JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 106, NO. D7, PAGES 7495–7503, APRIL 16, 2001

Arctic ozone loss in threshold conditions: Match observations in 1997/1998 and 1998/1999

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Abstract. Chemical ozone loss rates inside the Arctic polar vortex were determined in early 1998 and early 1999 by using the Match technique based on coordinated ozonesonde measurements. These two winters provide the only opportunities in recent years to investigate chemical ozone loss in a warm Arctic vortex under threshold conditions, i.e., where the preconditions for chlorine activation, and hence ozone destruction, only occurred occasionally. In 1998, results were obtained in January and February between 410 and 520 K. The overall ozone loss was observed to be largely insignificant, with the exception of late February, when those air parcels exposed to temperatures below 195 K were affected by chemical ozone loss. In 1999, results are confined to the 475 K isentropic level, where no significant ozone loss was observed. Average temperatures were some $8° - 10°$ higher than those in 1995, 1996, and 1997, when substantial chemical ozone loss occurred. The results underline the strong dependence of the chemical ozone loss on the stratospheric temperatures. This study shows that enhanced chlorine alone does not provide a sufficient condition for ozone loss. The evolution of stratospheric temperatures over the next decade will be the determining factor for the amount of wintertime chemical ozone loss in the Arctic stratosphere.

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Paper number 2000JD900653.
0148-0227/01/2000JD900653$09.00

1. Introduction

In the winter of 1994/1995, 1995/1996, and 1996/1997, the Arctic stratosphere experienced a series of unusually cold winters. Substantial chemical ozone loss was observed in these winters [*Manney et al.*, 1997; *Müller et al.*, 1997; *Rex et al.*, 1997, 1999; *Schulz et al.*, 2000a, and references therein], which was connected with large areas of synoptic scale temperatures that were low enough for polar stratospheric clouds (PSC) to exist. In contrast to this, the winter 1997/1998 was dynamically active, which led to higher stratospheric temperatures that only occasionally dropped below the threshold temperatures for PSC existence in small parts of the polar vortex [*Pawson and Naujokat,* 1999]. At the same time the polar vortex was much smaller than that in previous winters. Compared with preceding years, measured ozone values in winter 1997/1998 were higher and were much higher in the following Arctic stratospheric winter 1998/1999, which was even warmer. In both winters, the Match approach [*von der Gathen et al.,* 1995] which uses air parcels that are probed twice or more by ozonesondes was applied to determine chemical ozone destruction. These two warm winters provided the first opportunity to investigate Arctic chemical ozone loss with the Match technique under threshold conditions, where preconditions for chlorine activation and hence ozone destruction existed only intermittently.

2. Measurement Strategy and Analysis

The first passive Match analysis was made for the Arctic winter 1991/1992, where a large number of ozonesondes had been launched inside the polar vortex. Backward trajectories were calculated from these soundings in order to identify air parcels that had already been probed by an ozonesonde [*von der Gathen et al.,* 1995]. Since the winter 1994/1995, coordinated Match campaigns have been carried out each winter. During these campaigns, individual air parcels in different vertical levels are probed by an ozonesonde and then are tracked by means of forward trajectories. These near real time trajectories are calculated from European Centre for Medium-Range Weather Forecasts (ECMWF) horizontal wind field analyses and up to 3-day forecasts, and estimated diabatic cooling rates are used. If a trajectory passes over one of the participating stations, a second sonde is launched and a possible change in ozone mixing ratio within the air parcel can be detected. Each pair of soundings within the same air parcel is called a "match". In this way, chemical ozone loss is separated from dynamically induced ozone variations in the stratosphere.

In winter 1997/1998, a coordinated Match campaign was carried out in January and February. In total, 348 sondes were launched, with about 200 being inside the polar vortex. The stations that participated in the campaign are shown in Figure 1 (solid circles). The vortex

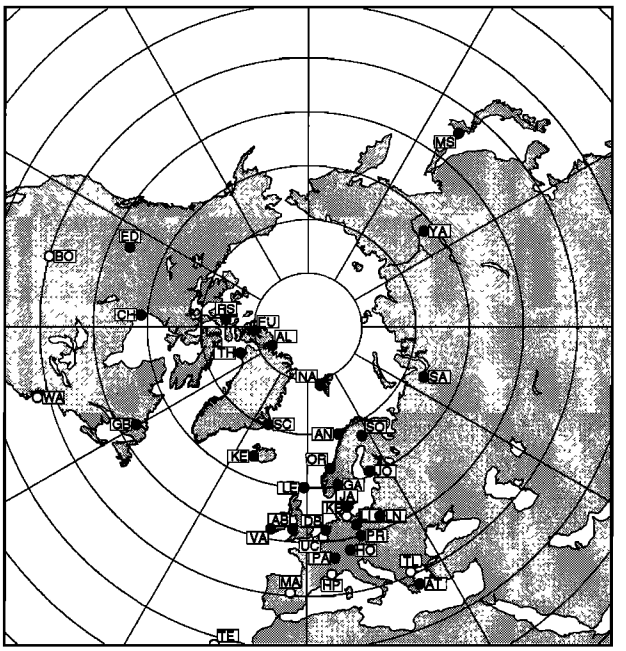

Figure 1. Map of all participating ozonesonde stations in the 1997/1998 and 1998/1999 Match campaigns. The solid circles mark the stations that participated in 1997/1998. In 1998/1999, the stations marked with open circles were also involved.

edge is chosen at $36\,s^{-1}$ normalized potential vorticity (PV) (see *Rex et al.* [1999] for a definition) corresponding to a value of $36 \cdot 10^{-6}\,K\,m^2\,s^{-1}\,kg^{-1}$ for Ertel's PV on the 475 K isentropic level for the winter 1997/1998, which is the same value as in former Match analyses.

In winter 1998/1999, the vortex was much weaker than in preceding years; so a vortex edge chosen at $36\,s^{-1}$ would be wrong by excluding large parts of the vortex. At 475 K, the highest gradient of PV in equivalent latitudes, averaged for January and February 1999, was determined to be $29.7 \pm 1.1\,s^{-1}$; so the vortex edge was chosen at $30\,s^{-1}$. This agrees with a study of *Kyrö et al.* [2000], who derived a vortex edge at 31 ± 3.8 (31.9 ± 3.5) potential vorticity units (PVU) in 475 K for January (February) 1999. The 1998/1999 campaign was carried out both inside and outside the polar vortex, with more than 900 sondes launched in total. All stations shown in Figure 1 participated in the campaign. Here only the results for inside the polar vortex are presented.

After the campaigns, the trajectories are recalculated from the analyzed wind fields, and diabatic descent rates from the SLIMCAT model [*Chipperfield,* 1999] are applied. For the final analysis, only matches that fulfill certain quality criteria are used [*Rex et al.,* 1999]. The present theory of polar stratopheric ozone loss indicates that chemical ozone loss occurs exclusively in sunlight; this was shown in former Match analyses. As a result, the loss rates are calculated by linear regression of the change of ozone mixing ratio versus the time the air parcel spent in the sunlight. Vortex-averaged loss rates

per day are obtained by multiplication of the loss rates per sunlit hour with the mean sunlit time per day inside the vortex. The error bars of the ozone loss rates given in Figures 2-6 represent the $1\,\sigma$ uncertainties of the regression coefficients and are purely statistical. They do not consider any possible systematic effects. A more detailed description of the method can be found in the work of *Rex et al.* [1999].

3. Results and Discussion

3.1. Winter 1997/1998

In Plate 1 the observed vortex-averaged ozone loss per day during January and February 1998 is shown. The thin lines are isonlines for A_{NAT}, the area with temperatures below T_{NAT} as calculated from ECMWF analyses. T_{NAT} was calculated after *Hanson and Mauersberger* [1988], using a HNO_3 profile as measured by the Limb Infrared Monitor of the Stratophere (LIMS) in January 1978 and assuming a constant H_2O mixing ratio of 4.6 ppmv. The results cover the vertical region from 410 K to 520 K between mid January and the end of February. Owing to a limited number of available ozonesondes in 1997/1998, the number of matches per day in the lower levels is less than that in former years, which makes it necessary for this analysis to include matches of a 20-day period for each regression instead of 14 days as was used in former Match analyses. Time resolution is therefore reduced, which might blur peak values. While little ozone loss was detected up to the end of January, loss rates at around 490 K increased at the beginning of February, and at the lower levels, toward the end of February. The maximum loss rate per day was 28 ± 8 ppbv/d at 450 K and 490 K at the end of February. This is lower than vortex-averaged values of previous Match campaigns, with maximum values between 40 ppbv/d and 60 ppbv/d at comparable vertical levels. The accumulated loss of ozone for the time period and vertical region shown in Plate 1 is 13 ± 7 Dobson units (DU). Even taking into account that the analysis is limited in time and vertical extent, this value is still low in comparison with accumulated ozone loss values determined in former years of 120-160 DU in 1994/1995 and 1995/1996 and 43 DU in 1996/1997 [*Schulz et al.*, 2000a, and references therein].

The highest vortex-averaged ozone loss rates per sunlit hour were observed in the third week of February on the 450 K and the 490 K levels with 2.8 ± 0.8 and 2.9 ± 1.1 ppbv/sunlit hour, respectively. These loss rates are also smaller than those in the preceding years, when maximum loss rates were found to be 10 ± 1 ppbv/h in 1994/1995, 10 ± 3 ppbv/h in 1995/1996, and 3.9 ± 0.8 ppbv/h in 1996/1997 [*Rex et al.*, 1997, 1999; *Schulz et al.*, 2000a].

In Figure 2 (top panel) the temporal evolution of the vortex-averaged ozone loss rate per day in the 475 K isentropic level is shown. Each data point includes

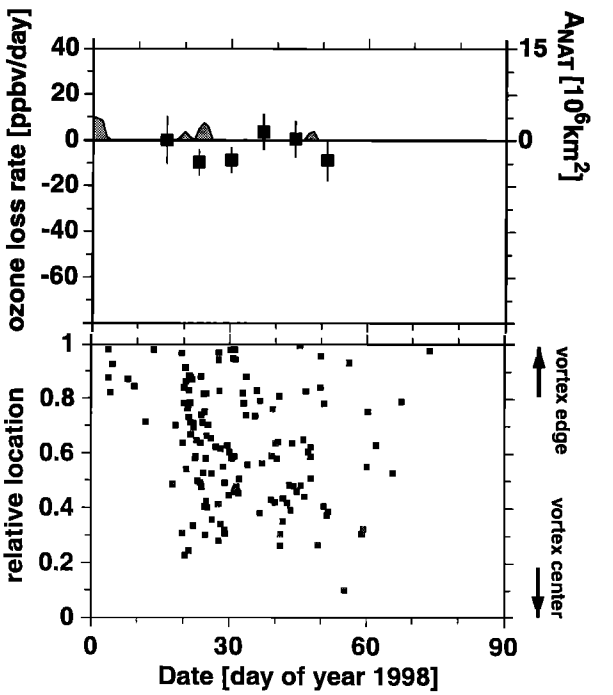

Figure 2. (top) Ozone depletion rates per day on the 475 ± 10 K potential temperature level as a function of time. Each data point represents a linear regression between ozone change and sunlit time of matches in a 14-day period around the given date. The shaded curve represents A_{NAT}, the area with temperatures below T_{NAT}. (bottom) Relative position of the corresponding match events inside the polar vortex. Zero refers to the vortex center (highest PV value), while 1 represents the vortex edge. On a given day, equal intervals on the scale correspond to equal area fractions of the vortex, while decreasing numbers represent increasing PV (e.g., 0.3 represents the PV isoline enclosing 30% of the vortex area).

matches of a ±7-day period around the given value. The shaded curve represents A_{NAT}. The bottom panel shows the relative location of the contributing match events inside the polar vortex. Between mid January and the end of February, matches at 475 K cover the polar vortex homogeneously (except for the innermost 20%); so the calculated loss rates can reasonably be regarded as vortex averages. At this level, the vortex-averaged loss rates do not exceed 10 ± 6 ppbv/d, which is barely statistically significant. For comparison, the same analysis for the 490 K isentropic level is shown in Figure 3. Here loss rates reach clearly statistically significant values in February.

Langer et al. [1999] deduced chemical ozone depletion from ground-based millimeter wave observations of ozone with a vertical resolution of 8 km, which at 475 K roughly corresponds to 150 K vertical resolution. Loss rates on the 475 K level were found to be not significant in December and the beginning of February. For the end of February they give a loss rate of 32 ± 10 ppbv/d or 4 ± 1.25 ppbv/sunlit hour. These values agree well with

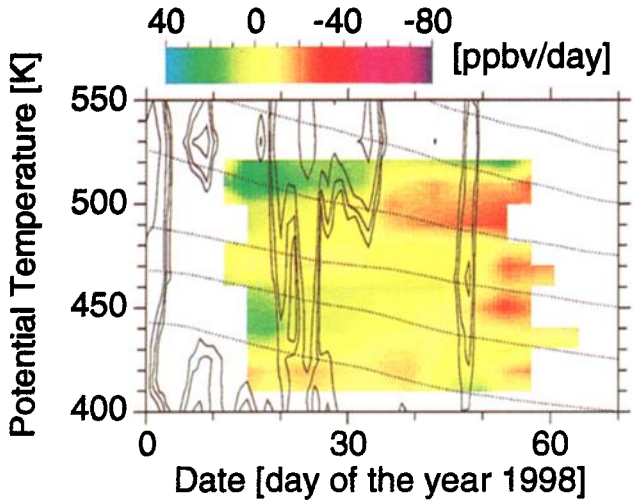

Plate 1. Ozone loss rates per day as a function of time and potential temperature. The thin solid lines indicate the 0.3, 0.7, 1.5, 4.0, and $8.0 \cdot 10^6$ km^2 isolines for the area with temperatures below T_{NAT} as derived from ECMWF analysis. The dashed lines show the diabatic descent (vortex average) of the air masses during the winter as determined by the SLIMCAT model.

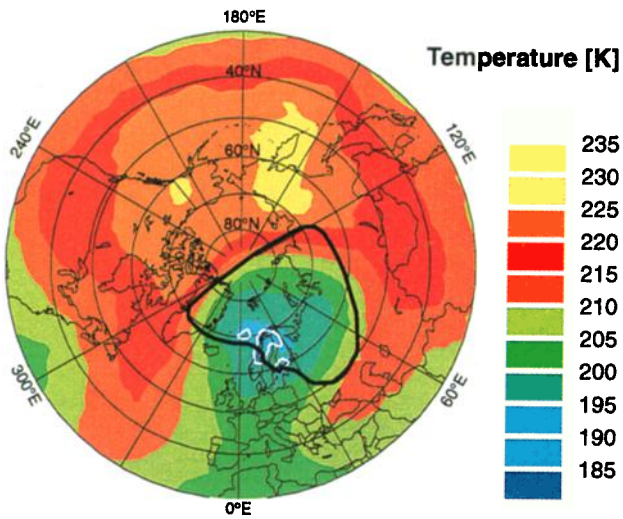

Plate 2. Synoptic temperatures at 475 K on February 18, 1998, from ECMWF analysis. The white line is the T_{NAT} isoline, and the black line marks the isoline of 36 s^{-1} normed potential vorticity, representing the vortex edge.

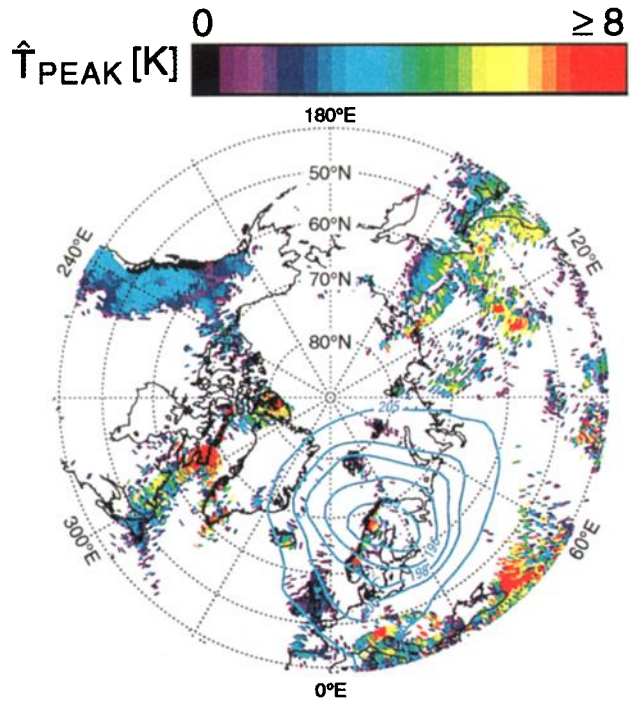

Plate 3. Map of the peak temperature amplitudes of mountain lee waves $\hat{T}_{PEAK}$ at 50 hPa on February 1998 at 1200 UT. The data are obtained by simulations with a mountain wave forecast model [*Bacmeister et al.*, 1994]. The blue contours are the synoptic temperature isolines (absolute temperature [K]).

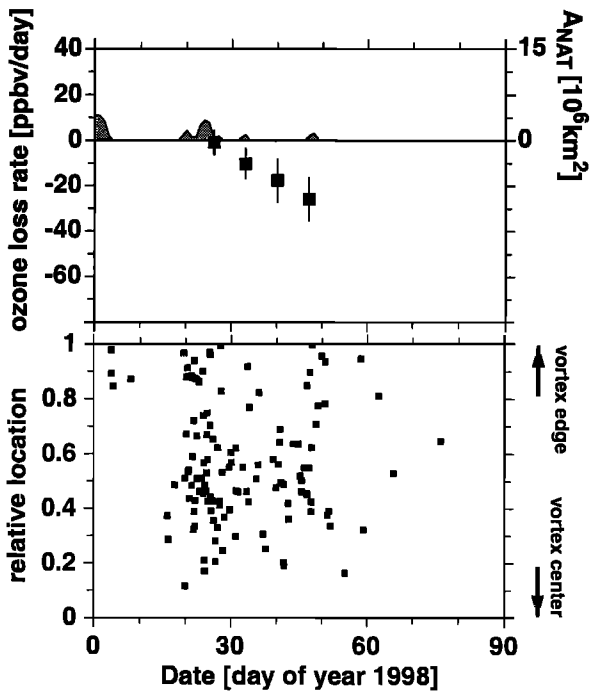

Figure 3. Same analysis as in Figure 2, but for the 490 ± 10 K potential temperature level.

the vortex-averaged rates we determine for the same period around 450 K and around 490 K.

To study the localization of the ozone depletion, the matches between February 10 and March 10 and between 430 K and 500 K potential temperature (covering the region with significant loss rates) were chosen and binned according to their relative position inside the vortex. In Figure 4 (top panel), the loss rates per sunlit hour calculated from this subset of match events are shown as a function of the relative location inside the vortex. The loss rates thus determined are small or zero throughout most of the vortex but significant at the vortex edge, where ozone loss rates of 6 ± 2 ppbv/sunlit hour are reached. In Figure 4 (bottom panel), the minimum temperatures (T_{min}) in the history of the corresponding air parcels are shown. T_{min} is defined as the lowest temperature found along a 10-day backward trajectory from the first sounding of the respective match and along the trajectory that links the two soundings. The averages of the minimum temperature are above T_{NAT} (dashed line) for all parts of the polar vortex, but individual air parcels experienced lower temperatures, especially at the vortex edge. Since the higher loss rates observed in February 1998 are found at the vortex edge, where mountain wave activity is to be expected owing to the geographical distribution of high mountains, the influence of mesoscale temperature reductions leading to PSC formation and hence ozone destruction as discussed by *Carslaw et al.* [1998] seems possible.

Figure 5 shows ozone loss rates for matches with different temperature histories. The open squares include matches from the beginning of January until February 10, and the solid squares include matches starting from February 10, which represent the same sample of matches as in Figure 4. Each match is assigned a minimum temperature T_{min} as defined above. Here ozone loss rates per sunlit hour are calculated for ensembles of matches with a minimum temperature inside a 2 K wide bin. The horizontal lines mark the temperature bin; the horizontal position of the markers describe the average value of the individual minimum temperatures. A clear correlation between the minimum temperature and the ozone loss rate is observed for the February data, with a maximum loss rate of 6 ± 1 ppbv/h for those air parcels that experienced minimum temperatures between 193 and 195 K. It should be noted that these temperatures are synoptic temperatures. Owing to possible mesoscale temperature fluctuations, the temperatures given are upper limits of the lowest temperatures experienced by the air parcels.

Matches obtained in January and the beginning of February (open squares) showed no ozone loss, including those with minimum temperatures between 193 and

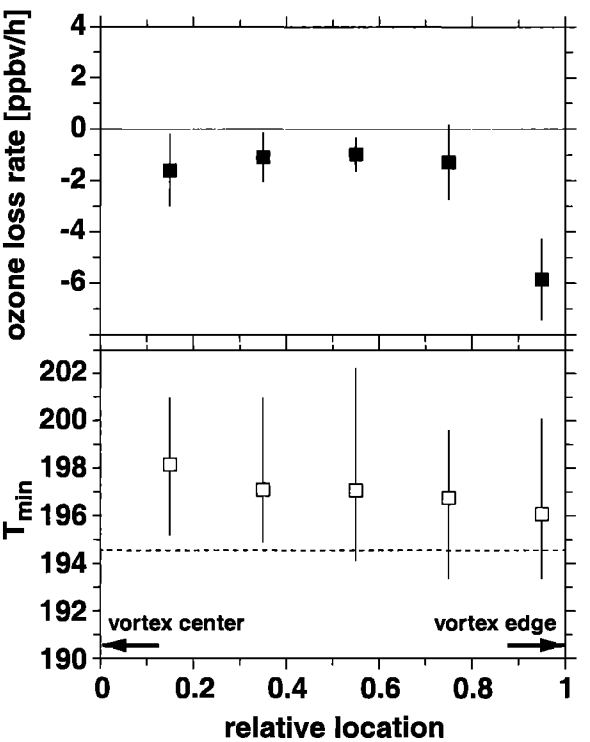

Figure 4. (top) Ozone loss rates per sunlit hour as a function of relative location (see Figure 2 for an explanation) inside the vortex. The data symbolize linear regressions of matches between February 10 and March 10, 1998, and between 430 K and 500 K, each of them including matches of a ± 0.1 range around the given relative location. (bottom) Corresponding average of the minimum temperatures T_{min} for air parcels contributing to loss rate calculations. For each matching pair of sondes, T_{min} represents the lowest temperature on a 10-day backward trajectory from the first sounding and the trajectory which links the two soundings. Vertical lines indicate the range of the T_{min} values.

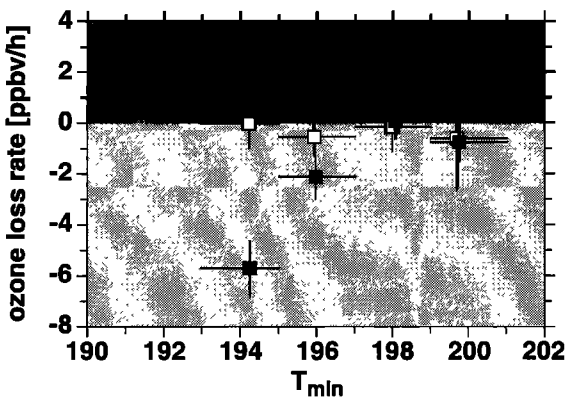

Figure 5. Ozone loss rates for different minimum temperatures T_{min} in the air parcel histories as defined in Figure 4. Open squares include the data from January 1 until February 10; solid squares include those from February 10 until the end of the campaign in mid March 1998. The horizontal position of the markers is determined by the average T_{min}; the horizontal line marks the limits of the T_{min} values included in the data point.

195 K in the 10-day history. The average time spent below 195 K for the 43 trajectories in the earlier period (open squares) with the lowest T_{min} is 12 hours, with 6 of those hours being between the two match soundings. This is comparable to the 13 trajectories in the later period (solid squares), that on average encountered temperatures below 195 K for 6 hours, 5 of them between the soundings. As was observed in other, colder Match winters, T_{min} values between 192 K and 195 K do not necessarily lead to major ozone loss at 475 K [*Schulz et al.*, 2000b], suggesting that mesoscale temperature fluctuations are needed to actually trigger ozone loss for T_{min} values in this range. The possible influence of orographic lee waves on the observed ozone loss in February is discussed below.

Eleven of the 13 air parcels with considerable ozone loss experienced their lowest temperatures between 480 K and 500 K potential temperature above Scandinavia between February 17 and February 19, either between the two ozone soundings or shortly before the first sounding. None of these 11 encountered temperatures below 195 K in the rest of the 10-day backward trajectory. In Plate 2 the synoptic temperatures at 475 K are shown for February 18. The lowest temperatures are observed at the vortex edge above Scandinavia, where the white line indicates the T_{NAT} isoline. In fact, PSCs were observed by Lidar in Andøya (northern Norway) on the night of February 16-17 (G. Hansen, private communication, 2000), and none were observed at this location on several other occasions in January and February. On February 17, PSCs were also observed at Sodankylä (Finland) and were between 18.5- and 19.5-km altitude, including a thin layer of solid PSC type I (R. Kivi, private communication, 2000). This altitude range corresponds roughly to 440-465 K, as inferred from radiosonde data, which is slightly lower than the ensemble of matches discussed here (480-500 K). However, the relation between geometrical height and potential temperature is highly variable within a lee wave event, which makes the comparison difficult.

It is therefore likely that this ensemble of air parcels experienced chlorine activation between February 17 and February 19 above Scandinavia. Model simulations with a mountain wave forecast model [*Bacmeister et al.*, 1994] indicate that between February 17 and February 19 the lowest temperatures at 50 hPa coincided with high mountain wave activity above the Scandinavian mountains, which was not the case on the days before or after. Plate 3 shows the simulation for February 18, 1200 UT, at the 50-hPa pressure level. This indicates that the true minimum temperatures seen by these air parcels may have been significantly lower than that indicated in Figure 5, and that lee waves might have been responsible for this observed ozone loss.

The corresponding data point of the early period (open squares) in Figure 5 showing no ozone loss includes 43 matches that are not confined to a small vertical region but are distributed vertically between 430 K and 500 K. The air parcels experienced their lowest temperatures at different times and locations and therefore cannot be brought in connection with any single lee wave event. However, mountain wave induced PSCs have been observed in January 1998 [*Behrendt et al.*, 2000; R. Kivi, personal communication, 2000], and the Match results at 475 K give indications for slight ozone loss in January (Figure 2); so it is likely that small-scale ozone loss confined to limited vertical regions also occurred in January.

3.2. Winter 1998/1999

The winter 1998/1999 was the warmest Arctic stratospheric winter examined with the Match technique so far. During most of the winter the stratospheric temperatures were well above T_{NAT}, and orographic lee wave activiy was weak and cannot be expected to have lowered the temperatures enough for PSCs to form. To our knowledge, the only PSC measurements were made on December 2 in Sodankylä [*Kivi et al.*, 2000] between 545 K and 590 K. Visual observations were made at Kiruna, Sweden during the December 1-3 period [S. Kirkwood, private communication, 2000]. Figure 6 (top panel) shows the temporal evolution of the vortex-averaged ozone loss rate per day in winter 1998/1999 at 475 K. No statistically significant ozone loss was observed. There was only one short period in mid February when temperatures were below T_{NAT} in the Northern Hemisphere. However, no T_{min} values below 195 K are found among the matches used for this analysis (Figure 6, middle panel), indicating that the small geographical region with synoptic temperatures below T_{NAT} was not sampled with Match during the time of interest. Small-scale ozone loss induced by this colder

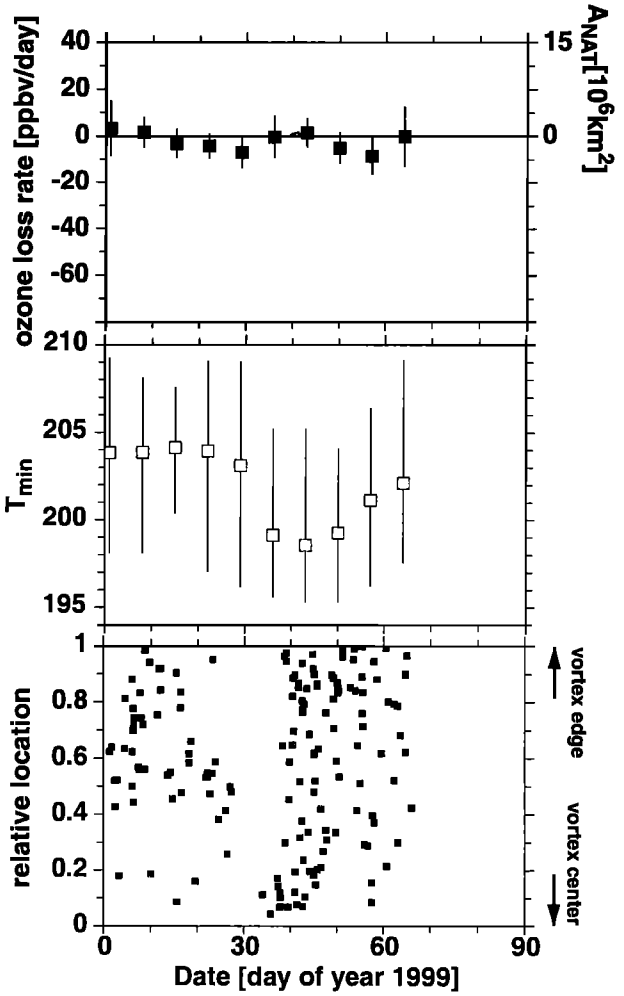

Figure 6. (top) Ozone loss rates per day in the 475±10 K potential temperature level as a function of time for early 1999. Each data point represents a linear regression between ozone change and sunlit time of matches in a 14-day period around the given date. (middle) Corresponding T_{min} values of air parcels. The vertical position of the marker is the average T_{min}, the vertical line represents the range of the individual T_{min} values. (bottom) relative position of corresponding match events inside the polar vortex (see Figure 2 for an explanation).

event may have been missed by Match. Still, the observation of no ozone loss stands out from the preceding winters with significant ozone loss and provides further experimental evidence that the present chlorine loading in the Arctic stratosphere does not necessarily lead to chemical ozone loss, provided the temperatures are high enough. At 475 K, the average temperature in the coldest 20% of the vortex during January, February and March 1999 was 204±5 K, which is 6 K higher than that in 1998 and 8-10 K higher than that in 1995, 1996 and 1997. These temperature differences remain even when other quantities such as the minimum temperature inside the vortex or the mean vortex temperature are compared.

Corresponding to the absence of major chemical ozone loss, the observed ozone mixing ratios in winter 1998/1999 were unusually high in comparison with recent preceding winters. The average mixing ratio in February and March at 475 K inside the polar vortex was 3.7 ± 0.3 ppmv and 3.8 ± 0.3 ppmv, as calculated from all sondes with a normed PV value higher than 30 s^{-1} and PV $\geq$ 36 s^{-1}, respectively. For comparison, the vortex-averaged (PV $\geq$ 36 s^{-1}) values for February and March of former years were 3.1 ± 0.4 ppmv in 1997 and 3.0 ± 0.3 ppmv in 1998.

4. Conclusion

The 1997/1998 Match results give a detailed picture of chemical ozone loss rates in the Arctic stratosphere with respect to time and altitude and to relative location inside the vortex. In 1997/1998 stratospheric temperatures were relatively high. They were close to T_{NAT} for a long period and occasionally dropped below T_{NAT} for a couple of days. This situation offered the unprecedented opportunity to study the chemical ozone loss under threshold conditions. The vortex-averaged ozone loss rates were barely significant for most of the winter throughout most of the vortex. Some clearly significant ozone loss occurred in February mainly confined to the vortex edge. This may have been connected to local PSC formation in lee wave events. The highest loss rates in February were observed in those air parcels with minimum temperatures below 195 K between the two soundings or in a 10-day history prior to the first ozonesonde. In 1998/1999, temperatures stayed well above T_{NAT} and no significant ozone loss was observed. Compared with earlier winters during the 1990s, the absence of substantial ozone loss during these two relatively warm winters shows how sensitive the chemical ozone loss is to changes in stratospheric temperature. As a rough estimate, the Match results show that a mean stratospheric temperature increase of 2-4 K inside the polar vortex can account for the difference between severe ozone loss and ozone loss that is confined to sporadic events such as those in winter 1997/1998, while a temperature increase of 8-10 K with respect to a situation with substantial ozone loss led to no chemical ozone depletion. These observations support the hypothesis that during the next decades, while the chlorine loading is still expected to be high [World Meteorological Organization, 1999], the evolution of the stratospheric temperatures will be the determining factor for the amount of wintertime chemical ozone loss in the Arctic stratosphere.

Acknowledgments. We are grateful to the operating staff of all participating stations for having made these campaigns possible. We thank H. Deckelmann, AWI Potsdam, for computer work, as well as the the European Centre for Medium-Range Weather Forecast (ECMWF) and the German Weather Office (DWD) for providing meteorological data. This work was supported by the European Commis-

sion within the project THESEO-O_3loss, contract ENV4-CT97-0510, and by national agencies of all the institutions involved. THESEO publication 33.

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(Received June 13, 2000; revised September 19, 2000; accepted September 29, 2000.)