that the odd oxygen loss in the polar winter stratosphere is dominated by the ClO dimer and ClO–BrO catalytic cycles, which is quite in line with our current theoretical understanding, and are consistent with the findings of previous studies.

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6.2.2 Middle stratosphere

In contrast to what is found at 475 K, the halogen catalysed cycles play little role in the Arctic ozone depletion at 675 K, as demonstrated in Fig. 8 (right panel). At this level, the ozone loss is essentially due to the NO–NO₂ cycle, which represents 50–70% of the total depletion in February–March, complemented by the ClO–O cycle that contributes about 10–20% to the total loss during the period. The ClO–O contribution is found to be as large as 20–55% in January. However, during this period the ozone loss at this altitude is very small (0–0.3 ppmv). The contribution of HO_x cycle, which is about 10–20% in January, increases during the course of the winter to become equal or larger than that of ClO–O in late winter. The rate limiting step in all these cycles is the combination of a specific molecule with oxygen (HO₂+O for HO_x, NO_x+O for NO_x and ClO+O for ClO_x). Therefore, availability of O-atoms mainly determines the efficiency and duration of these cycles and thus, the accumulated ozone loss at 675 K.

The inter-annual variability is relatively strong for ClO–O and NO–NO₂ cycles, markedly in January. The variability of ClO–O contribution is linked to the formation of PSCs. The NO–NO₂ cycle contributes 10–20% in 2010, 2009, 2008 and 2006, but 30–45% in 2005 and 2007. The maximum ozone depletion simulated around 675 K in 2007 is in agreement with the comparatively higher contribution of NO–NO₂ during the winter. However, similar contribution of this cycle in other winters is compensated by large ozone production as discussed in Sect. 6.1.

Unlike for lower stratosphere, only a few studies are performed on the aspect of contribution of different chemical cycles to the total loss above 650 K. Moreover, the available studies on previous winters address the aims of cycles in some specific issues such as ozone loss due to additional NO_x loading during solar proton events or warming events (Groß et al., 2005a; Vogel et al., 2008). On the one hand, a box model calculation by Konopka et al. (2007) noted the efficiency of NO_x, HO_x, ClO–ClO and BrO–ClO cycles as 76, 12.5, 3.5 and 1% respectively during the warm Arctic winter 2003. Interestingly, large loss of ozone at higher altitudes with a double peak structure

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(as found in 2005) was simulated in this winter too (Grooß et al., 2005a). The simulated ozone loss was comparable to that of 2005, with a maximum of around 1.4 ppmv at 475 and 675 K. They linked the higher loss above 600 K also to the exposure of vortex air to sunlight that began early during this dynamically disturbed winter, consistent with 5 their analyses for southern winter 2002 (Grooß et al., 2005b). These results are in agreement with our analysis for warm winters, during which the contribution from NO_x is larger than that of cold winters with higher ozone above 600 K. Therefore, in PSC-free polar stratosphere at 600–850 K, the $\text{NO}-\text{NO}_2$ cycle plays a major role in ozone depletion.

10 6.3 Impact on ozone column loss

To complement our analysis, we have evaluated the ozone column loss for each winter from both the Mimosa-Chim simulations and MLS observations. For the integration, the model ozone and tracer profiles were interpolated to MLS sampling points inside the vortex ($\geq 65^\circ \text{EqL}$). The MLS profiles were then interpolated to the vertical levels of 15 the model in order to have the same column computation procedure for both data sets. Partial column ozone loss was evaluated for the range 350–550 K and total column loss for 350–850 K. Except for the warm winters 2006 and 2009, the accumulated column loss were estimated from December through March. Calculations for these warm winters were restricted to 10 February due to the absence of vortex afterwards.

20 The resulted column loss for each winter is shown in Table 2.

As noted from the table for the total column loss, the largest loss is found in 2005 and the lowest loss in 2006 in accordance with our previous discussions. In 2005, the results based on MLS observations yield a loss of about 111 DU, while those from the model simulations provide a loss of about 94 DU. In warm winters, a limited loss of 25 ~40 DU is estimated with both MLS and the model data. The total column loss estimated from Mimosa-Chim/MLS profiles compares well with that of the ground-based observations for all winters (Goutail et al., 2005, 2010). As shown by the simulations, large loss in 2005 and 2010 and relatively small loss in 2006 and 2009 are estimated

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as illustrated in Fig. 5. The reason for the large difference between simulated and measured ozone loss in 2010 is discussed in Sect. 5.2.4. In addition, while the ground-based estimations depend on six or seven stations in the vortex edge region, the MLS sampling covers quite well the vortex area. Therefore, the slight offset among the analyses could be partly due to different sampling of the vortex by the ground-based and satellite measurements. Additionally, the ground-based analysis uses slightly different vortex criterion and the column measurements do not sample vortex air masses at all altitude, while only vortex air is considered in our analysis.

The difference in ozone total and partial column loss averaged between 2005 and 2010 for the MLS observations and Mimosa-Chim simulations are, respectively, 11.3 and 12.1 DU. Such a difference, mainly due to the effect of NO_x chemistry in the middle stratosphere has to be taken into account when comparing polar ozone loss computed from total ozone observations with that derived from profile measurements.

6.3.1 Ozone loss in 2005

Since the winter 2005 was one of the coldest in the decade, a number of ozone loss estimations from simulations and observations have been published. Table 3 compiles the vortex averaged ozone column loss calculated from various data sets. As shown by the table the loss estimated by different studies generate, different results. The Mimosa-Chim analysis shows a good agreement with Singleton et al. (2007). The model/MLS comparisons with Jin et al. (2006) and Tsvetkova et al. (2007) yield a good agreement within the error limits of each data set. The larger loss simulated in Feng et al. (2007) is due to the higher vortex descent and concomitant increased chlorine loading in the lower stratosphere of their model. The ozone loss computation from in-situ measurements by von Hobe et al. (2006) shows the lowest value among these analyses. This can be due to the fact that the accumulated loss in von Hobe et al. (2006) was estimated on 7 March, which is much earlier than other studies and there was a strong vortex and sustained loss afterwards. Most works register their peak ozone loss around 23–25 March 2005. Also, the ozone loss was estimated only up to

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460 K which is much lower than other studies. Such a discrepancy in the altitude range used for the analyses is one of the reasons for the spread in the results. Therefore, as stated previously, the selection of column length for the computation of partial column ozone loss is an important factor to be considered for the comparisons, particularly in 5 cold winters with large loss throughout the lower and middle stratosphere. Moreover, most works use their own vortex criterion for the ozone column loss estimation. However, the partial column loss estimates from all studies underestimate the loss found by Rex et al. (2006), the reasons for which are unclear at present. Finally, the ozone loss analysis by Grooß and Müller (2007) underestimates the results based on observations 10 (see Table 3), except von Hobe et al. (2006). This discrepancy could be due to a different sampling of the vortex by the model grid of Grooß and Müller (2007) study, as compared to the sampling by satellite observations. As a conclusion, the differences in vortex sampling, altitude range, time period and vortex definition of the analyses have to be taken into account when comparing different ozone loss estimations in the Arctic 15 winters.

7 Conclusions

The evaluation of Arctic winter ozone loss in recent winters from model and satellite observations shows that its variability is very large, in accordance with analyses performed for previous northern winters. The cold winters 2005 and 2008 record the 20 highest loss with peak ozone loss around 475 K. Large ozone loss is also observed in the most recent winter 2010. In 2007, the maximum loss is estimated at higher potential temperature, around 650 K and minimal loss among the winters is obtained in the warm winters 2006 and 2009. At 475 K, the cumulative ozone loss ranges from about 0.7 ppmv in 2006 to 1.7 ppmv in 2005. At 675 K it ranges from 0.4 ppmv in 2009 to 25 1.3 ppmv in 2005 and 2010. In general, the ozone loss estimates from the Mimaso-Chim model and MLS observations, combined with the model passive ozone tracer, are in good agreement and the differences are mostly within the estimated accuracy of

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the observations.

Model runs with specific chemical cycles suggest that the halogen cycles ClO–ClO contributes ~40–50% and BrO–ClO contributes ~30–40% to the total loss in December–February at 475 K. These cycles depend on temperatures in the lower stratosphere, PSCs, heterogeneous reactions on PSCs and thus the Arctic meteorology. The NO–NO₂ cycle is the key mechanism that depletes about 60–75% of ozone in the middle stratosphere, which is essentially predominant in January–March period.

The ozone total column loss estimated from Mimosa-Chim calculations at the MLS footprints inside the vortex shows about 94, 40, 79, 85, 43, and 89 DU in 2005, 2006, 10 2007, 2008, 2009 and 2010, respectively, and are in good agreement with that of the MLS and other ground-based observations.

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Table 1. The vortex averaged ozone loss estimated in volume mixing ratio (ppmv) by different studies for the Arctic winter 2005. The passive method is denoted by PT and the vortex averaged/profile descent method is denoted by VAO. The ozone loss analyses based on assimilated data are indicated by §.

Study	Method	Loss/ppmv	Peak altitude	period	measurements
This study	PT	1.7	475 K	Dec/Mar	MLS
Singleton et al. (2007)	PT	1.8	450 K	Jan/mid-Mar	MLS
Jin et al. (2006)	Various	1.8–2.3	475–550 K	1–7 Jan/mid-Mar	ACE/FTS
Rex et al. (2006)	Match	1.3–2.1	450–475 K	Jan/25 Mar	Ozonesondes
Tsvetkova et al. (2007)	VAO	1.7	450 K	Jan/25 Mar	SAGE-111
Manney et al. (2006)	VAO	1.5	450 K	Jan/10 Mar	MLS
Amraoui et al. (2008)	VAO	1.5	425 K	Jan/10 Mar	MLS
Rösevall et al. (2007)	VAO	1.3	450 K	Jan/14 Mar	MLS §
Jackson and Orsolini (2008)	VAO	1.2	450 K	early-Jan/early-Mar	MLS/SBUV2 §
Rösevall et al. (2007)	VAO	0.6–0.9	450 K	Jan/14 Mar	SMR/Odin §
Simulation					
This study	PT	1.7	475 K	Dec/Mar	Mimosa-Chim
Groß and Müller (2007)	PT	1.4	475 K	Jan/mid-Mar	CLAMS
Singleton et al. (2007)	PT	2.4/2.3	450/475 K	Jan/mid-Mar	SLIMCAT

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Table 2. The total (350–850 K) and partial (350–550 K) column ozone loss (DU) estimated from MLS sampling inside the vortex and corresponding Mimosa-Chim simulations interpolated to the observed points for each winter (121 Days from December to March). The calculations for the warm winters 2006 and 2009 are performed until vortex breakdown (72 days from 1 December to 10 February).

Total column	2005	2006	2007	2008	2009	2010
Mimosa-Chim	94	40	79	85	43	89
MLS	111	37	91	101	42	100
SAOZ	102	49	95	102	48	108
Partial column	2005	2006	2007	2008	2009	2010
Mimosa-Chim	85	29	60	72	39	72
MLS	84	35	71	83	41	98

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Table 3. Vortex averaged ozone partial column loss (DU) estimated for Arctic winter 2005. The error estimation provided by the respective studies are given together with the ozone loss values.

Study	Data	Column	DU
This study	Model/MLS	350–550	85/84
Singleton et al. (2007)	Satellites	400–550	$\sim 90 \pm 15$
Tsvetkova et al. (2007)	SAGEIII	350–625	116 ± 10
Jin et al. (2006)	ACE-FTS	375–650	116
Rex et al. (2006)	Match	350–550	127 ± 21
Feng et al. (2007)	Model	380–550	~ 140
Groß and Müller (2007)	Model	380–580	69 ± 20
von Hobe et al. (2006)	in-situ	344–460	62 ± 8

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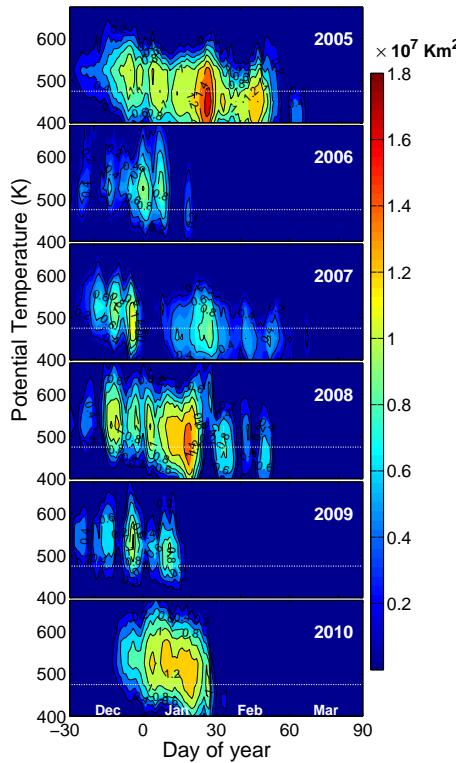


Fig. 1. The area (km^2) covered by polar stratospheric clouds – PSC – (between 400 and 675 K) inferred from the ECMWF temperature analyses for the Arctic winters 2005–2010. PSCs are assumed to form at the NAT frost point. The dotted line represents the 475 K and the topmost boundary stands for 675 K potential temperature level.

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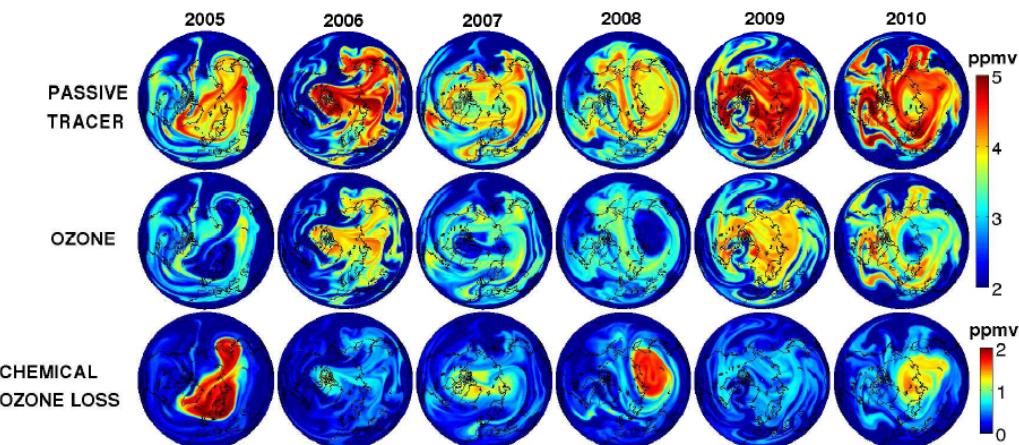


Fig. 2. Maps of passive tracer, ozone, and chemical ozone loss (passive tracer–ozone) calculated by Mimosa-Chim at 475 K on 15 March 2005–2010.

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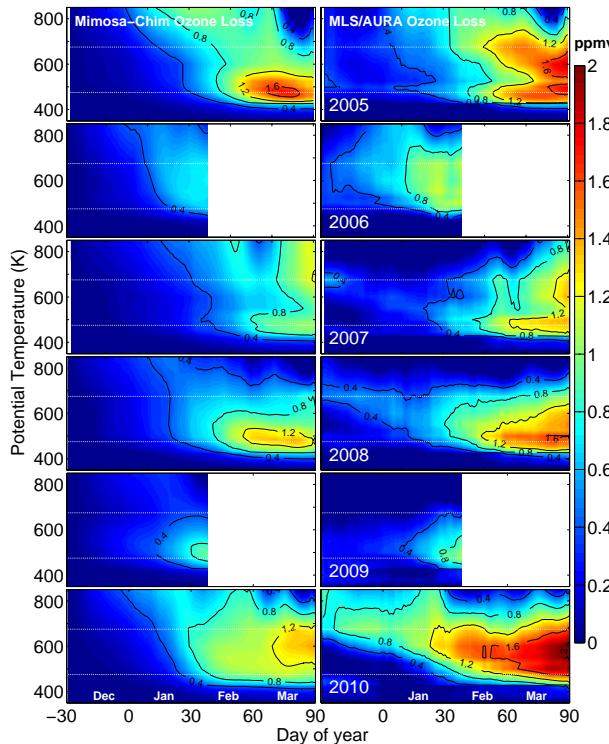


Fig. 3. Vertical distribution of the vortex averaged ozone loss (ppmv) estimated for the Arctic winters 2005–2010. The model fields are sampled at location of the MLS observations. Left: the ozone loss derived from the difference between the passive tracer and the chemically integrated ozone by Mimosa-Chim. Right: the ozone loss derived from the difference between the Mimosa-Chim passive tracer and the ozone measured by MLS. Due to early vortex dissipation caused by the major warmings, the analysis does not extend beyond 10 February in 2006 and 2009. Both data are smoothed for seven days. The white dotted lines represent the study altitudes 475 and 675 K.

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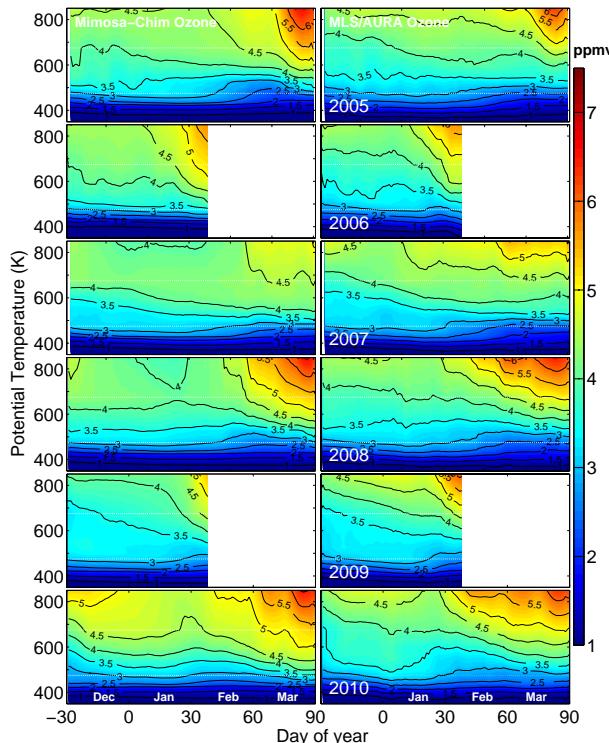


Fig. 4. Same as Fig. 3, but for ozone (ppmv).

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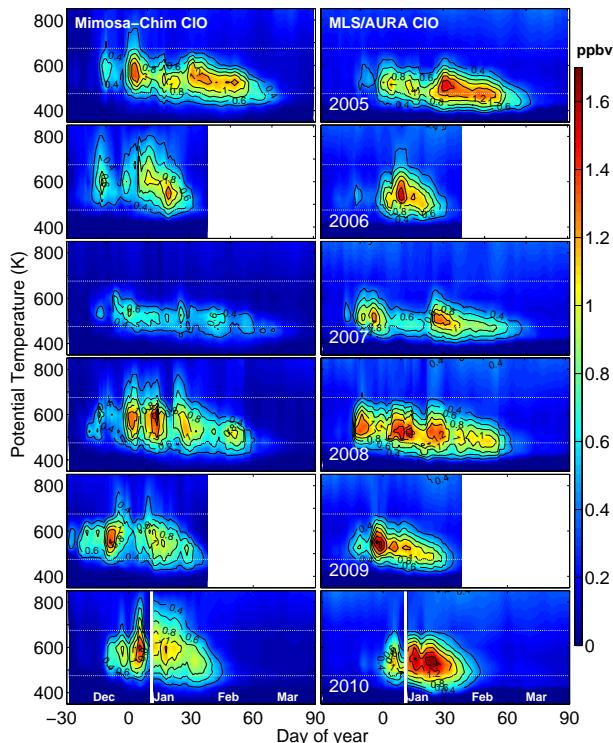


Fig. 5. Same as Fig. 4, but for CIO (ppbv). Model and MLS CIO coincident profiles are selected for solar zenith angles $<89^\circ$ and local time between 10 h and 16 h. Both data are smoothed for three days. MLS CIO values are corrected for the negative bias identified by Santee et al. (2008).

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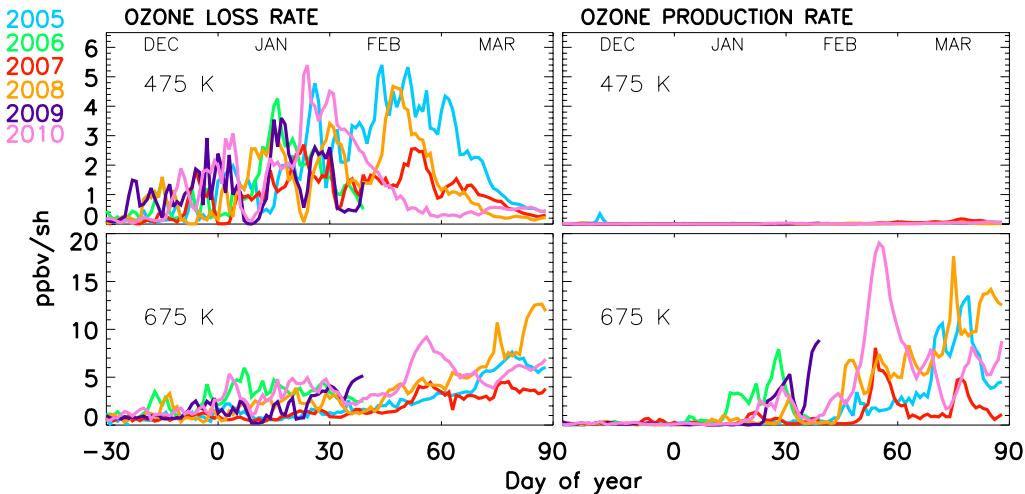


Fig. 6. Vortex averaged chemical ozone loss and production rates at 475 and 675 K, expressed in ppbv per sunlit hour (ppbv/sh), for the Arctic winters 2005–2010.

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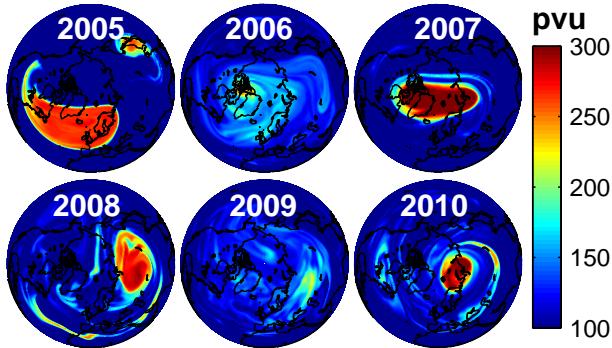


Fig. 7. Maps of potential vorticity (1 PV units (pvu)) = $10^{-6} \text{ Km}^2 \text{ kg}^{-1} \text{ s}^{-1}$ calculated from ECMWF data on 15 March 2005–2010 at 675 K.

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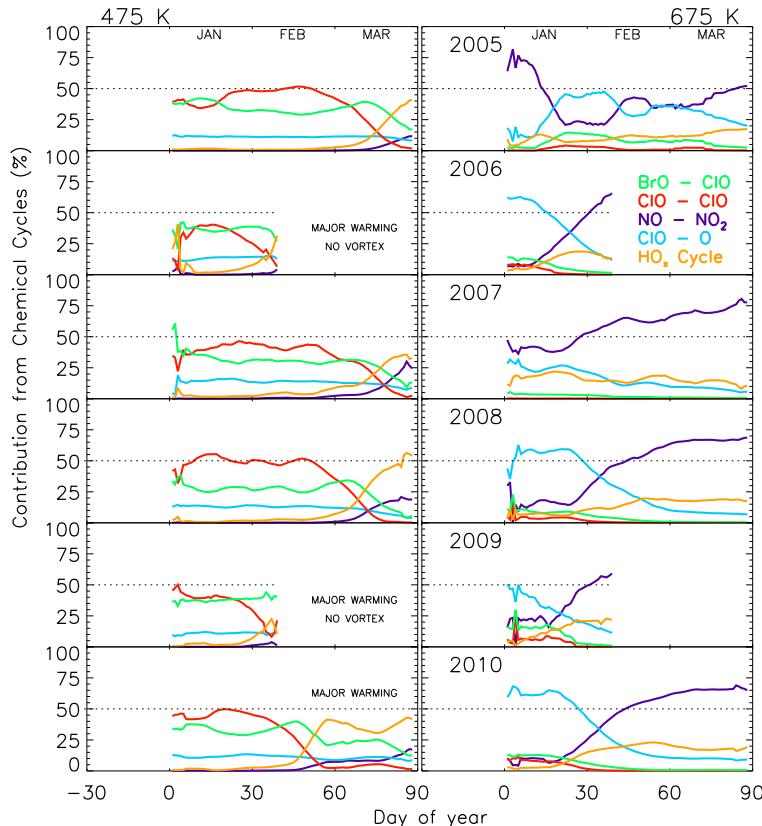


Fig. 8. Vortex averaged relative contribution of selected ozone depleting chemical cycles to the total chemical ozone loss at 475 and 675 K for the Arctic winters 2005–2010. The dotted lines represent 50% of the contribution.

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