

VARIABILITY OF OZONE AND AEROSOLS IN THE POLAR ATMOSPHERE

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Abstract: Since 1980 the appearance of spring ozone depletion directly caused by chemical removal has significantly changed the pattern of vertical ozone distribution in the antarctic stratosphere, and has become a principal feature of a changing atmosphere. In recent years chemical ozone loss has also been found in the arctic stratosphere. Transient events like the presence of volcanic aerosols can additionally remove ozone in the lower polar stratosphere. Balloon-borne ozone observations and sun photometer measurements performed in the Antarctic and Arctic as well as satellite data are used to review interannual and seasonal changes in the polar stratosphere. For the arctic region tropospheric ozone and aerosol variations are briefly discussed. Some features of polar ozone and the impact of aerosols are discussed by using original results.

1. Introduction

The southern polar ozone depletion in spring is one of the strongest signals of a changing atmosphere. This phenomenon was first noticed by CHUBACHI (1984) in ground-based total ozone measurements and a few balloon-borne data recorded at Syowa station (69°S; 40°E) and reported in more detail by FARMAN *et al.* (1985) from ground-based total ozone measurements recorded at Halley station (75°S; 27°W). From that time on scientific findings have accumulated experimental evidence for chemical ozone losses caused by elevated concentrations of reactive chlorine and bromine in the arctic and antarctic stratosphere, which are activated by heterogeneous reactions at the surface of polar stratospheric cloud (PSC) particles (*e.g.*, WMO, 1992, 1995).

Additional ozone perturbations have been observed in the lower stratosphere above the Antarctic after the eruptions of Mt. Pinatubo and Cerro Hudson in 1991 (HOFMANN and OLTMANN, 1993; RODRIGUEZ *et al.*, 1994). Ozone was depleted down to values never observed before since the beginning of regular recording due to the chemical perturbations caused by volcanic sulphate aerosols, acting in addition to the well-recognized chlorine and bromine reactions on polar stratospheric clouds (McCORMICK *et al.*, 1995).

In the Arctic, tropospheric ozone sensitively responds in a broad range of time scales to the chemical composition of air masses transported from mid-latitudes toward high latitudes. The mean annual variation of ozone has changed over decades (LOGAN, 1985; ANLAUF *et al.*, 1994). Strong ozone depletion events may occur within the boundary

layer (BARRIE *et al.*, 1988; OLTMANS *et al.*, 1989). Evidence have been found that this sudden ozone depletion appears when elevated concentrations of bromine are recorded (STAEBLER *et al.*, 1994). Up to now the meteorological conditions and chemical processes are not sufficiently studied in order to understand this phenomenon completely.

The global distribution of ozone is mainly controlled by stratospheric photochemistry and the global atmospheric circulation. Ozone is produced in the equatorial stratosphere by photochemical reactions and then transported poleward and downward toward high latitudes by the meridional circulation (ELUSZKIEWICZ *et al.*, 1996; SUTTON, 1994). This transport depends on the seasonal variation of planetary wave activity. In the lower stratosphere the lifetime of ozone is long during winter and spring, so as it is transported poleward it begins to accumulate there (SCHOEBERL and DOIRON, 1993).

The seasonal ozone distribution in the polar stratosphere is normally controlled by a circumpolar stratospheric vortex circulation which develops due to the increasing meridional temperature gradient between equator and pole in the winter hemisphere. This strong zonal circulation inhibits the advection of ozone into the polar stratosphere. So the ozone content in the winter polar stratosphere remains more or less constant until mid-winter. Inside the stratospheric vortex air masses subside slowly due to diabatic cooling and ozone is slightly displaced downward from the middle stratosphere to the lower stratosphere (SCHOEBERL *et al.*, 1992; TOON *et al.*, 1992). Increasing fluxes of heat and momentum by planetary wave activity lead to the final breakdown of the stratospheric vortex in late winter and spring. During this period ozone is advected into the middle and lower polar stratosphere by strong meridional exchange of air masses between mid-latitudes and polar latitudes. Therefore the stratospheric ozone content normally has a maximum in polar latitudes due to these dynamical processes. The transition from winter to summer circulation in the stratosphere dramatically slows the accumulation of ozone. In summer, planetary wave activity is weak and ozone decreases toward photochemical equilibrium. This photolysis of ozone molecules is forced by solar radiation in the 400 nm–800 nm range (RÖTH, 1994). This radiation penetrates deeper into the polar stratosphere and subsequently photolytical destruction of ozone leads to a minimum stratospheric ozone content in the middle stratosphere down to about 20 km in late summer.

The natural variability of atmospheric dynamics on the global scale and transient natural events, as well as the growing anthropogenic impact, control ozone in the polar atmosphere. Significant ozone changes on seasonal and interannual time scales depend on the occurrence of natural and anthropogenic aerosols or the formation of polar stratospheric cloud particles. Those aerosols may force heterogeneous chemical reactions yielding to chemical ozone losses. They can also change radiative transfer conditions and with them ozone-related photochemical reactions. Observations from the Antarctic and Arctic are used to discuss the variations of ozone and aerosols in the stratosphere and troposphere.

2. High Latitude Data for Ozone and Aerosols

2.1. Data

The variability of ozone in the polar stratosphere and troposphere is discussed by

using balloon-borne ozone observations from three antarctic sites and one arctic site. Information on the occurrence and seasonal variation of aerosols in the polar atmosphere are obtained from ground-based photometer measurements at several polar stations as well as from satellite instruments (*cf.* Table 1).

Electrochemical ozone sensors are flown with standard radiosondes. During the balloon ascent up to about 30 km altitude, *in-situ* data of ozone partial pressure, temperature, air pressure, humidity, and wind are transmitted on-line to the ground receiving station. Ozone concentrations or ozone mixing ratios can be calculated by using pressure and temperature data. The vertical resolution of the ozone profile is about 50 m. Ozone sonde data from Syowa Station, Georg Forster station, and Neumayer station provide the vertical ozone distribution for nearly the same latitude, *i.e.* at 70°S. Ozone sonde data from Ny-Ålesund in the Arctic refer to northern latitudes at 80°N. At these polar latitudes the tropospheric and stratospheric vertical ozone distribution, including the polar night, can be obtained for all seasons of the year by using balloon-borne ozone sounding techniques.

The total spectral optical depths of the atmosphere in the short wave range between 390 and 1045 nm can be measured by sun photometers with interference filters. By using the information from 8 channels, the aerosol optical depth can be calculated within this spectral range. The same type of sun photometer is operated at three antarctic stations and one arctic station (Table 1). Thus, the aerosol optical depths obtained by this network can be compared each other (HERBER *et al.*, 1996). Data from the Stratospheric Aerosol Monitoring experiment (SAM II) and the Stratospheric Aerosol and Gas Experiment (SAGE II) (McCORMICK *et al.*, 1995) provide spectral extinction coefficients of aerosols with high vertical resolution of about 1 km for altitudes from the middle troposphere up to the stratosphere. According to THOMASON and POOLE (1993), the calculation of zonal mean values with a latitudinal resolution of 2.5 degrees provides reliable strato-

Table 1. Observations of the vertical ozone distribution and total spectral optical depth of aerosols.

Station	Location	Data/instrument/sensor
Syowa	(69°S; 40°E)	vertical ozone distribution (balloon-borne ozone sensor (KC 79))
Georg Forster	(71°S; 12°E)	vertical ozone distribution (balloon-borne ozone sensor (OSE)) spectral optical depth (sun photometer)
Neumayer	(71°S; 08°W)	vertical ozone distribution (balloon-borne ozone sensor (ECC)) spectral optical depth (sun photometer)
Mirny	(66°S; 93°E)	spectral optical depth (sun photometer)
Ny-Ålesund	(79°N; 12°E)	vertical ozone distribution (balloon-borne ozone sensor (ECC)) spectral optical depth (sun photometer)

spheric aerosol extinction coefficients at high latitudes.

Sun photometer data were used to calculate weekly means of the total aerosol optical depth of the atmosphere at the wavelength 1000 nm for all observations, and additionally monthly means were calculated for October in the Antarctic from 1988 to 1995 and March in the Arctic from 1991 to 1994. Satellite data of the same wavelength range were used to calculate monthly zonal means of the stratospheric aerosol optical depth between 67.5°S and 72.5°S for October and between 70.0°N and 75.0°N for March from 1979 to 1994.

2.2. Basic data plots of the mean annual vertical ozone distribution

The mean annual patterns of the vertical distribution of ozone concentration were calculated for certain periods (Table 2). Some results are shown as altitude-time cross sections for the Antarctic and Arctic in Figs. 1 and 3. Figure 1 shows a plot with color coded ozone concentrations for antarctic stations, *i.e.* Syowa Station (upper panel) for the period 1967 to 1979, Georg Forster station (central panel) for the period 1985 to 1991, and Neumayer station (lower panel) for the period 1992 to 1995. Contributing individual ozone soundings are indicated above each panel. Figure 3 shows another plot for the arctic station Ny-Ålesund, *i.e.* the upper panel for the period 1988 to 1995 and the central panel for the period 1992 to 1995.

In order to discuss the long-term changes for the Antarctic, mean annual vertical ozone distribution has to be compared with respect to the oldest available data set for 70°S recorded at Syowa Station. Considering all available data, a comparison with the mean pattern before 1980 is made for those stations where the highest number of ozone soundings has been performed (Table 2). Three periods were selected, *i.e.* the period 1980 to 1984 when the ozone depletion was first recorded by balloon soundings, the period 1985 to 1991 when regular observations became available, and the period 1992 to 1995 for the recent mean state. Relative changes of the mean annual pattern of vertical ozone distribution in percent are obtained by calculating the ratios of the selected period versus the mean annual pattern of Syowa Station for the period 1967 to 1979, which is used as the reference for detecting changes in the southern polar stratosphere. The results are shown in Fig. 2. Yellow color (100%) means no significant change in the

Table 2. Calculation of the mean annual variation.

Location	Period	Number of years	Number of profiles
Syowa (SY)	1967–1979	13	141 (see Fig. 1)
Syowa (SY)	1980–1984	5	77 (not shown)
Syowa (SY)	1986–1991	6	171 (not shown)
Syowa (SY)	1992–4/1995	3.5	173 (not shown)
Georg Forster (GF)	1985–1991	7	356 (see Fig. 1)
Georg Forster (GF)	1986–1991	6	291 (not shown)
Neumayer (NM)	1992–4/1995	3.5	204 (see Fig. 1)
Ny-Ålesund (NA)	1988–4/1995	7.5	641 (see Fig. 3)
Ny-Ålesund (NA)	1988–1991	4	245 (not shown)
Ny-Ålesund (NA)	1992–4/1995	3.5	396 (see Fig. 3)

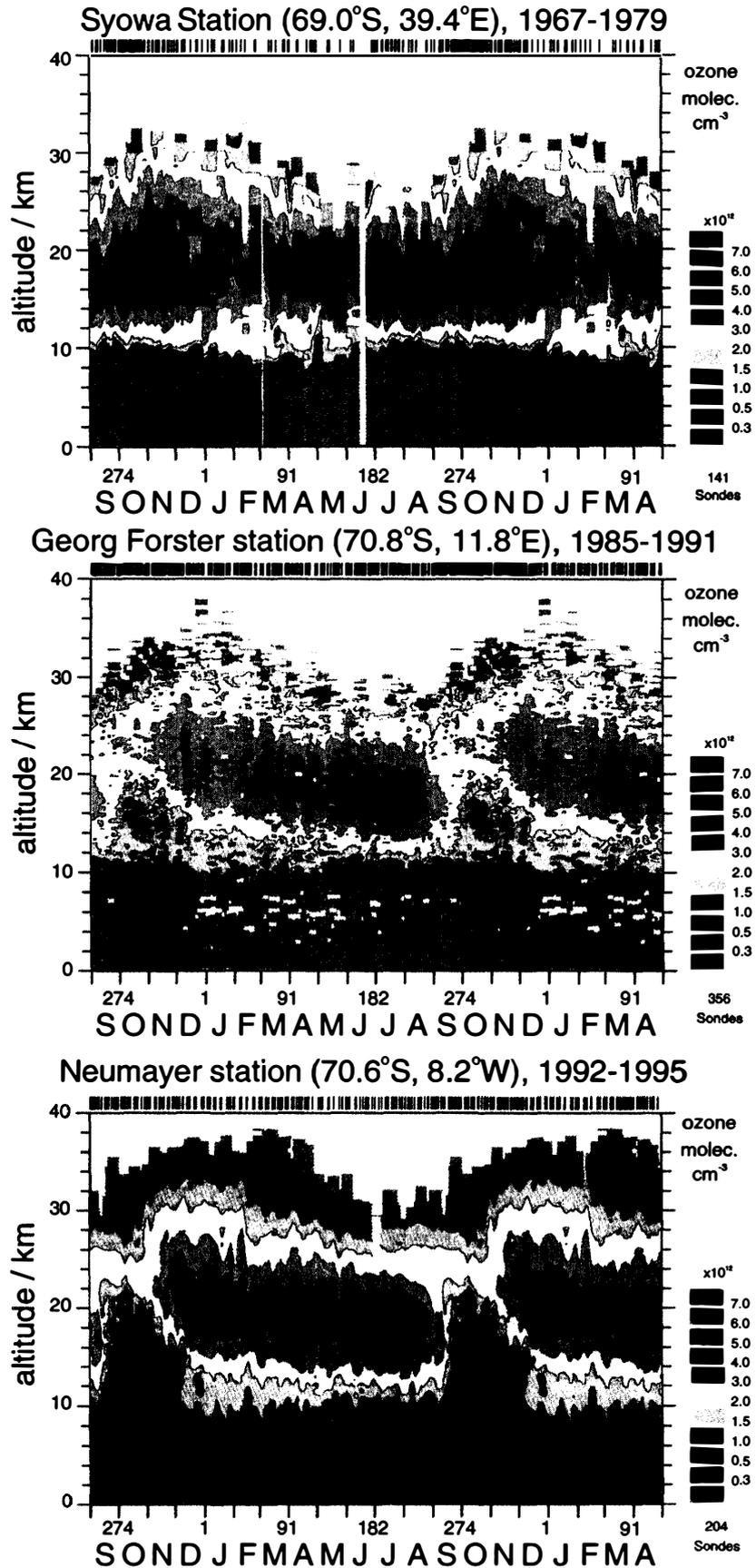


Fig. 1. Altitude-time cross sections of ozone concentration (color coded $\times 10^{12}$ molec/cm⁻³) illustrating mean annual variation for Syowa Station by 141 soundings (upper panel), for Georg Forster station by 356 soundings (central panel), and for Neumayer station by 204 soundings (lower panel). Altitude range is 0–40 km. Time range is 1.5 years (Julian day and month). Contributing soundings are indicated above each panel.

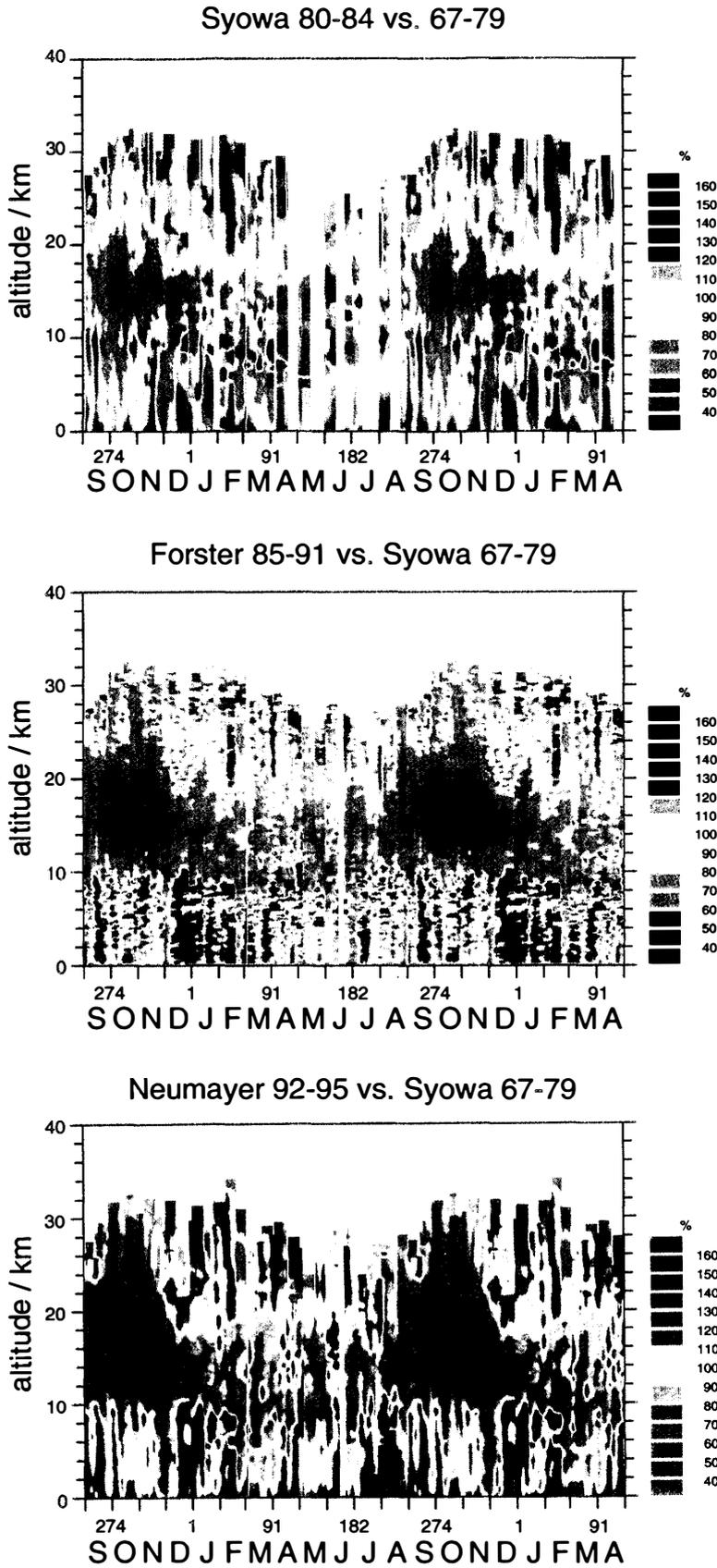


Fig. 2. Altitude-time cross sections for percentage change (%) of mean annual variations referred to the mean annual variation for Syowa Station (1967–1979). Upper panel: Syowa (1980–84) versus Syowa (1967–79); central panel: Georg Forster (1985–91) versus Syowa (1967–79); lower panel: Neumayer (1992–95) versus Syowa (1967–79). Values of 100% (yellow color) means no change, values greater than 100% (red color) indicate an increase and values less than 100% (blue color) indicate a decrease in ozone concentrations.

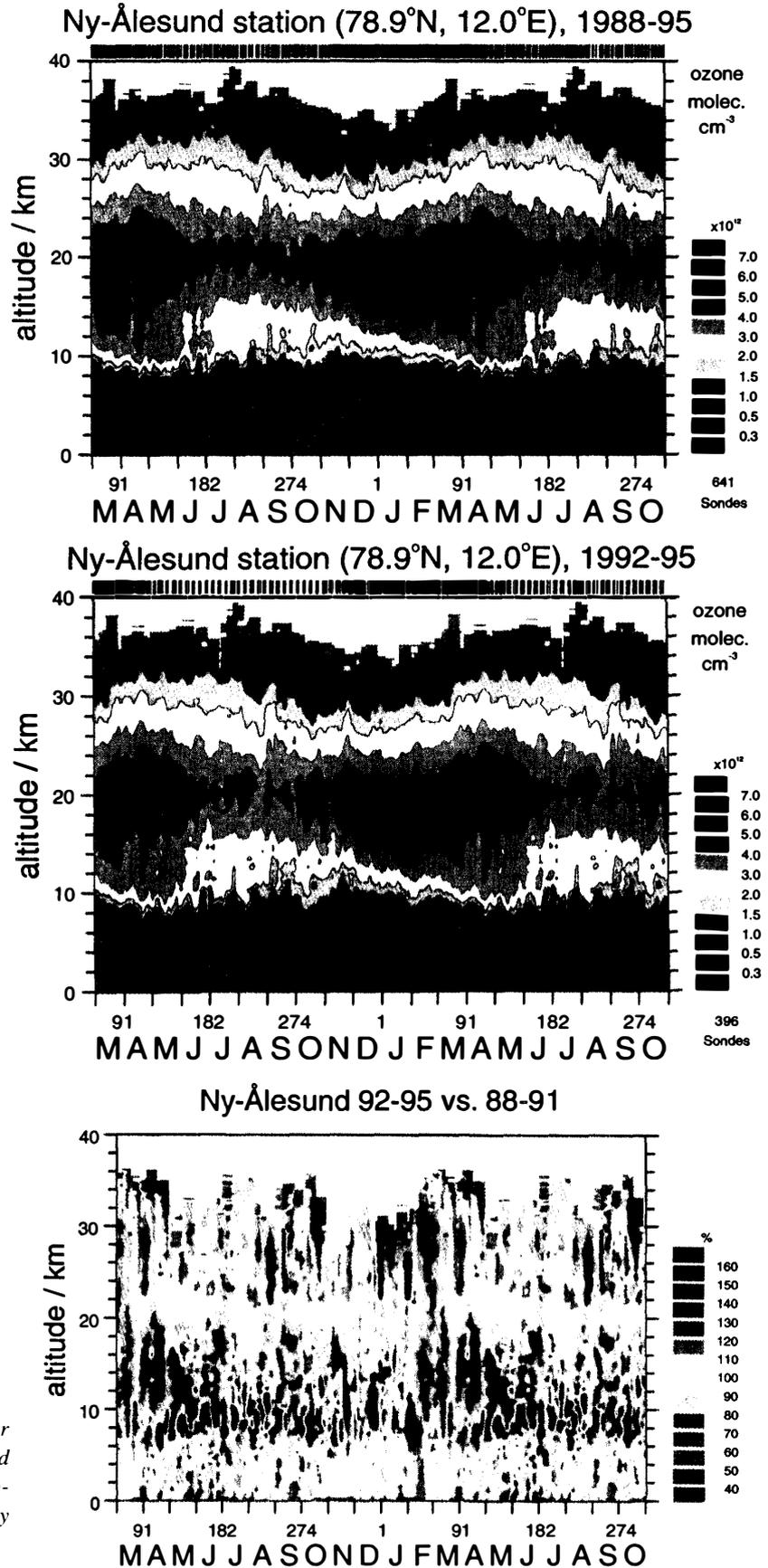


Fig. 3. Same as Figs. 1 and 2 for Ny-Ålesund station; period 1988-95 by 641 soundings (upper panel); period 1992-95 by 396 soundings (lower panel).

mean ozone concentration with respect to the Syowa annual mean before 1980. Red color indicates an increase by more than 30% while green and blue colors indicate a decrease by more than 30%, respectively.

A similar calculation was done for Ny-Ålesund. Changes of the recent mean annual pattern of ozone concentration for the period 1992 to 1995 with respect to the period 1988 to 1991 are shown in Fig. 3 (lower panel).

It has to be considered that antarctic ozone data were recorded at different locations by different ozone sonde types (Table 1). Furthermore, the mean annual variations of ozone concentration were calculated from ozone sounding data which are not uniformly distributed over the considered periods of years. So this attempt for discussing altitude-dependent long-term changes within the southern polar vortex is only a qualitative one. But recently no other data exist at the same latitude for a period of almost 30 years. Assuming that probably latitudinal differences of the annual vertical ozone distribution in stratospheric altitudes have been small at 70°S before 1980, the Syowa reference is assumed to show the normal mean pattern for all three locations at this southern latitude before 1980. Furthermore, the comparability of the mean annual patterns obtained by different sonde types has been checked for Georg Forster data and Syowa data which have a temporal overlap in the years 1986 to 1991 (Table 2). The ratio Forster 86–91 versus Syowa 86–91 was calculated. In stratospheric altitudes this ratio does not systematically decline by more than –10% for Georg Forster data with respect to Syowa data. In tropospheric altitudes the decline increases up to 50% in the summer season. Therefore tropospheric data cannot be used for intercomparisons without considering the local conditions.

3. Stratospheric Ozone in the Polar Regions

3.1. Ozone changes in the Antarctic

Mean annual patterns of the vertical ozone distribution and their long-term changes are shown for the antarctic stations Syowa, Georg Forster and Neumayer located near 70°S for a period of almost 30 years from 1967 to 1995 (Fig. 1). Before 1980, stratospheric ozone shows the normal maximum in spring, i.e. in October and November when the stratospheric vortex breaks down. During this time ozone is advected into the polar stratosphere mainly at greater altitudes above 20 km. From mid-summer until fall ozone decreases. Lowest ozone concentrations occur in the stratosphere in late summer just before the new formation of the winter stratospheric vortex (Fig. 1, upper panel). Later the maximum of ozone concentration changed to a pronounced minimum in the lower stratosphere. The decline begins at altitudes around 20 km in late August. Lowest values appear in October. The recovery begins at altitudes above 20 km. Low ozone is recorded below 16 km until December (Fig. 1 central and lower panel). The loss rate in September and the final total loss in October are strongest for the period 1992 to 1995. Low ozone is recorded below 16 km until December (Fig. 1, lower panel).

The development of the antarctic ozone depletion in spring is shown for three specified periods in more detail in Fig. 2. The altitude range for more than 50% loss in ozone concentration extended from a few kilometers to about 10 km in the early eighties. The largest vertical extent occurs in October. Simultaneously with the appearance of

ozone depletion in the lower stratosphere, air masses enclosed in the polar vortex subside due to diabatic cooling. So ozone is displaced from the middle stratosphere above 20 km downward. Later, in late October and November until the final breakdown of the vortex in late spring, ozone is additionally transported toward high latitudes.

In parallel with the intensification of spring depletion, significant ozone decrease is also seen in the lowest part of the polar stratosphere between approximately 8 km and 16 km in summer and even in winter.

In the middle stratosphere at altitudes above 26 km, another decrease of ozone can be identified from February until October for the period 1992 to 1995 (Fig. 1, lower panel) with respect to the mean pattern for Georg Forster and Syowa in Fig. 1. This ozone decline is smaller. It is significant by about -30% in October (Fig. 2, lower panel). It appears when the formation of the stratospheric vortex begins in fall, and it suddenly disappears when the stratospheric vortex breaks down at these altitudes at the end of October.

But these striking changes of stratospheric ozone recorded with respect to Syowa before 1980 are mainly confined to the lower polar stratosphere. The strongest change happened until the late eighties. The spring ozone minimum disappears in connection with the dynamical processes during the reversal to summer circulation. Obviously this dynamically controlled recovery did not change significantly for the periods considered here.

3.2. Chemical ozone loss in the lower polar stratosphere

In winter the stratosphere radiatively cools and temperatures may drop down to -90°C at altitudes between 18 and 22 km (KAYE, 1993; WMO, 1995). When temperatures are colder than -78°C at altitudes around 20 km, polar stratospheric cloud (PSC) particles consisting of nitric acid and water, or a ternary composition of sulphur and nitric acid with water, are formed. At temperatures below thresholds between -83°C and -88°C water ice forms another type of PSC particles (TABAZADEH *et al.*, 1994).

At the surface of such particles gaseous hydrochloric acid (HOCl) and chlorine gas (Cl_2) are formed from the reservoir gases ClONO_2 and HCl by heterogeneous reactions (WEBSTER *et al.*, 1993). When sunlight returns to the antarctic stratosphere, the molecular chlorine Cl_2 and HOCl are photolyzed to chlorine atoms, which react with ozone. Due to the high concentrations of Cl, the reaction with ozone yields a hundred fold elevated concentration of ClO compared to the global mean. If sufficient chlorine monoxide is present it can react with itself to form the dimer Cl_2O_2 (SOLOMON, 1990). This dimer photolyzes into chlorine and molecular oxygen. So the chlorine is again free to react with ozone. So the formation of the dimer at elevated ClO concentration forces the chemical ozone loss by catalytic reactions (RÖTH, 1994; KAYE, 1993). The chemical ozone loss stops if reactive chlorine reacts with NO_x to form the reservoir gases ClONO_2 and HCl again. If nitric acid were removed from the lower stratosphere by sedimentation of PSC particles the formation of reservoir gases would only occur when air masses with sufficient NO_x concentration are mixed into the polar stratosphere during the vortex breakdown. That explains the long duration of ozone depletion until the reversal of stratospheric circulation in the Antarctic.

The chemical ozone loss within the spring season is a consequence of certain me-

teorological conditions which force PSC-related chemical processes. This chemistry became more efficient due to the continuous increase of man-made source gases (SOLOMON, 1990; ENGEL *et al.*, 1995) which have yielded to higher concentrations of the reservoir gases ClONO_2 and HCl in the polar stratosphere since 1979 (WMO, 1992, 1995). So the net ozone loss in the lower stratosphere depends on the surface density of PSC particles, the duration of their existence, the elevated concentrations of reservoir gases and the temperature history within the stratospheric vortex. For a given composition of these parameters the net ozone loss within an air mass is then directly proportional to the duration of sunlit exposure (WEBSTER *et al.*, 1993).

3.3. Arctic ozone distributions

The mean annual variation of ozone concentration in the Arctic is shown in Fig. 3 (upper panel) for the period from 1988 to 1995. In winter and spring a maximum of stratospheric ozone concentration is observed similar to that in the Antarctic before 1980 (Fig. 1, upper panel). Lowest ozone values appear in late summer. The recent mean pattern for the period from January 1992 to April 1995 is shown in Fig. 3 (central panel). In comparison to the mean for the whole period small changes can be noticed at altitudes around 20 km where ozone concentration decreased by about 1×10^{12} molecules per cm^3 in August.

The ratio (Fig. 3, lower panel) between the annual means 1992/95 versus 1988/91 reveals less ozone by about 30% at altitudes above 26 km in winter from December until March, and at altitudes below 20 km from late winter until mid-summer. The ozone decrease in the middle stratosphere is similar to that in the Antarctic in late winter and spring (Fig. 2, lower panel).

In the lower arctic stratosphere the ozone decrease is less than in the Antarctic in the corresponding season, *i.e.* from late winter until summer. The stronger planetary wave activity causes perturbations of the stratospheric vortex circulation which lead to considerable variations of ozone in the lower arctic stratosphere described as "ozone mini-hole" events (ORSOLINI *et al.*, 1994).

But chemical ozone loss based on the same PSC related processes can also occur within the arctic stratospheric vortex where temperatures are also low enough to form PSC's (NEUBER *et al.*, 1994). The superimposition of both dynamical variation and chemical loss of ozone probably being of the same magnitude makes it more difficult to recognize chemical loss in the arctic stratosphere. It is impossible to separate both processes by ozone observations from one location.

First approaches to identify chemical ozone loss in the arctic stratosphere have been indirect. Based on satellite observations MANNEY *et al.* (1994) discussed the relationship between ozone and long-lived tracers. They concluded that the observed ozone decrease of about 20% was caused by chemical, rather than dynamic, processes in the lower stratosphere in February and March 1993. VON DER GATHEN *et al.* (1995a) reported on results obtained by calculating the chemical ozone destruction rates from ozone sonde data.

During the winter 1991/92, about 1.200 ozone sondes (ECC) were launched from 26 launching sites as part of the European Arctic Stratospheric Ozone Experiment (EASOE) and from Canadian stations at middle and high northern latitudes. In order to identify cases where ozone measurements were made at two different times in the same

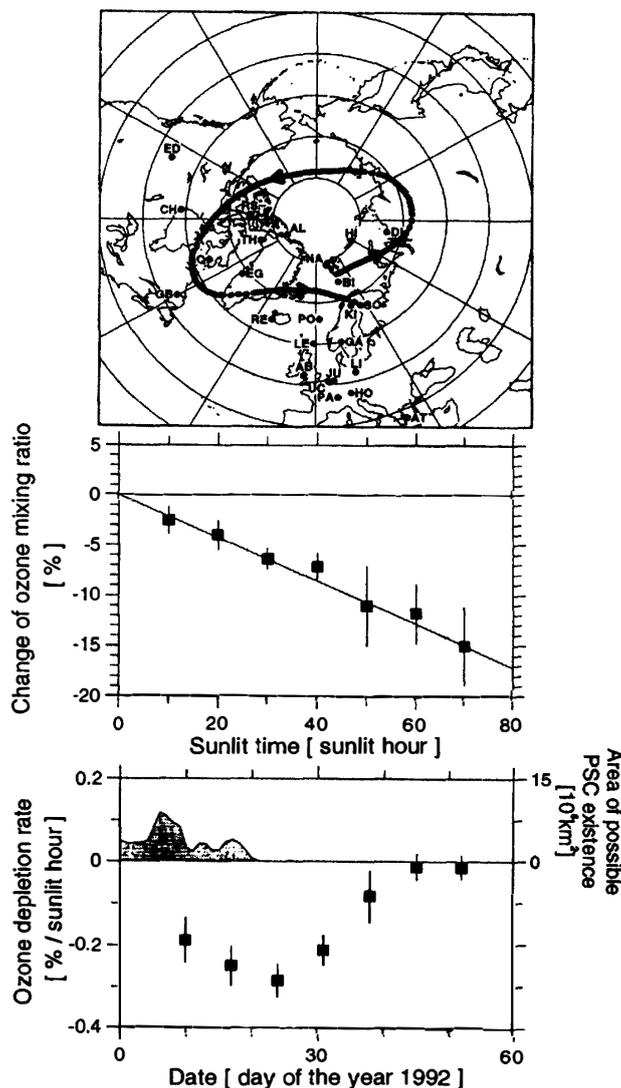


Fig. 4. Upper panel (reprinted with permission from *Nature* (VON DER GATHEN et al., 1995a): Pattern of an isentropic trajectory inside the polar vortex at the 475 K level. Dashed trajectory parts indicate sunlit exposure.

Central panel (reprinted with permission from *Nature* (VON DER GATHEN et al., 1995a): Running means over 20 sunlit hours (full squares) for percentage change in ozone mixing ratio (%) as a function of sunlit hours.

Lower panel: Running mean (full squares) over 14 days for the ozone depletion rates (% per sunlit hour, left scale); area of possible PSC formation $\times 10^6$ km² (right scale) as function of time; time scale (Julian day for 1992).

air mass, 10-day isentropic air parcel trajectories were calculated from the wind and temperature analyses of the European Centre for Medium Range Weather Forecasts (ECMWF). An example for the observation of ozone change along an isentropic trajectory for the 475 K level is shown in the upper panel of Fig. 4. In this case the ozone

sonde measurements from Ny-Ålesund and Sodankylae are matched by the trajectory, and the change of ozone mixing ratio is related to the same air parcel. That part of the trajectory, when the air parcel was exposed to sunlight, is indicated by the dashed line. For the Arctic vortex in the 1991/92 winter, VON DER GATHEN *et al.* (1995a) and REX *et al.* (1995) found that percentage changes in ozone mixing ratios are proportional to the exposure to sunlight along trajectories within the arctic stratospheric vortex (Fig. 4, central panel). The vortex edge is defined by the quantity of potential vorticity (PV), which is $PV > 27 \text{ Km}^2\text{kg}^{-1}\text{s}^{-1}$ for 475 K. Between 18 to 27 trajectory matches inside the vortex contribute within ± 10 sunlit hours to the first four data points, and between three and five matches contribute to the last three data points. The fit is shown by the solid line. The largest total ozone loss is found for matches which received the most sunlight. This correlation between decrease of ozone mixing ratio and sunlit exposure is crucial evidence that chemical ozone loss has been measured. It is consistent with the existing understanding of photochemical ozone depletion processes in the polar stratosphere in late winter and spring. Fig. 4 (lower panel) shows the time evolution of the ozone depletion rate (% per sunlit hour) during January and February 1992 inside the polar vortex at the 475 K potential temperature level (about 18 km). In addition, the area of the Northern Hemisphere where temperatures were cold enough to form PSC's, assuming NAT formation for constant volume mixing ratios of $\text{H}_2\text{O}=5 \text{ ppm}$ and $\text{HNO}_3=5 \text{ ppb}$, is plotted (REX *et al.*, 1995). There was found a mean ozone loss related to the chemical ozone depletion of about $0.23 \pm 0.03\%$ per sunlit hour for the period January 4 to February 9, 1992.

A more sophisticated approach was performed during the Second European Stratospheric and Mid-latitude Experiment in the 1994/95 winter. The coordination of ozone soundings and trajectory calculations was performed in the MATCH campaign (REX *et al.*, 1995). About 35 ozone sonde launching stations have been participating as the European ozone launching sites in mid and high latitudes as well as Canadian, Siberian and Japanese stations. Altogether about 1.000 ozone sondes were launched. These launches were on-line coordinated by using forward trajectories in order to achieve the matches. First preliminary results showed a similar mean ozone loss attributed to chemistry as was obtained from the winter 1991/92. The mean ozone loss rate amounted to $0.23 \pm 0.045\%$ per sunlit hour for the period from January 1 to February 1 (REX *et al.*, 1995).

In summary these results established chemical ozone loss in the arctic stratospheric vortex for short periods lasting about 10 to 20 days in late winter. It occurred only during sunlit periods along the trajectories, and it rose during periods when the probability for PSC existence was indicated by the related stratospheric temperatures. During the whole winter about 20% to 30% of total ozone loss can be attributed to catalytic ozone destruction in the lower stratosphere (VON DER GATHEN *et al.*, 1995a; REX *et al.*, 1995). So the changes of the vertical ozone distribution in the mean annual pattern (Fig. 3, lower panel) for the period 1992 to 1995 might be concerned with both the chemical ozone loss within the stratospheric vortex and the mesoscale dynamical variations causing mini-hole events of low ozone concentration in the lower stratosphere (ORSOLINI *et al.*, 1994).

3.4. Recent vertical ozone distributions in the Arctic and Antarctic during spring

Different mean annual patterns of ozone concentration are established in the stratosphere and troposphere of both polar regions (*cf.* Fig. 1 (lower panel) and Fig. 3 (central panel)). While nowadays a minimum appears in the Antarctic in late winter and spring, a maximum of stratospheric ozone is recorded in the Arctic for the corresponding season. However, it is interesting to notice, that the mean annual pattern recorded at Syowa station before 1979 (Fig. 1–upper panel) also shows an ozone maximum in spring similar to that observed in the Arctic in recent years. In summer the ozone minimum in the arctic stratosphere is more pronounced than the antarctic one. In the Antarctic at 70°S the chemical ozone loss occurs when sunlight returns after the polar night at the end of July (Fig. 1). In the Arctic at 79°N air masses with chemically depleted ozone even occur in the middle of January although sunlight returns later at the end of February. This early onset of ozone depletion is due to the stronger planetary wave activity pushing air parcels toward lower latitudes where sunlight can force the chemical removal of ozone.

The PSC related chemical processes within the vortex explain the strong ozone losses in the lower polar stratosphere. However, the decreasing ozone concentrations recently recorded with respect to the Syowa reference at greater altitudes above 26 km mainly in October and November (Fig. 2, lower panel) and for Ny-Ålesund above 26 km from December to March cannot be explained by the meteorological and chemical framework discussed for the lower stratosphere. At these altitudes the formation of PSC's is not likely because temperatures are higher even during the polar night. The simultaneous appearance of lower ozone content in the middle stratosphere of both polar regions for winter and spring suggests that probably less ozone is recently transported from the equator to the Arctic (VON DER GATHEN *et al.*, 1995b) and also to the Antarctic. Possible reasons are not studied in detail. The simultaneous decrease might be caused by some changes of the global dynamics within the winter hemisphere. But on the other hand ozone decrease by gas-phase chemical reactions in the upper stratosphere at lower latitudes might result in less ozone transported toward polar latitudes when the stratospheric vortex develops in fall.

The actual differences of the ozone depletion in the lower stratosphere between the Antarctic for September 1994 (Neumayer station) and the Arctic for March 1995 (Ny-Ålesund station) are shown in Fig. 5. Two ozone profiles observed in the Antarctic on September 13, 1994 and September 24, 1994 are compared with the mean profile for the polar night period from May, 20 to July, 20 in 1994. Corresponding ozone profiles observed in the Arctic on March 14, 1995 and March 23, 1995 are compared with the mean profile for the polar night period from November, 1 to January, 31 in 1995. The comparison for corresponding seasons, *i.e.* the spring months March and September, clearly shows the stronger ozone depletion in the South. Significant deviations of ozone from the winter mean profiles occurred between 380 K (14 km) and 500 K (20 km) in the Arctic, and between 350 K (13 km) and 620 K (24 km) in the Antarctic. Ozone continued to decrease in the Antarctic from September, 13 to September, 24, while in the Arctic ozone increased from March 14 to March 24. The greater vertical extent of the ozone loss region by about 4 km in the Antarctic might be explained by the greater vertical extent of very low temperatures in the antarctic stratospheric vortex, by smaller exchange

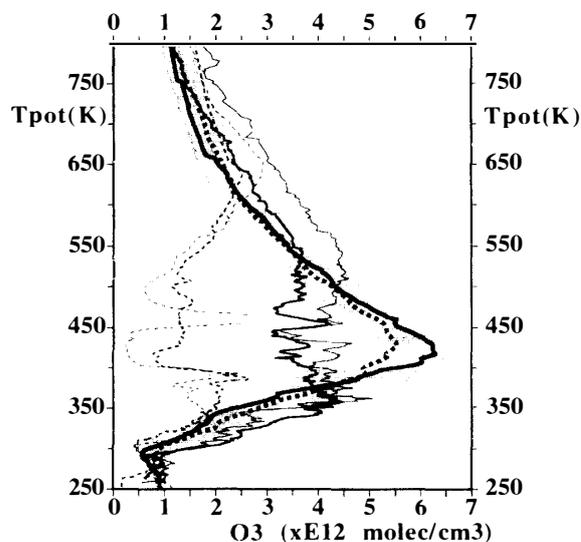


Fig. 5. Antarctic and arctic vertical distributions of ozone concentration ($\times 10^{12}$ molec/cm³). Individual profiles for corresponding months in spring: Antarctica on September 13 (medium dashed line) and 24 (thin dashed line) in 1995 and Arctic on March 14 (medium full line) and March 24 (thin full line) in 1995. Mean profiles with standard deviation for corresponding winter seasons: Ny-Ålesund (bold thick line), Neumayer (bold dashed line). Winter season for Ny-Ålesund station (November 1, 1994 to January 31, 1995), and for Neumayer station (May 20 to July 20, 1995). Altitude coordinate is potential temperature T_{pot} (K).

of air through the vortex margin, and, probably to some extent, also by the smaller vertical downward component of the residual circulation in the Antarctic compared to the Arctic (GELLER *et al.*, 1992).

Lowest ozone concentrations occur in the altitude range between 400 K and 450 K (14 km–16 km). With regard to the ozone concentrations for polar night conditions the total ozone loss at the 420 K level, *i.e.* chemical loss and dynamical variation, amounts to about –87% at 79°S and –39% at 79°N, respectively. The ozone loss rate per sunlit hour for the corresponding latitude is as high as –0.11% at 79°N and –0.15% at 71°S. These values are smaller than those obtained for the actual chemical ozone loss in the arctic stratosphere along the trajectories of air masses (Fig. 4). Maxima of ozone concentration within thin layers indicate small scale advection across the vortex margin below the 500 K level for both polar regions. This dynamic noise may also cover the retrieval of the actual chemical ozone loss from one location.

4. Natural Perturbations of Stratospheric Aerosols and Ozone

4.1. Volcanic aerosols in the polar stratosphere

Natural events such as volcanic eruptions may also modulate the ozone chemistry in the polar stratosphere. According to McCORMICK *et al.* (1995) volcanic eruptions can produce a significant perturbation to the atmosphere, which may persist for several years. The injected materials may include ash, which typically does not remain for more than a few months in the atmosphere, and gaseous compounds including water vapor, sulphur dioxide and hydrochloric acid. Aerosols are produced when the sulphur dioxide (SO₂) is chemically transformed into sulphuric acid (H₂SO₄) which rapidly condenses into aerosol droplets. This aerosol of volcanic origin increases the atmospheric albedo by reflecting solar radiation back into space, can warm the stratosphere by absorbing upwelling infrared radiation (KARL and BAKER, 1995) and may affect the stratospheric ozone content.

The eruptions of Mt. Pinatubo in June 1991 and Cerro Hudson in August 1991 caused large perturbations within the stratosphere. The volcanic cloud has been globally moni-

tored by the satellite-borne Stratospheric Aerosol Monitoring experiment (SAM II) as well as by the Stratospheric Aerosol and Gas Experiment (SAGE II) (TREPTE *et al.*, 1993, 1994; McCORMICK *et al.*, 1995). The Mt. Pinatubo aerosol cloud immediately dispersed throughout the tropics between 30°N and 20°S. In the arctic lower stratosphere first aerosols occurred about 4 months later from December 1991 to April 1992. This northward transport was forced by planetary waves in the zonal westerlies at mid-latitudes (TREPTE *et al.*, 1993; HERBER *et al.*, 1996). It was primarily responsible for the initial appearance of aerosols in the lower stratosphere of high arctic latitudes as reported from lidar observations (NEUBER *et al.*, 1994). In the southern hemisphere the initial aerosol arriving in the polar stratosphere was formed by the Cerro Hudson eruption (McCORMICK *et al.*, 1995; HERBER *et al.*, 1993). Stratospheric aerosol from Cerro Hudson was concentrated at low altitudes below 16 km (SCHOEBERL and DOIRON, 1993). The transport of this aerosol through the prevailing zonal westerly wind was also forced by planetary waves in the lower stratosphere during the southern hemispheric winter and spring in 1991 (HERBER *et al.*, 1996). So volcanic aerosols penetrated deeply into high southern latitudes below the base of the stratospheric vortex soon after that eruption. During the southern winter and spring in 1992 the Mt. Pinatubo aerosols also occurred in the southern polar stratosphere (McCORMICK *et al.*, 1995).

In polar latitudes the appearance of volcanic clouds has also been monitored by ground-based sun photometers (KANETO *et al.*, 1994), lidars (NEUBER *et al.*, 1994; BEYERLE *et al.*, 1995), and balloon-borne aerosol sondes (HOFMANN and OLTMANN, 1993; HOFMANN *et al.*, 1994). Regular ground-based sun photometer measurements performed at three

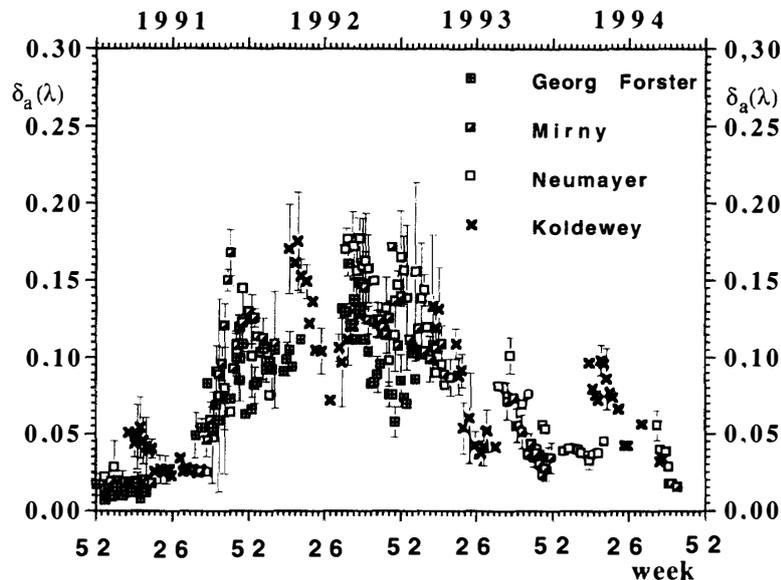


Fig. 6. Antarctic and arctic sun photometer observations of the total aerosol optical depth at 1000 nm from 1991 to 1994: weekly means of the total aerosol optical depth; Antarctic: Georg Forster (crossed squares), Mirny (half full squares), Neumayer (open squares); Arctic: Koldewey/Ny-Ålesund (crosses); years and months are indicated in the upper scale, and number of week per year in the lower scale.

antarctic stations and one arctic station (Table 1) have completed these observations (HERBER *et al.*, 1996). Weekly means of the retrieved aerosol optical depths for 1000 nm recorded at the antarctic stations Georg Forster, Mirny, and Neumayer, and the arctic station Koldewey in Ny-Ålesund are compiled in Fig. 6.

Coinciding with the dispersion of the volcanic aerosols, an increase of the optical depth at 1000 nm is seen in both polar regions. The increase at Mirny, Georg Forster, and Neumayer in late September 1991 confirms the arrival of Cerro Hudson aerosol particles above the Antarctic. In mid-November 1991 a second increase of the aerosol optical depth was recorded after the vortex breakdown. At this time, the aerosol optical depth at 10000 nm was approximately 0.12, *i.e.* 10 times as high as the background level. This second increase is attributed to the Mt. Pinatubo aerosols. A similar perturbation of the aerosol optical depth was recorded in the Arctic after the return of sunlight in March 1992, because sun photometer measurements need sufficient sunlight. However, the increased loading of the Arctic stratosphere by Mt. Pinatubo aerosol particles was already evident during the polar night (NEUBER *et al.*, 1994). Maximum optical depths appeared in 1992 and the beginning of 1993. In 1994 the aerosol optical depth of the atmosphere dropped down almost to the pre-volcanic level in both polar regions. The polar latitudes are particularly interesting because they are the last region to “clear out” following a major injection of volcanic material into the stratosphere (McCORMICK *et al.*, 1995).

The greater values of the optical depth in arctic spring 1993 as well as in 1991 are associated with the tropospheric Arctic Haze phenomenon (HERBER *et al.*, 1995).

4.2. Volcanic aerosols and ozone in polar regions

In mid-latitudes, a reduction of ozone in response to volcanic aerosol loading in the stratosphere was reported from lidar observations above the Observatoire de Haute Provence in Southern France (43.9°N, 5.7°E). The change in the ozone profile was correlated with the presence of volcanic aerosols in the lower stratosphere down to tropopause altitudes. At some altitudes this reduction of ozone approached about -40%, yielding a 10% reduction in the total ozone column (McGEE *et al.*, 1994).

In southern polar latitudes additional ozone loss was found in association with aerosols formed after the eruption of Cerro Hudson in August 1991 (DESHLER *et al.*, 1992). This aerosol was concentrated at altitudes below 14 km (350 K to 400 K). It appeared at southern polar latitudes in late September 1991. In the densest portions of the Cerro Hudson aerosol layer between 11 and 13 km, ozone decreased by about 50% for a period of 30 days following the arrival of this aerosol above South Pole station in Antarctica. Such low ozone concentrations below the base of the vortex have not been observed in the Antarctic before 1991. The aerosol surface-area density in this layer was as high as 100 mm²/cm³, *i.e.* 20–30 times higher than the background observed in 1990. Aerosol levels also remained unusually high for the following two years (HOFMANN *et al.*, 1994).

Ozone sounding data from the stations Syowa, Georg Forster, Neumayer, and Ny-Ålesund confirm such similar ozone losses caused by volcanic aerosols in the lowest part of the polar stratosphere for both polar regions in spring. Monthly means of the ozone mixing ratios were calculated for the 350 K isentropic level, *i.e.* about 12 to 13 km altitude. From antarctic data monthly means were obtained for October 1979 and

from 1982 to 1994. These values are compared with monthly means for March obtained from Ny-Ålesund ozone sounding data available since 1989. For spring these months were selected because they are then most ozone soundings have been performed (Fig. 7, lower panel). Except for the years 1991, 1992, and 1993 when volcanic aerosols were recorded in the southern polar stratosphere (McCORMICK *et al.*, 1995), the linear regressions were calculated for Syowa data for the period 1979 to 1994 and Ny-Ålesund data for the period 1989 to 1994. Both regressions are similar, with a negative trend of -0.50% per decade for Syowa ($r^2=0.896$), and a similar negative trend of -0.48% per decade for Ny-Ålesund ($r^2=0.701$) although the arctic record is short. With respect to these interannual negative trends the monthly mean of ozone mixing ratio is smaller by about one standard deviation for October 1991, and by more than one standard deviation for October 1992 and 1993 in the Antarctic. In the Arctic the monthly mean is smaller by

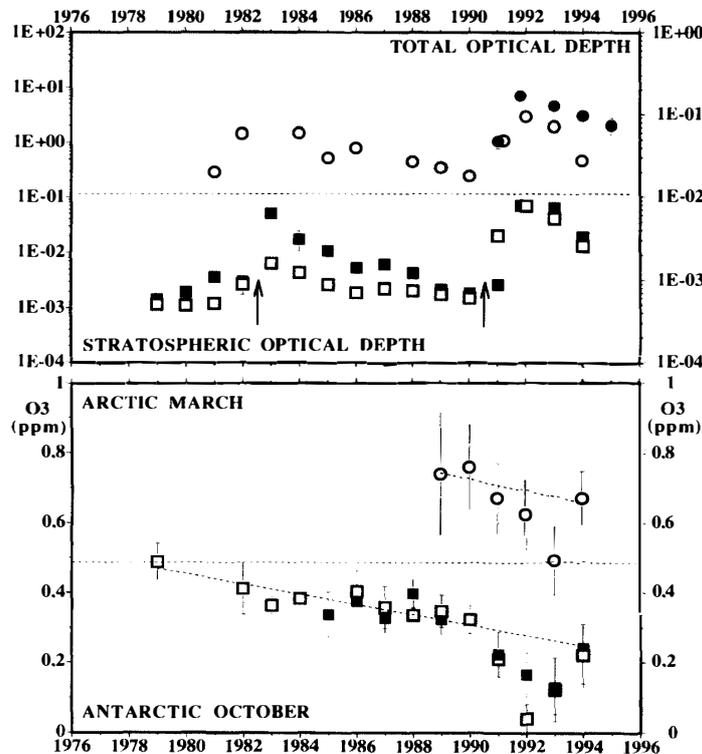


Fig. 7. Upper panel: monthly means of total aerosol optical depths by ground-based actinometers and sun photometers (right scale): Antarctic for October (open dots), Arctic for March (full dots); zonal monthly means of stratospheric aerosol optical depths by SAM II and SAGE II (left scale): Antarctic for October (open squares) Arctic for March (full squares). The eruptions of El Chichon and Mt. Pinatubo/Cerro Hudson are indicated by arrows.

Lower panel: monthly mean ozone mixing ratios (ppmm) at 350 K (12–13 km); Antarctic for October: Syowa Station (open squares), Georg Forster station from 1985 to 1991 and Neumayer station from 1992 to 1994 (full squares); Arctic for March: Ny-Ålesund station (open dots). Standard deviation is shown for all mean values. It is smaller than dots and squares for optical depth values.

more than one standard deviation only for March 1993.

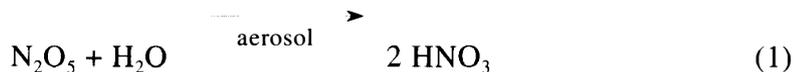
Monthly means of stratospheric aerosol optical depth and total aerosol optical depth for 1000 nm are shown for the corresponding months where ozone data are presented (Fig. 7, upper panel). In the Antarctic, sun photometers were operated at Georg Forster station from 1988 to 1992, and at Neumayer station since 1991. These records of the total aerosol optical depth of the atmosphere have been extended back to 1981 by including actinometric observations from Mirny station (Table 1). In the Arctic, sun photometers have been operated since 1991. Due to the contributions by tropospheric aerosols, the values of the total aerosol optical depth of the atmosphere measured by ground-based sun photometers are approximately 10 times greater than the corresponding stratospheric values (HERBER *et al.*, 1993). The interannual variation of the stratospheric aerosol optical depth retrieved from SAM II and SAGE II satellite data is shown for October for the Antarctic and March for the Arctic since 1979 (Fig. 7, upper panel).

As suggested by McCORMICK *et al.* (1995), the 1979 stratospheric aerosol optical depth values in 1979 probably show the stratospheric background in both polar regions. Later, elevations of stratospheric aerosol by volcanic eruptions from 1980 to 1981, and the stronger eruption of El Chichon in April 1982, yielded an increase of stratospheric aerosol optical depth. The recovery almost lasted until 1988. In general the total atmospheric aerosol optical depths follow the pattern of the stratospheric values. The greater monthly mean values of total aerosol optical depth in the Arctic for March 1991 to 1995 are mainly caused by the regular maximum of the tropospheric aerosol concentrations during arctic spring (HERBER *et al.*, 1995). Maximum stratospheric aerosol optical depths occurred after the eruptions of El Chichon one year later in 1983 as well as after the eruptions of Cerro Hudson and Mt. Pinatubo two years later in 1993. The early arrival of the Cerro Hudson aerosols in the antarctic lower stratosphere is seen from the elevated aerosol optical depth for October 1991. While the stratospheric aerosol optical depth has been almost equal in both polar regions since the volcanic eruptions in 1991, a significant asymmetry with a greater aerosol optical depth for the arctic stratosphere was recorded in March 1983 with respect to the October value in the Antarctic.

In the lower stratosphere at about 12 to 13 km altitude a significant negative correlation between the stratospheric aerosol optical depth and the monthly mean ozone mixing ratios for the 350 K isentropic level was recorded in both polar regions after the eruptions of Mt. Pinatubo and Cerro Hudson. In the Antarctic, the arrival of Cerro Hudson aerosol in late September 1991 yields a small decrease of the mean ozone mixing ratio in October 1991. The ozone decline is stronger in 1992, but different in magnitude for the stations Syowa and Neumayer. That might be explained by stronger longitudinal gradients in the aerosol distribution. In 1993 a strong ozone decline is evident for both polar regions during spring. In spring 1994 the ozone values recovered with respect to the long-term negative trends, although the stratospheric aerosol optical depth values are still elevated but already below the level for October 1991. As these additional ozone losses in the lower polar stratosphere for October 1991, 1992, and 1993 can be attributed to the presence of volcanic aerosol particles at these altitudes, the ozone reducing processes probably depend on a critical level of particle surface area density. This assumption is supported because significant ozone losses could not be observed in the antarctic lower stratosphere after the eruption of El Chichon in April 1982 when the

stratospheric optical depth became significantly smaller. On the other hand McCORMICK *et al.* (1995) argued that these ozone losses depend not only on the particulate surface density of the volcanic aerosols but also on the elevated chlorine and bromine levels in the stratosphere as a result of CFC emissions. Following that, there is a striking example of a changing coupling between human and natural perturbations. The delay by one year for the maximum ozone loss in the arctic and antarctic lower stratosphere with respect to the record of maximum stratospheric aerosol optical depth is another striking feature not yet understood completely.

Two dimensional model simulations for 60°N (TIE *et al.*, 1994) show that the maximum of ozone depletion occurs at about 15 km altitude in March, *i.e.* in spring. The observations at Ny-Ålesund in March 1993 confirm this model result (Fig. 7, lower panel). It is suggested that the ozone loss might be the consequence of heterogeneous reactions on the surface of volcanic aerosols. Laboratory experiments have shown that nitrogen oxides, N_2O_5 , and $ClONO_2$ react on the surface of sulphuric acid solutions the composition of which is similar to stratospheric aerosol particles (FAHEY *et al.*, 1993).



For stratospheric water vapor partial pressures and temperatures these reactions might be important in the lower stratosphere when the photolysis of HNO_3 is suppressed due to low solar elevations at higher latitudes in spring. In that case the low photolysis rate of HNO_3 back to NO_2 decelerates the formation of $ClONO_2$. Thus, concentrations of chlorine species remain high and can additionally remove ozone at altitudes where it is too warm for PSC's to occur (McCORMICK *et al.*, 1995).

As is seen in Fig. 7 (lower panel) the maximum ozone loss for spring in 1993 amounts to about -0.2 ppmm in the Arctic but only -0.14 ppmm in the Antarctic with respect to the long-term trends. The maximum solar elevation is about 10° at $80^\circ N$ in March. But it is about 25° at $70^\circ S$ in October. At the 350 K level the mean sunlight exposure is about 13 hr/day at $80^\circ N$ in March and about 17 hr/day in October at $70^\circ S$. These differences in solar elevation and sunlit exposure suggest a smaller photolysis of HNO_3 for Ny-Ålesund in the Arctic. According to McCORMICK *et al.* (1995) activated chlorine species then might remove more ozone in March at $80^\circ N$ than in October at $70^\circ S$, even if all other conditions would be equal for both polar regions.

5. Aerosols and Ozone in the Arctic Troposphere

5.1. Anthropogenic aerosol and ozone in the arctic troposphere

The mean annual variation of ozone concentration is significantly different between the Arctic and Antarctic (Figs. 1, 3). In the Antarctic, tropospheric ozone decreases after the polar night, and it remains low during sunlit conditions until fall. Low ozone concentrations first appear in the upper troposphere in October when the stratospheric ozone minimum is well pronounced. In contrast to the Antarctic, ozone concentrations are elevated in the lower arctic troposphere during spring.

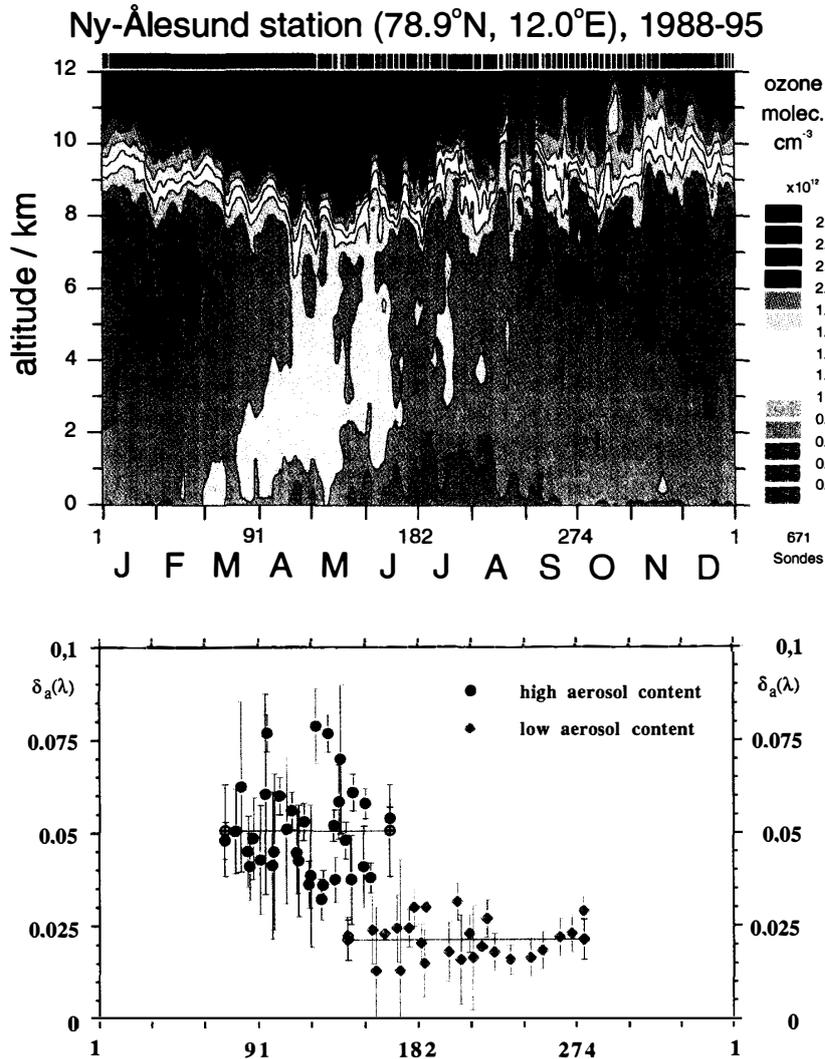


Fig. 8. Mean annual variation of tropospheric ozone and tropospheric aerosol optical depth at Ny-Ålesund station.

Upper panel: height-time cross section of ozone concentration (color coded $\times 10^{12}$ molec./ cm^3), mean annual variation for the period 1988 to 1995 by 671 ozone soundings; altitude range (0–12 km); time scale one year (Julian day, month).

Lower panel: Weekly means of the tropospheric aerosol optical depth for the period from 1991 to 1995; time scale (Julian day). Seasonal means for spring and summer are indicated by horizontal black lines.

In Fig. 8 a detailed plot of the mean annual variation of ozone concentration is shown for the arctic troposphere as it was observed at Ny-Ålesund for the period 1988 to 1995. At tropopause altitudes a downward displacement of stratospheric ozone is seen in late winter and spring. This downward stretch is evident down to about 7 km altitude. In fall ozone first decreases in the upper troposphere toward winter. This mean annual pattern is consistent with results obtained by LOGAN (1985) which also show that ozone concentrations in the arctic middle troposphere exhibit a maximum between March and May below the tropopause. This can be explained, in part, by increased transport of

stratospheric ozone into the upper troposphere during the reversal of circulation from winter to summer. Estimations of ozone fluxes from the stratosphere into the troposphere were made by GRUDZEV and SITNOV (1992). These results show that the tropospheric ozone content is controlled by stratospheric ozone down to the middle troposphere in polar regions.

In the lower troposphere ozone concentration increases after sunrise at 79°N on the end of February (Fig. 8, upper panel). First the ozone increases in the boundary layer. Later, with increasing solar elevation, the higher tropospheric ozone concentrations extend upward by June, while in the boundary layer ozone concentration again become smaller by July. This maximum of ozone concentration in the lower troposphere is partially attributed to the influence of air masses from mid-latitudes, assuming a tropospheric source, to form additional ozone (LOGAN, 1985).

Besides the tropospheric ozone, the arctic tropospheric aerosol composition is also affected by the transport of polluted air masses toward high northern latitudes. These aerosols consist of sea salt particles and sulphate aerosols, the latter mainly of anthropogenic origin, and are transported from mid-latitudes toward the arctic region mainly in spring, discussed as the "Arctic Haze" phenomenon (HEINTZENBERG, 1989). These aerosols elevate the optical depth of the troposphere, and cause a pronounced seasonal variation. By calculating the stratospheric optical depth from the SAGE II satellite data and removing this value from the total optical depth recorded with the sun photometer at the ground, the obtained residual value is the tropospheric optical depth (HERBER *et al.*, 1995). The mean seasonal pattern of the tropospheric optical depth for spring, summer, and early fall is shown in Fig. 8 (lower panel). Weekly means of the tropospheric optical depth were retrieved from ground-based sun photometer measurements and satellite observations for the period from 1991 to 1995. Despite the large scatter of the weekly mean values and their standard deviations the tropospheric optical depth is significantly higher in spring than in late summer when smallest values and smallest standard deviations occur.

Both the greater ozone concentrations in the lower troposphere and the increased tropospheric optical depths coincide very well in their temporal behavior. As discussed by LOGAN (1985) and HEINTZENBERG (1989), the formation of tropospheric ozone as well the appearance of elevated aerosol concentrations are connected with the transport of tropospheric air masses from mid-latitudes into the Arctic. Considering this, an interaction might be possible. The aerosol mainly resides in the boundary layer. Higher ozone concentrations appear above. The extinction coefficient of tropospheric aerosol with anthropogenic species consists of a scattering and an absorbing component. The latter one is due to the soot content of anthropogenic aerosols. The scattering of solar radiation in the short wave range by aerosols increases the backward radiative flux which might force photochemical reactions, and subsequently, more ozone might be produced in the lower and middle troposphere.

A rough estimate of possible change for the upward directed radiative flux was made for the 5 km level by using the radiative transfer model LOWTRAN 7 (KNEIZYS *et al.*, 1988). This model is considered for molecular line absorption, molecular continuum absorption, molecular scattering, aerosol extinction, cloud and rain droplet extinction, and multiple scattering. The radiances for the wavelength range 280 nm to 350 nm of the

Table 3. Estimated optical properties for Arctic Haze aerosols.

Month	Extinction coefficient	Absorption coefficient	Scattering coefficient
March	0.051 ± 0.012	8.98×10^{-3}	6.69×10^{-2}
July	0.021 ± 0.005	6.50×10^{-4}	2.51×10^{-2}

direct and scattered solar radiation were calculated with the aerosol parameters compiled in Table 3. The aerosol extinction values were retrieved from sun photometer measurements. The absorbing properties were estimated in order to obtain a reasonable fit for arctic aerosols (HERBER *et al.*, 1994). Following the model results the scattering on aerosols in the boundary layer increases the upward directed UV radiative flux by about 11% at 5 km altitude for low surface albedo ($A=0.1$) in March. For high surface albedo ($A=0.8$) the net upward directed UV radiative flux is decreased by -3% in March. For July with decreased aerosol content the upward directed radiative flux does not change significantly for both albedo cases. The surface albedo is decreasing at Ny-Ålesund from April to June when the aerosol optical depth remains high. So an increased upward directed radiative flux up to 11% might affect photochemical reactions in the troposphere.

5.2. Ozone effects in the boundary layer

By going down to the boundary layer another interesting feature of ozone variability in the arctic region can be shown. In spring surface ozone concentrations sometimes drop from normal values of about 40 ppbv down to the detection limit for periods of several hours to a few days. This phenomenon was first reported by BARRIE *et al.* (1988) and OLTMANS *et al.* (1989) for the arctic stations Alert (Canada) and Barrow (Alaska). Up to now the reasons are not yet completely understood. Therefore, during the spring seasons in 1994 and 1995 special campaigns were performed at Ny-Ålesund station in order to observe such events in more detail (WESSEL *et al.*, 1996). Measurements of surface ozone, balloon-borne ozone profiles, and particle number densities for selected size modes of surface aerosols have been performed. In Fig. 9 the variation of the ozone mixing ratio (ppbv) in the lower troposphere up to 4 km altitude is shown for the period from March to June in 1994 (upper panel) and in 1995 (lower panel). These plots clearly show four events of strong ozone depletion which are limited to an altitude of about 800 m. The upper boundary of depleted ozone coincides with pronounced temperature inversions. These ozone minima within the boundary layer are typically associated with marine arctic flow conditions in the lower troposphere as can be concluded from trajectory calculations and analysis of weather charts (WESSEL *et al.*, 1996). This is in good agreement with similar measurements performed at Alert (ANLAUF *et al.*, 1994), although the ozone depleted air mass only extended up to about 400 m.

First evaluations of these sudden ozone depletions show some relation to the particle number densities of surface aerosols in the accumulation mode, *i.e.* diameter range from 0.09 to 0.15 μm . Extremely low aerosol densities are observed during the ozone minima around Julian days 107 and 153 in 1994, and during Julian days 111 and 113 in 1995. For the weaker ozone depletion event around Julian day 121 in 1994 no significant

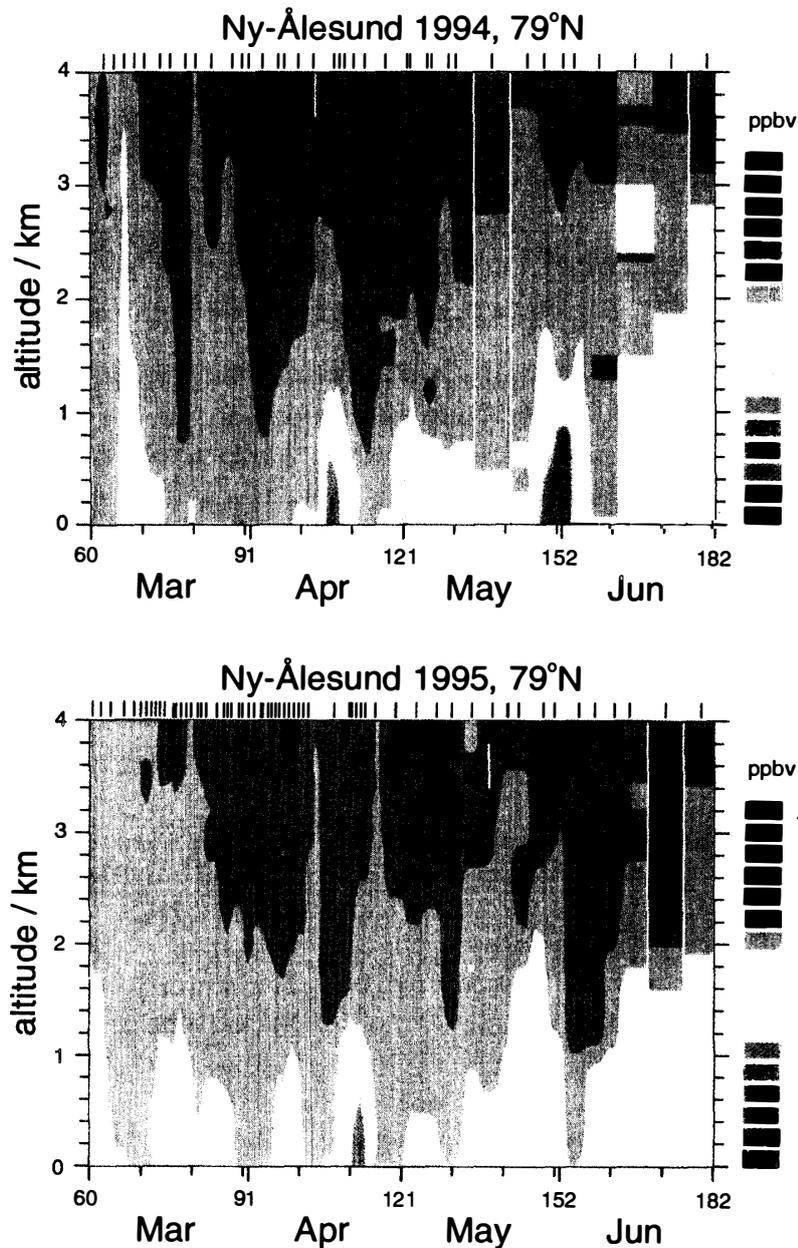


Fig. 9. Altitude-time cross section of seasonal variation of ozone concentration (color coded $\times 10^{12}$ molec/cm³) from March till June in 1994 (upper panel), and in 1995 (lower panel); altitude range (0–4 km); time scale (Julian day, month).

coincidence could be found with the occurrence of low particle number densities in the accumulation mode. The coincidence is more striking in 1994 with particle concentrations dropping down from 12 particles per cm³ to 0.4 particles per cm³ on Julian day 107, and from 10.8 particles per cm³ to 1.58 particles per cm³ on Julian day 153. The decrease of particle number density is more than one standard deviation of the mean value with 6.2 ± 3.92 particles per cm³. In 1995 there was a weaker decrease with particle concentrations dropping down from 16.39 particles per cm³ to 3.17 particles per cm³ on Julian

day 113, while the mean values were 6.95 ± 3.48 particles per cm^3 . These correlations were also found by similar but more detailed investigations during the Polar Sunrise Experiment at Alert (Canada) in 1992 (STAEBLER *et al.*, 1994).

It can be concluded that the sudden ozone depletion occurs in relatively clean air masses where the aerosol content in the accumulation mode is low. Further evidence was given by the observations at Ny-Ålesund that air masses enriched with anthropogenic aerosols in the diameter range between 0.1 and $1.0 \mu\text{m}$, advected from the eastern part of the Arctic Ocean, never showed any significant ozone depletion in the boundary layer (WESSEL *et al.*, 1996).

At antarctic coastal stations similar surface ozone losses may occur in late winter and spring from August until October. Different from arctic regions, ozone concentrations only drop from normal values of about 30 ppbv down to detectable values around 8–15 ppbv for several hours up to one or two days, as can be seen in a preliminary analysis of surface ozone observations from Syowa Station (S. AOKI, 1995, private communication) and at Neumayer station. Ozone sonde data also show that the vertical range for depleted ozone is limited to the temperature inversion in the lower troposphere. Those air masses containing low ozone concentrations have their origin from the ice covered Antarctic Ocean moving to the antarctic coastal regions as can be concluded from trajectory calculations and analysis of weather charts (WESSEL *et al.*, 1996).

6. Conclusion

Ozone sonde observations are useful to discuss different processes causing interannual and seasonal changes of the vertical ozone distribution in the polar troposphere and stratosphere. By using recently available data an approach was made to summarize long-term changes of the vertical ozone distribution for 70°S and 80°N . Stratospheric and tropospheric ozone in the polar region sensitively depends on the global circulation controlling the transport of air toward polar latitudes. They sensitively respond to changing trace gas composition and aerosol loading.

Aerosols may play an important role in forcing both chemical ozone loss and ozone formation processes. PSC related ozone losses were recorded in the lower stratosphere in both polar regions. Volcanic aerosols may strengthen chemical ozone removal.

In the Arctic tropospheric ozone responses were briefly shown which might be forced by natural and anthropogenic aerosols. On the other hand, sudden ozone losses in the boundary layer of the arctic troposphere occur when the particle number density in the accumulation mode drops significantly, indicating natural ozone depletion within clean arctic air masses. A detailed analysis is required to study the chemical and meteorological conditions yielding these irregular ozone losses in both polar regions during spring.

Preliminary observational evidences was found for decreasing ozone concentrations in the middle stratosphere above 26 km altitude during winter and spring, probably occurring simultaneously in both polar regions, as far as that can be determined from balloon-borne ozone observations. That decline seems to not be connected with the chemical ozone removal in the lower stratosphere. It is assumed to be mainly caused by a change of meridional transport conditions for ozone toward polar latitudes. Those changes might also be connected with the long-term internal variability of the global atmospheric system.

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