

Climatology of UTLS ozone and the ratio of ozone and potential vorticity over northern Europe

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[1] Annual and interannual variations of ozone in the upper troposphere and lower stratosphere (UTLS) region have been studied using ozonesonde data collected between 1994 and 2001 at several northern European stations. The climatology of ozone exhibits a prominent annual cycle in the UTLS region. The observed change in the phase of the annual cycle from late spring-early summer at 500 hPa to spring at 200 hPa and to winter-early spring at 100 hPa shows the switching of the ozone control from photochemical to dynamical. Traces of interannual variation in the lower stratosphere are seen not only in the upper troposphere but also in the middle troposphere (not necessarily always) indicating the dynamical influence on tropospheric ozone budget. Further, the correlation between ozone mixing ratio and potential vorticity (PV) is studied at three northern high-latitude stations. As expected, a good correlation is found in the lower stratosphere, while the correlation is fair in the middle troposphere, except during summer over the European Arctic. This weak correlation at high latitudes indicates the dominance of photochemistry over dynamics in the presence of prolonged hours of solar illumination. The correlation coefficients derived at high latitudes are smaller than those reported at midlatitudes. This could be due to the greater number of tropopause folds at midlatitudes than at high latitudes and this eventually leads to the conclusion that the downward cross-tropopause flux is greater at midlatitudes than at high latitudes. Absence of a significant north-south gradient in the ozone/PV ratio in the lower stratosphere suggests that a single ozone/PV ratio (however, the ratio varies with month) can be used to convert global PV fluxes to ozone fluxes. A few cases of tropopause folds (only one case study is reported in the present study) are selected and studied in detail with the help of a very high frequency radar and meteorological analysis. The ratio between ozone and PV for these case studies agrees reasonably well with the climatological ratios. *INDEX TERMS:*

0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334); 3334 Meteorology and Atmospheric Dynamics: Middle atmosphere dynamics (0341, 0342); 3362 Meteorology and Atmospheric Dynamics: Stratosphere/troposphere interactions; 6952 Radio Science: Radar atmospheric physics; *KEYWORDS:* UTLS ozone, potential vorticity, tropopause folds

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1. Introduction

[2] There has been growing interest in the upper troposphere and lower stratosphere (UTLS) region in the atmospheric scientific community over the past few years for various reasons. They include concerns about increase in emissions, in particular NO_x , from subsonic and supersonic aircraft, which normally fly in this region, and variations of

ozone trends with height as well as spatial variations. Different three-dimensional (3-D) global chemistry and transport models have estimated an NO_x perturbation of 20–70% in the 8–12 km layer, causing an increase in O_3 concentration by 2–9% through photochemical reactions [Schumann, 1995]. Observed trends in ozone show interesting features with large decreases in the lower stratosphere at high and midlatitudes in both hemispheres, while the trends in the troposphere show regional differences with a positive trend over Europe, a negative trend over Canada, and no statistically significant trend over United States

[Logan, 1994; Logan *et al.*, 1999; Oltmans *et al.*, 1998; Staehelin *et al.*, 2001]. Further, now it is known that ozone-depleting heterogeneous and multiphase reactions occur in the lower stratosphere in midlatitudes and high latitudes with peak ozone losses between 15 and 22 km. The World Meteorological Organization (WMO) has reviewed the current state of the ozone problem through a series of assessments [WMO, 2002, and references therein]. However, most of the results on trends and seasonal variation of ozone are based on the data collected at midlatitudes and Canadian Arctic stations. There are no reported reliable long-term measurement series of ozone in the UTLS region over the European Arctic. Most of our understanding on European Arctic ozone is based on results from a series of large campaigns, for instance, European Arctic Stratospheric Ozone Experiment (EASOE), Second European Stratospheric Arctic and Midlatitude Experiment (SESAME), THESEO, SOLVE/THESEO 2000, conducted in the European Arctic augmented by some other small campaigns. Talaas and Kyrö [1992] reported characteristics of ozone and temperature at various levels using 2 years of ozone soundings from Sodankyla. Fortuin and Kelder [1998] and Logan [1999a, 1999b] used 3–4 years of two European Arctic soundings stations (Ny-Ålesund and Sodankyla) data while preparing a global climatology of ozone to test 3-D models. However, several years are needed to provide adequate statistics for monthly mean values. A minimum of 20 soundings are required for 95% confidence intervals of the ozone monthly means to be less than $\pm 30\%$ near the extratropical tropopause [Logan, 1999a]. So in this paper, an attempt has been made to study ozone variations (annual as well as interannual) over northern Europe using a database of 8 years.

[3] Further, stratosphere-troposphere exchange (STE) processes play a major role in altering the distribution of ozone in stratosphere as well as in troposphere. For instance, chemicals of importance for ozone-depletion processes, such as chlorofluorocarbons (CFCs), enter the stratosphere from the troposphere at low latitudes and descend while advecting isentropically to the poles through the Brewer-Dobson circulation. On the other hand, transport from stratosphere to troposphere constitutes not only an important removal mechanism of many stratospheric species but also a significant input of ozone into the troposphere. Holton *et al.* [1995] argued that this global-scale meridional circulation is driven by wave-induced forces, which exerts a nonlocal control over the transport of mass across lower stratospheric isentropic surfaces as a kind of fluid dynamical suction pump. In contrast to this slow global-scale diabatic circulation, faster alternative processes of STE are associated with meteorological phenomena, such as penetrative cumulus convection, tropopause folds, which occur along the edges of upper level troughs, and cutoff lows [Price and Vaughan, 1993; Ancellet *et al.*, 1994]. Various issues related to STE have been reviewed by Holton *et al.* [1995].

[4] It is generally accepted that much of the ozone transport from the lower stratosphere into troposphere occurs in tropopause folds [Price and Vaughan, 1992]. However, their episodic nature makes them difficult to assess. It was Danielsen [1968], who found a good correlation between ozone mixing ratio and potential vorticity (PV) and demonstrated that PV can be used as a tracer of

Table 1. Location of the Stations and the Total Number of Ozonesoundings Used for the Present Study

Station	Latitude	Longitude	Total Number of Soundings
Lerwick	60°07'N	1°10'W	533
Jokioinen	60°49'N	23°30'E	79
Kiruna	67°31'N	20°08'E	74
Sodankyla	67°22'N	26°38'E	496
Scorebysund	70°28'N	21°57'W	218
Bear Island	74°25'N	19°04'E	65
Ny-Ålesund	78°55'N	11°52'E	657

stratospheric air in the troposphere even when that air is only a component of a mixture. The good correlation between ozone mixing ratio and PV is plausible as both have similar gradients in UTLS region and this correlation holds both synoptically [Danielsen, 1968; Danielsen *et al.*, 1987; Browell *et al.*, 1987] and statistically [Danielsen, 1985; Beekmann *et al.*, 1994]. Gidel and Shapiro [1980] estimated the stratospheric part of tropospheric ozone by converting PV fluxes to ozone fluxes using zonal mean PV (from general circulation model (GCM)) and zonal mean ozone values. Beekmann *et al.* [1994] built a climatology of ozone/PV ratio at Observatoire de Haute Provence (OHP), southern France (44°N, 6°E), and found some discrepancy between these ratios and zonal mean ratios of Gidel and Shapiro [1980], suggesting that constructing a climatology of ozone/PV ratio for each vertical ozone sounding station is more meaningful than using zonal mean ozone/PV ratio. In the present study, the correlation between ozone and PV is studied in more detail, and a climatology of ozone/PV ratio is constructed for three northern European stations using 8 years of ozonesounding data. In addition, few events of tropopause folds are studied in detail, and the ratio between ozone mixing ratio and PV is estimated for these case studies to validate the climatological ratio. However, only one case study is reported in this paper.

[5] This paper is organized as follows. In section 2, the data used for the present study and data analysis are briefly described. Annual and interannual variations of ozone are discussed in section 3. Correlations between ozone and PV are studied in section 4, which also contains climatology of ozone/PV ratio for three northern European stations. In section 5, a brief description of a tropopause folding event is presented. Section 6 summarizes the present study and contains some conclusions.

2. Data Analysis

[6] The data used for the present study are primarily measurements of the vertical distribution of ozone using ozonesondes. The ozonesondes are the only archives of ozone measurements of the best available data quality in the UTLS region. Ozonesonde data are obtained mainly from two sources: Norwegian Institute for Air Research (Norsk Institutt for Luftforskning (NILU)) and World Ozone and Ultraviolet Data Center (WOUDC). Eight years of data from 1994 to 2001 at seven northern European ozonesonde stations (Table 1) are considered for the present study. Figures 1a and 1b show the location of the selected stations and number of soundings per month used for the present study. As shown in Figure 1b, the data are not uniform for

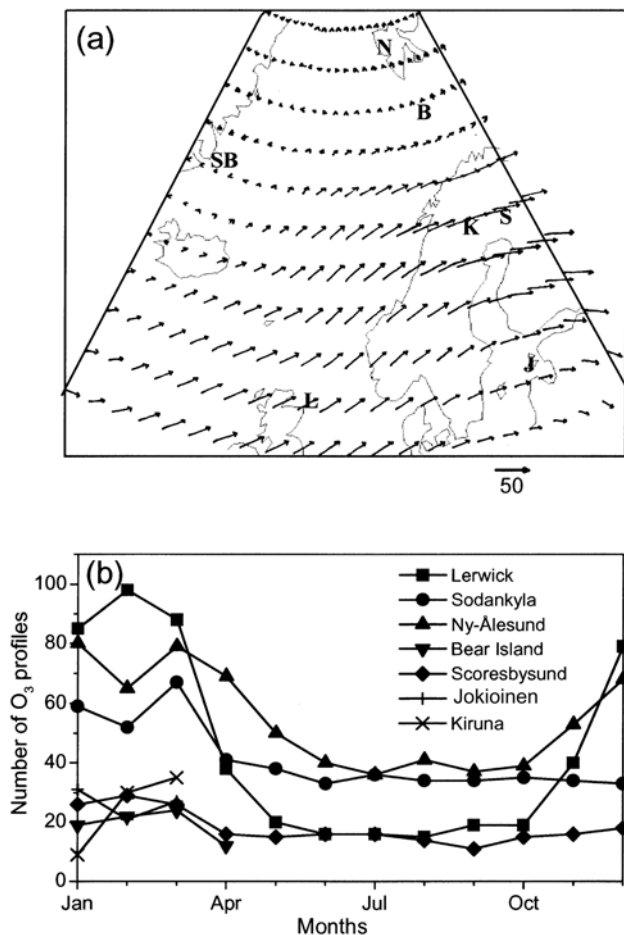


Figure 1. (a) Location of ozonesonde stations used for the study (L, Lerwick; J, Jokioinen; K, Kiruna; S, Sodankyla; SB, Scoresbysund; B, Bear Island; N, Ny-Ålesund). Also included is the wind pattern on 200 hPa surface observed on 23 February 1997. (b) Number of available ozonesonde profiles in each month.

all the months and for all the stations. Lerwick, Sodankyla, Scoresbysund, and Ny-Ålesund stations have somewhat regular program of at least one ascent per week with few exceptions (for example, only two to three soundings are available in May–October period for some years in Lerwick and in most of the months in Scoresbysund). Larger numbers of soundings are available for most of the stations during the November–April period. Data are available only in January, February, and March for Kiruna, Jokioinen, and Bear Island stations, and most of the ozone measurements at these sites have been made during campaigns. So these measurements are more representative of the years in which campaigns have been conducted. However, these measurements are included in the present analysis to see how well they represent the annual cycle of ozone obtained from long-term ozone measurements.

[7] Vertical profiles of ozone are obtained for all the stations using the electrochemical concentration cell (ECC) ozonesonde. Vaisala PTU sondes are flown simultaneously with ozone sensors to measure pressure, temperature, and relative humidity. Several studies have proved that ECC

sondes are more stable than any other sondes and can measure ozone with an accuracy of $\pm 5\%$ in the troposphere and the stratosphere [Komhyr *et al.*, 1995]. The estimated average correction factor (CF) (ratio between total ozone measured by standard instrument and the sum of integrated ozone from balloon measurements and residual ozone from climatological tables) for Sodankyla and Ny-Ålesund is very close to 1 with a standard deviation of 0.05, which indicates that the stability is, indeed, very high for this type of sondes. The residual ozone is estimated from the climatological tables of ozone residuals from solar backscatter ultraviolet (SBUV) measurements [McPeters *et al.*, 1997]. Dobson/Brewer spectrophotometers and Total Ozone Mapping Spectrometer (TOMS) (preference is given in the same order) measurements of total ozone are used to estimate CF. Simultaneous total ozone measurements are available for more than 50% of the ozonesonde profiles. However, the correction has not been applied to ozone profiles, since this procedure mainly depends on the accuracy of the ozone measurement in the stratosphere, where 90% of the ozone resides, and the accuracy of total ozone measurements. Error in any of those measurements will distort the ozone profile.

[8] European Centre for Medium-Range Weather Forecasts (ECMWF) data are used to estimate PV and for trajectory analysis. ESRAD, a very high frequency (VHF) radar, located at Erange, Kiruna, Sweden, is also used mainly to identify tropopause folds. More details of PV estimation and a brief description of the radar are given in the following sections.

3. Annual and Interannual Variations

[9] For the present study, climatological means of ozone mixing ratio are calculated using pooled data (for example, January mean is the average of all the profiles in January irrespective of year). Standard deviations, σ , and standard errors, $SE = \sigma/\sqrt{N}$, where N is total number of profiles, are evaluated at different pressure levels. Climatological means are also evaluated using monthly means of all years to check whether large weights are given to months having large number of sondes. However, the difference between these two means is found to be within the statistical error. Seasons are divided as follows: December–February: winter; March–May: spring; June–August: summer; and September–November: fall. These notations are followed throughout the paper.

[10] Monthly variations of ozone mixing ratio (in parts per billion by volume) at different pressure levels in the UTLS region are shown in Figure 2. It shows ozone variation in the course of the annual cycle from 500 to 100 hPa and also in space from about 60°N to 80°N . Some of the features observed in the ozone distribution are similar to those already reported by others [Logan, 1999a, 1999b, and references therein]. The annual cycle at 500 hPa clearly shows a late spring-early summer maximum at all the stations under consideration. This maximum could be attributed to a significant photochemical production of ozone during this period associated with anthropogenic sources of NO_x , CO, and hydrocarbons [Logan, 1985]. Further, equal monthly mean ozone mixing ratios (within the statistical error) at these sites indicate the importance of long-range transport in transporting ozone and its precursors to as north

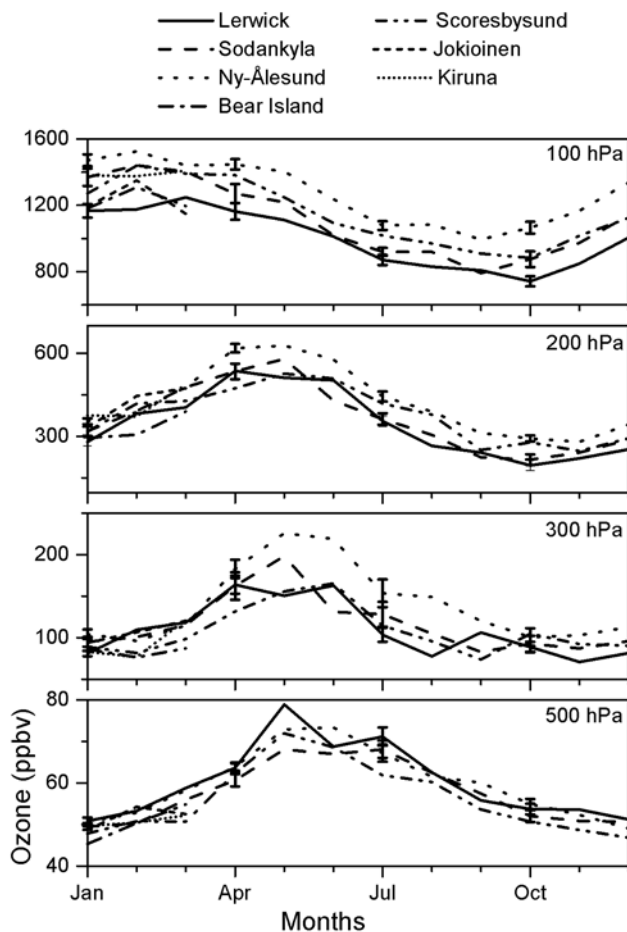


Figure 2. Annual cycle of ozone at different pressure levels and for different stations in the UTLS region. Error bar denotes 2 SE.

as 79°N (Ny-Ålesund). The observed annual cycle of ozone mixing ratio is similar to that seen at European midlatitudes, while it is different from that seen at Canadian Arctic station, Resolute [Logan, 1985; London and Liu, 1992; Beekmann et al., 1994; Fortuin and Kelder, 1998; Logan, 1999a]. The annual cycle at the Resolute shows a spring maximum, thought to be associated with large influx from stratosphere during winter and spring [Logan, 1985]. At 300 hPa, higher ozone values are seen at northernmost station (Ny-Ålesund) throughout the year, except for winter. This is due to, at least a part, the higher frequency of sampling stratospheric air at 300 hPa at high latitudes. A shift in the ozone maximum to April–May is seen at 200 hPa, as also observed by Logan [1999a]. A clear positive gradient of ozone with latitude is seen during most of the year at 100 hPa with high values of ozone mixing ratio at Ny-Ålesund followed by Scoresbysund, Sodankyla, and Lerwick. A broad winter-early spring maxima is seen at 100 hPa as a result of the stratospheric circulation outlined in section 1, which peaks in winter. The variation of ozone at Kiruna, Jokioinen, Scoresbysund, and Bear Island, in general, agrees well with that obtained from the long-term database.

[11] Figure 3 shows the interannual variability of ozone at different pressure levels at Sodankyla and Ny-Ålesund. The number of profiles are less than four for most of the

months, particularly in summer, in Lerwick, thus they may not provide meaningful monthly mean profiles and are omitted in the present analysis. Even at the other two stations, the monthly means are more valid in winter as the number of profiles is more than 8–10 in a month. The interannual variability is mainly driven by variations in planetary wave forcing of stratospheric circulation. Fusco and Salby [1999] found a good correlation between EP flux, a measure of the wave driving of the diabatic circulation, and total ozone in the extratropics. A part of the interannual variation in ozone can be related to solar cycle, quasi-biennial oscillation (QBO) and North Atlantic Oscillation (NAO) [refer WMO, 2002; Logan et al., 1999; Staehelin et al., 2001]. From Figure 3, it can be seen very clearly that ozone mixing ratio in 1998, 1999, and 2001 is higher than in other years in most of the months (particularly in winter and spring). In contrast, the ozone values in 1996 and 1997 are less in most of the months. The high ozone in 1998 and 1999 and low ozone in 1996 and 1997 are a result of the strong and weak wave-driven Brewer-Dobson circulation, respectively [Newman et al., 2001]. The stronger wave forcing will not only bring more ozone from source regions but also disturbs the polar vortex which eventually leads to a warm stratosphere with less chemical ozone depletion. On the other hand, reduced wave driving will lead to cooler polar temperature, stable

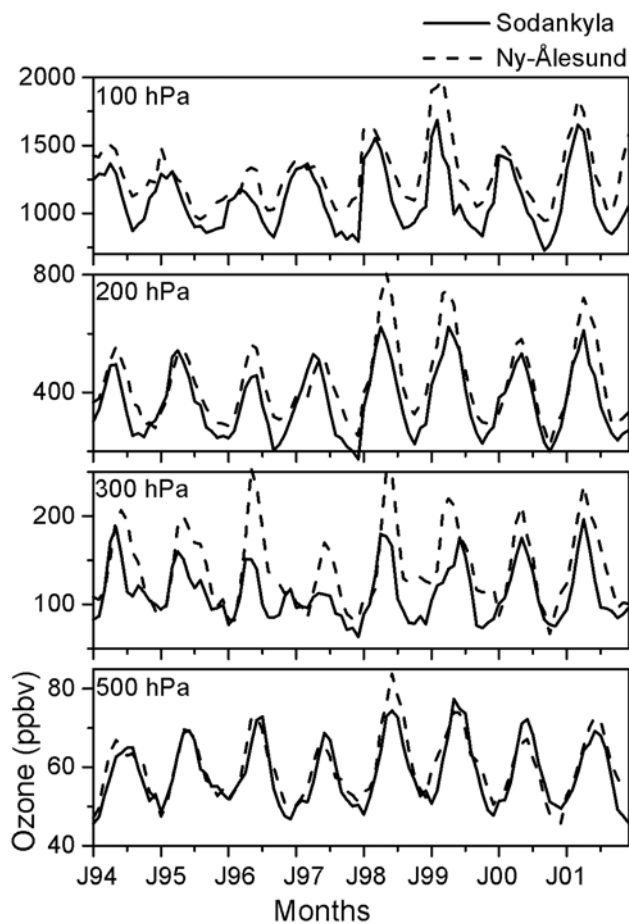


Figure 3. Time series of monthly mean ozone showing interannual variations at selected pressure levels in the UTLS region at Sodankyla and Ny-Ålesund.

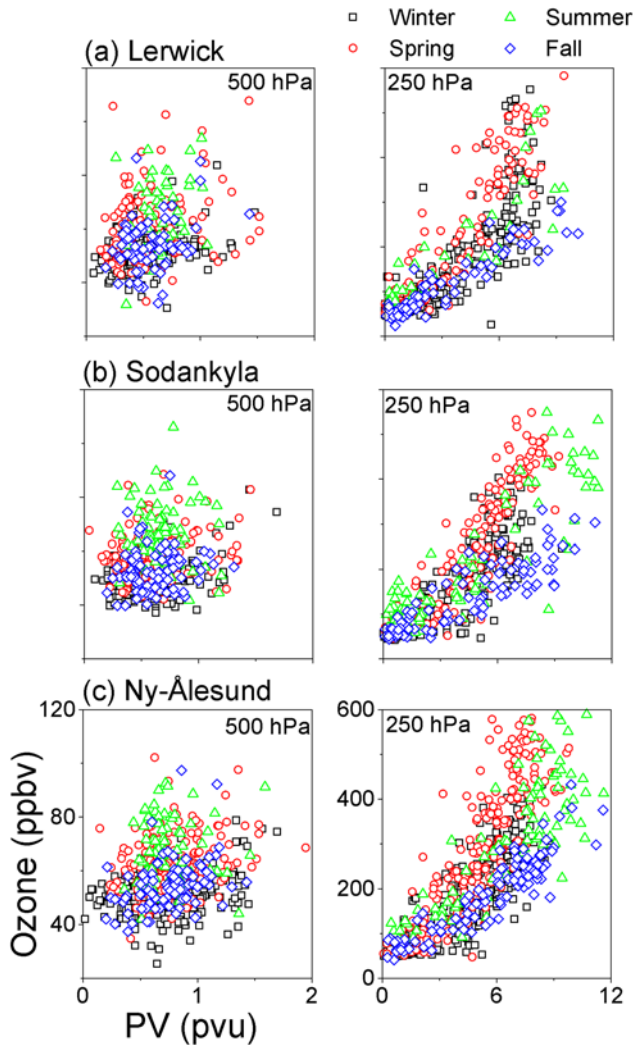


Figure 4. Scatterplots between ozone and potential vorticity at (a) Lerwick, (b) Sodankyla, and (c) Ny-Ålesund at (left) 500 hPa and (right) 250 hPa for different seasons.

vortex, and more chemical ozone loss [Newman *et al.*, 2001]. Similar large interannual variation of ozone (total as well as local ozone) was observed in 1990s not only at the two stations shown in Figure 3 but also at other Arctic stations [Anderson, 1999; Chipperfield and Jones, 1999; WMO, 2002, and references therein]. This indicates that these interannual variations, on average, are associated with large-scale planetary wave-driven circulation. Further, similar interannual variations are often seen in the lower stratosphere and in the upper troposphere, a few occasions even at 500 and 700 hPa (not shown in the figure), indicating a strong dynamical influence even at these heights. It is plausible that the decrease (increase) in stratospheric ozone could contribute a smaller (larger) stratospheric source to the tropospheric ozone budget.

4. Correlation Between Ozone and PV

[12] Potential vorticity, a conserved dynamical parameter in the absence of diabatic and frictional forces, is the product of static stability and absolute vorticity, $PV =$

$-g(\partial\theta/\partial p)$ ($\zeta_\theta + f$), where ζ_θ is the relative vorticity evaluated on the surfaces of potential temperature, θ , and f is Earth's vorticity.

[13] As already discussed earlier, both PV and ozone have similar distributions and gradients in the lower stratosphere and thus are highly correlated. A positive correlation between PV and O_3 can be expected in the troposphere during the process of the tropopause fold. However, there are certain other factors, which will effect the correlation between PV and O_3 . They include mainly (1) PV change due to radiative cooling at the tropopause, heating at the surface, and latent heating (cooling) due to water vapor condensation (melting of ice or evaporation of water) and (2) photochemical ozone production [Beekmann *et al.*, 1994].

[14] For this study, PV values have been evaluated at each station and at each pressure level using the ECMWF (T106 spectral data) wind and temperature fields. The horizontal resolution of the analysis is 1.25° latitude \times 1.25° longitude, and the vertical resolution is about 1.5 km. First, PV values are estimated on isentropic surfaces at model grid points and later interpolated to pressure surfaces and selected stations. Figure 4 shows the scatterplots between ozone and PV at 500 and 250 hPa for Lerwick, Sodankyla, and Ny-Ålesund. The correlation of ozone and PV is studied using linear regression analysis. The results of the linear regression analysis are tabulated in Table 2. It can be seen from Figure 4 and Table 2 that a strong correlation, with correlation coefficient >0.8 (significant at $>99\%$), exists between PV and ozone at 250 hPa at all stations and in all seasons. As expected, the correlation at 500 hPa level is weak at all stations and there is almost no correlation in summer at Sodankyla and Ny-Ålesund. Beekmann *et al.* [1994] found a correlation of 0.40 between ozone and PV at 500 hPa for all seasons with $>99.0\%$ significance. However, the correlation observed at 500 hPa in the present study for all seasons is small in comparison with that of observed at midlatitudes [Beekmann *et al.*, 1994] and varies from 0.21 at Sodankyla to 0.29 at Lerwick (significant at $>99\%$).

[15] Beekmann *et al.* [1994] made use of the positive correlation of PV with ozone in separating tropospheric ozone sources on the basis of high and low PV. In other words, ozone with high PV group indicates that the ozone is from the stratosphere. However, the diabatic heating can

Table 2. Correlation Coefficient Between PV and Ozone at 250 and 500 hPa

	250 hPa	Significance, %	500 hPa	Significance, %
<i>Lerwick</i>				
Winter	0.83	>99.9	0.29	99.9
Spring	0.91	>99.9	0.24	99.2
Summer	0.88	>99.9	0.23	85
Fall	0.93	>99.9	0.28	98.3
<i>Sodankyla</i>				
Winter	0.81	>99.9	0.37	99.9
Spring	0.90	>99.9	0.27	98
Summer	0.87	>99.9	0.08	56
Fall	0.87	>99.9	0.23	86.5
<i>Ny-Ålesund</i>				
Winter	0.81	>99.9	0.29	99.9
Spring	0.84	>99.9	0.32	99.9
Summer	0.82	>99.9	0.04	27
Fall	0.91	>99.9	0.36	99.9

change the PV values without changing ozone. So in the present study one more constraint is added to separate the ozone database. The ozone is treated as of stratospheric origin, if the PV at 500 hPa is higher than the climatological monthly value and if the relative humidity is less than 20%. The number of cases with high PV and low relative humidity is small (9% and 6% of the total data for Sodankyla and Ny-Ålesund, respectively), particularly in summer. The total contribution of cross-tropopause flux and photochemistry (in terms of magnitude) to tropospheric ozone is beyond the scope of the present paper. Figure 5 shows the annual cycle of ozone at 500 hPa for the two groups (high PV with low relative humidity class, hereafter high PV class, and low PV class). The high PV class shows a peak in May–June, while the low PV class shows a broad maximum from May–August. The seasonal variation of the high PV class is in agreement with the seasonal variation of cutoff lows [Price and Vaughan, 1992], which peaks in late spring and early summer and also of tropospheric Sr90 [WMO, 1986 for more references]. Appenzeller et al. [1996] formulated time series of air mass flux across extratropical tropopause and found that the maximum transport occurs in late spring. Seo and Bowman [2001] used a Lagrangian trajectory model to estimate the downward flux of stratospheric air and observed a prominent annual cycle with largest downward flux in late spring to summer and smallest in fall. These studies are also consistent with the annual cycle of the high PV class. On the other hand, as explained in earlier sections, the dominance of photochemistry in summer over other seasons is also in agreement with the ozone maximum for low PV class, seen in May–August period, from the present study.

[16] The ratio between ozone and PV is useful to initialize chemistry transport models through modeled potential vorticity fields and to calculate the ozone fluxes from PV fluxes [Gidel and Shapiro, 1980; Ebel et al., 1991]. It has also been used to reconstruct the hemispheric ozone fields from aircraft measurements [Kyrö et al., 2000] and from satellite measurements [Randall et al., 2002]. Monthly averages of ozone mixing ratio and PV are used to evaluate their ratio for the data at Lerwick, Sodankyla, and Ny-Ålesund. Figure 6 shows the annual cycle of ozone/PV ratio for selected stations at 500 and 250 hPa. These pressure levels are chosen as they represent the

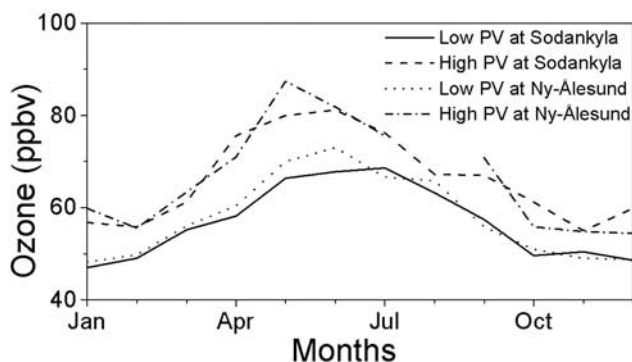


Figure 5. Annual cycle of tropospheric ozone sources (high PV and low humidity represent the downward cross-tropopause flux and low PV represents photochemistry) at European Arctic stations.

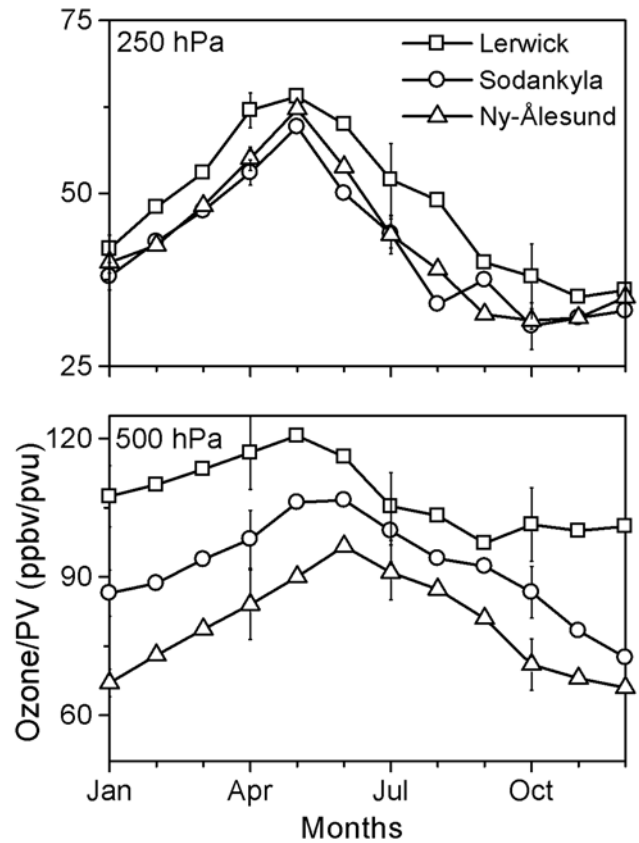


Figure 6. Annual variation of ozone/PV ratio at Lerwick, Sodankyla, and Ny-Ålesund at 500 and 250 hPa.

middle troposphere and lower stratosphere, respectively. In the present study, the 250-hPa level is preferred to represent lower stratosphere even though it is too close to (just above) the tropopause in summer and fall. Reasons for choosing the 250-hPa level include among others the following: (1) backward trajectories ending in tropopause folds revealed that for most of the cases air descends from altitudes between 200 and 300 hPa; (2) it allows us to compare the climatological ozone/PV ratios reported, by Beekmann et al. [1994], at midlatitudes, where ozone/PV ratios are evaluated at 225 hPa level; (3) ozone/PV ratios are also evaluated at three other pressure levels (200, 225, and 275 hPa) to understand the sensitivity of the ratio to the choice of the pressure surface and found that the difference of the ozone/PV ratio evaluated at these pressure levels is not more than 5 ppb/potential vorticity unit (pvu). Further, with the help of a Lagrangian particle dispersion model, FLEXPART, James et al. [2003] have shown that the stratospheric air originating above 4 pvu spends considerable time in the tropopause region before crossing 2 pvu surface (tropopause). Thus most of the air masses with stratospheric characteristics in the troposphere will have a relatively recent origin in the tropopause region only, and not in the stratosphere above.

[17] The ozone/PV ratio at 250 hPa level shows a prominent annual cycle with high values in late spring to early summer. In general, the middle troposphere values are two times more than the lower stratospheric values. In the middle troposphere, the north-south gradient is seen in

Table 3. Annual Mean of Ozone/PV Ratio at Lerwick, Sodankyla, and Ny-Ålesund

Station	Ozone/PV Ratio, ppb/pvu		PV Average, pvu	
	500 hPa	250 hPa	500 hPa	250 hPa
Lerwick	104.2 ± 6.2 ^a	48.4 ± 5.6	0.58 ± 0.03	4.13 ± 0.34
Sodankyla	91.7 ± 7.5	41.4 ± 5.9	0.63 ± 0.04	5.12 ± 0.40
Ny-Ålesund	79.6 ± 6.1	42.9 ± 5.5	0.78 ± 0.03	5.35 ± 0.42

^aDenotes 2 SE.

ozone/PV ratio with the northernmost station (Ny-Ålesund) having lower values than the southernmost station (Lerwick). Note that the ozone mixing ratio at 500 hPa (Figure 2) are almost equal for all the stations. The PV values (not shown as a separate figure) are more at Ny-Ålesund than at Lerwick and are responsible for the observed latitudinal gradient in ozone/PV ratio. In the lower stratosphere, the ozone/PV ratio varies from about 30–34 ppbv/pvu ($\text{pvu} = 10^{-6} \text{ K kg}^{-1} \text{ m}^2 \text{ s}^{-1}$) in November to about 60–64 ppbv/pvu in May. Annual means of ozone/PV ratio at selected locations for 500 and 250 hPa are shown in Table 3. These values are compared with those reported in the literature. Annual mean of ozone/PV at OHP for 500 hPa level is, about 111 ppb/pvu [Beekmann *et al.*, 1994], more than the values observed at the northern stations and is consistent with the latitudinal gradient discussed above. In the lower stratosphere, no significant difference between the annual means of ozone/PV is found at the stations of interest. The seasonal variation of ozone/PV observed at OHP [Beekmann *et al.*, 1994] is similar to that observed in the present analysis and the ozone/PV values at OHP, if interpolated linearly to 250 hPa, and is also in agreement with the present study. The climatological values of ozone/PV deduced from zonal means of O_3 and PV by Gidel and Shapiro [1980] are a factor of 2 more than those observed in the present analysis, as also observed, at least for January values, in Western Europe, by Beekmann *et al.* [1994].

However, the ratio of ozone/PV observed here is in good agreement with the values reported from case studies of tropopause folds [Browell *et al.*, 1987; Ancellet *et al.*, 1994]. The former used airborne LIDAR data to measure ozone and radiosounding analysis for PV estimation and reported an average value of 50 ppb/pvu in March, while the latter used LIDAR and ECMWF analyses to arrive at a ratio of 30–40 ppb/pvu for the PV range of 1.6–3 pvu in November. Ravetta *et al.* [1999], instead of using a linear fitting to ozone and PV, used a parabolic fit to the data taken in a tropopause fold in March at OHP and found a ratio of about 45 ppb/pvu for PV values between 2 and 6.

5. Case Study of Tropopause Fold

[18] Climatological ozone/PV ratios obtained in the study are compared with the ozone/PV ratio obtained in case studies of tropopause folds at high latitudes. A VHF radar, ozonesonde data, and ECMWF trajectory analysis are used for this purpose. Radar has the advantage of providing wind and turbulence information continuously and can be used to monitor and study tropopause folds. The radar (ESRAD) used in this study is located at Esrange (67°53'N, 21°06'E), about 35 km from Kiruna, in the northern Sweden. Its main characteristics are frequency, 52 MHz, peak transmitted power, 72 kW, range resolution, 300 m, and antenna aperture, 1600 m². A complete description of the radar can be found in the work of Chilson *et al.* [1999].

[19] The synoptic situation over northern Europe on 23 February 1997 at 1200 UT can be seen (Figure 1a) from the wind pattern on 200-hPa surface. It clearly shows a jet streak with velocities, predominantly westerlies, more than 50 m s^{-1} over Kiruna and Sodankyla. A downward sloping tropopause fold beneath the jet streak extending toward equator can be anticipated from this scenario. Indeed, a fold has been observed with the radar and also in the meteorological analysis as can be seen in Figure 7. The figure shows

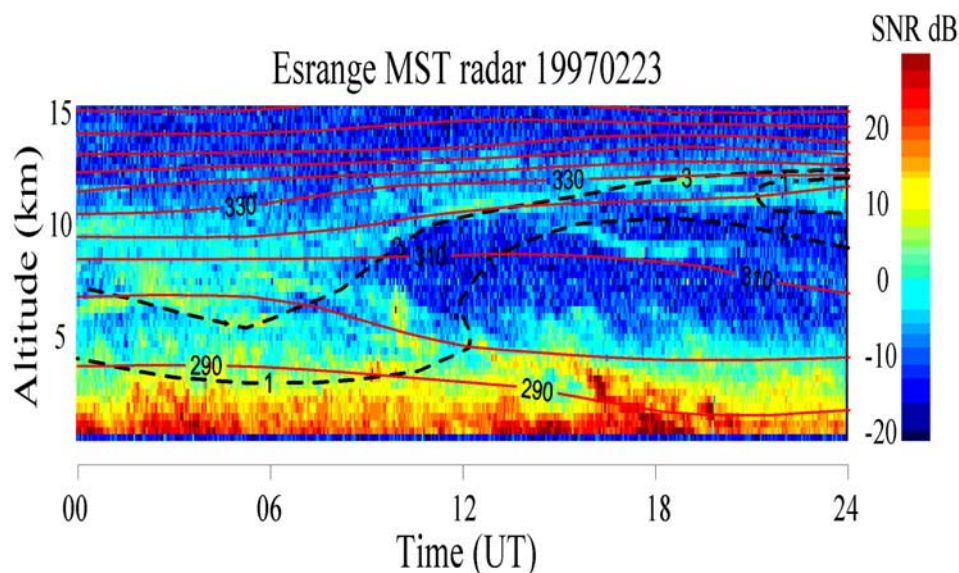


Figure 7. Range-time-intensity plot of ESRAD observed on 23 February 1997 during the passage of an upper level frontal system over the radar site. It also includes the isentropes (solid lines) and PV = 1 and 3 surfaces (dashed lines) derived from ECMWF fields.

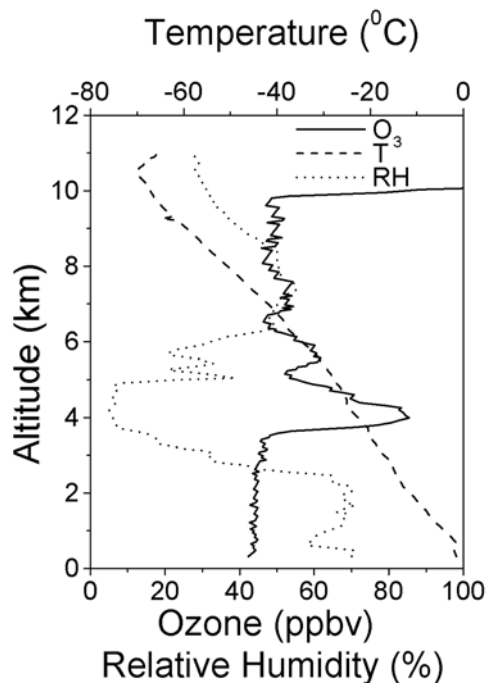


Figure 8. Vertical profiles of ozone (solid line), temperature (dashed line), and relative humidity (dotted line) measured at Esrange on 23 February 1997 in a tropopause fold.

the range-time-intensity plot obtained with ESRAD on 23 February 1997 during the passage of the upper level frontal zone. It also contains isentropes (solid lines) and isolines of potential vorticity (dashed lines) deduced from the balloon soundings and ECMWF analysis. PV = 1- and 3-pvu surfaces represent the boundary of the fold and tropopause, respectively. The choice of PV representing the tropopause is somewhat arbitrary and has appeared with different values from 1 to 3 in the literature. It can be seen from Figure 7 that the PV = 3-pvu surface is very close to the tropopause seen by the radar [see Gage and Green, 1979, for radar tropopause detection from returned echo power]. The upper level frontal zone can be seen as a layer of strong signal-to-noise ratio (SNR) sloping downward from tropopause to the lower troposphere. Following the detachment of frontal structure from the tropopause, the tropopause is lifted significantly. The PV = 1-pvu surface also exhibits a folded structure. However, as expected, the fold observed with radar is much deeper and contains more details than the PV analysis. The 300 K isentropic surface crosses the tropopause, slopes downward, and almost follows the frontal structure observed by radar. An ozonesonde released at 1200 UT from ESRANGE shows a layer of enhanced ozone (solid line) at around 4 km (Figure 8). The position of this high ozone layer corresponds quite well to a stable dry layer with relative humidity (dotted line) less than 10% and also to the sloping structure seen with radar and corroborates the tropopause folding. However, the air intruded into the troposphere can transfer back reversibly into the stratosphere. To know the fate of the air in the fold, forward trajectories (3-D) have been calculated using ECMWF wind fields starting on 23 February 1997 at 600 hPa. The

trajectories remain in the troposphere for 5 days indicating that (figure is not included) the ozone-rich stratospheric air, indeed, transferred irreversibly into the troposphere. The average (average is taken over values in the fold) ozone/PV ratio estimated for this case study (50 ppb/pvu) is found to be close to the lower stratospheric climatological value of 45 ppb/pvu in February at Sodankyla. Similar comparisons have been made for several case studies and found a close agreement between ozone/PV ratios observed in case studies and the lower stratospheric climatological value.

6. Summary and Conclusions

[20] Long-term observations of ozone with ozonesoundings are used to study annual and interannual variations in the UTLS region over northern Europe. The ozone mixing ratio shows a significant annual cycle with maximum values in late spring-early summer and minimum values in winter in the middle troposphere. Logan [1985] noticed different annual cycles of ozone at remote sites (spring maxima) and polluted regions (summer maxima) and argued that the cross-tropopause flux and photochemical production are responsible for the observed difference in the annual cycle. Note that in the present study, remote sites like Ny-Ålesund (79°N) also shows a peak in late spring-early summer indicating the importance of long-range transport of ozone and its precursors. However, the role of cross-tropopause flux cannot be neglected in other seasons, in particular in spring. Van Haver *et al.* [1996] speculated that the maximum impact of cross-tropopause flux (associated with tropopause folds) is in spring, when the lower stratospheric ozone mixing ratios are at a maximum. A shift in ozone maximum from late spring-early summer at 500 hPa to late spring at 200 hPa and to winter-early spring at 100 hPa shows the switching of ozone control from photochemical to dynamical. The distribution of ozone over northern Europe shows a clear latitudinal gradient above the tropopause with highest values at the northernmost station, while ozone values at 500 hPa are almost the same at all the stations. Significant interannual variability is seen in the lower stratosphere and is mainly attributed to the variations in large-scale wave-driven stratospheric circulation. Traces of interannual variation in the lower stratosphere are seen not only in the upper troposphere but also in the middle troposphere (not necessarily always) and indicate the role of the stratosphere in controlling the upper tropospheric ozone budget. The annual cycle of tropospheric ozone for different sources (separated on the basis of PV and humidity values) shows a peak in May–June for stratospheric sources and in summer for photochemical sources. The observed maximum cross-tropopause flux in May–June is consistent with the reports available in the literature [Appenzeller *et al.*, 1996; Seo and Bowman, 2001].

[21] At all sites and in all seasons, as expected, a significant (at >99%) positive correlation is seen in the lower stratosphere (250 hPa) between ozone and PV. On the other hand, a fair correlation at 500 hPa is seen at all stations, except in summer at high-latitude stations. The correlation coefficient between ozone and PV at midlatitude stations is, in general, more than at high-latitude stations in the middle troposphere. It is generally accepted that the cross-tropopause flux, which favors the positive correlation

between ozone and PV in the troposphere, in the extra tropics depends strongly on the strength and location of synoptic disturbances such as tropopause folds and cutoff lows, associated with large-scale wave amplification and cyclogenesis. Thus weaker correlation between ozone and PV at 500 hPa at high latitudes than at midlatitudes suggests that the stratospheric flux of ozone into the troposphere is more at midlatitudes than at high latitudes. Ebel *et al.* [1996] used the **Q**-vector divergence and PV maximum criteria to identify the folds and found the occurrence to be maximum at midlatitudes (40°–70°N). A 1-year climatology of mass flux across a 2-pvu tropopause following a Lagrangian approach revealed maximum downward fluxes between 35° and 50°N in the Northern Hemisphere [Wernli and Bourqui, 2002]. The polar-type cutoff lows [Price and Vaughan, 1992], which are efficient agents of STE (through tropopause folds) and centered at midlatitudes, are more in number than the subtropical and polar-vortex-type cutoff lows. All these studies corroborate the idea of more downward cross-tropopause flux at midlatitudes than at other latitudes. Further, in summer, no correlation is seen between ozone and PV at high-latitude stations. It indicates, as explained, the predominance of photochemistry over STE in polar summer. On the other hand, a fair correlation exists between ozone and PV at OHP [Beckmann *et al.*, 1994] and also at Lerwick in summer. This could be due to the maximum occurrence of polar-type of cutoff lows in summer at midlatitudes [Price and Vaughan, 1992]. It can be inferred from the above discussion that the number of folds appears to be less in summer at high latitudes. In contrast, studies of the climatology of tropopause folds at midlatitude stations using long-term ozonesounding data reveal that the frequency of tropopause folds is similar for all seasons [Van Haver *et al.*, 1996; Beckmann *et al.*, 1997]. A similar study at high latitude should throw some light on this aspect and will be pursued in the near future.

[22] The climatology of the ozone/PV ratio at middle troposphere and lower stratosphere heights shows high values in the middle troposphere and a seasonal variation which is similar to that of ozone with late spring-early summer maxima. A large discrepancy is seen between the observed ozone/PV ratio and zonal mean ozone/PV ratio reported by Gidel and Shapiro [1980]. However, the lower stratospheric ozone/PV ratios are close to the ratios reported in case studies of tropopause fold and climatological ratios at midlatitudes reported by Beckmann *et al.* [1994]. Nearly equal values of ozone/PV in the lower stratosphere at all latitudes indicate a lack of a significant latitudinal gradient in the extra tropics. In other words, a single ozone/PV ratio (as a representative for that month) can be used to convert global PV fluxes to ozone fluxes. However, the ratio varies significantly over the course of the annual cycle. The ratio between ozone/PV is estimated for several case studies and compared with the climatological ratio for validation of the climatological ratio. A case study of a tropopause fold has been presented. A fairly good comparison is found between the estimated value in the fold and the climatological value.

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