

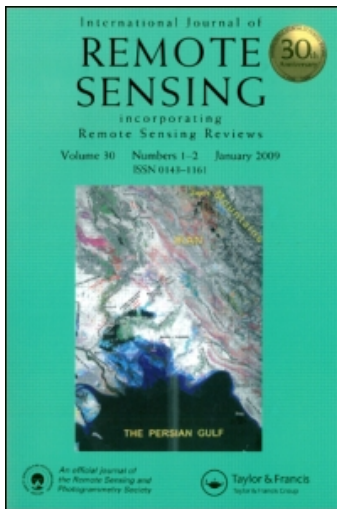
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## Ozone depletion in the Arctic winter 2007–2008

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The Arctic winter 2007–08 was characterized by cold temperatures and a strong vortex. Potentials for large areas of ice and Polar Stratospheric Clouds (PSCs) are observed during the winter. A vortex wide denitrification (removal of 60–80% of  $\text{NO}_y$ ) and intense chlorine activation (0.6 to 1.05 ppb of ClO) are found inside the vortex at 475 K. This chemical morphology triggered a high rate of ozone loss during the winter. The simulated results from MIMOSA-CHIM show a large loss of ozone at 425–550 K from January to March, about 1.5–2.3 ppm. The vortex averaged loss at 475 K is about 2.5 ppm in mid-March, which is in very good agreement with the estimated loss (2.3 ppm) from the Microwave Limb Sounder (MLS) measurements. Similar analyses from MIMOSA-CHIM for recent winters show a cumulative loss of 2.1 ppm in 2006–07 and 2.0 ppm in 2004–05 in tune with the measurements. The measured and simulated results show the highest loss in 2007–08 in comparison with the analyses for the last four winters at 475 K.

### 1. Introduction

Curiosity over the developments of the ozone layer in the Arctic stratosphere was raised when the first stamp of human activities on our environment appeared in the form of an ozone hole in the Antarctic (Farman *et al.* 1985). Even though not severe as it is seen in the south, a large loss of ozone is generally observed in unusually cold Arctic winters, like 1995–96, 1999–00, and 2004–05 (WMO 2007). The moderately warm winter of 1991–92 also witnessed a catastrophic loss of ozone due to the heterogeneous chemistry on sulphate aerosols, which were emitted from the Mt. Pinatubo volcanic explosion (WMO 2007, von der Gathen *et al.* 1995). Unprecedented variability in Arctic ozone loss has been observed in the last decade and studies reveal its impact on mid-latitudes too. (Reid *et al.* 1998, Goutail *et al.* 1999, Schulz *et al.* 2000, 2001, Rex *et al.* 2006, Singleton *et al.* 2008). The high inter-annual variability in ozone depletion hints at the necessity of constant monitoring of the stratosphere to predict future climate scenarios. This is also important to assess the impact of international treaties like the Montreal Protocol that aimed to phase-out ozone depleting substances (ODS) (WMO (2007) and references therein). In addition, continued and consistent monitoring of the ozone layer is inevitable for implementing further amendments to the treaties with a vision for a better environment and a more comfortable climate.

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Ozone loss simulated by models generally underestimates measurements (WMO 2002). Nevertheless, tremendous improvements have been made in the last few years in reducing the gap between measurements and model simulations of ozone depletion (WMO 2007) due to improvements in the kinetic data (Sander *et al.* 2003, 2006). Coarse resolution of models is another flaw that induces low bias, because of problems in isolating vortex boundaries, though the Prather scheme (Prather 1986) helps to reduce the bias to some extent. Tripathi *et al.* (2006) addressed both issues in detail and showed how the ozone simulations improve or deteriorate with respect to the data/parameters used.

This study deals with the polar processing and ozone loss of the Arctic winter 2007–08. It uses simulations from the MIMOSA-CHIM chemical transport model (CTM) and measurements from the Microwave Limb Sounder (MLS) on the National Aeronautics and Space Administration (NASA) Aura satellite (Waters *et al.* 2006). The model and MLS features are outlined in Section 2 and 3 followed by the method used for the ozone loss analyses. The meteorological situation during the Arctic winter 2007–08 is presented in Section 5. Since denitrification and chlorine activation play a pivotal role in Arctic ozone loss (Waibel *et al.* 1999, WMO 2002), these chemical processes during the winter are inspected in Section 6.1. The vertical and horizontal behaviour of the simulated ozone loss in 2007–08, together with other recent winters, are given in section 6.2. The simulated results are compared with the MLS data in Section 6.3. The final part presents a critical discussion on the ozone loss analyses and Arctic ozone trends with concluding remarks.

## 2. The MIMOSA-CHIM CTM

The MIMOSA-CHIM model has been proved to be a successful tool for estimating polar processing and ozone loss studies (Tripathi *et al.* 2006, 2007). MIMOSA-CHIM combines the MIMOSA (Modélisation Isentrope du transport Méso-échelle de l'Ozone Stratosphérique par Advection) PV advection model (Hauchecorne *et al.* 2002) with the REPROBUS chemistry module (Lefèvre *et al.* 1994). The model has a horizontal resolution of  $1^\circ \times 1^\circ$  with 25 isentropes as vertical levels between 350 and 950 K. The vertical resolution in the lower stratosphere is 5 K (between 400 and 500 K). The model runs on an azimuthal equidistant orthogonal projection centred at pole. Advection starts with potential vorticity (PV) interpolated on to the MIMOSA grids, where the PV is fed from the calculations using European Centre for Medium-Range Weather Forecasts (ECMWF) meteorological analyses. The PV is advected using the ECMWF winds on each MIMOSA grid and isentrope. As the run progresses with time, the grid is stretched and deformed by horizontal gradients in the wind fields. After the time step of 6 h, the PV field is advected again from the original grid to make sure that the distance between two adjacent grids is constant. The expected change in the distance between the grids during the advection is 10–15% at 400–675 K. Since the regridding induces numerical diffusion, an interpolation scheme based on the preservation of the second order momentum of PV perturbation has been implemented to reduce its effects on the simulated results (Hauchecorne *et al.* 2002). The diabatic transport through vertical levels is computed from the heating rate calculations by the MIDRAD radiation routine (Shine 1987), for which the climatological  $\text{H}_2\text{O}$ ,  $\text{CO}_2$  and interactive  $\text{O}_3$  are taken from the model itself.

The REPROBUS chemical module uses 55 chemical species and 160 gas-phase, heterogeneous and photolytic reactions (Lefèvre *et al.* 1994). Chemical kinetics data

and absorption cross-sections are in general taken from Sander *et al.* (2003, 2006). For  $\text{Cl}_2\text{O}_2$  the model uses the absorption cross sections by Burkholder *et al.* (1990), which are extrapolated to longer wavelengths on a log linear grid (Stimpfle *et al.* 2004). REPROBUS includes a comprehensive treatment of liquid binary ( $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ ), liquid ternary ( $\text{HNO}_3\text{-H}_2\text{SO}_4\text{-H}_2\text{O}$ ) and solid polar stratospheric clouds, using the analytic expressions given by Carslaw *et al.* (2002). Liquid supercooled aerosols, NAT and ice particles are assumed to be in equilibrium (Lefèvre *et al.* 1998).

The chemical fields are initialized from a long term simulation of the REPROBUS CTM interpolated on to the MIMOSA-CHIM grid.  $\text{Cl}_y$  is set to 3.6 ppb above 30 km as recommended by WMO (2007). The  $\text{H}_2\text{O}$  initialization is done from MLS climatology.  $\text{Br}_y$  in the model is based on the correlation with CFC-11, which considers the bromine output from  $\text{CH}_3\text{Br}$ ,  $\text{CH}_2\text{Br}_2$ ,  $\text{CH}_2\text{BrCl}$  and halons (Wamsley *et al.* 1998).

### 3. MLS aboard Aura

In order to diagnose the simulated results, measurements of ozone ( $\text{O}_3$ ) and chlorine monoxide (ClO) from MLS (version 2.2) are used. The standard ozone retrieval makes use of a line centred at 240 GHz and the ClO retrieval uses a cluster of lines centred at 640 GHz (Froidevaux *et al.* 2006, Santee *et al.* 2008). The retrieved ozone profiles for v2.2 have a vertical range of 215–0.02 hPa and a vertical resolution of  $\sim 3$  km, while the horizontal resolution is  $\sim 200$  km. The precision is about 2–4% and the accuracy is  $\sim 8\text{--}10\%$  for the altitude considered in this study. In the lower stratosphere, the MLS ozone shows some positive bias too, in particular at high and mid-latitudes (Froidevaux *et al.* 2006). The vertical range of ClO v2.2 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 precision of the product is  $\pm 0.1$  ppb and the accuracy is 10–20% with a negative bias of 0.24 ppb at 68 hPa (Santee *et al.* 2008).

### 4. Ozone loss analyses with the transport method

We performed MIMOSA-CHIM simulations for the 2007–08, 2006–07, 2005–06 and 2004–05 Arctic winters. The chemical depletion of ozone inside the vortex is quantified by the method used by Goutail *et al.* (1999). Using a passive tracer ( $\text{Tracer}_{\text{model}}$ ) initialized identically to ozone at the beginning of each simulation, the ozone loss computed by the model is defined as  $\text{Loss}_{\text{model}} = \text{Tracer}_{\text{model}} - \text{Ozone}_{\text{model}}$ .  $\text{Ozone}_{\text{model}}$  is replaced with  $\text{Ozone}_{\text{MLS}}$  to calculate the loss from the MLS measurements. In order to make the comparisons, the model calculations are interpolated on the MLS sampling points and are sorted with respect to the PV values to differentiate the measurements inside the vortex. A constant PV value of 35 pvu (1 PV unit =  $10^{-6} \text{ m}^2 \text{ s}^{-1} \text{ K kg}^{-1}$ ) is taken as the vortex edge criterion for these analyses after a sensitivity test. The higher PV criteria make the data sparse and the lower PV criteria add noise, hence the aforesaid value is considered as the optimum. The daily average of the MLS and model ozone are subtracted from the corresponding daily mean of the tracer to find the ozone loss as given by the formula.

### 5. Meteorology of the Arctic winter 2007–08

Figure 1 shows the daily minimum temperature north of  $70^\circ\text{N}$  analysed by ECMWF between 2005 and 2008. In 2007–08, temperatures are below  $T_{\text{NAT}}$  from December to

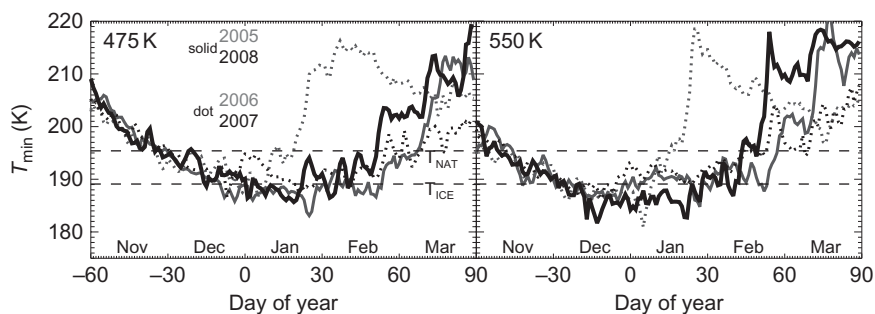


Figure 1. The daily minimum temperature observed at 70–90°N estimated from the ECMWF analyses at 475 and 550 K.

mid-February at 475 K. Temperatures are below the  $T_{ICE}$  threshold for a few days in mid-December, early and mid-January and in early February, which are the lowest among the recent winters too. A minor warming is observed in late January and the final warming started around mid-February at 475 K. At 550 K, temperatures are below 195 K from mid-November to mid-February and below 189 K from December to late January. The winter experiences the coldest December and January in recent years at 550 K.

## 6. Results

### 6.1 Denitrification and chlorine activation in the winter 2007–08

Figure 2 shows the  $NO_y$  distribution computed by MIMOSA-CHIM at 475 K for the Arctic winters 2004–05 to 2007–08. The calculations show a massive change in the distribution of  $NO_y$  from December to January, where the temperature shows the lowest values. The mixing ratios from January to March are about 60–80% lower than the December values. Since  $NO_y$  is a good indicator of denitrification (more than 50% or complete removal of  $NO_y$  is termed as denitrification), it can be said that the process is severe in the same period. The scale of denitrification is similar in all winters too, except 2005–06. However, the calculations show comparatively higher values in October–December of 2006 and 2007 and hence, the removal of  $NO_y$  (denitrification) is more explicit in these winters.

Figure 3 depicts the daily mean ClO mixing ratios averaged inside the vortex (where  $PV \geq 35$  pvu) at low zenith angles from MIMOSA-CHIM and MLS for 2005–2008. There is hardly any activation up to January 2008. The ClO mixing ratios show values less than 0.4 ppb during the period, which is a general feature in all winters too. High values of ClO ( $\sim 0.4$ – $1.05$  ppb) in the late winter indicate a strong chlorine activation inside the vortex and is in accord with the temperature in 2007–08. As compared to 2005 and 2007, the ClO values in February 2008 are slightly higher and suggest a stronger chlorine activation. The activation is not that explicit in 2006 except in January because of warmer temperatures and early final warming. It should be noted that the simulations generally overestimate the measurements, especially in February and March.

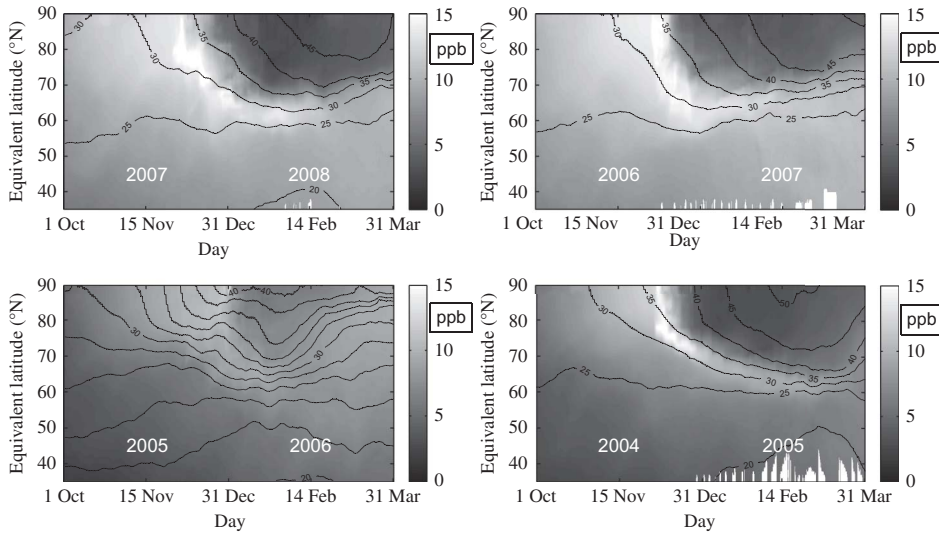


Figure 2.  $\text{NO}_y$  mixing ratio at 475 K for the Arctic winter 2007–08 compared to other recent Arctic winters. The dark contours represent the PV values.

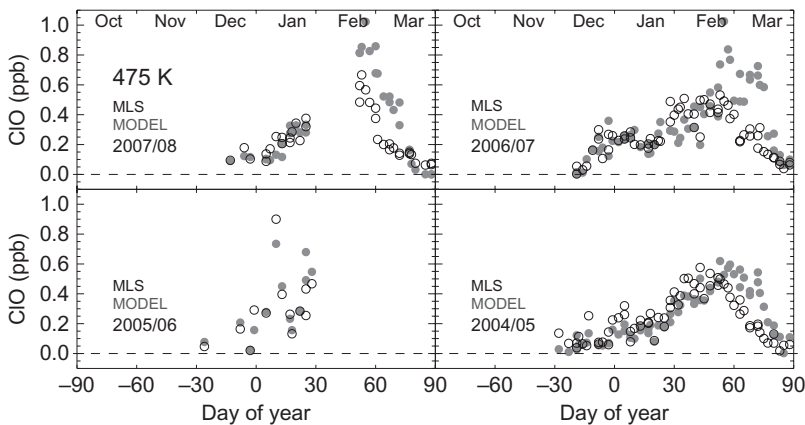


Figure 3. The observed and simulated vortex averaged ClO at 475 K for the Arctic winter 2007–08 in comparison with the same in 2006–07, 2005–06 and 2004–05 winters.

## 6.2 Simulated ozone loss and comparison with previous years

Figure 4 represents the vertical structure of ozone loss in the Arctic winter 2007–08. The loss in November and December are confined to the altitudes above 450 K and are less than 0.5 ppm. In January, due to very cold temperatures and high potentials for PSCs, heterogeneous reactions are initiated on the surfaces of the clouds. They induce a strong ozone loss in January through March. The ozone depletion rapidly increases to 2.0 ppm by mid-February and reaches a maximum of 2.3 ppm in early March around 475 K.

Figure 5 shows the ozone loss at 475 K (as the peak loss is observed around this altitude) in 2007–08 as a function of equivalent latitude and time. The loss starts in

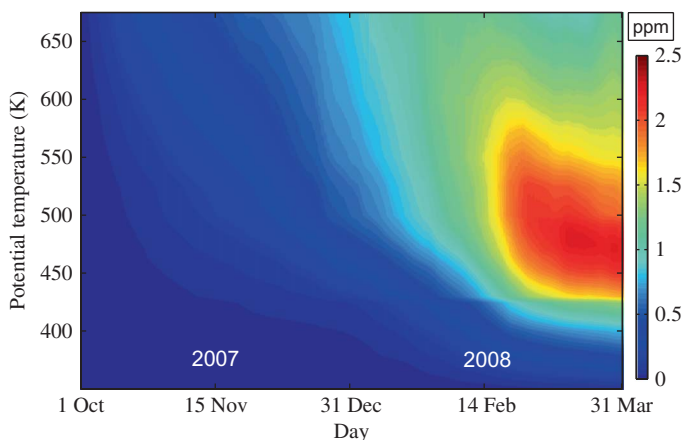


Figure 4. The vertical distribution of ozone loss averaged over equivalent latitudes  $\geq 65^\circ$  for the Arctic winter 2007–08.

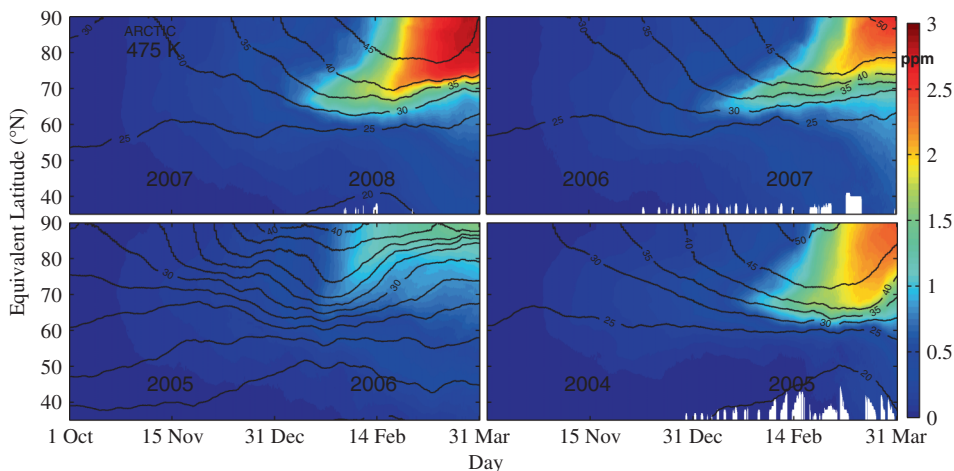


Figure 5. The simulated ozone loss at 475 K for the recent Arctic winters. The black contours on the plots represent the PV values.

November and is  $\leq 0.5$  ppm during the November–December period as registered by figure 4. The depletion peaks as the sun returns over the Arctic and increases to 2.7 ppm by the end of the winter inside the vortex. The loss is well-timed with the high chlorine activation observed during the same period.

Figure 5 also illustrates the simulated ozone loss at 475 K for the recent Arctic winters 2004–05, 2005–06 and 2006–07. It shows that, as discussed for 2007–08, the loss from October to December are around 0.5 ppm. Large loss of ozone is simulated in January and February; about 0.5 to 1.2 ppm. The loss is maximum in mid-February to March period and is varied from year to year. The reduction in ozone during this period is 1.5–2.7 in 2007–08, 1.5–2.15 in 2006–07, 1–1.5 in 2005–06 and 1.5–2.1 ppm in 2004–05. The cumulative ozone loss is highest in

2007–08 as compared to other recent winters. The higher loss in 2007–08 can be explained by high chlorine activation as compared to the other winters due to coldest temperatures in mid-December, mid-January and early February in 2007–08. In addition, the potential PSC and ice area in 2007–08 is similar or higher than the estimated area in 2004–05 at 550 K (not shown). The ice clouds at high altitudes can sediment particles and thus, induce higher denitrification and chlorine activation to cause high ozone loss at lower levels like 475 K.

### 6.3 Comparison with ozone loss deduced from MLS measurements

The ozone loss quantified from the MLS measurements and MIMOSA-CHIM simulations inside the vortex at 475 K are shown in figure 6. The observed loss until January 2008 is below 0.5 ppm, which is consistent with the results shown in figure 5. The loss increased during January–March period and gained the maximum of 2.3 ppm at the end of the winter. The simulated results slightly overestimate the measurements in tune with the relatively higher chlorine activation in the model as shown in figure 3. The observed loss from December to January is within 0.65 ppm in all winters and the maximum loss in this period for each winter is 0.4, 0.4, 0.5 and 0.65 ppm in 2007, 2006, 2005 and 2004 respectively. The observed cumulative maximum is 2.2 ppm in 2004–05 and is 1.95 ppm in 2006/07. Since the vortex was dissipated early in 2005–06, only a few samples were available for the analyses and the loss is about 1.0 ppm by January 2006.

Figure 6 also demonstrates the comparison between MLS and MIMOSA-CHIM ozone loss at 475 K for the recent winters. The simulated loss is within 0.5 ppm until January and the maximum cumulative loss is 2.5, 2.1 and 2.0 ppm in 2008, 2007 and 2005 respectively, which are in good agreement with the MLS results. The measurements and calculations show the highest loss in 2007–08 and the lowest loss in 2005–06. The model slightly underestimates the measurements in 2005–06, which could be due to sporadic vortex sampling. The other difference between the measurements and simulations is the higher loss in 2006–07 as compared to 2004–05 in MIMOSA-CHIM. This can be due to the higher chlorine loading in the model, as

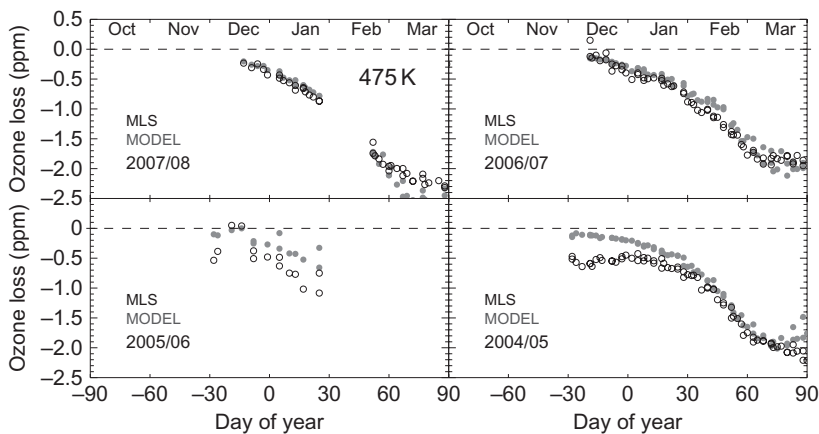


Figure 6. The vortex averaged ozone loss at 475 K from MIMOSA-CHIM and MLS in recent Arctic winters.

indicated by the high ClO mixing ratios in 2006–07, for which the measured values are overestimated by 0.5 ppb in late February and March.

## 7. Discussion

Substantial ozone loss has been measured in 2007–08, 2006–07 and 2004–05 with similar scale. The comparatively higher loss in 2007–08 is justified by the coldest temperatures in mid-December, early and mid-January and in mid-February. The situation brought about stronger chlorine activation and thus, higher ozone depletion.

The analyses with different methods produced different results for the ozone loss estimations in the Arctic winter 2004–05. Singleton *et al.* (2007) use the same method that applied in this study to quantify ozone loss from MLS. They estimate a loss of 1.8 ppm by mid-March 2005, which is similar to the loss obtained from MIMOSA-CHIM and MLS (~1.9 ppm), as concluded in this study. Manney *et al.* (2006) used vortex-averaged descent method to find the ozone loss from MLS for the same winter. They found the vortex averaged ozone loss as 1.2–1.5 ppm from January to 10 March at 450–550 K. This is slightly lower (about 0.4 ppm) than the estimated loss from the work presented here. The method used in Manney *et al.* (2006) has some difficulties in isolating the vortex air from mid-latitude air and hence, the ozone inside the vortex gets mixed, which obviously leads to underestimation of ozone loss (Müller *et al.* 2005). This could be the reason for the lower values of ozone loss in Rösevall *et al.* (2007) and in Groöß and Müller (2007) as well. On the other hand, the Match technique estimates a loss of 1.3–2.1 ppm at 450–550 K from 5 January to 25 March (Rex *et al.* 2006) and Jin *et al.* (2006) find a loss of 2.1 ppm at 450–550 K from 1–7 January and 8–15 March, close to the findings of this work.

Rösevall *et al.* (2008) estimate a depletion of 0.5–1.0 ppm at 475 K in the Arctic winter 2006–07, which is slightly lower than the loss found in MIMOSA-CHIM. The difference can be due to the uncertainties in the loss estimation by the particular method (Müller *et al.* 2005) as discussed above.

## 8. Conclusions

### 8.1 Ozone loss in 2005–2008

A sizable amount of ozone is depleted inside the vortex in the Arctic winter 2007/08 and appeared to be centred at 450–550 K. The loss is found to have started in November and is within 0.5 ppm during November and December. The vortex averaged cumulative ozone loss at 475 K from MIMOSA-CHIM is about 2.5 in 2007–08, 2.1 in 2006–07 and 2.0 ppm in 2004–05. The ozone depletion measured by MLS at the same altitude level also shows similar values in all four winters: 2.3 in 2007–08, 1.95 in 2006–07 and 2.2 ppm in 2004–05. Both the simulated and measured values show the highest loss in 2007–08 and is well timed with the high chlorine activation and pronounced denitrification in the winter. The simulations show slight overestimation of ozone loss and ClO especially during the late winter at 475 K, which hints that the chlorine defined in the model is slightly large. The assessment points out that the model is now able to catch the measured ozone loss with high accuracy by using improved chemistry input on high resolution model domain.

## 8.2 Trends in the Arctic ozone loss

On the twentieth anniversary of the Montreal Protocol, it would be appropriate to discuss about the trends of ozone loss in the Arctic with respect to the available data presented in this study. Though a four year data set is inadequate to make an analysis on trends, the same can be used as an extension to the analyses performed in the past by various data sources. So the WMO (2007) report is used as a reference in this context.

The inter-annual variations of ozone loss are very large in the Arctic and are well correlated with the year-to-year fluctuations in polar stratospheric temperatures. For instance, though the 2004–05 winter was one of the coldest and witnessed significant amounts of ozone loss, the succeeding year (2005–06) was one of the warmest and the ozone loss was relatively small. Further, the 2006–07 and 2007–08 Arctic winters were again characterized by very low temperatures inside the vortex and hence, the ozone loss was substantial. In addition, the ozone loss in 2007–08 was also the largest among the recent Arctic winters at 475 K. It has already been proved that there is a near-linear relation between ozone loss and volume of PSCs (Rex *et al.* 2006, WMO 2007), which are directly connected to polar temperatures as well.

Therefore, the observed and simulated ozone loss during the recent winters are also in line with analyses done in the past (i.e. years with large and small ozone loss episodes). A specific change is hardly visible and the large inter-annual variability masks any tangible trend in the data too. This is consistent with the results obtained from other analyses (WMO 2007) and is in accord with our present understanding of the trend of Arctic ozone loss (WMO 2007).

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