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Vertical distribution of tropospheric ozone in Antarctica and in the European Arctic

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ABSTRACT

Ozone soundings performed since 1988 on the Antarctic Peninsula at Marambio (64.2°S, 57.7°W) and in the European Arctic at Sodankylä (67.4°N, 26.6°E), Bear Island (74.5°N, 19.0°E) and Ny Ålesund (78.9°N, 11.9°E) have been studied. Regular ozone soundings have never been made at these locations. A comparison of monthly mean tropospheric ozone profiles at high southern and northern latitudes has been made. In midwinter, differences between the monthly means in the northern and southern hemispheres are small, i.e., <1 mPa. The general photochemical activation in springtime leads to larger differences: the partial pressures decrease in Antarctica whereas in the Arctic, pronounced production of ozone is seen. In spring and summer, the hemispheric differences are about 3 mPa. Exceptionally high and low partial pressures at Sodankylä and Marambio have been studied using 3-dimensional trajectories based on the ECMWF analyses. Episodes of upper tropospheric ozone loss at Marambio have been observed during the stratospheric ozone depletion period in spring. The only cause for especially high tropospheric ozone concentrations in Antarctica has been stratospheric intrusions. Low partial pressures in the boundary layer have been connected with advection of marine airmasses to Marambio. At Sodankylä, low boundary layer values have been observed during an advection from the Northern sea areas and also during the transport of airmasses from industrial source areas of Eastern Europe. Especially strong stratospheric ozone intrusion into the middle troposphere was observed at Sodankylä in September 1989. Partial pressures exceeding 7 mPa in the lower troposphere have been observed at Sodankylä during the summer in connection with the long-range transport of NO_x/HC-containing air from Western Europe to Northern Scandinavia.

1. Introduction

The importance of tropospheric ozone has been recognised since the discovery of photochemical reactions in the background troposphere (Crutzen, 1970). There is strong evidence of a pronounced increase of tropospheric ozone in the Northern Hemisphere during the most recent decades (Staehelin and Schmid, 1991; Feister and Warmbt, 1987; Wege et al., 1989). The discovery of the Montsouris surface ozone record measured a century ago has confirmed, that the surface ozone concentration may have doubled, at least in Central Europe (Bojkov, 1986; Volz and Kley, 1988). This is important because of the infrared absorption properties of ozone and its strong general oxidising capacity in the troposphere.

High surface ozone concentrations have been observed in remote areas with relatively low local NO_x- and HC-emissions, e.g., the Nordic Countries (Grennfelt, 1976; Schjoldager et al., 1978; Hakola et al., 1989). These episodes have been caused by long-range transport from Central Europe and the photochemical production of ozone (Taalas, 1988). These ozone concentrations may be harmful to European forests (Sutinen, 1987).

The discovery of the Antarctic ozone "hole" has led to the establishment of new ozone sounding stations in the Arctic and at the Antarctica. The

Finnish Meteorological Institute (FMI) started ozone soundings in Northern Finland in March 1988 and has continued a regular 1–2 soundings a week programme since January 1989. The Norwegian Institute for Air Research (NILU) started ozone soundings at Bear Island in October 1988 and ozone soundings began in Ny Ålesund in January 1989 by the Alfred-Wegener Institute (AWI) and the FMI. No regular ozone soundings have ever before been made in the European Arctic. The FMI and the Argentine Weather Service started ozone sounding activities on the Antarctic Peninsula at Marambio in November 1988. The station was destroyed by a fire in May 1989 but the regular sounding programme was resumed in June 1990.

It is well-known fact that the emissions of NO_x-compounds are of an order of magnitude higher in the Northern Hemisphere compared with the Southern Hemisphere (Ehhalt and Drummond, 1982). Such emissions south of 60° S are very small. It is therefore interesting to compare the behaviour of tropospheric ozone at roughly the same latitudes in different hemispheres, where the concentrations of precursors for ozone differ, but the intensity of short-wave radiation is of the same order of magnitude. It may be assumed that Antarctica represents a more natural, less perturbed troposphere, although there is evidence for a negative tropospheric trend in summer, which may have been caused by the stratospheric ozone depletion and an increase in the intensity of uv-radiation (Schnell et al., 1991).

2. Observations and methods

The basic information concerning ozone soundings is seen in Table 1. All stations use the Vaisala MicroCora or DigiCora sounding equipment and Vaisala PTU-sondes. The ozone sondes are of ECC5A type, manufactured by Science Pump Co. Errors caused by oxidants other than ozone are estimated to be negligible. In this study, soundings have only been made during the ascent phase. In principle the descent phase can also be used, but at the stations presented here no parachutes were employed during the descent phase. The sounding frequency has been normally 1–2 per week at Sodankylä, Bear Island and Marambio. The sounding frequency at Ny Ålesund has been more variable. The stations of Bear Island, Ny Ålesund and Marambio are located on islands and represent marine surroundings. Sodankylä is located in Northern Scandinavia with surroundings of marshland, forest and tundra. At Sodankylä and at Marambio it is possible to correct the integrated ozone profiles against total ozone instruments, but this is not possible at Bear Island and at Ny Ålesund. To permit the different stations to be comparable, however, no such total ozone corrections have been made. The lack of a total ozone

Table 1. *Information on the Arctic and Antarctic ozone sounding stations used in this study*

Station	Institute responsible	Location	Instrumentation	Length of available record	No. of soundings	Correction against
Sodankylä	Finnish Meteorological Institute (FMI)	67.4° N 26.6° E	Vaisala + ECC	March 88–May 91	146	(Brewer)
Bear Island	Norwegian Institute for Air Research (NILU)	74.5° N 19.0° E	Vaisala + ECC	Oct. 88–June 90	87	no
Ny Ålesund	Alfred-Wegener-Institute (AWI) and FMI	78.9° N 11.9° E	Vaisala + ECC	Jan. 89–Oct. 89, March 90–April 90	65	no
Marambio	Argentine Weather Service (SMN) and FMI	64.2° S 57.7° W	Vaisala + ECC	Nov. 88–March 89, June 90–July 91	71	(Dobson)

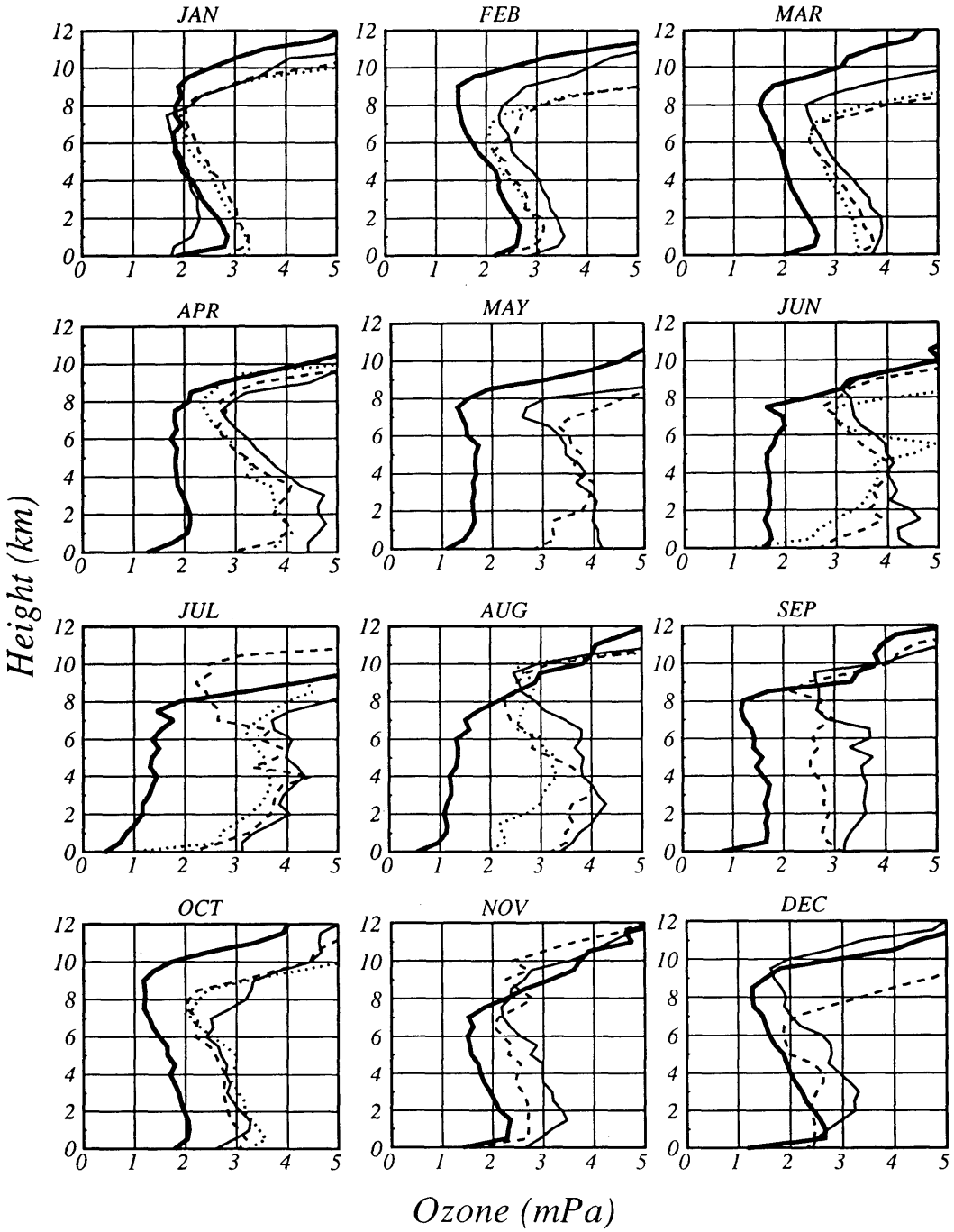

Fig. 1. Monthly mean profiles of the partial pressure of tropospheric ozone in the European Arctic and on the Antarctic Peninsula. The values for Marambio have been shifted 6 months. The thick solid line denotes Marambio (64°S), the thin solid line Sodankylä (67°N), the dashed line Bear Island (74°N) and the dotted line Ny Ålesund (79°N).

correction is not very significant. Taalas and Kyrö (1992a) have shown that the average total ozone correction factor at Sodankylä during 1989 and 1990 was less than 3%.

Three-dimensional trajectories have been calculated for the analysis of tropospheric ozone anomalies at Sodankylä and Marambio. These trajectories are based on the global horizontal and vertical wind analyses of the European Centre for Medium Range Weather Forecasts (ECMWF). These winds are given every 6 hours on a 1.5° latitude-longitude grid at 17 σ-levels. The trajectory calculation has been done using a model developed by the Royal Netherlands Meteorological Institute (KNMI). See Reiff et al. (1986) for further details. These trajectories have been calculated 5 days backwards, and a time step of 2 h has been used.

3. Monthly means

Monthly mean ozone profiles are seen in Fig. 1. The curves have been calculated as 500 m means. The monthly means for Marambio have been shifted by 6 months to be able to compare the behaviour of ozone in the two hemispheres during the same seasons. From Fig. 1 is seen that the differences between the partial pressures of ozone at Marambio and in the Arctic are less than 1 mPa in midwinter. After the return of short-wave radiation to high latitudes of both hemispheres (February–March in the Northern hemisphere) large differences, i.e., of the order of 3 mPa (April, accordingly), are seen.

It is interesting to notice the strong ozone inversion near the surface at Ny Ålesund and on Bear Island in June and July. The vertical gradient has been about 1 mPa/km. This is most evident in the former month.

The yearly trend of tropospheric ozone in three 500 m thick layers at the four stations is seen in Fig. 2. At the lowest level at Marambio the yearly minimum (0.5 mPa) is observed in January (July in Fig. 2) and maximum (2.2 mPa) in October (February in Fig. 2). The shape of the annual trend is rather similar to that observed at Syowa, 69°S (Logan, 1985). The partial pressures are well below those observed at the South Pole in spring and summer (Oltmans and Komhyr, 1986). The seasonal variation is smaller in the upper

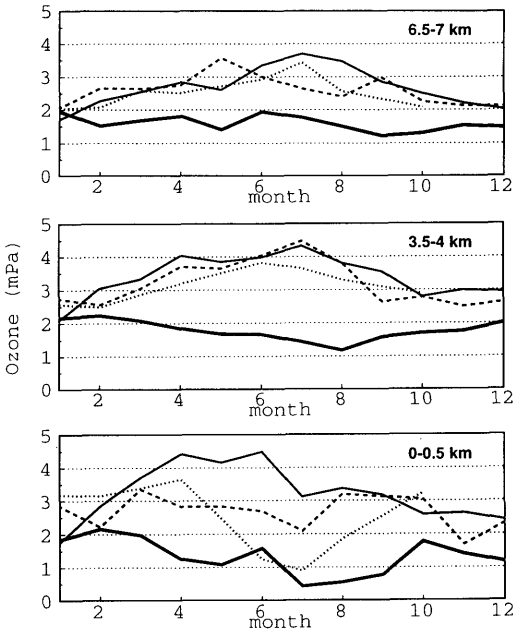

Fig. 2. Annual mean trend of the partial pressure of tropospheric ozone at three altitudes in the European Arctic and on the Antarctic Peninsula. The values for Marambio have been shifted 6 months. The thick solid line denotes Marambio (64°S), the thin solid line Sodankylä (67°N), the dashed line Bear Island (74°N) and the dotted line Ny Ålesund (79°N).

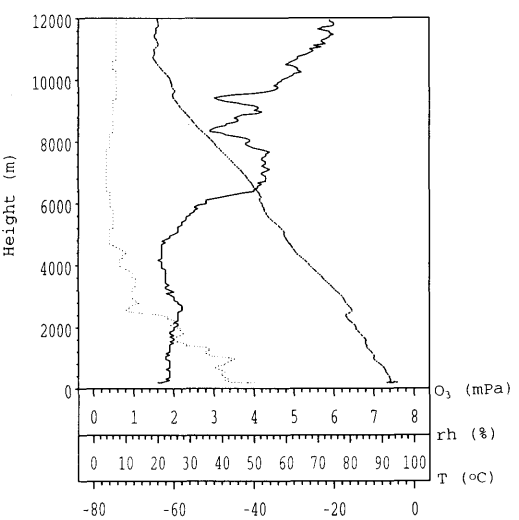

Fig. 3. Stratospheric intrusion at Marambio (Antarctic Peninsula) on 17 October 1990. The vertical distributions of the tropospheric partial pressure of ozone (solid line), temperature (dashed) and relative humidity (dotted) are indicated.

troposphere. The August minimum and February maximum are similar to those reported by Logan (1985) from Syowa.

In the Northern Hemisphere, spring maxima are observed in the lower and middle troposphere at all stations. We have observed a strong surface level ozone maximum in Sodankylä, whereas the more Northern stations Bear Island and Ny Ålesund do not show any maxima in summer.

As was mentioned earlier, there are large differences between the two hemispheres. This is most evident at the surface level and in the middle troposphere.

4. Characteristic anomalies

Seasonal means and standard deviations have been calculated for the ozone profiles of Marambio (1988–91, see Table 1) and for those of Sodankylä (1989). All tropospheric ozone anomalies greater than 2σ from the seasonal means have been analysed by calculating 3-dimensional trajectories 5 days backwards. Typical anomalies will be presented in this section.

4.1. Antarctic minima and maxima

Of the 71 soundings analysed, 6 cases were identified in the upper troposphere, where stratospheric intrusions are estimated to have caused the maxima. These maxima were always connected with local relative humidity minima, which were below 5%. According to Danielsen (1985) such dryness is characteristic of stratospheric air. An example of a maximum caused by stratospheric intrusion is shown in Fig. 3. A highest partial pressure of 4.4 mPa was observed at 7 km altitude with a relative humidity value of 4%.

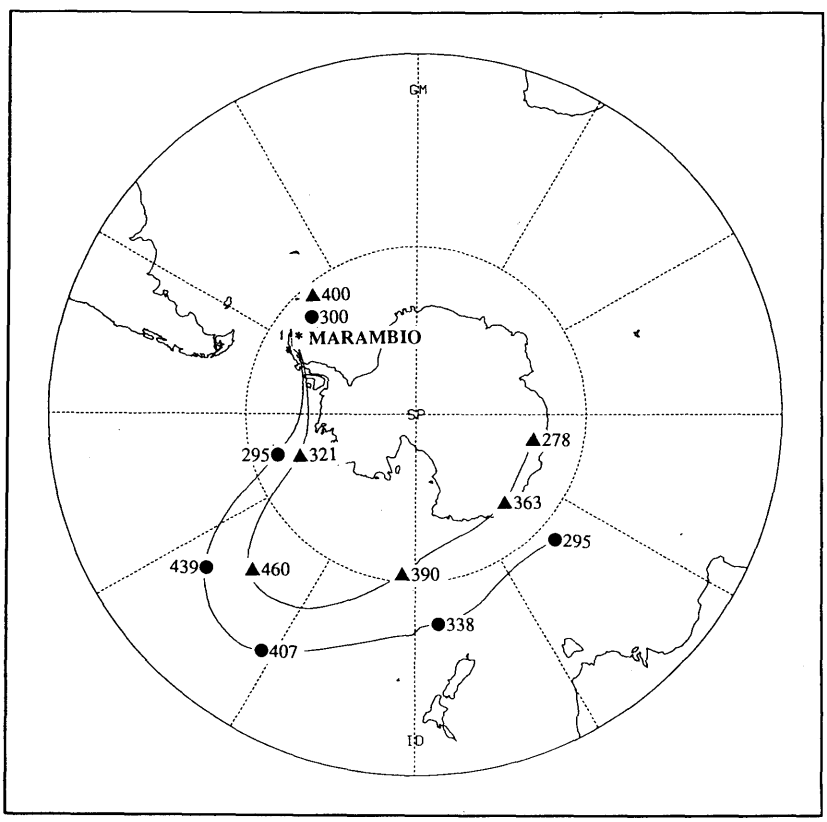

Fig. 4. 3-dimensional trajectories 5 days backwards ending at the 400 and 300 mb levels during stratospheric intrusion. The vertical coordinate (mb) is plotted every 24 h.

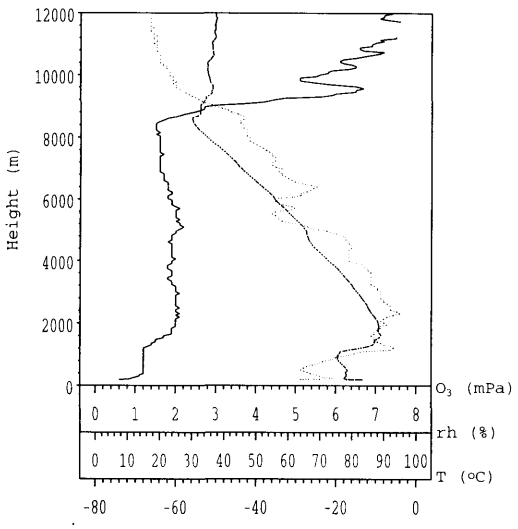

Fig. 5. As Fig. 3, but showing ozone advection from sea area at Marambio on 27 October 1990.

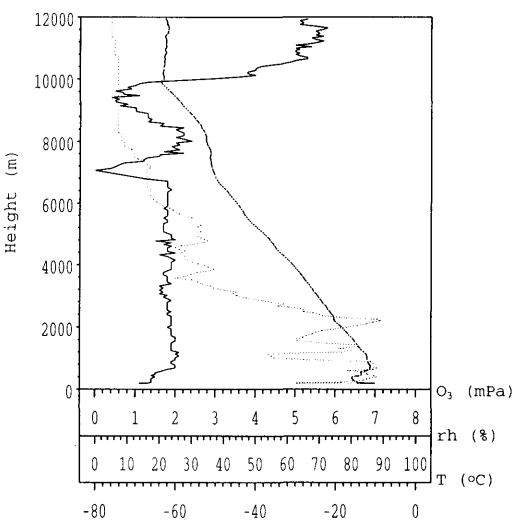

Fig. 6. As Fig. 3, but showing tropospheric ozone depletion on 24 October 1990.

3-dimensional trajectories 5 days backwards reaching the 400 and the 300 mb levels at Marambio are seen in Fig. 4. The strongest maximum was observed near the 400 mb level. The 400 mb trajectory shows cyclonic curvature, which is thought to be typical for tropopause folding episodes during cyclogenesis (Danielsen, 1985). Sinking motion, which has reached tropopause level (278 mb), is also seen. It may be assumed that the grid and the observing system do not allow one to see tropopause foldings in the ECMWF analysis data. The relative shortness of our time series does

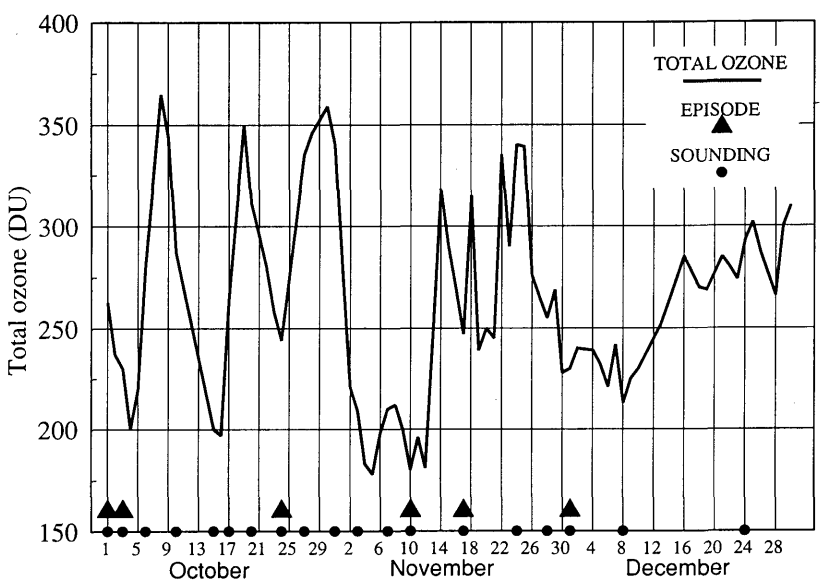

Fig. 7. Total ozone, upper tropospheric ozone loss episodes and the sounding frequency at Marambio during October–December 1990.

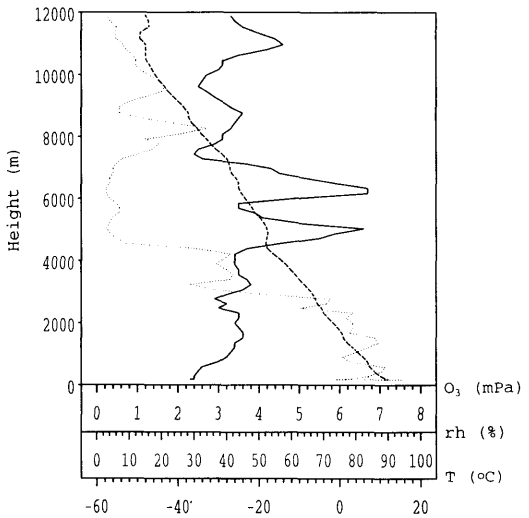

Fig. 8. Stratospheric intrusion at Sodankylä (Finland) on 6 September 1989. The vertical distributions of the tropospheric partial pressure of ozone (solid line), temperature (dashed) and relative humidity (dotted) are indicated.

not yet allow one to obtain valid statistics on the seasonal dependence of the stratospheric intrusions.

An example of a minimum in the lower troposphere is seen in Fig. 5. The lowest partial pressure was 0.6 mPa near the surface. An especially strong temperature inversion was observed at 1500 m/800 mb (−8.7°C). Below this level temperatures of −17 to −20°C were observed with partial pressures of ozone of 0.6–1.2 mPa. Above the inversion level the partial pressures were 1.9–2.1 mPa. The 925 mb trajectory analysis indicated the influence of marine mid latitude air. Four similar cases on the Antarctic Peninsula were observed in our study.

A totally new type of tropospheric ozone anomalies is seen in Fig. 6. We have observed altogether six cases during October–December 1990 in which pronounced ozone depletion ($P_{O_3} < 0.5$ mPa) in the upper troposphere has been present. The partial pressure at 7 km height (370 mb) was 0 mPa and at 9.5 km (250 mb)

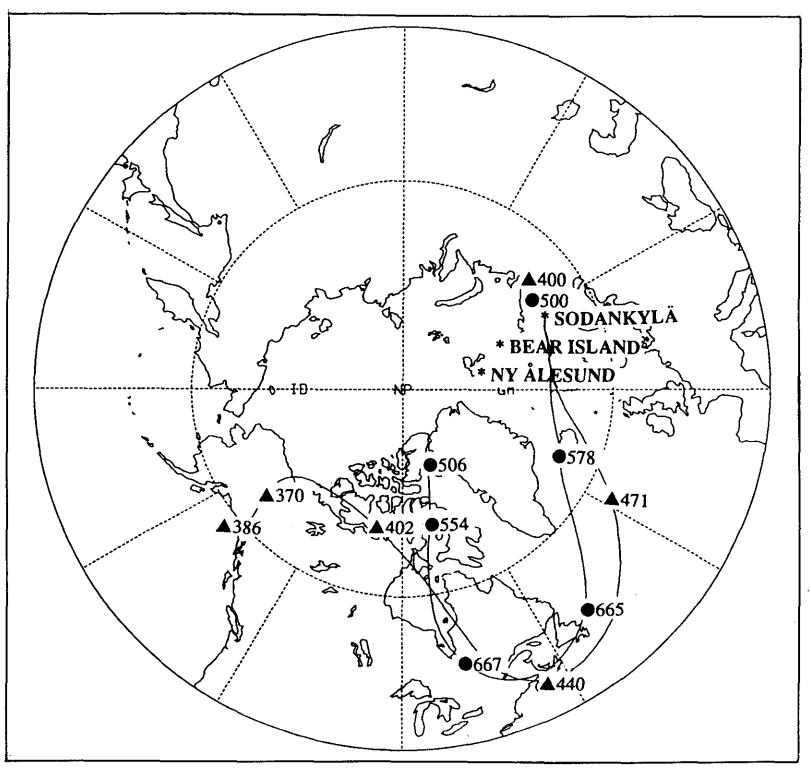

Fig. 9. 3-dimensional trajectories 5 days backwards ending at the 500 and 400 mb levels during stratospheric intrusion. The vertical coordinate (mb) is plotted every 24 h.

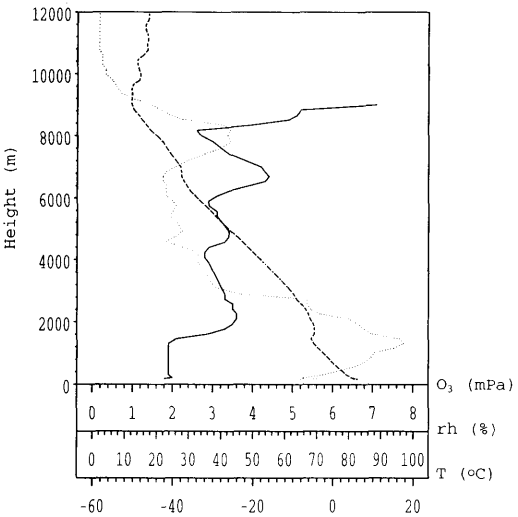

Fig. 10. As Fig. 8, but showing Arctic advection on 31 May 1989.

0.4 mPa. The tropopause showed a somewhat folded structure with inversions at the 7.5 km and 9.9 km levels. Relative humidity values were 16% and 7%, respectively. Trajectories reaching Marambio at the 400, 300 and 200 mb levels indicated that the situation at the 300 and 400 mb levels was very stagnant. All trajectories indicated that the observed airmasses have originated in the Antarctic region, where stratospheric ozone depletion may be present. Schnell et al. (1991) have previously estimated that the increase in uvb-radiation may have caused the observed decreasing trend in surface level ozone. Our observations indicate that this loss is strongest in the upper troposphere, where the intensity of shortwave radiation is much stronger than at the surface level. The relative humidity mean for these minima has been 39% (range 16–56%), which does not suggest that these episodes might have

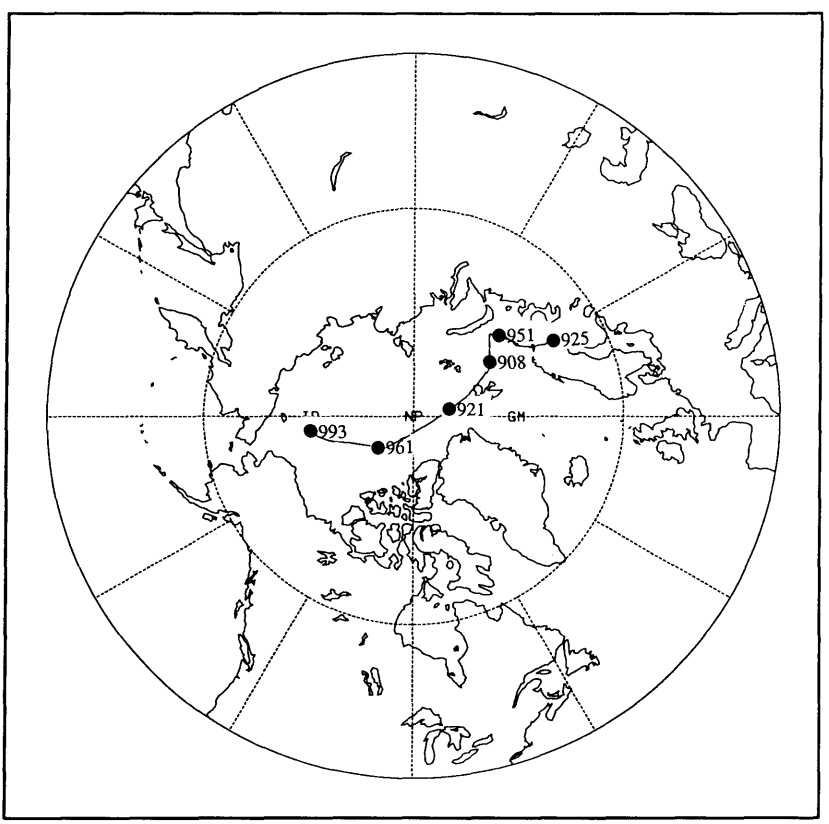

Fig. 11. The 3-dimensional trajectory 5 days backwards ending at the 925 mb level during Arctic advection. The vertical coordinate (mb) is plotted every 24 h.

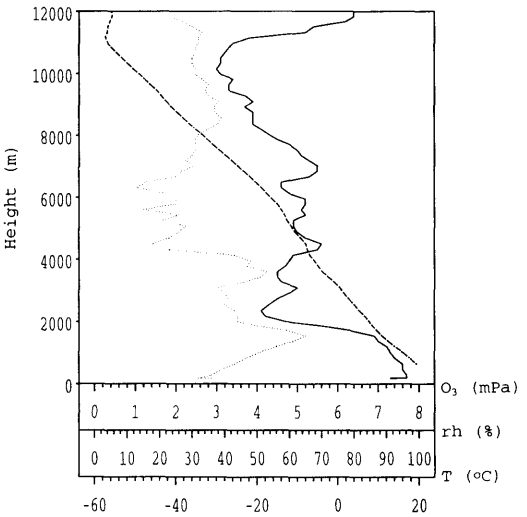

Fig. 12. As Fig. 8, but showing the photochemical production of ozone during long-range transport from major European source areas of NO_x and hydrocarbons on 28 June 1989.

caused by low-ozone-bearing stratospheric air, which directly intruded into the troposphere. The total ozone values for October–December are seen in Fig. 7. The occurrence of low upper tropospheric ozone episodes and the dates of the ozone soundings are also plotted. One may see that the episodes are observed, when the total ozone content has been lower than normally. The total ozone mean for the episode days was 235 D.U., whereas the October–December mean was 264 D.U.

4.2. Arctic minima and maxima

An example of an airmass of stratospheric origin is shown in Fig. 8. This middle tropospheric ozone maximum was first observed at Sodankylä on 6 September 1989. Altogether 9 stratospheric intrusion episodes were identified in 1989 at Sodankylä. The dual-peaked maximum was 6.6 mPa at the 5 km level (530 mb) and 6.7 mPa at the 6.2 km level (450 mb). These maxima were characterised

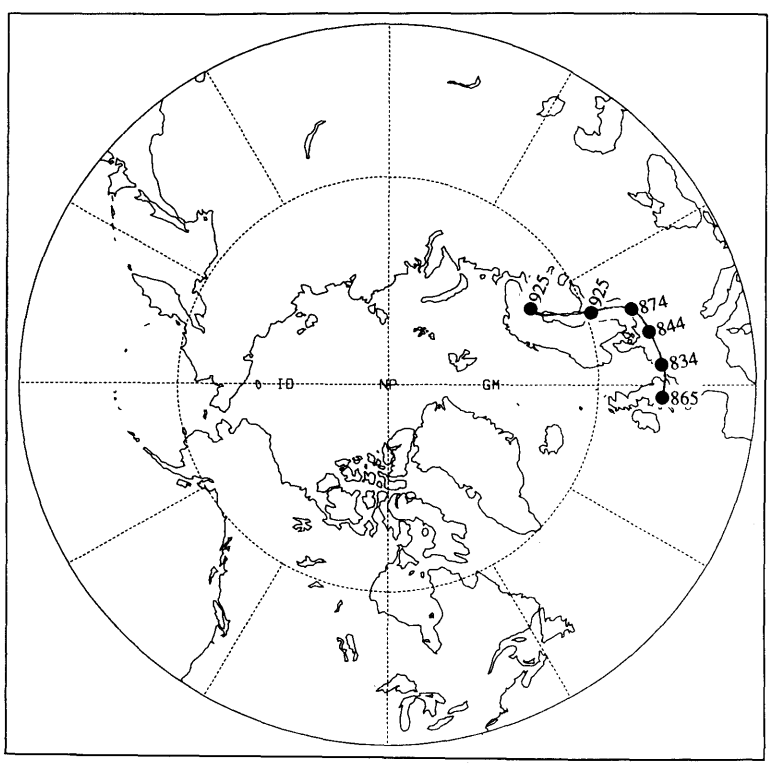

Fig. 13. The 3-dimensional trajectory 5 days backwards ending at the 925 mb level during photochemical production of ozone. The vertical coordinate (mb) is plotted every 24 h.

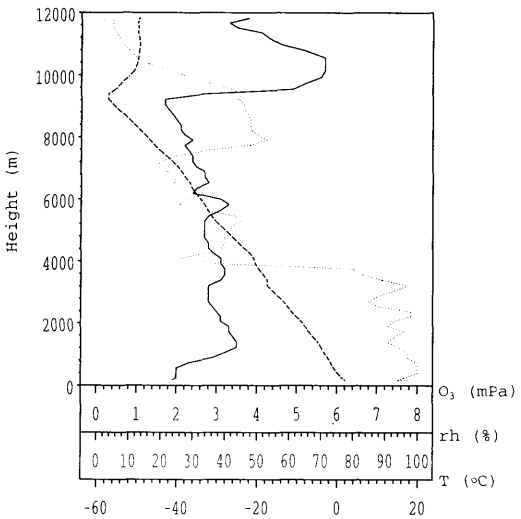

Fig. 14. As Fig. 8, but showing the long-range transport from industrialised areas of Russia on 11 October 1989.

by extreme dryness, i.e., 3%, at both levels. Trajectories for the 500 and 400 mb levels are seen in Fig. 9. These indicate curvature typical of cyclogenesis over North-eastern Canada. Fairly similar maxima were observed in a sounding made two weeks later at Sodankylä. The maxima were a little weaker, i.e., 5.0 and 5.2 mPa respectively, but the vertical location was almost the same. The airmass was now more humid, 22% and 15%. If our assumption concerning the same airmass is right, we may deduce a decay rate due to irreversible mixing of 0.11 mPa/day.

We have observed 6 cases in which the ozone content of the lowest troposphere was low, while advection from Northern sea areas (the Arctic Ocean, Northern Atlantic Ocean) has been occurring. An example of this kind of profile is seen in Fig. 10. The partial pressure of ozone was below 2 mPa in a layer below 1.4 km, where an inversion was observed. The airmass of low ozone content

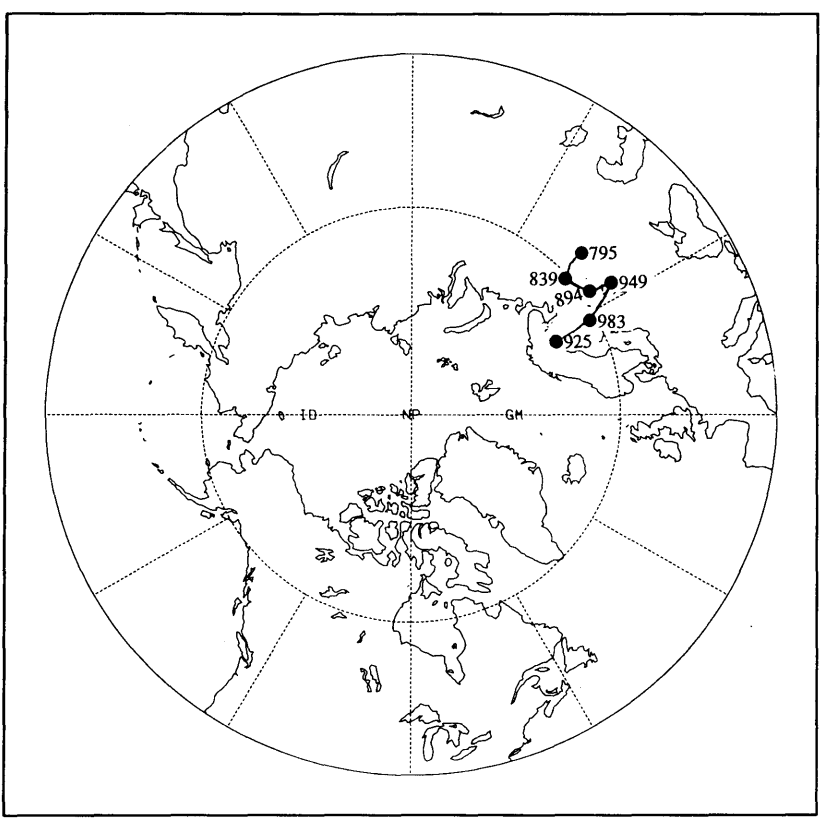

Fig. 15. The 3-dimensional trajectory 5 days backwards ending at the 925 mb level during advection of polluted air from Russia. The vertical coordinate (mb) is plotted every 24 h.

was characterised by high relative humidity (up to 97%) and low temperatures (5°C at the surface and −5.4°C at the inversion level). The trajectory analysis for 925 mb indicates advection across the polar region (Fig. 11).

In contrast to the Antarctica one can see direct indications of anthropogenic emissions in the Arctic. We have observed 4 cases, in which advection from Western Europe has coincided with high ozone concentrations in the lower troposphere. We have also observed one such case with advection from the East coast of the United States. An example of very high partial pressures is seen in Fig. 12. Partial pressures of ozone of up to 7.7 mPa were observed in a layer below 2 km. Especially high temperatures (26°C) were observed in connection with advection from Germany and the Benelux-countries, where the highest European emissions of NO_x and hydrocarbon species are observed (Iversen et al., 1991; Simpson, 1991). See Fig. 13 for trajectories. This kind of advection together with photochemically favourable weather conditions is the most common cause for surface-level ozone enhancements in Finland (Taalas, 1988). Similar episodes have been reported earlier in Southern Sweden (Grennfelt, 1976) and Norway (Schjoldager et al., 1978). The vertical structure of these episodes has never previously been observed in Northern Europe.

Another type of episode is seen in Fig. 14. The partial pressure of ozone was about 2 mPa in a layer below 800 m. No temperature inversions were observed in the lower troposphere. The relative humidity was 94–100% in this lowest layer. A trajectory analysis for the 925 mb level is seen in Fig. 15. The airmass has passed some heavily industrialised areas of the Russia (for example St. Petersburg). We have identified 11 cases in 1989 in which a low ozone content of the lower troposphere coincided with advection from Eastern Europe, where the NO_x and HC emissions are relatively low compared with those of Western Europe ones, but where the SO_2 emissions are high (Iversen et al., 1991; Simpson, 1991).

5. Discussion

The seasonal comparison of tropospheric ozone in different hemispheres, shown in Figs. 1 and 2 has revealed considerable differences after the return of solar radiation in spring and during the summer months. These interhemispheric differences are relatively small during the photochemically less active winter season. The springtime differences may result from the general activation of NO_y/HC compounds throughout the whole troposphere. The mechanism was first proposed by Penkett and Brice (1986). The balance between photochemical production and destruction is highly dependent on the concentration of NO_x (Fishman et al., 1979). Destruction of ozone is estimated to occur at NO_x concentrations of less than 20 pptv. Measurements of NO_x in high southern latitudes have revealed concentrations below 30 pptv, whereas values in high northern latitudes have been of the order of 100 pptv (Gerhardt et al., 1989).

The effect of large-scale photochemical activation in the Northern hemisphere is best seen in the middle troposphere. Taalas and Kyrö (1992b) have studied the variability of ozone in the lower, middle and upper troposphere based on ozone soundings from Sodankylä. They have shown that the variability is weakest in the middle troposphere. Local sink effects and the variability caused by the dynamics and chemistry of the atmospheric boundary layer are well seen in the lower troposphere, whereas in the upper troposphere stratospheric intrusions are the cause of variability. The considerable difference between the hemispheres decreases towards the autumn with the decreasing intensity of photochemistry.

An ozone minimum is seen at Marambio in spring whereas at the Arctic stations maxima are found (see Figs. 1, 2). This kind of surface level maxima has not been observed at remote sites in Northern Canada (Logan, 1985) nor in Alaska (Oltmans et al., 1989). Oltmans et al. (1989) observed a spring minimum, which was thought to be connected with an annual bromine maximum. Our observations in the Arctic do not show indications of a pronounced surface level ozone sink in springtime.

Strong surface-layer inversions (Fig. 1) and low partial pressures of ozone have been observed at island stations (Marambio, Bear Island and Ny Ålesund) in summer. This may be an indication of an ozone sink in the remote marine boundary layer. These stations may be influenced by cleaner marine air with a fairly low NO_x content. The marine boundary layer is generally thought to

be a photochemical sink for ozone due to low NO_x concentrations and high OH concentrations (e.g., Liu, 1988). Decreased lower tropospheric ozone concentrations have also been observed at Sodankylä during advection from the Arctic sea area.

Trajectory analysis of increased lower tropospheric ozone episodes has revealed that during summer Sodankylä is occasionally influenced by the long-range transport of ozone and its precursors from Central Europe, followed by the photochemical formation of ozone. The summer minima observed at the other stations is not therefore seen at Sodankylä. On the other hand, long-range transport of airmasses from industrialised areas of Russia may lead to low ozone contents in the lower troposphere in Northern Scandinavia. Russian NO_x and HC emissions are relatively low compared with those of Western Europe, but SO_2 emissions are high (Iversen et al., 1991; Simpson, 1991). It is therefore possible that the oxidation of these emissions by ozone has led to the decrease of ozone observed at Sodankylä.

A totally new type of decreased upper tropospheric ozone content has been found at Marambio (see Fig. 5). The increased UVB-radiation (caused by stratospheric ozone loss) followed by intensified photochemical sink reactions may have been at least partly responsible for the low partial pressures in the upper troposphere. On the other hand it is worth noting that these episodes are not always observed when the total ozone content is low. A definitive explanation for our observations needs in situ measurements of the more important chemical species. The possibility of errors caused during preparation or production of the ECC sondes cannot be totally excluded as an explanation for our observations.

6. Summary and conclusions

The ozone soundings performed during 1988–91 on the Antarctic Peninsula (64° S) and in the European Arctic (67–79° N) have been studied. No regular ozone soundings have ever before been made at these locations. Former tropospheric ozone studies at these locations have been based on surface level measurements, which give a very limited view of tropospheric ozone, due to the surface sink of ozone and other local effects. Besides seasonal mean behaviour we report characteristic ozone minima and maxima at these high latitudes. The 3-dimensional wind fields of the ECMWF and 3-dimensional trajectories 5 days backwards at various levels have been used to find out the advective factors behind the anomalies.

Differences in the partial pressure of ozone in the two hemispheres are fairly small in winter, when photochemical production or destruction is not important. The photochemical activation in spring leads to large differences between the hemispheres. The activation of NO_y and hydrocarbon species stored in the Northern hemisphere leads to pronounced photochemical production in the whole troposphere. A summer maxima is observed in the free troposphere of Northern Europe. This is an indication of a large-scale photochemical production of ozone, for which the European and the North-American NO_x/HC-emissions are of importance. On the other hand this increase in the intensity of short-wave radiation leads to the destruction of ozone due to low NO_x concentrations in Southern high latitudes. A sink of ozone in the marine boundary layer has been identified in both hemispheres in summer. We have not seen indications of pronounced ozone loss due to bromine reactions in spring, as has been the case in the American Arctic. In contrast to the surface ozone measurements in Alaska and Northern Canada, we have observed springtime ozone maxima in the lower troposphere at our Arctic stations. The surface level concentrations on the Antarctic Peninsula are much lower than at the South Pole station in spring and summer.

The analysis of single tropospheric ozone anomalies in the Arctic and Antarctica has been done by calculating 3-dimensional trajectories. We have observed stratospheric intrusions in both hemispheres, which have been characterised by extremely dry air and cyclonic curvature of the trajectories.

We have found that airmasses of remote marine origin are characterised by low ozone concentrations in the lower troposphere in both hemispheres. Similar episodes in Northern Scandinavia have also been connected with the advection of polluted air from Eastern Europe, where the emissions of ozone precursors are relatively low, but SO_2 emissions are high. In contrast, extremely high lower tropospheric ozone concentrations have been observed with an advec-

tion from Western Europe, where the NO_x and HC emissions are the highest in Europe.

We have identified considerable ozone loss episodes in the upper troposphere during the stratospheric ozone depletion period in the spring. The total ozone concentration has been below the seasonal mean during these loss episodes. Neither the trajectory analyses nor the water vapour content for these minima indicate that these episodes might have caused by stratospheric ozone-poor air, that had intruded into the troposphere. Increased short-wave radiation and sink reactions for ozone are the most probable explanations for the phenomena.

After these conclusions we would like to add some warnings concerning our results. Due to the relatively short period of ozone soundings (excluding Sodankylä) too far-reaching conclusions should be avoided. The possibility of errors caused by ECC sondes cannot be totally excluded from the upper tropospheric ozone minima cases in Antarctica. There are also some sources of errors in the trajectory analysis. One is the grid of 1.5° and the other is the lack of observations from remote areas such as Southern Ocean areas and Northern high latitudes.

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