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## Model simulations of the northern extravortex ozone column: Influence of past changes in chemical composition

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[1] A high-resolution three-dimensional chemistry transport model is used to evaluate the contributions of different mechanisms (in situ chemistry, import of chemically activated or ozone-depleted polar air) to the extravortex chemical ozone loss during the winter-spring period and to show how their efficiencies might have changed in the last 2 decades. Two extreme, but somewhat typical, present-day winters are considered: the 1999–2000 winter (cold with a stable and persistent vortex) and the 2001–2002 winter (warm with an weak and distorted vortex). An ozone budget analysis is performed for the partial ozone column between 350 and 600 K using a combination of geographical and chemical ozone tracers. The results suggest that the interannual variability in the seasonal ozone loss at midlatitudes is mainly driven by the import of ozone-depleted polar air. The contribution from the import of chemically activated polar air is not found to be significant. The magnitudes of the polar contributions (entirely driven by the halogen cycles) differ by about a factor 4 between the simulations. The polar contribution is found to be responsible for 60% of the total extravortex ozone loss for the exceptional 1999–2000 Arctic winter. In contrast, the magnitude of the in situ destruction (dominated by HO<sub>x</sub> and NO<sub>x</sub> cycles) varies little from one winter to the other one. The evolution of the atmospheric chemical composition since 1980 may account for an additional midlatitude ozone loss of 3 to 10 Dobson units depending on the winter considered (representing up to about 40% of the observed midlatitude ozone trends). The polar contribution is found to increase by a factor 2 to 3 in the last 2 decades, whereas the in situ destruction increases by about 20% only. **INDEX TERMS:** 0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334); 1610 Global Change: Atmosphere (0315, 0325); 3334 Meteorology and Atmospheric Dynamics: Middle atmosphere dynamics (0341, 0342); **KEYWORDS:** extravortex ozone loss, transport of ozone-depleted polar air, filament

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### 1. Introduction

[2] It is now well established that the total ozone column at northern middle latitudes has declined substantially over the last decades [*World Meteorological Organization (WMO), 2002*]. Current chemistry transport models forced by past changes in halogen loading tend to predict ozone trends in the midlatitudes lower stratosphere which are smaller than those derived from observations [*Solomon et al., 1996*]. Some studies suggest that interannual variability in stratospheric circulation could be an important factor in midlatitude ozone trends depending on the timescale considered [*Weiss et al., 2001; Hadjinicolaou et al., 2002*]. Midlatitude ozone is also affected by polar processes

through the following sequence. During the winter and early spring, chlorine is first activated on the surface of polar stratospheric clouds (PSCs) in the polar lower stratosphere resulting in chemical ozone destruction in the presence of sunlight [*WMO, 1995*]. Air parcels are peeled off the vortex edge and mixed with midlatitude air throughout the winter and spring seasons [*Plumb et al., 1994*]. This influx of polar air leads to ozone reductions at middle latitudes when the polar air is chemically activated (i.e., highly enhanced levels of active chlorine) triggering in situ ozone chemical destruction [*WMO, 1998*]. The other possibility is when the polar air which is being mixed is depleted in ozone with respect to midlatitude air. For example, *Knudsen and Grooß [2000]* used high-resolution reverse domain-filling trajectory calculations to show that the breakup of the Arctic polar vortex and the associated dilution of ozone-depleted air at middle latitudes in the early

spring of 1995 and 1997 may represent up to 40% of the TOMS total ozone trend observed since 1979. This derivation of the contribution of polar ozone losses to midlatitude ozone trends appears to be rather crude. First, a more accurate comparison with changes observed since 1979 would take into account the polar contribution to midlatitude ozone loss in 1979. Second, considering only the final breakup of the Arctic vortex neglect the transfer of chemically activated and ozone-depleted polar air during the winter and the resulting impact upon midlatitude ozone.

[3] In this paper, a high-resolution chemistry transport model is used to study the impact of the past evolution in the stratospheric chemical composition (halogen and nitrogen loading) on the winter-spring lower stratospheric ozone budget at the northern midlatitudes. Particular attention is paid to quantifying accurately the contributions of in situ chemistry and Arctic polar vortex processes to seasonal extravortex ozone loss and how they might have changed in the last 2 decades. The way that middle latitude ozone is affected by polar processes strongly depends on the meteorological conditions prevailing during the winter considered [WMO, 1998]. In order to consider a range of possible meteorological conditions, the CTM simulations are performed for two extremes, but somewhat typical, present-day winters. The 1999–2000 winter was cold with a stable and persistent polar vortex, whereas the 2001–2002 winter was warm with an unstable and distorted vortex.

[4] In the first section, a brief description of the high-resolution model and of the methodology is provided. The origins of the model-calculated extravortex ozone loss in the two winters are diagnosed and contrasted in the second section. The role of different chemical ozone loss mechanisms such as the import of chemically activated polar air (i.e., air with high levels of ozone-destroying halogen radicals) is also assessed. The core of the paper is devoted to evaluating changes in the seasonal midlatitude ozone loss resulting from the past changes in stratospheric chemical composition. The changes in the efficiency of individual ozone-destroying chemical are also evaluated for the different ozone loss mechanisms. The focus is on the possible interannual variability and the sensitivity of the different ozone loss mechanisms (in situ versus polar vortex processes) to halogen and nitrogen loading. The model-calculated extravortex ozone changes are compared to midlatitude ozone trends derived from ground-based and satellite data. The final section summarizes the results and discuss the implications for the future evolution of stratospheric ozone.

## 2. Description of the Model and Numerical Experiments

[5] The high-resolution chemistry transport model MIMOSA-CHIM used here is already described in detail by Marchand *et al.* [2003]. A brief description is given here. The off-line model couples the advection scheme of MIMOSA [Hauchecorne *et al.*, 2002] and the chemical scheme of the REPROBUS model [Lefèvre *et al.*, 1994]. The model is forced by daily meteorological  $2.5^\circ \times 2.5^\circ$  analyses (European Centre for Medium-Range Weather Forecasts (ECMWF)) for the horizontal winds and temperatures. The vertical coordinate is the potential temperature. Sixteen isentropic levels are considered covering the 350–

950K altitude range with a resolution of 25K between 350 and 675K. The model horizontal resolution is set high ( $1^\circ$  latitude  $\times$   $1^\circ$  longitude) in an attempt to simulate as accurately as possible the exchanges between the polar vortex and midlatitudes. The model domain is centered onto the North Pole and covers the Northern Hemisphere down to  $30^\circ\text{N}$ . In order to take into account fluxes from the region below  $30^\circ\text{N}$  latitude, PV fields of ECMWF analyses and REPROBUS chemical fields are used to force this boundary of the model. Potential vorticity (PV) is advected within the model. Its diabatic evolution is taken into account by relaxing the model PV toward PV derived from ECMWF fields with a relaxation time of 10 days. The advection scheme of the model is semi-Lagrangian. The original orthogonal grid is stretched and deformed by horizontal gradients in the wind field. An interpolation onto the original grid is performed every 6 hours. The resulting numerical diffusion is reduced by the use of an interpolation scheme based on the preservation of the second order moments of the tracer distribution [Hauchecorne *et al.*, 2002]. The diabatic vertical transport across isentropic surfaces is derived from heating rates calculated by a radiative scheme [Shine, 1987; Chipperfield, 1999]. Climatological ozone and water vapor fields are used in the radiative calculations. The chemical scheme contains a detailed treatment of the stratospheric chemistry of the  $\text{O}_x$ ,  $\text{NO}_y$ ,  $\text{Cl}_y$ ,  $\text{Br}_y$ , and  $\text{HO}_x$  families [Lefèvre *et al.*, 1994]. The model also includes long-lived tracers such as  $\text{N}_2\text{O}$ ,  $\text{CH}_4$ , and  $\text{H}_2\text{O}$ . There is also a representation of the heterogeneous chemistry associated with liquid supercooled sulphuric acid aerosols, NAT and ice particles which are assumed to be in equilibrium with the gas phase [Lefèvre *et al.*, 1998]. The denitrification originating from the sedimentation of large NAT particles is also taken into account. The model is initialized by interpolating linearly PV fields from ECMWF analyses and REPROBUS chemical fields onto the finer MIMOSA-CHIM horizontal grid. In order to quantify the ozone changes of chemical origin, an ozone-like passive tracer is also included in the model. This tracer is initialized in the same way as active ozone at the beginning of the integration and then advected passively. Model performances have been evaluated by Marchand *et al.* [2003]. Comparisons between numerical simulations and lidar/sonde measurements have shown that the model is able to reproduce the broad features of the evolution of ozone on the 435 and 475K levels at middle and high latitudes during 1999–2000 winter.

[6] Different domains are distinguished using a PV-based analysis and an algorithm of filament recognition. Dynamical barriers are identified from the model PV fields. The model PV is first expressed as a function of equivalent latitude. Then the center of the polar vortex edge is defined as the PV contour corresponding to the maximum of the first-order derivative. The extrema of the second-order derivative provide the inner and outer limits of the vortex edge [Nash *et al.*, 1996; Bergeret *et al.*, 1998]. This method has already been used successfully for the detection of vortex intrusions and filaments over the OHP (Observatoire de Haute Provence) station [Godin *et al.*, 2002]. By opposition to extravortex air, air parcels with a PV greater than the PV threshold value of the vortex outer edge are considered to be polar (i.e., inside the vortex). To determine

whether an air parcel is located within a filament, the air parcel has to be polar and the variation of PV between the air parcel and the center of the vortex has to meet the following criteria. Essentially, the vortex outer edge is searched along the line connecting each polar air parcel to the center of the vortex (defined as the location of maximum PV). If the line crosses the vortex outer edge, the polar air parcel is considered to be within a filament. This algorithm of filament recognition largely overestimates the amount of filaments. Indeed, a strongly distorted vortex or a large midlatitude intrusion lead to parts of the vortex being counted as filaments although, unlike filaments, they do not peel off from the polar vortex.

[7] The model also includes a range of air tracers and chemical ozone tracers which monitor the transport and exchanges between the three domains (extravortex, polar, and filaments) and the amount of chemical ozone destruction/production within each domain [Marchand *et al.*, 2003]. This combination of geographical tracers (monitoring ozone loss originating from specific domains) and the ozone chemical tracers (monitoring ozone loss from the individual ozone-destroying processes) allows to trace back the origin of the air within each domain and the amount of ozone destruction it has experienced. This approach is similar to the one adopted by Millard *et al.* [2002]. However, there are two important differences. In the work of Millard *et al.* [2002], the geographical tracers monitor the total ozone loss within specified equivalent latitude bands, whereas our geographical tracers consider only three regions, the polar vortex, filaments and the extravortex. The polar vortex and the extravortex domain, also called the surf zone, are characterized by relatively rapid mixing and are separated by a region of strong potential vorticity gradients acting as a dynamical barrier to mixing. Characterizing the ozone loss in the extravortex region is a good way of identifying mechanisms responsible for ozone loss at middle latitudes. Our chemical tracers are also different from those of Millard *et al.* [2002]. In the work of Millard *et al.* [2002], the ozone chemical tracers describe the cumulative ozone loss from the different ozone-destroying cycles wherever the ozone loss occurs whereas, in the present study, our ozone chemical tracers are also geographical tracers (i.e., domain-specific). This allows to decompose the midlatitude ozone loss into contributions from different ozone-destroying cycles operating within the different domains. Our approach requires many more tracers than given by Millard *et al.* [2002] but allows us to characterize more accurately the polar vortex contribution to midlatitude ozone.

[8] For the 1999–2000 winter, the numerical simulation starts on 13 December 1999 and runs until the end of April. For the 2001–2002 winter, the numerical simulation starts on the 10 December 2001 and runs until the end of April. Each simulation is initialized with the corresponding REPROBUS chemical simulation with the chlorine, bromine, nitrogen oxides and methane loadings representative of the year of the simulation.

### 3. Results

#### 3.1. Present-Day Simulations

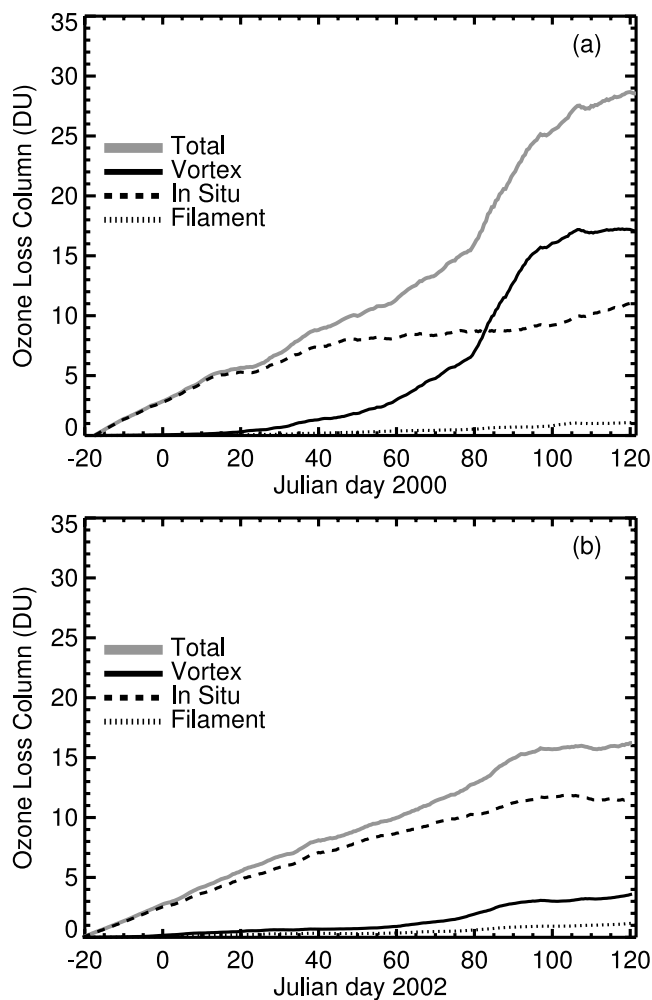
[9] The meteorological conditions of the two winters are first contrasted in this section. The 1999–2000 Arctic

stratospheric vortex was unusually cold, especially early in the winter, with a vast area below the temperature threshold for PSC formation in December and January. The lower stratospheric temperatures averaged over the November–January period were lower than those observed during any previous Arctic winters. The evolution of the polar vortex was characterized by a discontinuous vortex, large in the upper stratosphere and small in the lower stratosphere, in late December and a continuous, and relatively upright vortex by mid-January [Manney and Sabutis, 2000]. Rex *et al.* [2002] used the Match technique based on coordinated ozonesonde measurements to infer chemical ozone loss rates inside the Arctic polar vortex. They diagnosed a first period of ozone loss inside the vortex from mid-January to early February between 460 and 570K. A second period of rapid loss was observed below 520K from late February to late March. According to this technique, the cumulative chemical ozone loss reached 70% in March in a 1 km thick layer centered at around 450K [Rex *et al.*, 2002]. The performances of MIMOSA-CHIM for the 1999–2000 winter have been evaluated by Marchand *et al.* [2003]. The model calculates chemical ozone losses within the vortex of the order of 50% at 475K during this period, which is at the lower end of the ozone losses derived from observations [WMO, 2002].

[10] During 2001–2002 winter, the Arctic vortex formed in October–November with temperatures dropping below the long-term mean in November and December. Polar stratospheric clouds were observed on most days during mid-December. A strong stratospheric warming took place in the second half of December which significantly weakened the vortex. PSC conditions were briefly reestablished in early January, but a second strong warming then occurred. Despite a succession of minor warming, a weak warm vortex was present until mid-May when the summertime circulation was reestablished. The disturbed nature of the vortex resulted in many more intrusions of polar air into the midlatitudes compared to the 1999–2000 winter. However, the lack of sunlight during the initial period of PSC activation strongly limited the amount of chemical loss with reductions of about 10% in the total ozone column. Averaged over the January–March period, ozone levels were typical of those observed during other warm winters.

[11] The strong contrast between the meteorologies of the two winters provide an opportunity to gain some insight into the variability of the polar contribution to midlatitudes ozone loss. Midlatitude ozone is affected not only by background in situ chemistry but also by the amount of chemically processed polar air reaching this region during the winter and the magnitude of polar ozone depletion being mixed during the breakup of the vortex. The different components of the ozone losses outside the vortex predicted by the model for both winters are shown in Figure 1. The cumulative contributions of three domains (vortex, filaments, and extravortex) to the 350–600 K ozone column loss in extravortex air between 35°N and 90°N are plotted as function of time. Note that the term ozone loss in the text refers actually to the net chemical ozone loss, which is defined as destruction minus production. Figure 1 highlights where the chemical ozone loss seen in extravortex air originates from. The extravortex ozone loss increases throughout the winter reaching a maximum of 29 Dobson





**Figure 1.** Time evolution of the cumulative extravortex ozone loss (DU) averaged over the  $35^{\circ}\text{N}$ – $90^{\circ}\text{N}$  latitude band for the (a) 1999–2000 and (b) 2001–2002 winter. The different contributions to the total loss are also shown: total (shaded line), polar vortex (solid line), in situ (dashed line) and filaments (dotted line).

units (DU) at the end of April 2000 and a maximum of 16 DU at the end of April 2002. The magnitudes of the in situ destruction are very similar in both simulations, reaching about 11 DU by the end of April. In the 1999–2000 meteorology simulation, the polar contribution (which originates from polar vortex air which has experienced ozone destruction and been subsequently transported outside the polar vortex) becomes significant at the end of January. This contribution increases sharply at the end of March, outweighing the in situ contribution and leveling off at 17 DU after the final vortex dilution. By the end of April, the polar contribution represents 60% of the total extravortex ozone loss. In contrast, the polar contribution in the 2001–2002 meteorology simulation does not exceed 4 DU, which represents only 20% of the total extravortex ozone loss by the end of April. The results of the 1999–2000 meteorology simulation can be compared to a numerical photochemical study of that winter by Millard *et al.* [2002]. They used similar air tracers and ozone loss tracers but did not map

them to the three domains considered here (extravortex, polar vortex, and filaments) but rather to equivalent latitude bands such as the equivalent latitude  $30^{\circ}$ – $60^{\circ}\text{N}$  and  $70^{\circ}$ – $90^{\circ}\text{N}$  bands. The vortex edge was mostly positioned between  $60^{\circ}\text{N}$  and  $70^{\circ}\text{N}$  equivalent latitude during the 1999–2000 winter. They found that the total ozone loss within the  $30^{\circ}$ – $60^{\circ}\text{N}$  equivalent latitude band reached 25 DU by the end of April, which appears to agree with our estimate.

[12] The ozone loss in filaments is found to be negligible in both simulations. Since the transport of chemically activated polar air to the northern midlatitudes occurs through filamentary structures and has been previously identified as a potentially major ozone loss pathway [WMO, 1998], it is important to assess the level of confidence of this finding and its implications. First, it has already been established that, within the accuracy of the meteorological analysis, our advection model is able to predict the size and structures of polar filaments detected with airborne [Heese *et al.*, 2001] and ground-based lidar measurements [Godin *et al.*, 2002]. Therefore we are fairly confident that the model produces plausible filamentary structures. This also suggests that the amount of chemically processed polar air transported to midlatitudes via the filaments is somewhat reasonable. Second, the algorithm of filament recognition is likely to overestimate vastly the number of air parcel within polar filaments. Parts of a strongly distorted vortex or midlatitude polar intrusions can be counted as filaments although they often do not peel off from the vortex. This should result in a vast amount of ozone destruction being labeled as occurring within filaments when it actually occurs within the polar vortex. This bias is not affecting our conclusions because, even overestimated, the contribution of filaments to ozone loss is found to be negligible. The other critical concern is whether the model is able to reproduce the halogen deactivation in filaments. The deactivation rate is determinant for the overall ozone destruction within filaments. Previous calculations have shown that the lifetime of filaments of chemically activated polar air depend on the resolution of the model via the numerical diffusion [Searle *et al.*, 1998]. The higher the model resolution is, the lower the numerical diffusion is. Current CTMs have resolutions of the order of several hundreds kilometers. By using a model at high resolution ( $\sim 100$  km), the effects of numerical diffusion should be minimized. It is partly supported by a study by Tan *et al.* [1998], who found that, under conditions relevant to filamentation at the edge of the vortex, chlorine deactivation and ozone destruction is much less sensitive to model resolution at resolutions smaller than 100 km. Systematic comparisons with high-resolution ozone data [Heese *et al.*, 2001; Godin *et al.*, 2002] have shown that our advection model resolves well ozone filaments and does not appear to mix them too quickly with the surrounding midlatitude air. Therefore the numerical diffusion does not seem to compromise strongly the integrity and hence the deactivation of the filaments in the model. Only comparisons with high-resolution measurements of halogen radicals in filaments can confirm it. Even without any diffusion, numerical or real, the filaments would deactivate on a timescale of 10 days because of the photolysis of  $\text{HNO}_3$  [Tan *et al.*, 1998]. Overall, a negligible ozone destruction in filaments is not surprising. Since filaments are the main vehicle for

**Table 1.** Monthly Mean Polar and In Situ Contributions to the Cumulative Extravortex Ozone Loss and the Differences Between the Corresponding Simulations<sup>a</sup>

	January	February	March	April
<i>Meteorology 2000</i>				
Chemical composition 2000				
Polar	0.3	1.7	7.03	16.4
In situ	4.8	7.6	8.73	9.89
Chemical composition 1980				
Polar	0.2	1.03	4.1	9.7
In situ	4.0	6.4	6.74	7.3
Change 2000–1980 (DU)				
Polar	0.1	0.67	2.93	6.7
In situ	0.8	1.2	2.0	2.6
<i>Meteorology 2002</i>				
Chemical composition 2002				
Polar	0.43	0.73	1.7	3.2
In situ	4.3	7.5	10.7	11.6
Chemical composition 1980				
Polar	0.3	0.5	1.3	2.4
In situ	3.6	6.22	8.71	9.3
Change 2002–1980				
Polar	0.13	0.23	0.4	0.8
In situ	0.7	1.28	1.99	2.3

<sup>a</sup>Contributions are given in Dobson units. See text for details.

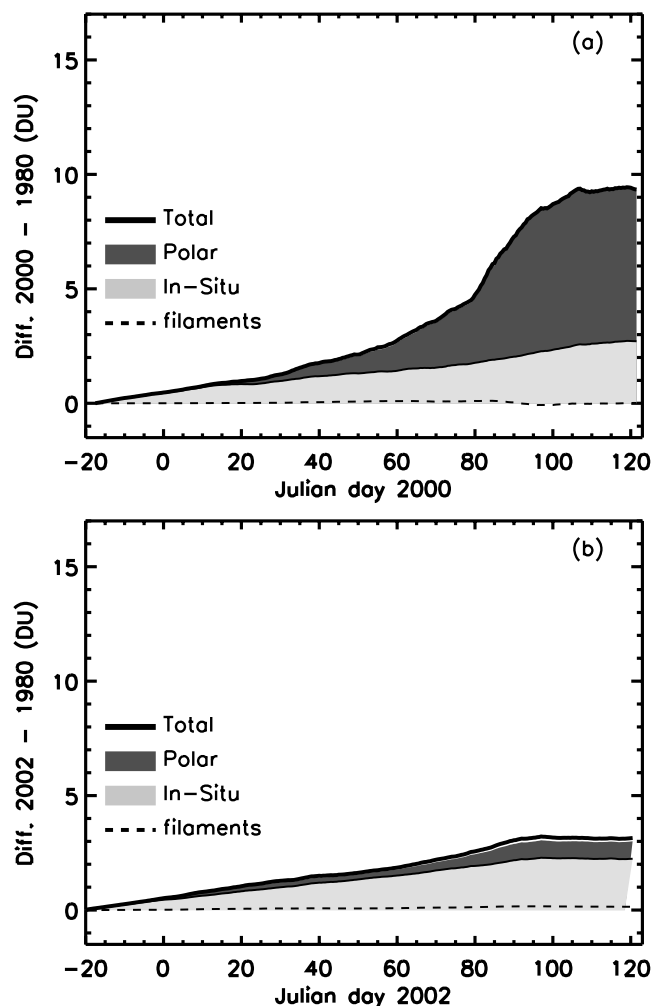
the transport and mixing of chemically activated polar air in the midlatitudes, our results implies that this process is certainly a marginal ozone loss pathway. This is consistent with the lack of differences between the in situ contributions of the two simulations. Indeed, in situ contributions include the ozone loss component from chemically activated polar air which has been transported outside the vortex, mixed and subject to in situ ozone destruction. The 2001–2002 winter was characterized by a disturbed vortex which was fairly activated early in the winter with substantial filamentation and mixing into the middle latitudes, whereas the vortex was fully activated but strong and stable during the winter 1999–2000 with little mixing with the middle latitudes. Therefore one might have expected some influence of the magnitude of the exchanges vortex/extravortex on the in situ contribution. This is not the case. The polar contributions to midlatitude ozone loss are very different with the contribution being about 4 times greater during the 1999–2000 winter than during the 2001–2002 winter. However, the time evolution and magnitude of the in situ contribution varies very little from one winter to the other one, at least until the end of February (see Figure 1 and Table 1). This strong similarity also suggests that the variability from year to year of the seasonal chemical ozone loss at middle latitudes is mainly driven by the transport and mixing of ozone-depleted vortex air into this region.

### 3.2. Impact of Past Changes in Halogen Loading

[13] In order to investigate the impact of past changes in the chemical composition of the stratosphere and, in particular, in the halogen loading since 1980 on midlatitude ozone, two other numerical simulations are performed forcing the model by the 1999–2000 meteorology and the 2001–2002 meteorology, but with a chemical atmospheric composition representative of 1980. The assumed 1980 atmospheric loadings of chlorine, bromine, nitrogen oxides and methane represents 68%, 64%, 94% and 90% of the 2000 and 2002 loadings respectively [WMO, 1998]. The

differences between the chemical composition of the 2000 and 2002 atmosphere are very small.

[14] Figure 2a shows the time evolution of the difference in the cumulative extravortex ozone loss and its various components between the two simulations forced by the same meteorology (1999–2000) but with different chemical composition (1980 and 2000). The same plot is shown in Figure 2b for the simulations forced by the 2001–2002 meteorology. The magnitude of the actual ozone destruction is the slope of the cumulative ozone loss. Table 1 summarizes the results with monthly means. The difference in ozone loss between the two 1999–2000 meteorology experiments, referred thereafter as the 1999–2000 meteorology experiment, increases throughout the winter to reach 9.5 DU by the end of April or 9.3 DU if monthly means are considered (see Table 1); the actual extravortex ozone loss increases by about half, from 19.5 DU in the 1980 simulation to 29 DU in the 2000 atmosphere simulation. During



**Figure 2.** Time evolution of the difference (DU) in cumulative ozone loss in the (a) 1999–2000 and (b) 2001–2002 meteorology experiments (see text). The extravortex ozone loss is averaged over the 35°N–90°N latitude domain and plotted according to the origin of the ozone loss: total (solid line), polar vortex (dark shaded area), in situ (light shaded area) and filament (dashed line).

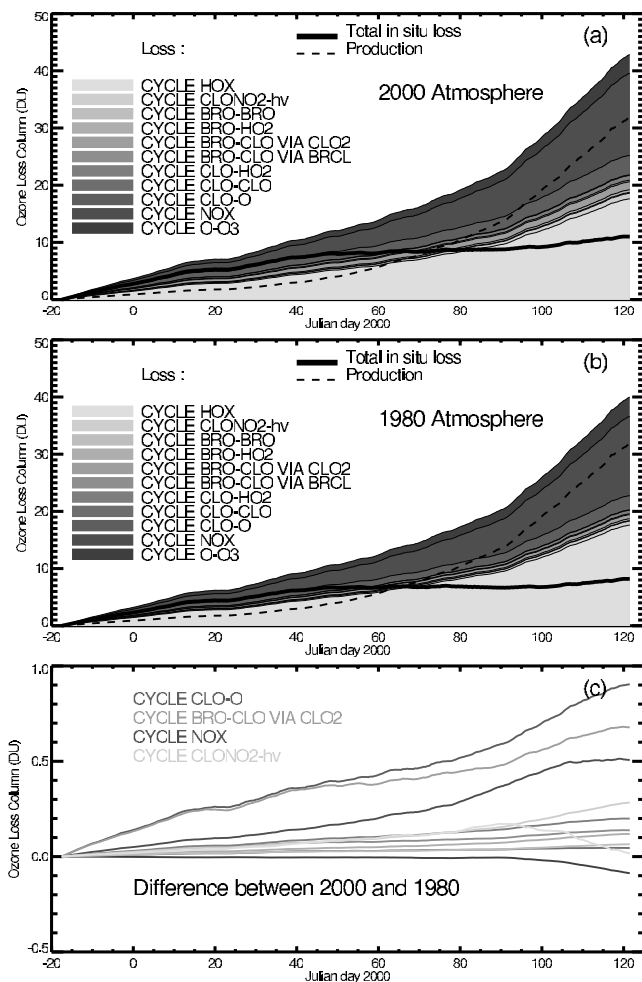
December and January, the larger ozone loss found in the 2000 atmosphere simulation compared to the 1980 atmosphere simulation is associated with its stronger in situ destruction. The difference between the contributions from polar destruction becomes significant in February and is the driving factor by the beginning of March. It causes a sharp increase in the difference in ozone loss at the end of March and beginning of April (see Figure 2a). The ozone destruction from halogen-driven cycles is responsible for most of the differences.

[15] The difference in the cumulative extravortex ozone loss in the 2002 meteorology experiment increases at the beginning of the winter in the same way as in the 2000 meteorology experiment. However, the difference carries on increasing steadily throughout the winter in the 2001–2002 meteorology experiment whereas the difference increases much more sharply from February onward in the 1999–2000 meteorology experiment. The difference in ozone loss reaches only 3.15 DU by the end of April or 3.1 DU in terms of monthly mean in the 2001–2002 meteorology experiment (see Figure 2b) compared to 9.5 DU in the 1999–2000 meteorology experiment. The differences in the in situ ozone destruction are very similar in both sets of experiments with contributions reaching about 2 DU at the end of April. The large differences between the two sets of experiments originate from the differences in the magnitude of the polar destruction. In the 2001–2002 meteorology experiment, the polar vortex contribution remains very small and a minor contributor to the ozone loss in the two numerical simulations; most of the differences in total ozone loss between the two simulations are due to the changes in the in situ component. In the 1999–2000 meteorology experiment, the polar contribution becomes dominant by the beginning of March in the two simulations. The changes in this dominant component are responsible for most of the differences in total ozone loss between the two simulations of the 1999–2000 meteorology experiment. Although the actual magnitude of the ozone destruction is substantial in January and February in all the simulations (see Figure 1), the differences in ozone loss for those months are relatively small in both sets of experiments compared to the differences in ozone loss for March and April. The relatively small changes early on are due to the fact that the cumulative extravortex ozone loss is then dominated by the in situ destruction which is much less sensitive to the halogen loading than the polar destruction (see thereafter).

[16] It is difficult to carry out a fully quantitative comparison with observed ozone trends. Indeed, our seasonal simulations attempt to diagnose the effect of the enhanced halogen and nitrogen loading from mid-December to April for a background aerosol loading whereas observed ozone trends include the influence of other factors varying over two decades. For example, it is likely that the interannual variability in dynamics has been making a contribution to midlatitude ozone trends in the lower stratosphere [Weiss *et al.*, 2001]. In addition, the September–October–November ozone trend is significant and so presumably not all the April trend is due to ozone loss since mid-December. Nonetheless, one can attempt to compare the results to observed ozone trends keeping in mind these caveats. The middle latitude ozone trend derived from ozonesondes is about  $-13.2 (\pm 2.8)$  DU/decade [WMO, 2002]. This trend

has been calculated from data at eight stations between  $36^\circ$  and  $59^\circ\text{N}$  for the period 1980–2000 and corresponds to the 10–25 km altitude range for the March–April–May period. As the ozone trends above 25 km and the tropospheric ozone trends are relatively small in terms of Dobson units (DU), the altitude range contains most of the contribution to the column ozone trend [WMO, 2002]. Our calculations cover approximately both the altitude range and the latitude domain of this ozonesondes trend. Our seasonal simulations indicate a change in the extravortex chemical ozone loss ranging from 1.55 DU/decade (in situ: 1.12 DU/decade, polar: 0.45 DU/decade for the 2002 winter) to 4.66 DU/decade (in situ: 1.3 DU/decade, polar: 3.3 DU/decade for the 2000 winter) depending on the meteorology of the winter. These values represent between 12% and 35% of the observed ozonesondes trend (in situ: 8.5–9.8%, polar: 3.4–25.0%). One caveat is that the ozonesonde trends are consistently larger than the more reliable total column trend derived from satellite data ( $-10.9 (\pm 2.8)$  DU/decade) between  $35^\circ$  and  $60^\circ$  [WMO, 2002]. The agreement is even worse if the ozone losses above 25 km are included. The most invoked reason for this disagreement is the sparse sampling of the ozonesondes (only eight stations) [WMO, 2002]. Our estimates represent between 14.5% and 42.7% of the total column trend derived from satellite data (in situ: 10.3–12.4%, polar: 4.1–30.3%).

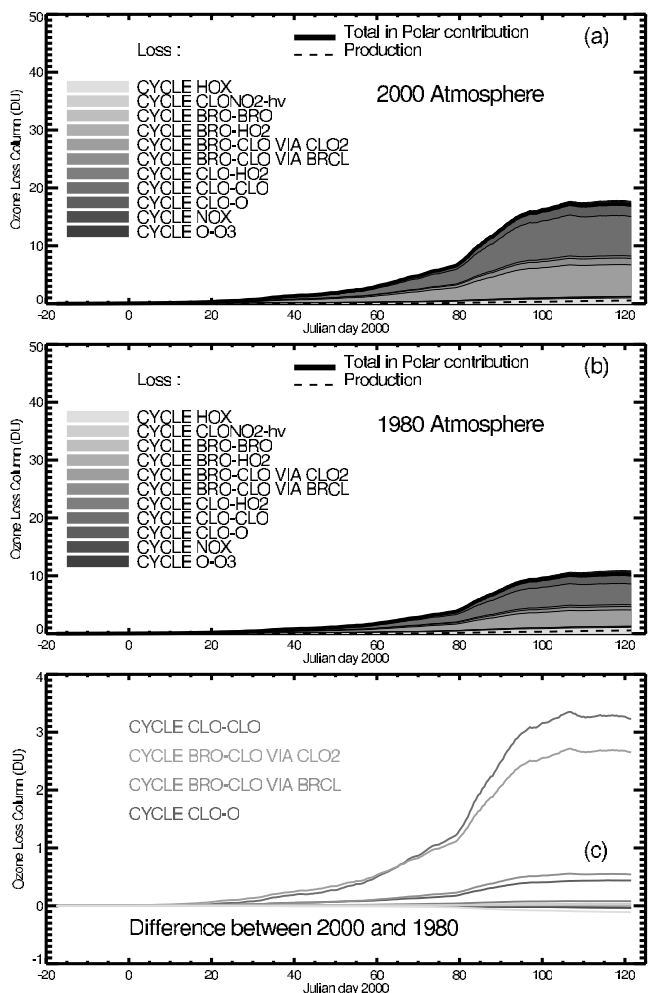
[17] In order to understand exactly how the changes in halogen and nitrogen loading influence midlatitude ozone, the ozone-destroying pathways are characterized by including in the model additional chemical tracers which monitor the production and destruction due to hydrogen, halogen, nitrogen and oxygen compounds within the polar and extravortex domain. Only the results for the extravortex ozone loss are presented here. The cumulative in situ ozone destruction from the main ozone-destroying cycles are plotted as a function of time in Figure 3 together with the cumulative production from  $\text{O}_2$  photolysis and the total cumulative loss (i.e., destruction-production) for the 1980 and 2000 simulations forced with the 1999–2000 meteorology. The same plots are shown in the Figure 4 for the contribution of ozone destruction within the polar vortex to extravortex ozone loss (i.e., polar vortex air which has experienced ozone destruction and been subsequently transported outside the vortex). The results of the 2000 atmosphere simulation will be first described in order to discuss later the differences with respect to the 1980 atmosphere simulation. The fraction of extravortex ozone loss (production-destruction) associated to in situ processes increases until the end of February and then levels off at 11 DU till the end of April (see Figure 1). The cumulative in situ ozone destruction increases throughout the winter/spring to peak at 43 DU by the end of the simulation. However, the in situ ozone destruction is more or less balanced by the in situ ozone production in March and April, resulting in the leveling off of the cumulative ozone loss at about 11 DU. The  $\text{HO}_x$  cycle is the largest contributor to in situ ozone destruction. Their relative contribution is relatively stable during the simulation and represents about 40% of the total ozone destruction. The  $\text{NO}_x$  cycle is responsible for about a third of the total ozone destruction by the end of April. Its efficiency increases in early spring reflecting the release of  $\text{NO}_x$  at middle altitudes from the enhanced photolysis of



**Figure 3.** Time evolution of the in situ contribution (DU) for the (a) 2000 atmosphere simulation, (b) 1980 atmosphere simulation and (c) difference to the cumulative extravortex ozone loss (solid line). The different components indicated are the contributions from the individual ozone-destroying cycles (see legend) and the ozone production from  $O_2$  photolysis (dashed line). See color version of this figure at back of this issue.

$HNO_3$ . In contrast, the efficiency of the halogen cycles tends to decrease at the end of the winter and the early spring. This is due to the release of nitrogen oxides, which favors the conversion of active halogen species (ClO and BrO) into their inactive forms (ClONO<sub>2</sub> and BrONO<sub>2</sub>). The polar destruction contributing to the extravortex ozone loss is dominated by different cycles (see Figure 4). Almost all of the destruction is driven by halogen cycles, mainly the ClO-ClO cycle and the ClO-BrO cycle. The quadratic dependency of the ClO-ClO cycle efficiency on ClO concentrations makes the polar contribution very sensitive to the chlorine loading. The ozone production within the polar vortex is very small compared to the ozone destruction. It is worth noticing that, although the polar ozone destruction contributing to extravortex ozone loss peaks strongly in late March/early April with the return of sunlight over the pole, almost half of the destruction has already taken place by mid-March.

[18] The results can be compared to the distribution of the ozone-destroying cycles found by Millard *et al.* [2002] within the 30°–60°N equivalent latitude for the 1999–2000 winter. Unlike the present study, Millard *et al.* [2002] did not decompose the midlatitude ozone loss into domain-specific contributions for each ozone-destroying cycle. They monitor the ozone loss from the different ozone-destroying cycles wherever the ozone loss occurs. Therefore their results can only be compared to the sum of our in situ and polar contributions. The sums of the in situ (Figure 3a) and polar (Figure 4a) contribution of the main ozone-destroying cycles for all the simulations are shown in Table 2. We find a more important contribution from the HO<sub>x</sub> and NO<sub>x</sub> cycles contributions. It is partly due to the more extended altitude range considered here (350–600K) compared to 342–500K altitude range considered by Millard *et al.* [2002]. Indeed, ozone destruction by the NO<sub>x</sub> cycles and, to lesser extent, the HO<sub>x</sub> cycle are strongly



**Figure 4.** Time evolution of the polar contribution (DU) for the (a) 2000 atmosphere simulation, (b) 1980 atmosphere simulation and (c) the difference to the cumulative extravortex ozone loss (solid line). The different components indicated are the individual ozone-destroying cycles (see legend) and the ozone production from  $O_2$  photolysis (dashed line). See color version of this figure at back of this issue.



**Table 2.** Polar and In Situ Contributions to the Cumulative Extravortex Ozone Loss From Individual Ozone-Destroying Cycles at the End of April for the 2000 Atmosphere and the 1980 Atmosphere Simulations Forced With 1999–2000 Meteorology<sup>a</sup>

	2000 Atmosphere			1980 Atmosphere		
	In Situ	Polar	Sum	In situ	Polar	Sum
O-O	00.0	00.0	00.0	00.0	00.0	00.0
O-O <sub>3</sub>	03.3	00.2	03.5	03.4	00.2	03.6
NO-NO <sub>2</sub>	14.4	00.6	15.0	13.9	00.7	14.6
ClO-O	03.4	01.8	05.2	02.5	01.4	03.9
ClO-ClO	00.1	06.8	06.9	00.1	03.6	03.7
ClO-HO <sub>2</sub>	00.8	00.4	01.2	00.6	00.3	00.9
BrO-ClO via BrCl	00.3	01.1	01.4	00.1	00.6	00.7
BrO-ClO via ClO <sub>2</sub>	01.4	05.4	06.8	00.7	02.8	03.5
BrO-HO <sub>2</sub>	00.5	00.1	00.6	00.3	00.0	00.3
BrO-BrO	00.1	00.1	00.2	00.1	00.0	00.1
ClONO <sub>2</sub> + <i>hnu</i>	01.0	00.2	01.2	00.7	00.1	00.8
HO <sub>x</sub>	17.6	01.0	18.6	16.6	01.1	17.7

<sup>a</sup>Contributions are given in Dobson units. The sum of the in situ and polar contributions is also given. See text for details.

favored over the 500–600K altitude range compared to the halogen cycles. The contributions to ozone loss from halogen cycles are somewhat comparable in both studies during the winter. However, in early spring, the ozone destruction from halogen chemistry is still very effective in the *Millard et al.* [2002] study, whereas it tends to level off in our study. They also noticed that the model predicted larger polar ozone losses when it was forced with United Kingdom Meteorological Office Analysis (UKMO) instead of ECMWF analyses, even when denitrification was ignored. This difference was due to the enhanced PSC formation with UKMO analyses which were consistently colder in the coldest regions of the lower stratospheric vortex than ECMWF analyses, especially in midwinter.

[19] Figure 3c shows the time evolution of the differences in in situ ozone destruction from the individual cycles for the 2000 meteorology experiment. The total in situ ozone destruction in the 1980 atmosphere simulation is slightly smaller than in the 2000 atmosphere simulation (see Figure 2). It is mostly due to the lower efficiency of the ClO-ClO and ClO-BrO cycle resulting from the reduced halogen loading in the 1980 atmosphere. The change in the efficiency of the NO<sub>x</sub> cycle also contributes to the reduced O<sub>3</sub> loss. The lower NO<sub>x</sub> levels in the 1980 atmosphere compared to the 2000 atmosphere would tend on their own (i.e., without the concomitant lower halogen loading) to boost the halogen and HO<sub>x</sub> cycles. However, the lower NO<sub>x</sub> levels also result in enhanced O<sub>3</sub> destruction at the higher levels of the vertical domain considered here where the NO<sub>x</sub> cycle dominates. Although the destruction from the halogen and NO<sub>x</sub> cycles is reduced, the efficiency of the HO<sub>x</sub> cycle increases slightly at the end of the winter in the 1980 atmosphere. It is mainly caused by the reduced formation of reservoir species as HNO<sub>3</sub> and HNO<sub>4</sub> particularly above 400K.

[20] The time evolution of the difference in the polar contribution to the extravortex ozone loss is plotted in figure 4c along with the individual components of the ozone-destroying cycles. The lower polar contribution in the 1980 atmosphere is almost entirely caused by the change in the efficiency of the halogen cycles, essentially the

ClO-ClO and BrO-ClO via ClO<sub>2</sub> cycles. It is not surprising because these cycles are already responsible for the bulk of the polar ozone destruction in all the simulations.

#### 4. Conclusion

[21] A three-dimensional high-resolution chemical transport model MIMOSA-CHIM is used to estimate the impact of the evolution of the stratospheric chemical composition on the winter-spring midlatitude ozone budget at northern midlatitudes. Since midlatitudes ozone is affected by the import of chemically activated or ozone-depleted polar air and by background in situ chemistry, the focus is on evaluating as accurately as possible the contributions of the different mechanisms to the extravortex chemical ozone loss and how their efficiencies might have changed in the last 2 decades. The contribution of polar processes being very sensitive to the meteorological conditions prevailing around the vortex, two extremes, but somewhat typical, present-day winters are considered: the 1999–2000 winter and the 2001–2002 winter. Four numerical simulations are performed forcing the model with ECMWF analyses from December to the end of April for a 2000 atmospheric chemical composition and a 1980 atmospheric chemical composition. A budget analysis is carried out for the partial ozone column between 350 and 600K using a combination of geographical and chemical ozone tracers.

[22] All the model simulations indicate that the transport of chemically activated polar air to northern midlatitudes during the winter does not represent a significant source of extravortex ozone loss. The midlatitude chemical ozone loss appears to be simply determined by the in situ destruction and the transport of ozone-depleted polar air (air which has experienced ozone destruction within the vortex before being transported and mixed at midlatitudes). The magnitudes of the polar contributions differ by about a factor 4 between the 1999–2000 meteorology simulations and the corresponding 2001–2002 meteorology simulations. The polar contribution is found to be responsible for 60% of the total extravortex ozone loss when the model is forced by the exceptional meteorology of the 1999–2000 Arctic winter. In contrast, the magnitude of the in situ destruction is found to vary little from one winter meteorology to the other one. The results suggest that the interannual variability in the winter-spring chemical ozone loss at midlatitudes is mainly driven by the magnitude of the polar contribution. The chemical ozone tracers experiments show that the in situ destruction is dominated by the HO<sub>x</sub> and NO<sub>x</sub> cycles whereas the polar contribution to extravortex ozone loss is almost entirely due to the halogen cycles.

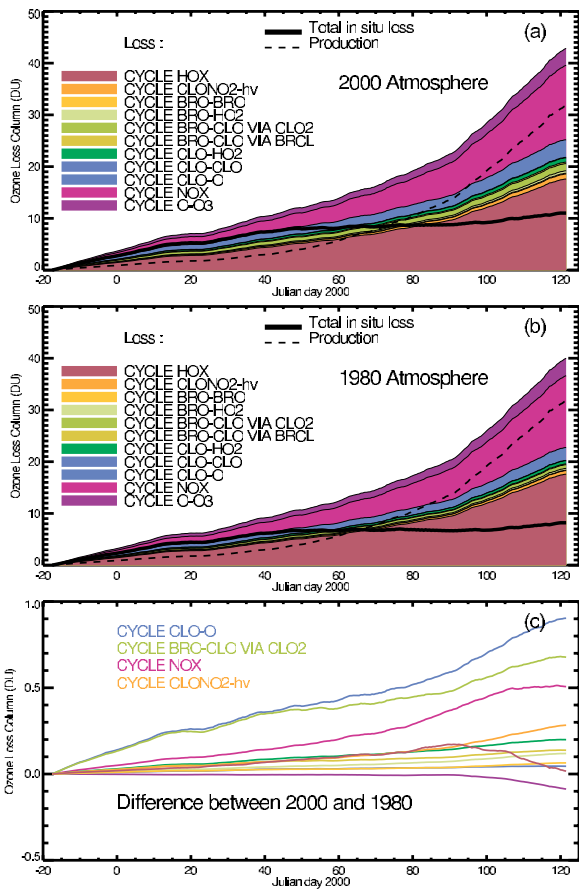
[23] According to the model simulations, the changes in the atmospheric chemical composition, in particular the halogen and nitrogen loading, since 1980 should have resulted in an additional midlatitude ozone loss of 3 to 10 DU depending of the meteorology of the winter. The model-calculated in situ destruction increases by about 20% which represents about 2 DU of the total additional ozone loss. This increase is more or less unaffected by the meteorology of the winter considered. In contrast, the model calculates a factor 2 to 3 increase in the polar contribution in the last 2 decades. This highlights the strong sensitivity of the polar contribution to the halogen loading. The polar

contribution accounts for between 1 and 7.5 DU of the additional extratortex ozone loss depending on the winter considered. The extratortex ozone loss is found to be maximized during the 1999/2000 winter when the polar contribution outweighs the in situ destruction. This type of meteorological conditions with a cold and persistent vortex is also very favorable to polar ozone destruction. The model-calculated ozone differences between the 1980 atmosphere simulations and the corresponding 2000/2002 atmosphere simulations represent up to 35% of midlatitude ozone trends derived from ozonesondes and up to 43% of midlatitude trends derived from satellite data. As the polar contribution is very sensitive to the halogen loading, the expected decay in chlorine loading following the implementation of various international protocols on the control of CFCs emissions could be accompanied by a large reduction in the polar contribution to the extratortex ozone loss and hence in the interannual variability of the ozone loss. One mitigating factor is the observed stratospheric cooling which may delay the recovery of Arctic ozone [Shindell *et al.*, 1998] by making more common the particular conditions of the 1999–2000 winter and hence boosting the importance of the polar contribution. Furthermore, as the HO<sub>x</sub> cycle is a major contributor to the extratortex ozone loss, the stratospheric water vapor trend [WMO, 2002] should directly enhance the in situ destruction. This would reduce further the interannual variability in the midlatitude ozone loss because the extratortex in situ destruction does not appear to vary much with the meteorological conditions.

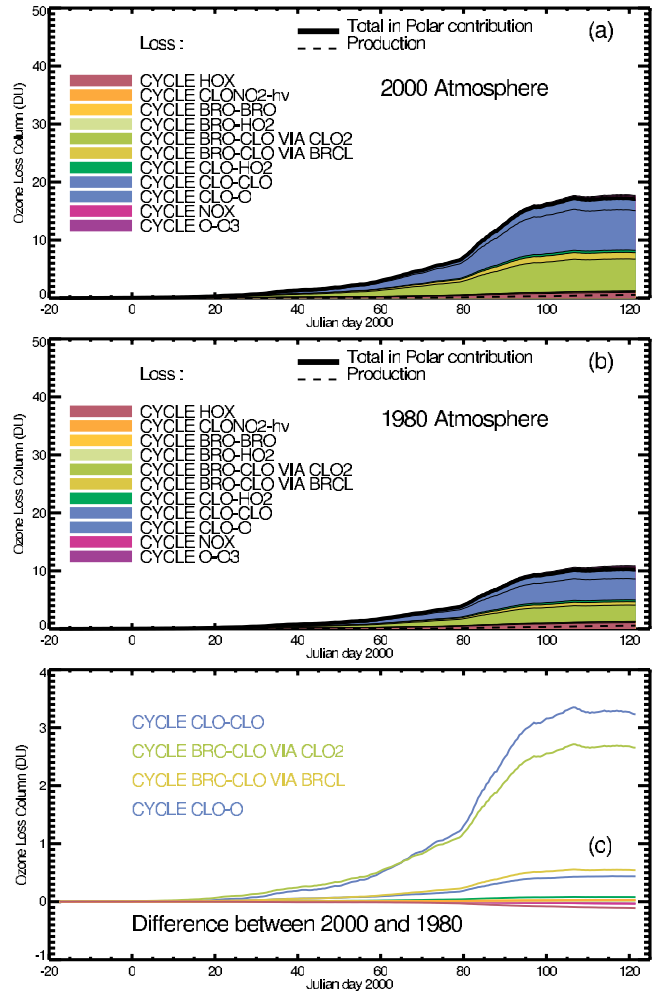
[24] **Acknowledgments.** We are grateful to ECMWF and NILU for providing meteorological data.

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**Figure 3.** Time evolution of the in situ contribution (DU) for the (a) 2000 atmosphere simulation, (b) 1980 atmosphere simulation and (c) difference to the cumulative extravortex ozone loss (solid line). The different components indicated are the contributions from the individual ozone-destroying cycles (see legend) and the ozone production from O<sub>2</sub> photolysis (dashed line).



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