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Analysis of tropospheric ozone measurements in Greenland

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ABSTRACT

The Danish Meteorological Institute (DMI) initiated continuous measurements of surface ozone concentrations in Greenland during spring 1994 as a part of the EU-project ARCtic Tropospheric Ozone Chemistry (ARCTOC). In 1995 and 1996, several events of deep ozone depletion were recorded at Thule and Søndre Strømfjord connected with passing frontal systems (cold fronts) from the west or north-west with rising pressure and falling temperature. At Scoresbysund, minor episodes were observed both in 1995 and 1996. The major episodes could definitely be assigned to cold air masses moving from the Arctic Ocean over the Canadian Archipelago across Baffin Bay and Davis Strait and further crossing Greenland. Analyses of backward trajectories calculated by a 3-D transport model developed at DMI utilising meteorological data from the numerical weather prediction model DMI-HIRLAM (HIgh Resolution Limited Area Model) have been performed. Results from these studies show that air parcels with low ozone values at the receptor point generally have passed the northern Canadian Archipelago or the Arctic Ocean a few days before the arrival at the receptor point.

1. Introduction

In the last 10 years, measurements of surface-ozone depletion episodes in spring at several locations in the Arctic have been reported; at Alert (Barrie et al., 1988; Bottenheim et al., 1986, 1990), at Barrow, Alaska (Oltmans et al., 1986), north of Canada (Hopper et al., 1994) and in the Norwegian Arctic (Solberg et al., 1994, 1996). Depletion episodes are often characterised by a sudden decrease in the surface ozone concentration from the normal level to a 10-times lower level, with a duration of hours to days.

Depletion of ozone at different sites in the Arctic is partly caused by chemical destruction such as catalytic reactions of BrOx radicals and photochemistry of bromoform, (Barrie et al., 1988), and by transport of air masses already deficient in ozone (Li et al., 1990). Analyses based on trajectory calculations for Svalbard, Norway, show that most depletion episodes occurred when air masses were transported from the West or North (Solberg et al., 1996).

In this study, measurements of tropospheric ozone at coastal sites in Greenland were analysed. A period of 3 years 1994–1996 is covered, and trajectories have been calculated in order to analyse the origin of the ozone-depleted air masses.

We will focus on the extent of the ozone-loss phenomena and the physical (meteorological) environment of the phenomena. The chemical environment and possible mechanisms causing the ozone loss will not be discussed in detail because we do not have any measurements of relevant trace constituents (as BrO, ClO and IO).

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2. Measurements

2.1. Location and instrumentation

Surface ozone concentrations in Greenland have been observed by the Danish Meteorological Institute (DMI) in Thule from April 1994, in Scoresbysund from May 1994 and in Søndre Strømfjord from May 1995.

The monitor in Thule, cf. Fig. 1, is located at the South Mountain 240 m above sea level just south of the Thule Air Base. In Scoresbysund, the monitor is located near the top of a small local hill about 65 m above sea level in connection with the telestation 500 m east of the village. In Søndre Strømfjord, the monitor is located 5 km south-east of the air base near the top of a small hill 320 m above sea level. The instruments are photometric ozone analysers, model 400 from Advanced Pollution Instrumentation, automatically calibrated once a day. Data averaged over 10 min are automatically collected on a PC and sent to DMI either directly (by ftp) or via diskette.

2.2. Results

Fig. 2 shows the daily mean ozone concentrations in Thule, Scoresbysund and Søndre Strømfjord for the years 1994, 1995 and 1996. There is a clear seasonal variation with average values of 35–40 ppb, during the months June to August the values decrease to about 25 ppb.

In 1994, it seems that Thule experienced a depletion episode at the start of the measurements in April, while minor episodes were recorded in June. In Scoresbysund, 3 minor episodes occurred in May and early June.

In 1995, 2 major episodes were recorded at Thule in March. A research group from The National Oceanic and Atmospheric Adminiatration (NOAA), USA, who had a campaign in Søndre Strømfjord measuring surface ozone concentrations and trace constituents (e.g. BrO, OClO), evidently measured the same 2 episodes observed in Thule (Miller et al., 1997). Both episodes were clearly connected with a cold air mass moving from the northern Canadian

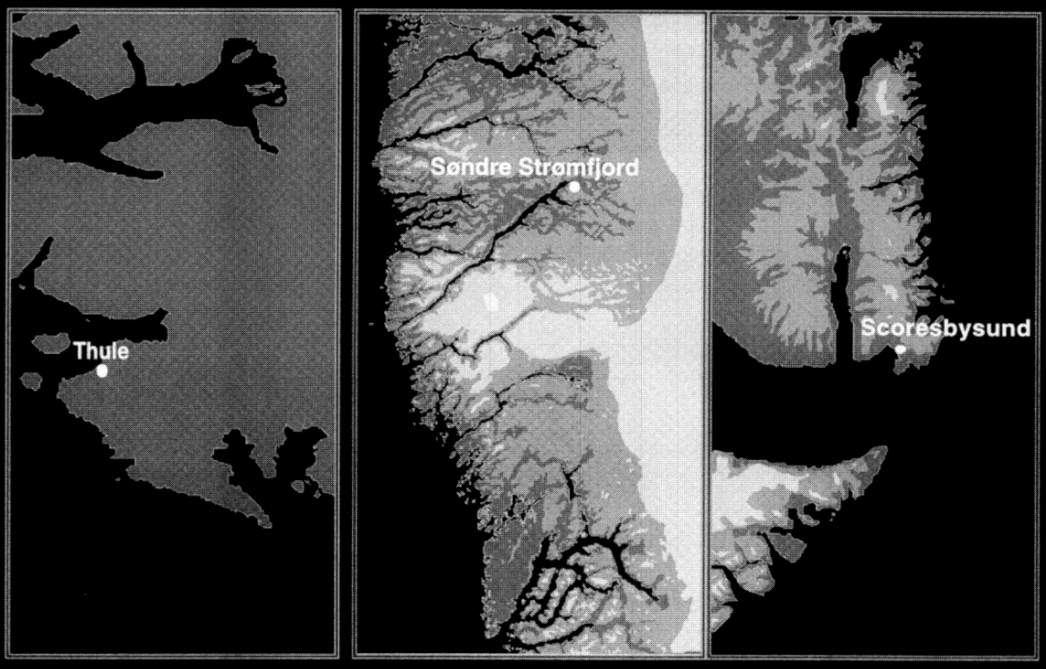

Fig. 1. Map showing the observation sites in Greenland measuring surface ozone concentrations: Thule (76°31′N, 68°50′W), Søndre Strømfjord (67°00′N, 50°48′W) and Scoresbysund (70°29′N, 21°58′W).

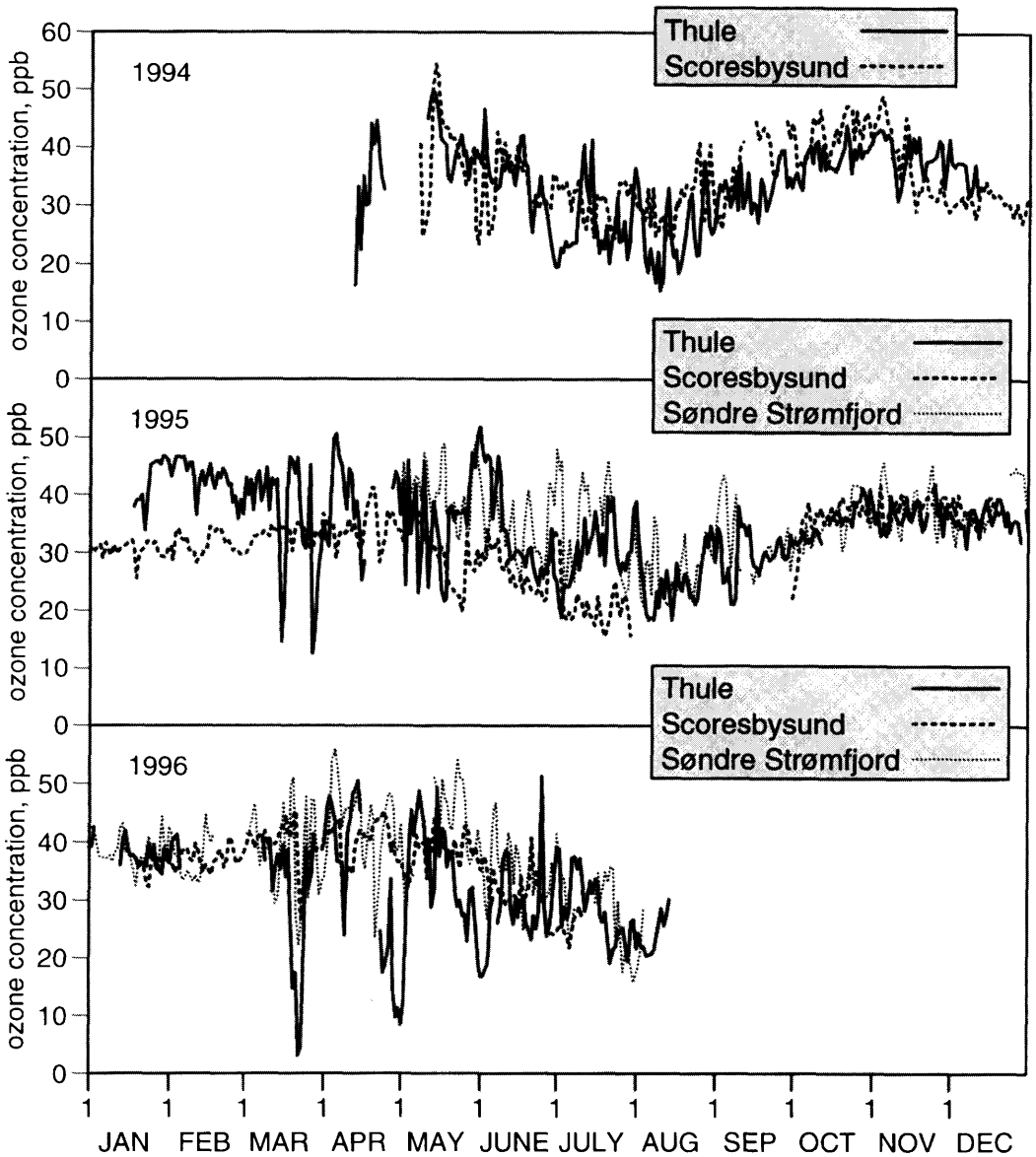

Fig. 2. Daily mean ozone concentrations (ppb) for Thule, Scoresbysund and Søndre Strømfjord for the years 1994, 1995 and 1996.

Archipelago to Greenland. In Scoresbysund, minor episodes were recorded in May and early July.

In 1996, a major episode was recorded around 20 March. This episode was measured at all 3 stations. A similar major episode was recorded in Thule in late April and again in late May.

Below we will discuss the major episode in March 1996 in detail. The other major episodes show very similar features; in fact, the episode in 1996 was forecasted 4–5 days before occurring based on the experience from the first 2 years measurements.

In Fig. 3, a closer look is given at the episode

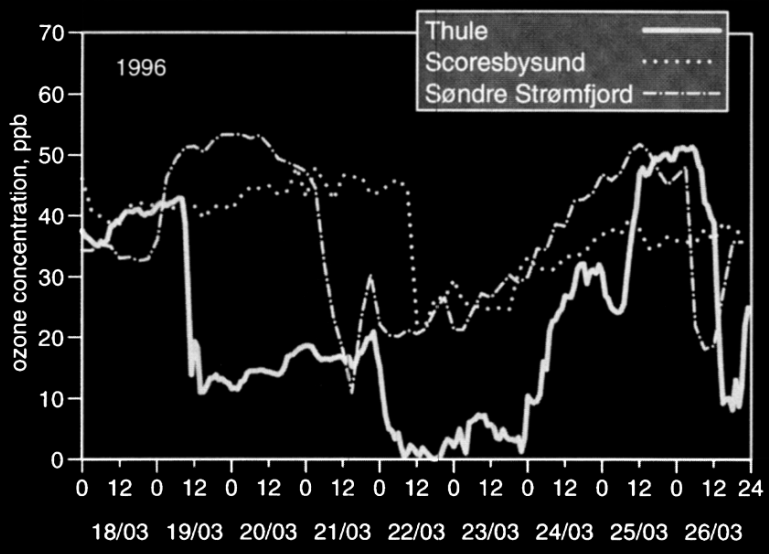

Fig. 3. Hourly mean ozone concentrations (ppb) for Thule, Scoresbysund and Søndre Strømfjord for the period 18 to 26 March 1996.

in March 1996. The hourly mean ozone concentrations are around 40 ppb until the 19th, but in Thule, the concentration falls sharply to about 12 ppb over 4–5 h. During the next 2 days, the concentrations in Thule slowly increase to nearly 20 ppb, then suddenly fall to below 5 ppb on the 22nd. On the 24th and 25th, the level in Thule increases to about 50 ppb. In Søndre Strømfjord, the concentration falls rather slowly to about 16 ppb on the 21st, while in Scoresbysund the concentrations fall sharply to 21 ppb during three hours on 22 March.

Inspection of weather maps shows that the episode is connected with a cold air mass moving from the Arctic Ocean and the northern Canadian Archipelago to east and south-east across Greenland, cf. Fig. 4.a-c. On 19 March (Fig. 4.a), a low-pressure area and cold arctic air is situated over the north-western parts of the Canadian Archipelago and over the Arctic Ocean just north of the Archipelago. The cold air reached Thule during the day with a marked drop in the ozone concentration.

On 21 March 12 UTC (Fig. 4.b), the cold air had just reached Søndre Strømfjord causing a drop in the ozone concentration, and on the 23 March (Fig. 4.c), the cold air had also reached Scoresbysund accompanied by a marked fall in the ozone levels. Note that very cold air is situated over Thule while it is experiencing deep ozone depletion.

Fig. 5. shows time series of hourly ozone concentrations, pressure and temperature in Thule for the same period as in Fig. 3. On the 19th the temperature drops from about −16°C to about −30°C, and on the 22nd with the most intense ozone depletion, the temperature falls to about −36°C. The pressure only increases slightly in connection with the first sharp decrease in ozone on the 19 March, while the pressure rises to above 1035 hPa during the most intense depletion period. The wind shift is quite marked in connection with the front passing on the 19th, while there is no windshift on the 21st. An interesting feature on the figure is the very short-lived low-ozone episode observed both in Thule and Søndre Strømfjord on 26 March. The episode occurred in connection with a cold air mass approaching from the west-south-west with precipitation. Note that there is no wind shift for this short episode, but a pronounced increase in wind velocity.

In Table 1, the correlation between ozone concentration and meteorological parameters is given. Generally, the correlation coefficients are rather small, but especially for 1996, there is a significant positive correlation between temperature and

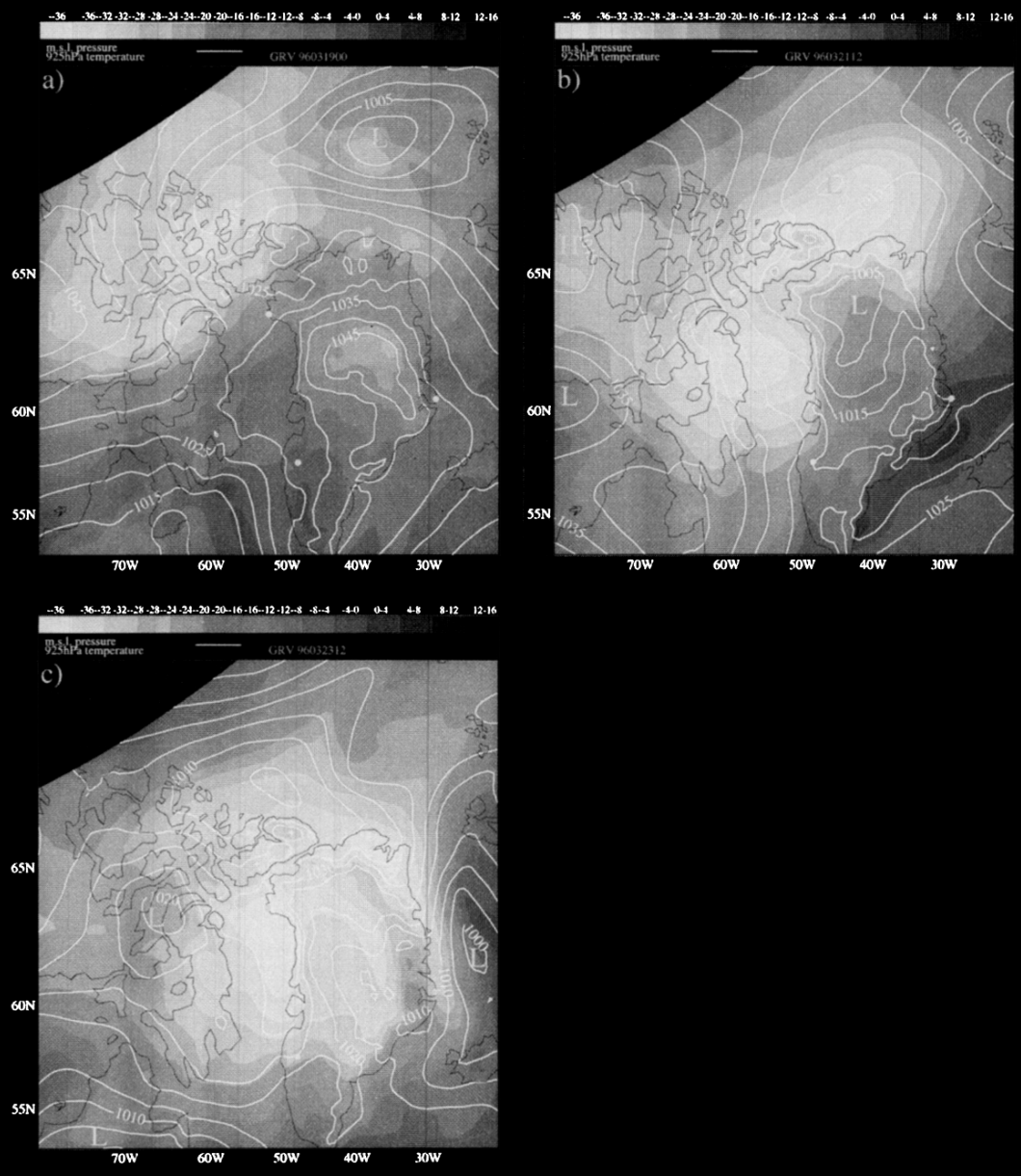

Fig. 4. DMI-HIRLAM maps of msl-pressure and 925 hPa temperature for (a) 19 March, 00 UTC, 1996, (b) 21 March 12 UTC, 1996 and (c) 23 March, 12 UTC, 1996.

ozone concentration, and anticorrelation between pressure and ozone concentration. The correlation coefficients between wind velocity and ozone concentration are very small, which is in accordance with findings based on data from Svalbard (Solberg et al., 1994).

Fig. 6a shows the ozone sounding profile from Thule for 29 March 1995 (taken during a major depletion episode in 1995 very similar to the episode in March 1996; unfortunately there was no sounding from the episode in 1996). The concentrations are very low, about 6 ppb, in a

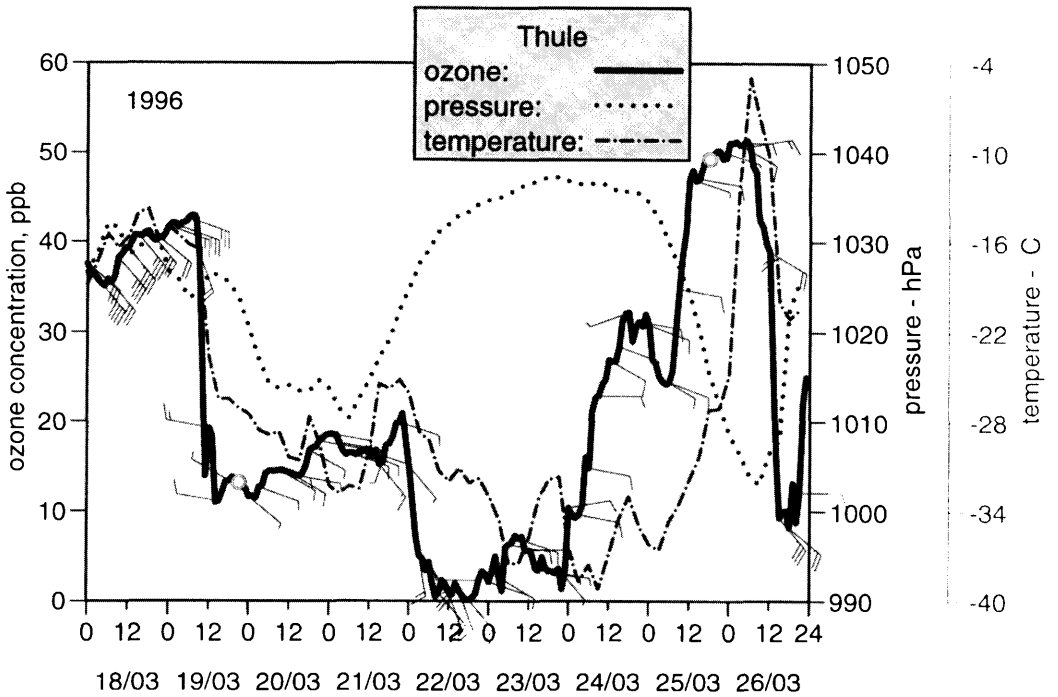

Fig. 5. Time series of surface ozone concentrations (ppb), pressure (hPa), temperature (°C) and wind (WMO plotting convention) at Thule for the period 18 to 26 March 1996. Meteorological parameters are taken from the synoptic station, 04202, at Thule Air Base.

Table 1. *Correlation coefficient between hourly mean surface ozone concentrations and the meteorological parameters: temperature, pressure, wind velocity and wind direction; Calculated for the month March and for the period March to May for 1995 and 1996; meteorological parameters are from synoptic stations near by*

	Temperature	Pressure	Wind-velocity	Wind-direction
Thu, 1995, March	−0.07	−0.03	0.16	0.04
Thu, 1995, March–May	0.10	−0.17	0.14	−0.02
Thu, 1996, March	0.52	−0.57	0.08	−016
Thu, 1996, March–May	0.25	−0.22	0.16	−0.05
Sco, 1995, March	0.30	−0.12	0.17	0.02
Sco, 1995, March–May	−0.15	−0.06	0.07	0.02
Sco, 1996, March	0.75	0.09	−0.18	0.00
Sco, 1996, March–May	0.26	0.02	−0.12	−0.05
SSt, 1996, March	0.60	−0.29	0.18	0.10
SSt, 1996, March–May	0.44	−0.03	0.08	0.08

well mixed layer up to about 250 m. Up to about 400 m, the concentration increases rather sharply to just below 20 ppb, slowly increasing to about 26 ppb up to 1350 m, then sharply increasing to above 40 ppb. The profile indicates downward flux of ozone from an entrainment zone at 250–400 m. The ozone destroying process is probably still active with positive concentrations of BrO.

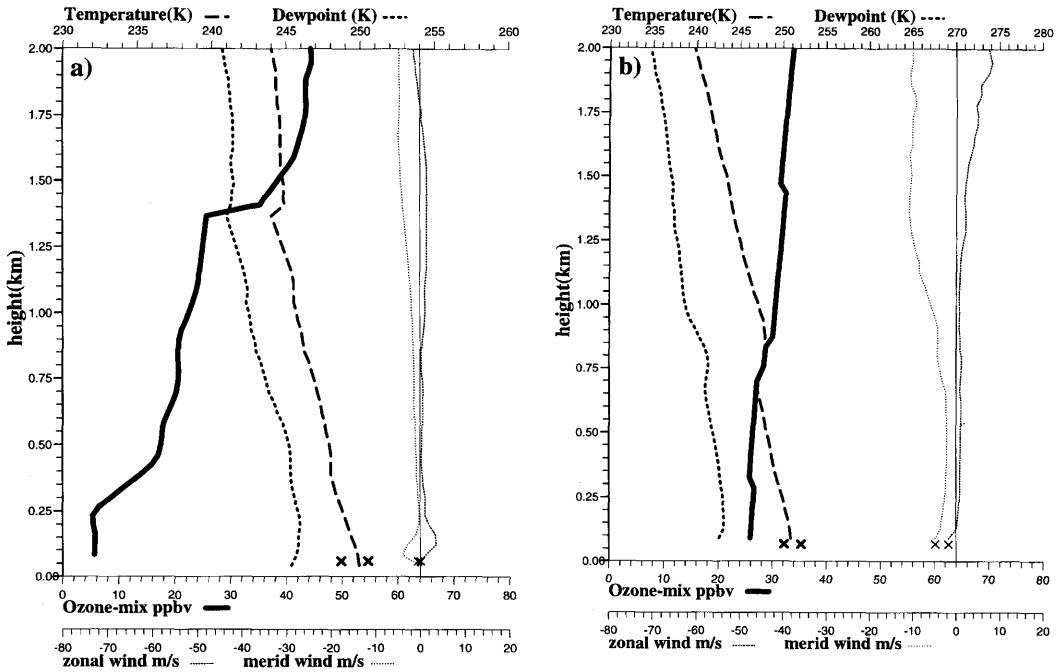

Fig. 6. Ozone soundings from (a) Thule for 29 March 1995 17:26 UTC and from (b) Scoresbysund for 23 March 1996 11:07 UTC. Profiles of temperature, dewpoint temperature, zonal, and meridional wind velocity are also shown.

In the ARCTOC measurement campaign at Ny-Aalesund, it was found that non-zero concentrations of ozone and BrO could co-exist for several days, while the chemistry with the measured level of BrO would indicate that the ozone should be destroyed in a short time.

The profile from Scoresbysund for 23 March 1996 is from the already mentioned episode affecting all 3 monitoring stations (cf. Fig. 3). It appears that the ozone in the whole column up to 2 km is fairly well mixed with concentrations slowly increasing from about 26 ppb to about 35 ppb at 2 km. The air mass trajectories (cf. Section 3) is from the East Siberian Sea over the Arctic Ocean passing north of the Canadian monitoring station Alert, and along the north-eastern coast of Greenland.

Inspection of several ozone profiles from Scoresbysund all show fairly well-mixed ozone up to at least 2 km. This can perhaps lead to the conclusion that the ozone destroying mechanism is not very effective along the coast of north-eastern Greenland, or alternatively that the vertical mixing near the monitoring station must be very strong (otherwise we should observe profiles with low concentrations near the surface, as in Thule).

3. Trajectory analysis

3.1. Description of the transport model

DMI's 3-D transport model is utilised for many purposes, e.g., as a part of the Danish Emergency Response Model for the Atmosphere (DERMA) (Sørensen and Rasmussen, 1995) and in the Danish Atmospheric Chemistry Forecasting System (DACFOS) (Jensen et al., 1996).

The trajectories are calculated by a standard iteration method (Kållberg, 1984) with a prescribed tolerance. The advection time step used is set to 15 min. The model can utilise data from the different versions of DMI-HIRLAM (see below) or from the global model at the European Centre for Medium-Range Weather Forecast (ECMWF).

Parameters along the trajectory are, e.g., horizontal wind, temperature, relative humidity, 10-meter horizontal wind, surface temperature, precipitation intensity, total cloud cover, surface

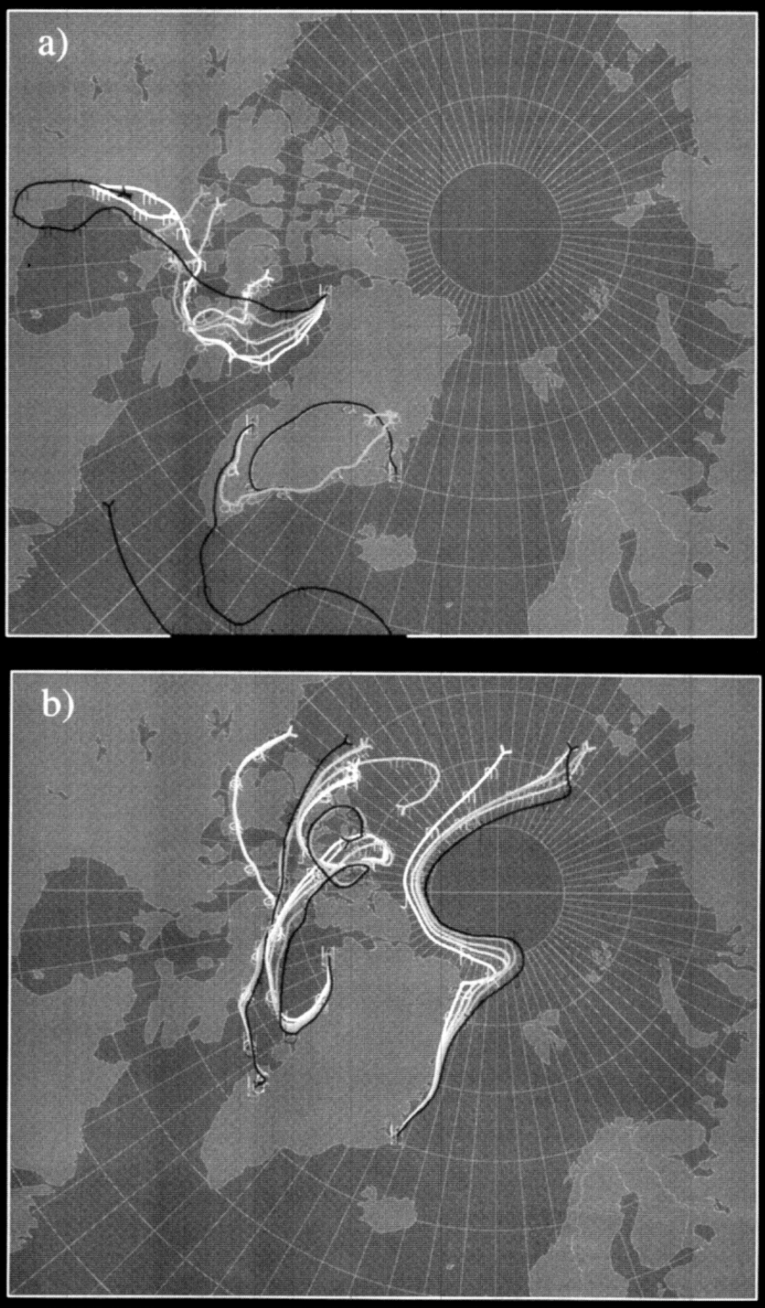

Fig. 7. Trajectories for Thule, Søndre Strømfjord and Scoresbysund for (a) 19 March, 00 UTC, 1996 and (b) 23 March, 00 UTC, 1996. The 5 trajectories for each station arrive at equidistantly distributed heights in the ABL above the receptor point; the lowest trajectory is black and the highest trajectory is white. The letters on the trajectories indicate the height of the trajectory relative to the ABL-height in situ: a <0.1, b: 0.1–0.2, ..., l: 1.1–1.2, m >1.2. The trajectories are marked each 12 h.

pressure, surface heat flux, surface momentum flux and the height of the Atmospheric Boundary Layer (ABL).

The ABL-height is calculated within the transport model from DMI-HIRLAM vertical profiles of temperature, wind and humidity using a bulk Richardson number method.

3.2. *The HIRLAM model*

The HIgh Resolution Limited Area Model (HIRLAM) (Källén, 1996) is a primitive-equation Numerical Weather Prediction (NWP) model. The horizontal grid is a regular spatially staggered latitude/longitude grid in a rotated spherical projection. The vertical coordinate is a terrain-following hybrid co-ordinate, which near the surface is identical with the sigma coordinate ($\sigma = P/P_{surface}$), and approaches the pressure P with increasing height.

The DMI-HIRLAM model, which is operational at the Danish Meteorological Institute (DMI), is run on two different limited areas (Sass, 1994). Data from a version covering Greenland and large parts of the North Atlantic, Canada and the Polar Basin has been used in this study. The horizontal resolution is 0.42° (46 km) and in the vertical, the model has 31 hybrid levels. The DMI-HIRLAM

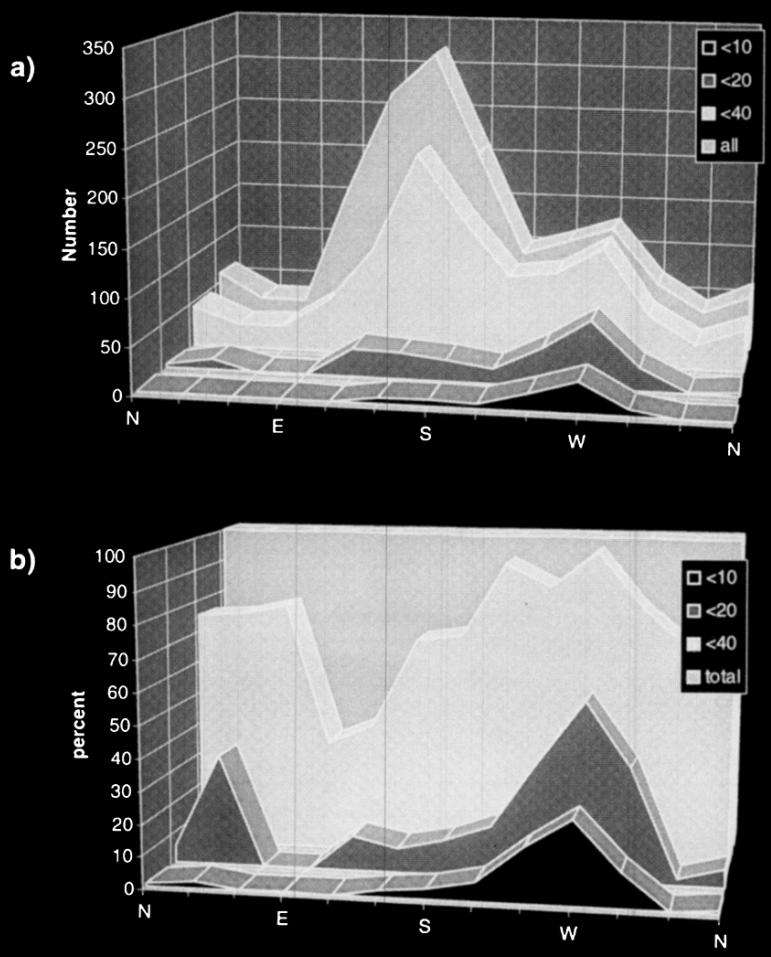

Fig. 8. Sector analysis of 72-h back-trajectories for Thule for the period March to April 1996. (a) The number of trajectories per sector (30°) for different ozone concentrations at receptor point; (b) as a, but as a % of the total number of trajectories in the sector.

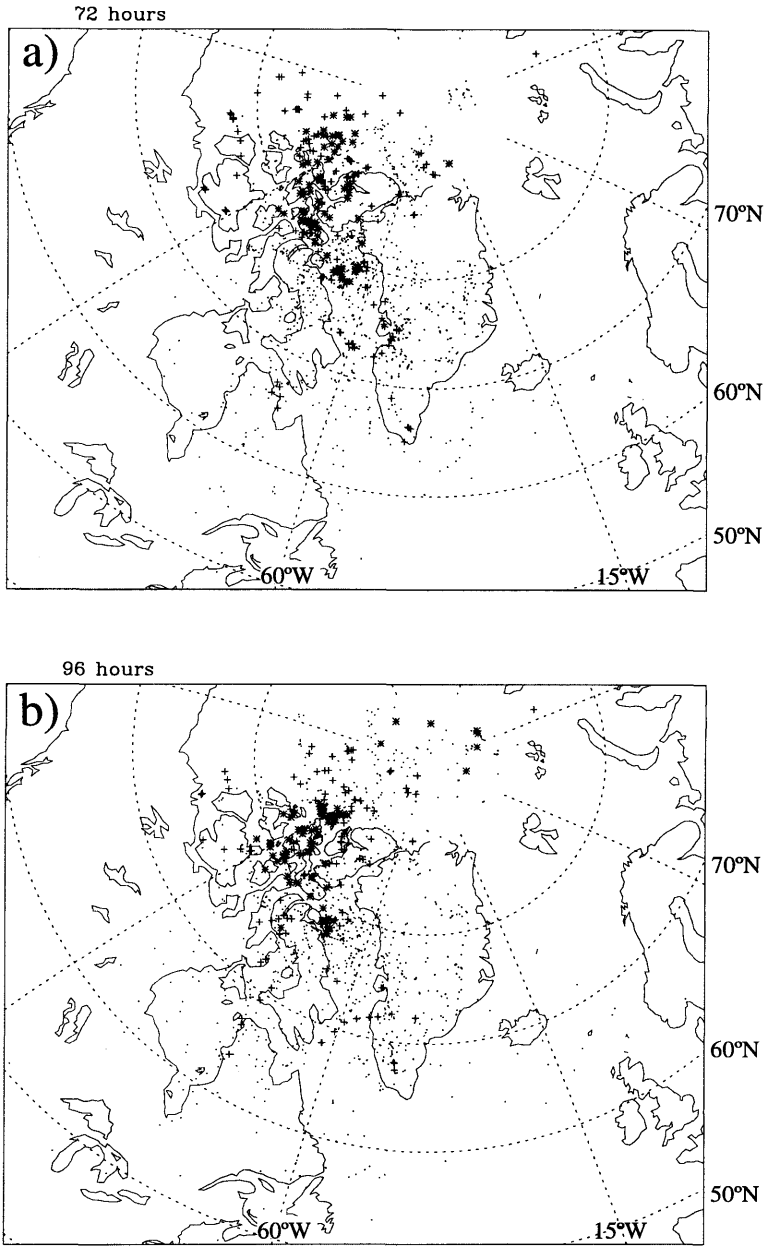

Fig. 9. Analysis of backward trajectories from Thule for the period March to May 1996, showing positions 72 h (a) and 96 h (b) before arriving at Thule. Positions are marked by * for observed ozone concentrations below 10 ppb, by + for concentrations between 10 and 20 ppb, and otherwise by a dot. Ozone concentrations are 6-h averages.

forecasting system consists of pre-processing, analysis, initialisation, forecast, post-processing and verification. Both model versions are run with their own 6-hourly data-assimilation cycle.

3.3. *Sector analysis*

In Fig. 7a,b, the trajectories for Thule, Søndre Strømfjord and Scoresbysund for 19 March 1996 and 23 March 1996 are shown. The two dates correspond to a non-depletion and a depletion episode, respectively (cf. Fig. 3). The 5 trajectories presented for each station are arriving at equidistantly distributed heights in the atmospheric boundary layer (ABL) over the receptor point. The length of the trajectories correspond to 5–6 days (provided they remain within the DMI-HIRLAM area) and the time resolution of the trajectory data is 15 min.

A sector analysis of the trajectories for the period March to May 1996 has been performed for Thule, Søndre Strømfjord and Scoresbysund. The analysis is based on 4 sets of trajectories each day (00, 06, 12 and 18 UTC), each set consisting of 5 backward trajectories arriving at the receptor point (e.g., Thule) at equidistantly-distributed heights between the ground and the top of the boundary layer.

In Fig. 8a,b, a result of the sector analysis of the 72-h backward trajectories for Thule for the period March to May 1996 is presented. The main part of the trajectories have their origin in the sector S–SE (Fig. 8a), but for the case of low ozone concentrations (<10 and <20 ppb), the largest number of trajectories come from the western sector.

In Fig. 9a,b, the start-positions of back trajectories, 72 h and 96 h respectively, for Thule for the period March to May 1996, are marked by symbols depending on the 6-h mean ozone concentration at the receptor point in Thule. It is clear from the figure that the main source region of low ozone concentrations going 72 h back (Fig. 9a) is the northern Canadian Archipelago. Going 96 h back, the conclusion is the same, but a number of low episodes now have their origin in the Polar Basin. Going further back (120 h and 144 h, not presented) gives a similar picture.

For Scoresbysund (not shown), the low ozone episodes (below 20 ppb) correspond to back-trajectories having their origin in the Polar Basin. For Søndre Strømfjord, the results are not so simple; the major part of the low ozone episodes (below 20 ppb) have their origin in the Canadian Archipelago, but some episodes have their origin around Hudson Bay, and others from the area south-west of Greenland.

4. Conclusions

The ozone measurements in Greenland have established the fact that the phenomenon of sudden arctic tropospheric ozone loss during spring also appears in Greenland. It is most pronounced at the west-coast of Greenland and generally confined to the period from March to May. Spatially, the phenomenon is very extended as a whole air mass from Thule to Søndre Strømfjord is seen to be affected by the phenomenon.

The major episodes are related to cold air masses moving from the Arctic Ocean north of the Canadian Archipelago across Baffin Bay and Davis Strait, and further crossing Greenland. Analysis of backward trajectories calculated by the DMI 3-D transport model utilising meteorological data from the numerical weather prediction model DMI-HIRLAM (High Resolution Limited Area Model) supports this finding.

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