Arctic and Antarctic ozone layer observations: chemical and dynamical aspects of variability and long-term changes in the polar stratosphere[1]

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The altitude dependent variability of ozone in the polar stratosphere is regularly observed by balloon-borne ozonesonde observations at Neumayer Station (70°S) in the Antarctic and at Koldewey Station (79°N) in the Arctic. The reasons for observed seasonal and interannual variability and long-term changes are discussed. Differences between the hemispheres are identified and discussed in light of differing dynamical and chemical conditions. Since the mid-1980s, rapid chemical ozone loss has been recorded in the lower Antarctic stratosphere during the spring season. Using coordinated ozone soundings in some Arctic winters, similar chemical ozone loss rates have been detected related to periods of low temperatures. The currently observed cooling trend of the stratosphere, potentially caused by the increase of anthropogenic greenhouse gases, may further strengthen chemical ozone removal in the Arctic. However, the role of internal climate oscillations in observed temperature trends is still uncertain. First results of a 10 000 year integration of a low order climate model indicate significant internal climate variability, on decadal time scales, that may alter the effect of increasing levels of greenhouse gases in the polar stratosphere.

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Since the mechanisms of the formation of the Antarctic ozone hole were discovered, a question of prime concern has been whether comparable chemical losses of ozone can also occur in the Arctic. During winter/spring, Arctic stratospheric air masses regularly drift above mid-latitudes. Any considerable ozone depletion in these air masses would influence the densely populated areas of North America, Europe and northern Asia and could have severe consequences for the biosphere in these regions (van der Leun 1998).

The rapid ozone loss in the polar lower stratosphere is due to chlorine and bromine catalysed reaction cycles (WMO 1999). Nearly all of the chlorine and a substantial fraction of the bromine in the stratosphere originates from the breakdown of man-made chlorofluorocarbons (CFCs) and

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halons (WMO 1999). The most effective ozone destruction cycles involve active chlorine radicals and can proceed only if sunlight is present. However, in mid- and low latitude stratospheric conditions a large fraction of the chlorine is in the form of passive compounds that are not effective in destroying ozone. The ozone destroying effect of enhanced chlorine levels in mid-latitudes is therefore limited, though probably an important reason for the observed negative trend of the thickness of the ozone layer in mid-latitudes (WMO 1999).

At polar latitudes the particular meteorological conditions can lead to a situation in which nearly all available chlorine is in its active form and ozone can be destroyed very rapidly. During polar winter the stratospheric temperatures fall and large low pressure systems form over the polar regions. These so-called polar vortices are encompassed by strong jet streams that isolate the air masses inside the vortices from air at mid-latitudes. Breaking atmospheric waves apply a drag on the jet stream

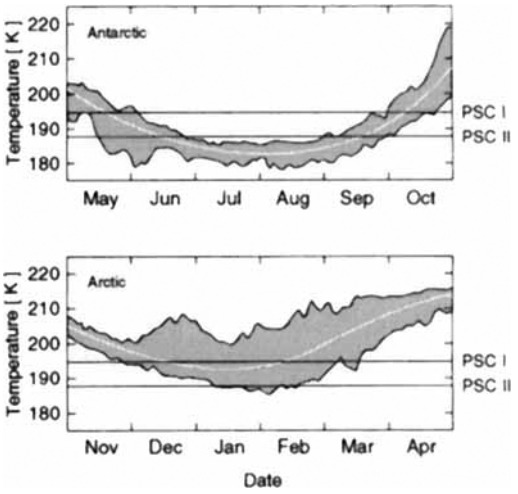


Fig. 1. Wintertime variation of the daily hemispheric minimum temperatures in 50 hPa based on NMC analysis between 1978 and 1994. The shaded area gives the spread of the curves for the individual years. The approximate threshold temperatures for PSC I and PSC II existence are marked by horizontal lines. (Reproduced from WMO 1995.)

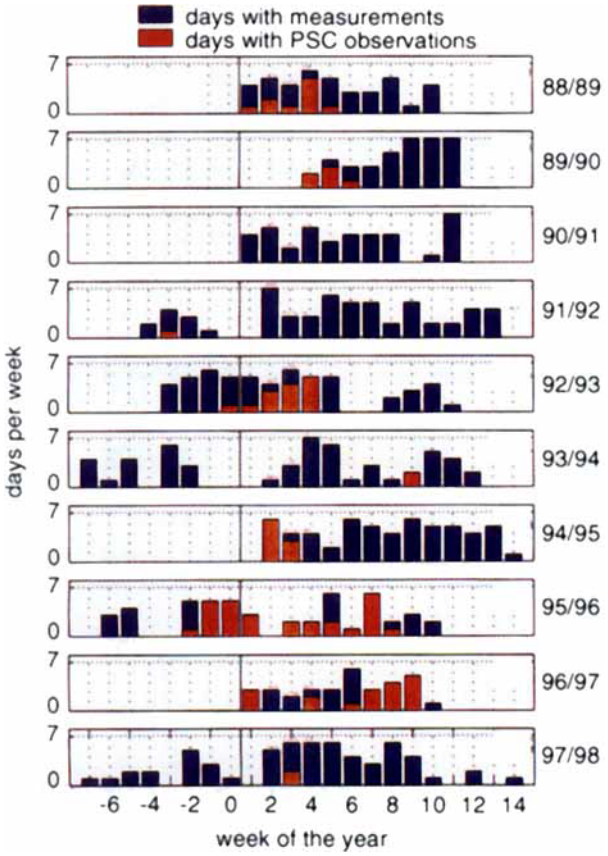


Fig. 2. Frequency of PSC I observations above Spitsbergen, as detected by a multi-wavelength lidar installed at the NDSC Station in Ny-Ålesund (78.9° N, 11.9° E).

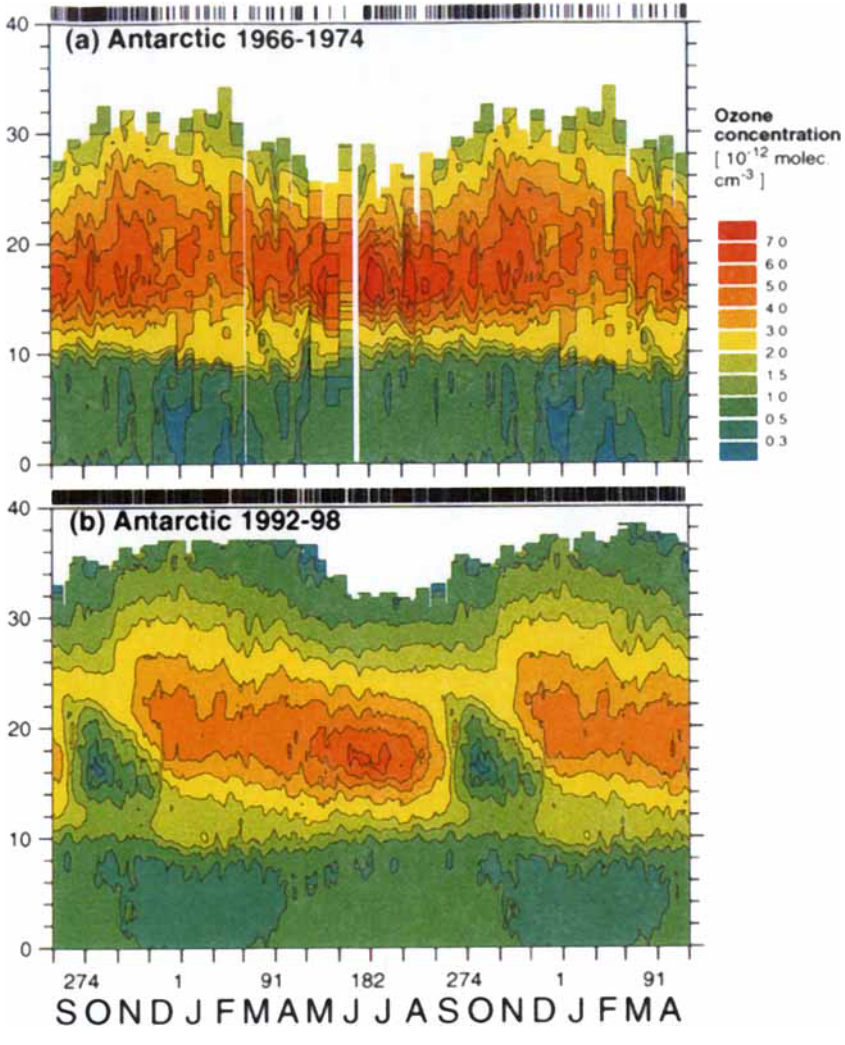
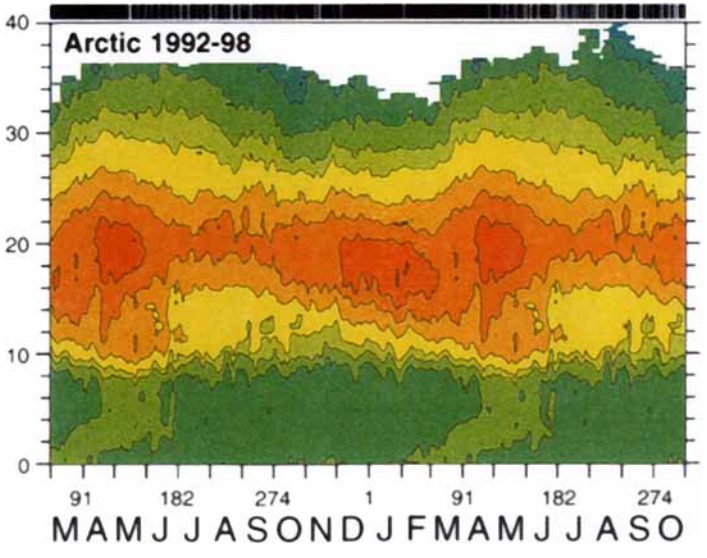


Fig. 3 (left). (a) Altitude-time section of the mean seasonal variation of the ozone concentration above Syowa Station (69.0° S, 39.4° E) between 1966 and 1974. The time axis denotes the day of the year, with the initial letters of the months also given. The months September to April are repeated on both sides of the diagram. 155 ozonesondes contributed to the plot. Each sonde is marked by a vertical black bar at the upper edge of the figure. (b) Altitude-time section of the mean seasonal variation of the ozone concentration above Neumayer Station (70.6° S, 8.2° E) between 1992 and 1998. 492 ozonesondes contributed to (b); all other information is the same as for (a).

Fig. 4 (below). Altitude-time section of the mean seasonal variation of the ozone concentration above Koldewey Station (78.9° N, 11.9° E) between 1992 and 1998. The time axis is shifted by six months compared to Fig 3; otherwise the information is the same. 693 ozone sondes contributed to the plot. For colour key, see Fig. 3.



and, against the purely geostrophic motion of the air, allow the air to move slowly poleward. This process leads to subsidence and diabatic cooling of air inside the polar vortex. The degree of subsidence in a given winter is controlled by the respective wave activity. A stronger wave drag leads to higher temperatures inside the vortex, larger diabatic cooling rates and faster subsidence (Holton 1997).

Dramatic ozone losses can take place in the isolated polar vortices. When temperatures fall below approximately −78°C (at 475 K), condensed aerosol particles consisting of mixtures of H_2O, H_2SO_4 and HNO_3 may form Type I Polar Stratospheric Clouds (PSC I). Rapid heterogeneous reactions on the surface of PSC I particles convert passive chlorine compounds into active chlorine radicals. The most effective mechanism to deactivate the chlorine radicals once they have formed requires the presence of gas phase HNO_3. However, when temperatures during the polar winter fall below ca. −85°C (at 475 K), the PSC I particles quickly take up water ice, grow much larger and form Type II PSCs. These particles can sediment with considerable velocities and can irreversibly remove water and HNO_3 from the stratosphere. In these so-called denitrified air masses, rapid ozone loss can proceed for longer periods of time, leading to more severe depletion (e.g. Rex et al. 1997; Waibel et al. 1999).

The wave activity in the Arctic is much stronger than in the Antarctic, and the Arctic polar vortex is less stable, warmer and breaks up earlier. Temperatures and vortex strength show a pronounced year to year variability (e.g. Pawson et al. 1995). In some Arctic winters the vortex breaks up in midwinter, causing dramatic increases in polar temperatures in those years (e.g. Scherhag 1952; Naujokat 1992). Figure 1 shows the development of the minimum temperature for both hemispheres. In the Antarctic, the temperatures fall below the PSC I and PSC II thresholds over many weeks in each winter. In the Arctic, PSC I temperatures are reached in most of the winters, but the length of the PSC I period and the area covered by PSC I temperatures is variable. PSC II temperatures are rarely reached in the Arctic. However, during the extremely cold Arctic winter of 1995/96, minimum temperatures fell below the PSC II threshold for about three weeks (Naujokat & Pawson 1996).

Figure 2 shows the frequency of PSC I observations above Spitsbergen, as detected by a multi-wavelength lidar installed at the NDSC Station in Ny Ålesund (78.9° N, 11.9° E). As this site is not directly influenced by orographical effects, the observation statistic can be regarded as representative of the inner stratospheric vortex. Figure 2 reveals PSC "poor" winters in 1990/91, 1991/92, 1993/94 and 1997/98. Otherwise the occurrence of PSCs is highly variable, including periods when every measurement detected PSCs as during the cold periods of the winters of 1995/96 and 1996/97, as well as others, when PSCs occurred transiently, as in January 1989 and 1993.

Ozone observations in the Arctic and Antarctic

Ozone profiles

We have used balloon-borne electrochemical ozonesondes to measure ozone profiles in the Antarctic since 1985 and in the Arctic since 1988. The ozone sensors are launched with standard radiosondes. During ascent up to 30–38 km altitude, in situ measurements of ozone partial pressure and meteorological parameters are transmitted on-line. Ozone concentrations or ozone mixing ratios can be calculated from the measurements with a vertical resolution of ca. 150 m. Ozonesondes are currently the only instruments capable of measuring profiles of ozone with detailed vertical resolution throughout the year (including polar night and polar day) and during nearly all weather conditions. Between 1985 and early 1992 the measurements in the Antarctic were taken at Georg Forster Station (70.8° S, 11.8° E) (Gernandt et al. 1989). In 1992 the soundings have been moved to Neumayer Station (70.6° S, 8.2° E) (Gernandt et al. 1996). For comparison we also show measurements from Syowa Station (69.0° S, 39.4° E) between 1966 and 1974 (Gernandt et al. 1996). Due to the similar latitude of all three stations and the high zonal symmetry of the mean ozone field in the Antarctic the three data sets can be compared directly. The soundings in the Arctic have been made at Koldewey Station (78.9° N, 11.9° E) (Gernandt et al. 1996). In winter the measurements of all stations reflect the conditions inside the polar vortex.

Figures 3 and 4 show vertical sections of the ozone concentrations in the Antarctic and Arctic throughout the year. The ozone concentrations in the Antarctic are plotted for two different time

periods: (a) shows the measurements from Syowa Station between 1967 and 1979, i.e. before the Antarctic ozone hole formed; (b) shows the measurements at Neumayer Station between 1992 and 1998.

In both hemispheres the stratospheric ozone concentrations increase below about 24 km after the formation of the respective polar vortex in fall (about September in the Arctic, March in the Antarctic). This is caused by the diabatic subsidence of air inside the vortex. In the lower stratosphere ozone mixing ratios systematically increase with altitude. Air masses with higher ozone mixing ratios descend and compression leads to higher ozone concentrations. This effect is limited to the region below ca. 24 km, since the vertical gradient of ozone mixing ratios is weak above this altitude.

In the Antarctic soundings from Neumayer Station (Fig. 3b) the ozone concentrations in the vertical region of the ozone maximum dramatically drop during the months August to December. The older data from Syowa Station (Fig. 3a) show no decrease in the ozone concentrations during these months. Figure 5 shows a number of recent profiles from these months. The ozone concentrations drop below the detection limit of the sonde in a broad vertical region. This effect, clearly caused by halogen catalysed ozone destruction (WMO 1999), is one of the strongest anthropogenic signals in the atmosphere. A profile from the mid-1980s is also shown in Fig. 5. In a smaller vertical region the ozone was nearly completely destroyed even in this early profile. Since then a further decrease of ozone concentrations occurred mainly at the upper and lower edge of the depleted region.

In the Northern Hemisphere no comparable decrease of the ozone concentrations in late winter is discernible. Instead, averaged over many years, the concentrations continue to increase until the polar vortex breaks up in spring. At this time air from mid-latitudes, where subsidence is weak and ozone concentrations are therefore lower, spreads into the polar region contributing to the observed decrease in the ozone concentrations during May to July. Part of this decrease may also be due to natural slow photochemical ozone destruction.

Figure 6a shows ozone profiles taken in the Arctic during late March of different years. The profiles from 1995, 1996 and 1997 show considerable ozone deficits in the region of the ozone maximum. In these years the Arctic winter was exceptionally cold. However, in the Arctic the dynamically induced variability of the ozone layer is too large to allow a straightforward interpretation of these deficits in terms of chemical ozone loss. Figure 6b shows one example of a dynamically driven ozone fluctuation above Koldewey Station. The short-term drop in the ozone concentration and subsequent recovery is caused by advection of air masses with different histories and is a common natural phenomenon in the Arctic. Given the large dynamical variability, more sophisticated approaches have to be used to detect and quantify chemically induced ozone loss in the Arctic. We will show that substantial chemically induced ozone loss did occur in recent Arctic winters and that even in 1992 some chemical ozone loss occurred which is not apparent in Fig. 6a.

Ozone column densities

A vertical integration of the ozonesonde profiles allows the calculation of ozone column densities. For the integration the ozone mixing ratios above the burst level of the balloon is assumed to be constant. To reduce the error introduced by this assumption, only ozone profiles that reached at least 25 km altitude have been used for the calculations of column densities. The estimated residual ozone above this level is generally smaller than 10% of the total ozone column.

Figures 7 and 8 show the variation of the ozone column density for different years above Neumayer Station, Antarctica, and Koldewey Station, the Arctic, respectively. In the Arctic (Fig. 8) the ozone column density reaches its maximum in spring. The general increase of the ozone column during winter is due to subsidence of air in the polar vortex. Air with high ozone mixing ratios replaces the subsided air masses at high altitudes, whereas air with lower ozone mixing ratios leaves the polar vortex in its lowest parts. The large year-to-year variability of the column densities in spring is mainly caused by the varying times of the vortex break-up in different years. After the break-up of the vortex, mid-latitude air masses with lower ozone column densities mix into the Arctic. Extremely high column densities in March/April were only reached in years when the vortex still existed during these months. In the winters of 1995, 1996 and 1997, the ozone column densities remained at low values, although the vortex still existed during March. During January to March

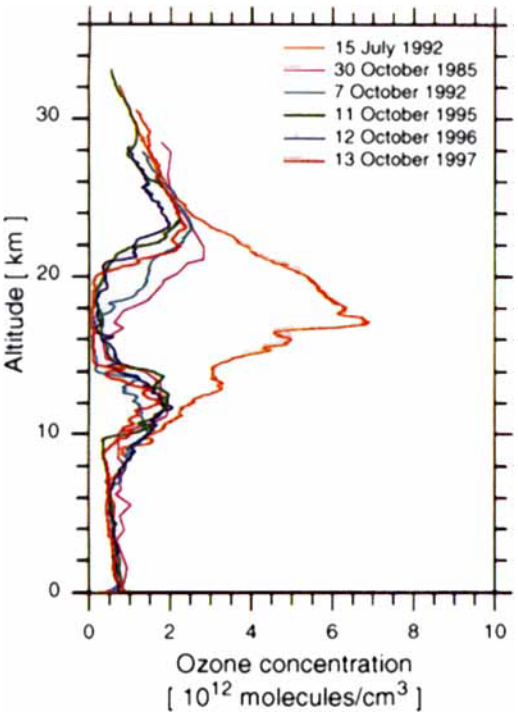
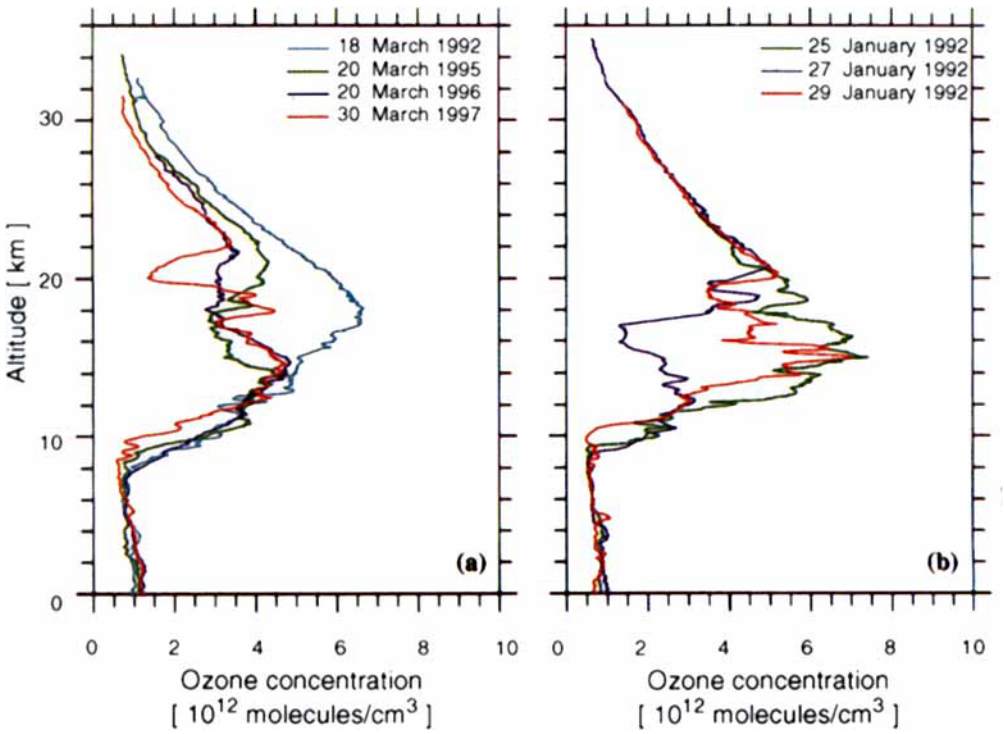


Fig. 5 (left). Ozone profiles measured during October at Georg Forster Station in 1985 and Neumayer Station in 1992–97. One early winter profile, July 1992, is also plotted.

Fig. 6 (below). Ozone profiles measured at Koldewey Station (78.9° N, 11.9° E): (a) profiles measured in late March during four different years; (b) profiles measured within four days in 1992.



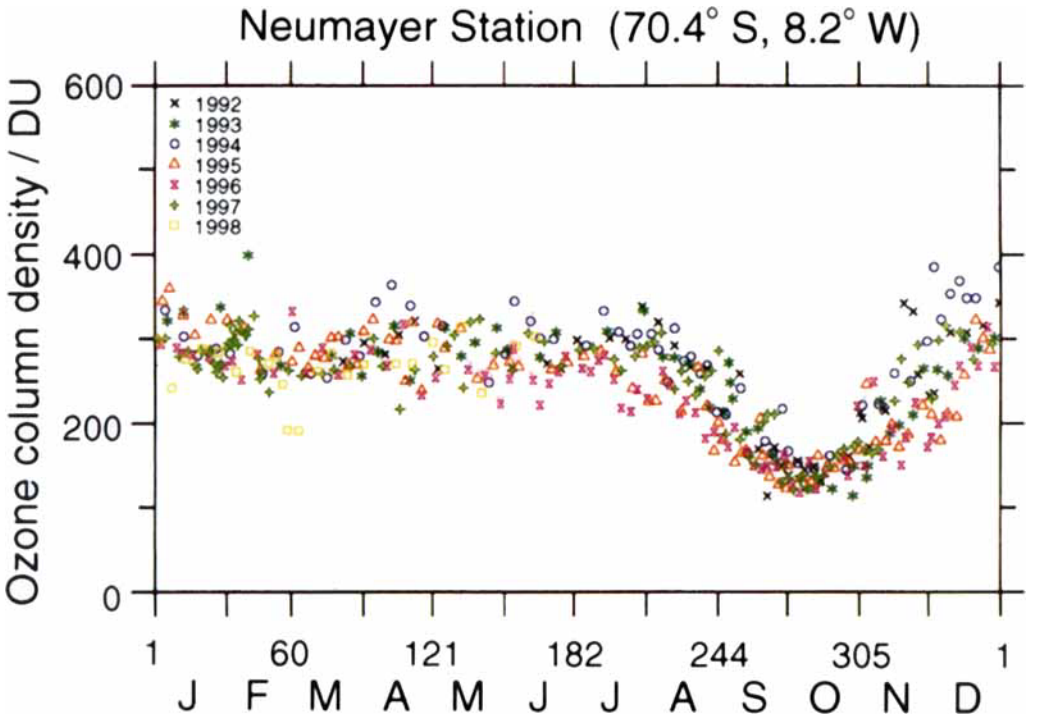


Fig. 7. Neumayer Station (70.6° S, 8.2° E). Data from the years from 1992 to 1998 are marked with different symbols and colours (see key). The time axis denotes the day of the year and the initial letters of the months are also given.

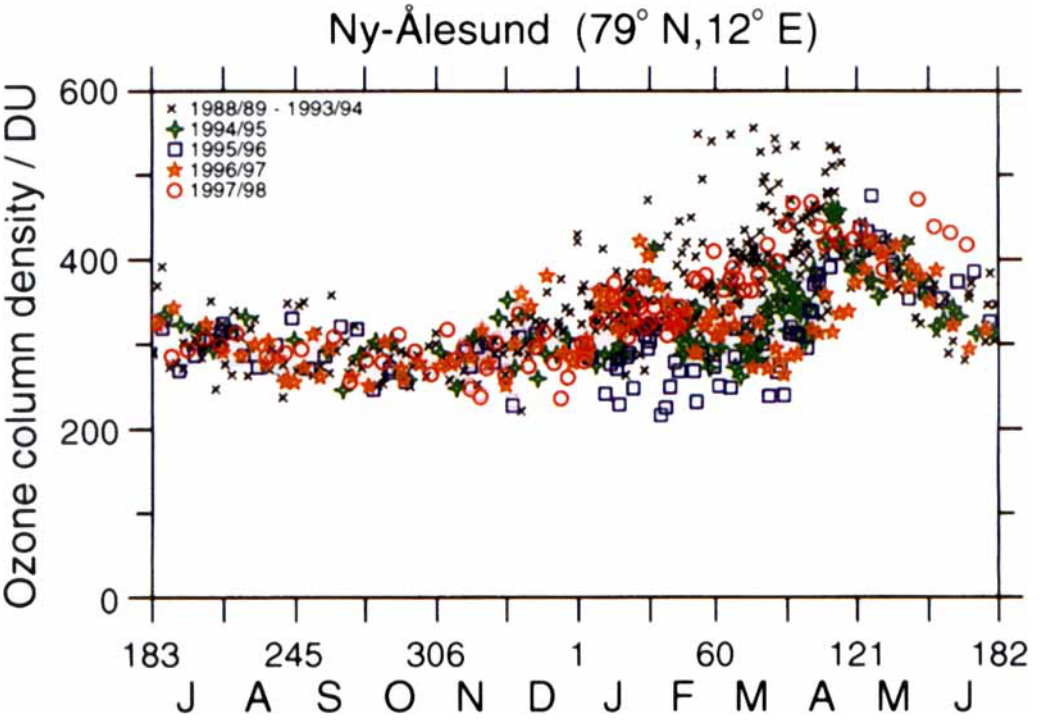


Fig. 8. Seasonal variation of the ozone column density above Koldewey Station (78.9° N, 11.9° E). See caption with Fig. 7.

1996, the lowest ozone column densities ever measured above Koldewey Station were reported. Possible explanations for the low ozone column densities during these years are: a weak wave activity connected with less subsidence in the polar vortex; chemical destruction of ozone; or a combination of both. The cold stable vortices of these years indicate that the wave activity was indeed weak. So it is reasonable to attribute a part of the unusually low ozone column densities during spring to a weaker diabatic circulation in these years. This is particularly true for the winter 1996/97, when the wave activity was exceptionally weak. However, we will demonstrate that a considerable fraction of the negative ozone anomaly in these Arctic springs was due to extensive chemical loss of ozone.

In the Antarctic no increase of the ozone column during winter is observed. This may partly be due to early chemical ozone loss, but is also caused by the weaker wave activity in the Southern Hemisphere resulting in a weaker diabatic circulation and less transport of ozone into the polar region during the southern winter. In late winter and spring rapid chemical destruction of ozone leads to a sharp drop in the ozone column densities. During these periods ozone column densities as low as 130 Dobson Units (1 DU = $2.69 \cdot 10^6$ cm^{-2}) are common.

Chemical ozone loss in the Arctic

Since the large variability of the Arctic ozone layer makes it difficult to detect chemical ozone loss, we developed the so-called Match approach. The technique is based on the statistical analysis of a large number of "matches." A match is defined as a pair of ozonesonde measurements, where both sondes probed the same air parcel at different times as it passed over their respective stations. The basic idea of the Match approach is illustrated in Fig. 9. To identify the matches, calculated air parcel trajectories that take into account modelled diabatic cooling rates were used to track the motion of the air parcels between the measurements. The approach can be applied in two ways: (1) for post campaign analysis of a very large number of uncoordinated ozonesonde soundings by selecting soundings which are linked by chance based on calculated trajectories (von der Gathen et al. 1995; Rex et al. 1998); or (2) much more effectively as a joint effort by a large number of stations that perform coordinated ozonesonde launches during a "Match campaign" (Rex et al. 1997; Rex et al. 1999; Schulz et al. 2000, in press). The analysis includes various quality controls on ozonesonde data as well as trajectory data. The main advantage of this Lagrangian approach is that chemical and dynamical effects can be separated to a high degree. The ability of the Match technique to account for dynamical changes in ozone without introducing a systematic bias is demonstrated by a statistical analysis that shows that no inferred chemical loss of ozone occurs during periods of darkness along the trajectories (Rex et al. 1998; Rex et al. 1999). Match results, based on the coordinated launches of 600 to 1400 ozonesondes per winter, are currently available for five winters between 1991/92 and 1997/98. A map showing the locations of the participating stations is shown in Fig. 9.

To derive the spatial and seasonal variation of the ozone loss rate, various subsets of matches can be analysed. Figure 10 shows an overview over the chemical ozone loss rates measured with the Match technique during specific Arctic winters. Additionally the areas of the Northern Hemisphere were temperatures were cold enough to allow PSCs to exist are indicated. Chemical ozone loss was detected in all winters other than the relatively warm winter of 1997/98, when the measured rates of chemical loss are hardly significant. In all instances periods of significant chemical ozone loss rates were initiated by periods with temperatures below the PSC threshold. Usually the ozone loss stopped about two weeks after temperatures warmed above the PSC threshold. The winter 1995/96 is an exception and is discussed below. This pattern and the fact that the ozone loss occurs only in sunlight is in qualitative agreement with the current theory.

Figure 11 shows a vertical section of the ozone loss rates measured during January to March 1995. At all levels the observed ozone loss periods are closely related with periods of possible PSC formation. The low loss rates in early January are due to a lack of sunlight close to mid-winter. During this winter, maximum local losses of approximately 60% occurred at ca. 20 km altitude. Larger local ozone losses (approximately 65%) have been found in the colder winter of 1995/96. Chemical loss rates in the ozone column reached about 2.5 DU per day during two periods. The accumulated loss in the ozone column is 127 ± 14 DU (ca. 30%). The observed local ozone

loss rates are similar to the loss rates observed over Antarctica (WMO 1999). The main differences between both hemispheres are the longer persistence and the larger vertical extent of the ozone loss in the Antarctic.

Although the qualitative pattern of the ozone loss rates agrees very well with model calculations, the rate of the measured ozone loss is considerably larger than found by models (Becker et al. 1998, 2000). This discrepancy reveals an important inability of chemical models to quantitatively reproduce the observed ozone losses and indicates that further research is needed to improve our understanding of the ozone loss mechanisms.

After the extremely cold Arctic winter of 1995/96, the ozone loss in a thin vertical layer continued long after temperatures warmed above the PSC threshold, causing the largest amounts of local ozone loss ever observed in the Arctic. Model studies and trace gas measurements suggest that this was probably due to denitrification, causing Antarctic-like conditions in these Arctic air masses (Rex et al. 1997). This shows how close Arctic conditions are to those in the Antarctic and that only slightly lower temperatures in the Arctic can lead to dramatic ozone losses there.

Future of the ozone layer

External effects

Due to the effective regulations of the CFC production the increase in stratospheric chlorine loading flattens. During the coming decades the large reservoir of CFCs in the atmosphere will continue to maintain high levels of stratospheric chlorine loading. Models predict a slow decrease of stratospheric chlorine loading, reaching "pre-ozone hole concentrations" around the year 2050 (WMO 1999). However, the results of the Match experiments show how sensitive the Arctic ozone loss is to small changes in stratospheric temperatures. During the next decades the effect of year-to-year variations of the Arctic stratospheric temperatures will have a far greater effect on the ozone loss in a given year than the slow variation of the total chlorine loading. Model calculations and observations indicate that climate changes connected with increased greenhouse forcing may lead to a cooling trend in the stratosphere, coupled with a trend towards more stable Arctic vortices (WMO 1999). This could lead to larger Arctic ozone losses in the near future and may substantially delay the recovery of the ozone layer (Shindell et al. 1998; Waibel et al. 1999).

During the next decades strong volcanic eruptions that lead to an increase in the stratospheric aerosol loading could increase the ozone loss by the enhanced chlorine loading (Solomon et al. 1996). Significant depletions of the ozone layer have been observed after the eruptions of El Chichon (1982) and Mt. Pinatubo (1991) (e.g. Rodriguez et al. 1994).

Internal variability

Predictions of the future of the ozone layer are complicated by the fact that observed trends of temperature and vortex strength in the Arctic can be attributed not only to changes in response to greenhouse gas forcing but is also possible due to oscillations as part of natural decadal climate changes. There is growing evidence that the Arctic stratospheric polar vortex is implicated in the long-term oscillations of Earth's climate. Perlwitz & Graf (1995), for example, showed a strong coupling between the strength of the Arctic polar vortex and the North Atlantic Oscillation. It is crucial to know the influence of internal long-term climate variability of Earth's climate before reliable predictions for the future can be made.

An approach to study the causes and physical mechanisms of long-term natural climate variability was developed by Dethloff et al. (1998). A nonlinear atmosphere-like dynamical system with a sparse horizontal resolution including orographical and thermal forcing in a long planetary wave and its interaction with both a baroclinic wave and the zonal flow has been constructed and integrated over 10 000 years. This long-term integration allows the determination of the dominating modes of atmospheric variability on decadal time scales with high statistical significance. Spectra of the prognostic variables show well-pronounced maxima at the decadal time scale, statistically significant at the 95% confidence level. An extension of this approach to a coupled atmosphere–ocean ice model of moderate complexity is underway.

The current poor knowledge of the complex nonlinear interactions in the tropospheric–stratospheric climate system coupled with ocean and sea ice makes assessment of future atmospheric trends difficult. The unknown long-term natural climate variability over decadal and centennial time scales

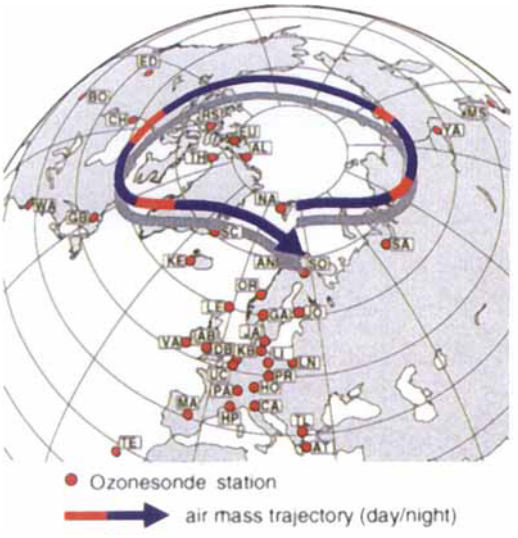


Fig. 9. Illustration of the Match technique and map of the participating ozonesonde stations.

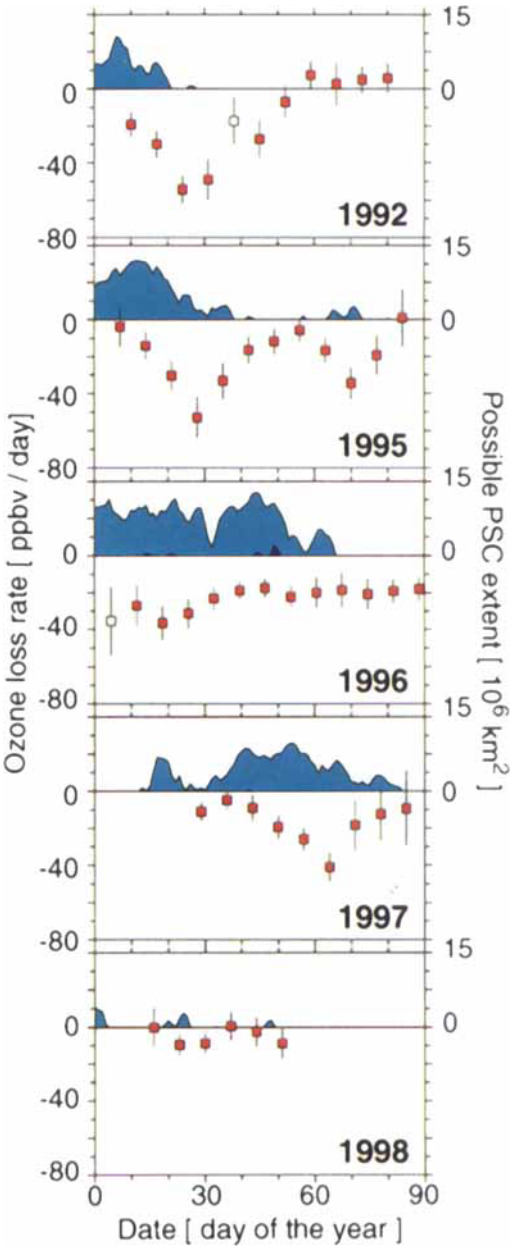


Fig. 10. Overview of the results of the different Match campaigns. The red points give the variation of the ozone loss rates through the different winters. The blue shaded curves give the potential PSC extent. Adapted from Rex et al. (1997, 1998, 1999) and Schulz et al. (2000).

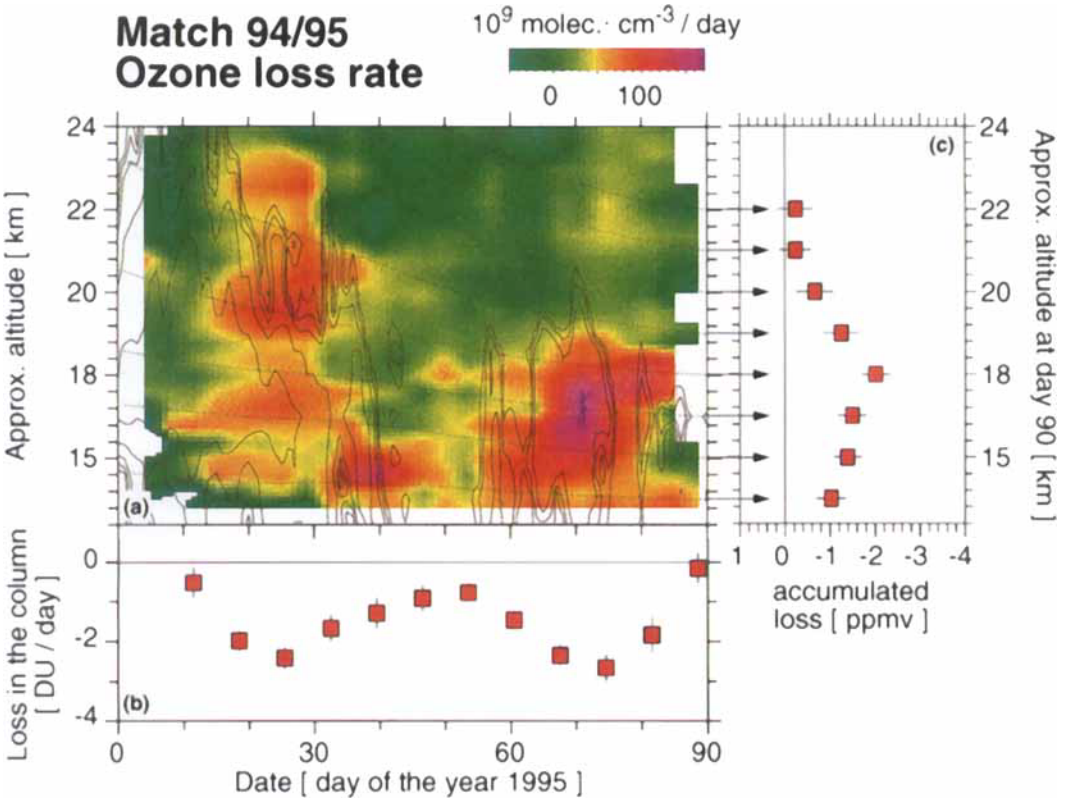


Fig. 11. (a) Altitude time section of the ozone loss rate during January to March 1995. 1470 matches contributed to the plot. The 0.3, 0.7 and 1.5 · 106 km^2 isolines of the potential PSC extent are plotted as thin black lines. The dotted lines give the descending motion of air masses through the plotted section. (b) Loss rates of the ozone column calculated by vertically integrating the local loss rates. The accumulated loss of the ozone column is 127 ± 14 DU (ca. 30%). (c) Accumulated ozone losses in subsiding layers of air as calculated by integrating the local loss rates along the dotted lines in panel (a).

may exert a significant impact on climate trends independent from greenhouse gases or coupled with them and has the potential for unexpected future changes.

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