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Monitoring of boundary layer ozone in Norway from 1977 to 2002

Sverre Solberg

Contents

Summary

An evaluation of the Norwegian ozone monitoring data is presented. A detailed investigation of the technical monitoring history was required to prepare a subset of data with acceptable data quality for long-term trend studies. These data indicated a reduction in the 99-percentile of the daily (daytime) ozone data in the southern part of the country in the summer half year of the order of 1 ppbv/year during the 1990-ies for some sites. For sites further north a statistically significant increase in the mean ozone concentration of 0.3-0.5 ppbv/year was found for the winter half year. An increasing trend in the mean ozone concentration was found also in summer but less clear. Based on trajectory analyses a significantly increasing ozone trend in background air masses was found.

A particular problem in the inspection of the data was the infrequent calibrations of the various instruments until 1997 when a more rigorous procedure for quality assurance, field inspections and field calibrations was started. Based on the monitoring history periods of the ozone data were rejected for the purpose of long-term trend studies, while regarded valid for other applications with less strict requirements for accuracy. This reduced the amount of data and the length of the time series considerably, decreasing the relevance of the trend study, but increasing the accuracy and reliability of the underlying data.

The trends at the station Prestebakke (in the southeast) differed from the other sites in that region and showed significantly increasing ozone concentrations, particularly marked since 1999. The reason for this is unclear, but must probably be explained by changes in local conditions (land use, nearby emissions etc).

It is unclear whether the increase in the ozone concentration in the background air masses could be explained by a steady growing ozone concentration in the background or due to a shift from one 3-4 years period with low background ozone in the mid 1990-ies to another period of high background ozone values at the end of the 1990-ies. Based on the trajectory sectors it does not seem likely that a major shift in the atmospheric transport pattern could explain the observed trends. Only at Jergul/Karasjok (in the far north) indications of such a shift is seen with a higher frequency of transport from E-SE during the last part of the 1990-ies and less periods with background air masses. For the other sites there was no corresponding trends in the frequency of transport sectors.

A main conclusion from this study is the need for long time series of measurement data with high and well-documented data quality. Without detailed knowledge of the data quality, any assessments of the compliance with the emission abatement protocols may become futile.

Monitoring of boundary layer ozone in Norway from 1977 to 2002

1 Monitoring network

Monitoring of surface ozone started in the southern part of Norway in 1975-1977 (Hanssen and Sivertsen, 1977; Schjoldager and Thorstad, 1978). By the time ozone was first included as part of EMEP's extended measurement programme in 1984, four ozone monitoring sites were in operation in Norway, although none were actually EMEP stations. In 1987, when a systematic collection and checking of ozone data were initiated in EMEP, about ten Norwegian ozone sites were operating (Pedersen, 1992). Table 1 shows the list of Norwegian background ozone monitoring sites that have been in operation through the history and the present network is shown in Figure 1. In addition to this, there are urban ozone monitoring in a number of cities in Norway, and there have been a few short-term based ozone monitoring activities e.g. connected to local industrial assessment studies and also air-craft measurements at a very early stage in the monitoring history (Schjoldager et al, 1981).

Table 1: Norwegian background ozone monitoring stations.

Code	Station name	Reported here	Start	End	Latitude	Longitude	Altitude (m)
-	Nordmoen		1986	1996	60 16 00 N	11 06 00 E	200
-	Klyve [1)]		1979	-	59 09 00 N	09 35 00 E	60
-	Haukenes [1)]		1979	-	59 12 00 N	09 31 00 E	20
-	Langesund [1)]		1979	-	59 00 01 N	09 45 00 E	12
NO01	Birkenes	X	1985	-	58 23 00 N	08 15 00 E	190
NO15	Tustervatn	X	1990	-	65 50 00 N	13 55 00 E	439
NO39	Kårvatn	X	1988	-	62 47 00 N	08 53 00 E	210
NO41	Osen	X	1990	-	61 15 00 N	11 47 00 E	440
NO42	Zeppelin Mtn.	X	1988	-	78 54 00 N	11 53 00 E	474
NO43	Prestebakke	X	1985	-	59 26 00 N	10 36 00 E	160
NO45	Jeløya	X	1979	2003	59 26 00 N	10 36 00 E	3
NO48	Voss	X	1990	2003	60 36 00 N	06 32 00 E	500
NO52	Sandve	X	1997	-	59 12 00 N	05 12 00 E	15
NO47	Svanvik		1986	1997	69 27 00 N	30 02 00 E	30
NO30	Jergul [2)]	X	1988	1997	69 27 00 N	24 36 00 E	255
NO55	Karasjok [2)]	X	1997	-	69 28 00 N	25 13 00 E	333
NO56	Hurdal	X	1997	-	60 22 00 N	11 04 00 E	300

[1)] Operated by the State Pollution Authority
[2)] Station Jergul was replaced by station Karasjok in 1997

During the 1990-ies NILU has been running of the order of ten ozone stations as part of EMEP and the national monitoring activity (Hjellbrekke and Solberg, 2003), and in addition SFT, the State Pollution Authority, has been running three

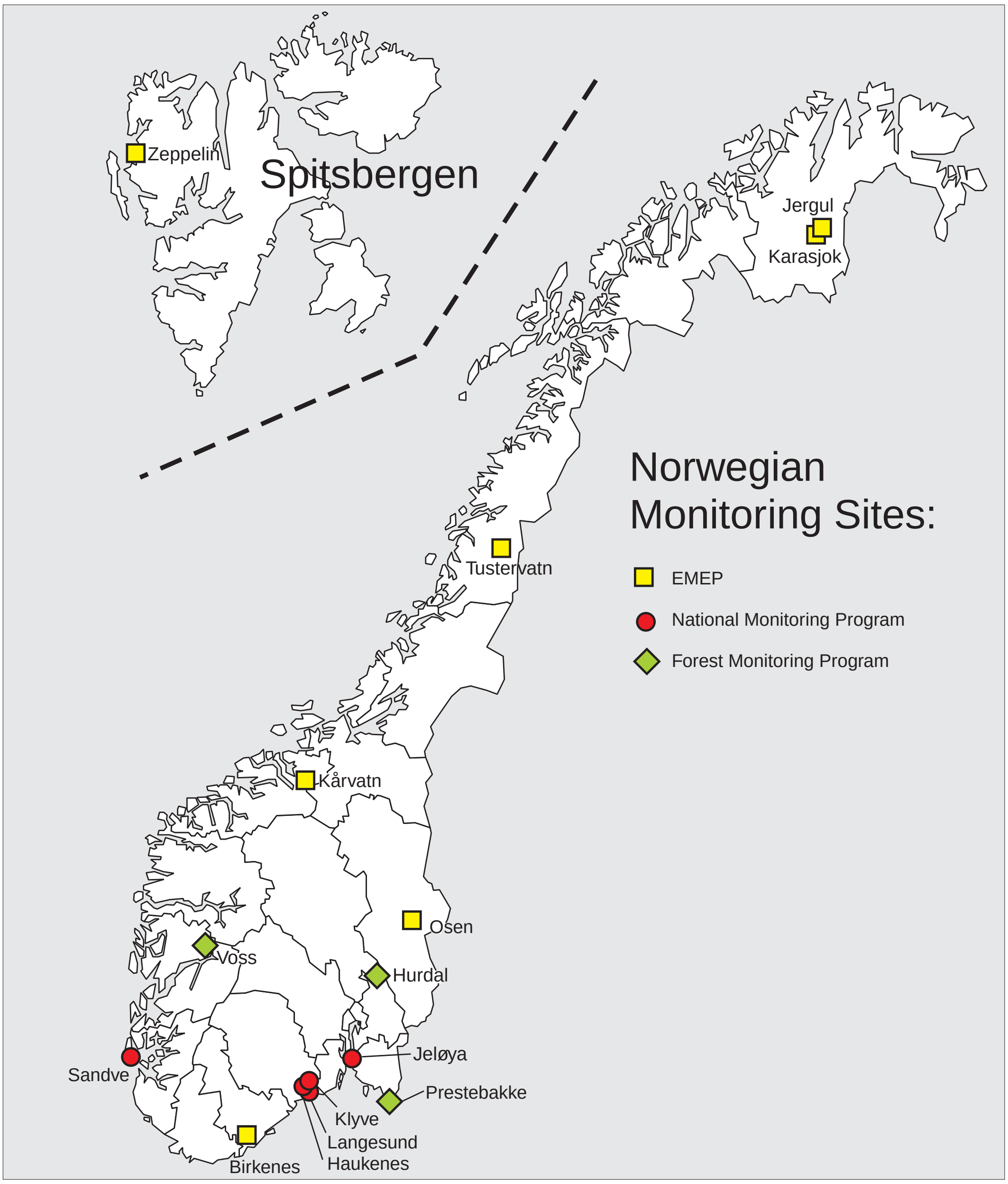


Figure 1: Map of Norwegian monitoring stations.

stations in the industrial area of Nedre Telemark in south Norway as indicated by Table 1. A few changes to the network have been made. Due to substantial influence of local emissions, the station Nordmoen (close to the new main airport at Gardermoen) was replaced by the research station Hurdal in the same district in 1997. Also in 1997, the station Jergul in North Norway was replaced by Karasjok. Norwegian ozone monitoring data have previously been presented by Pedersen and Lefohn (1994), Solberg et al. (1997) and in annual reports for EMEP and the national monitoring programme as listed in Appendix B.

2 Instrumentation calibration procedures etc.

During the first years the monitoring were based on chemiluminescence using two types of instruments. At the stations Klyve, Haukenes and Langesund a Phillips type, based on chemiluminescence between ozone and the compound "Rhodamin B", and for the other sites a Bendix type based on chemiluminescence between ozone and ethylene was used.

In 1981 an improved method for calibration of the ozone chemiluminescence monitors was introduced, and it was noted that data calibrated with the previous method could be multiplied by a factor 0.76 to harmonize with the new data series (Schjoldager et al., 1981). It was also noted, however, that the old method for calibration had a poor reproducibility so that this kind of correction was not really recommended.

In 1983 NILU started using a DASIBI UV-monitor as reference method. A comparison between the old KIBRT and the new UV method was carried out and a correlation coefficient of r = 0.999 was found. A deviation of less than 5% in the interval 100-300 ug/m3 was found and a best-fit regression line was determined to be:

$$O_3^{UV} = 0.92\ O_3^{KBIR} + 10.8$$

These results were, however, based on a few comparisons in the lab only, and not on a long-term parallel monitoring in the field. During the following years the chemiluminescence monitors were gradually replaced by UV-monitors at the various stations. Annual calibrations with NILU's standard UV-monitor were carried out during these years. From the mid of the 1980-ies NILU's reference monitor was calibrated against a primary standard at a National Bureau of Standard (NBS) UV-monitor in the US and in Sweden with very satisfactory result. The calibration with the US standard in 1989 revealed an uncertainty of ≤1 ppb for concentrations below 100 ppb and ≤ 1% for concentrations above 100 ppb (Roemer, 1998). By 1988 all ozone measurements in Norway were done by use of UV-monitors.

In the late 1980s and early 1990s the monitors were usually checked in the field more or less regularly about every third month. These routine checks in the field consisted of a standard program: The Teflon inlet filter was replaced, and checks were made of the sample and control frequency of the UV lamp, temperature and pressure in the measurement cell, and the calibration span and zero. A leak test was also carried out. In later years the zero-span checks as well as the checks of the UV lamp and the temperature and pressure were performed automatically and remote.

In the beginning of the 1990-ies the DASIBI monitors were gradually replaced by Monitor Labs ML8810 UV monitors and from the mid of the 1990-ies the ML8810 monitors were in turn replaced by API400 UV monitors.

Calibration of the field monitors was, however, done at infrequent and irregular intervals, and normally when the instrument was taken to NILU for maintenance or repair. Thus, a long-term drift in the monitors may have taken place unnoticed between the few calibrations until a malfunctioning forced the monitor to be brought back to NILU for repair. Then, the calibration of the previous data may not have been possible due to a damaged instrument. This is discussed in more detail below. The procedure with infrequent calibrations complicates any trend evaluations of ozone for the Norwegian network, particularly considering that the trend looked for is small and masked by varying meteorology from year to year.

3 Ozone monitoring data

The monthly medians, 25- and 75-percentiles for the period 1986-2002 for the 12 sites indicated in Table 1 are shown in Figure 2. These plots are based on daytime (10:00-18:00) averages. A marked seasonal cycle is evident with highest median values in April-May and lowest in late autumn. The peaks in the 75-percentiles are displaced towards the summer, with highest values in May-July. Table 2 gives the annual hourly maximum concentrations reported for the years 1985-2001.

Table 2: Annual hourly maximum ozone concentration (ppbv) at NILU's monitoring stations during 1985-2001. Note that changes in instrumentation, calibration procedures etc has not *been taken into account in this table.*

	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
Prestebakke	-	127	102	64	85	94	80	58	71	91	67	79	72	64	77	86	72
Jeløya	133	134	85	105	86	97	77	86	68	75	74	73	71	64	71	70	60
Hurdal													76	65	66	64	65
Osen						78	70	72	75	69	72	86	71	63	64	69	59
Birkenes	58	72	98	83	81	84	71	86	77	89	80	83	69	65	73	77	62
Sandve													75	64	69	61	70
Voss						101	80	86	74	79	71	73	81	66	71	71	59
Kaarvatn					65	65	63	98	82	82	68	67	71	67	73	82	63
Tustervatn						69	54	62	70	68	60	78	57	64	66	69	62
Jergul				74	72	73	53	70	63	68	49	50					
Karasjok													69	58	56	61	59
Svanvik				91	86	63	73	79	68	63	45	50					
Zeppelin						58	61	58	50	51	47	52	54	51	61	48	53

A visual inspection of these time series indicates a downward trend at some sites, e.g. Jeløya, Birkenes and Voss, while at other sites there is no clear tendency. The high peak values of the 75-percentiles above 80 ppbv seen in the late 1980-ies at Birkenes and Jeløya did not occur after 1990.

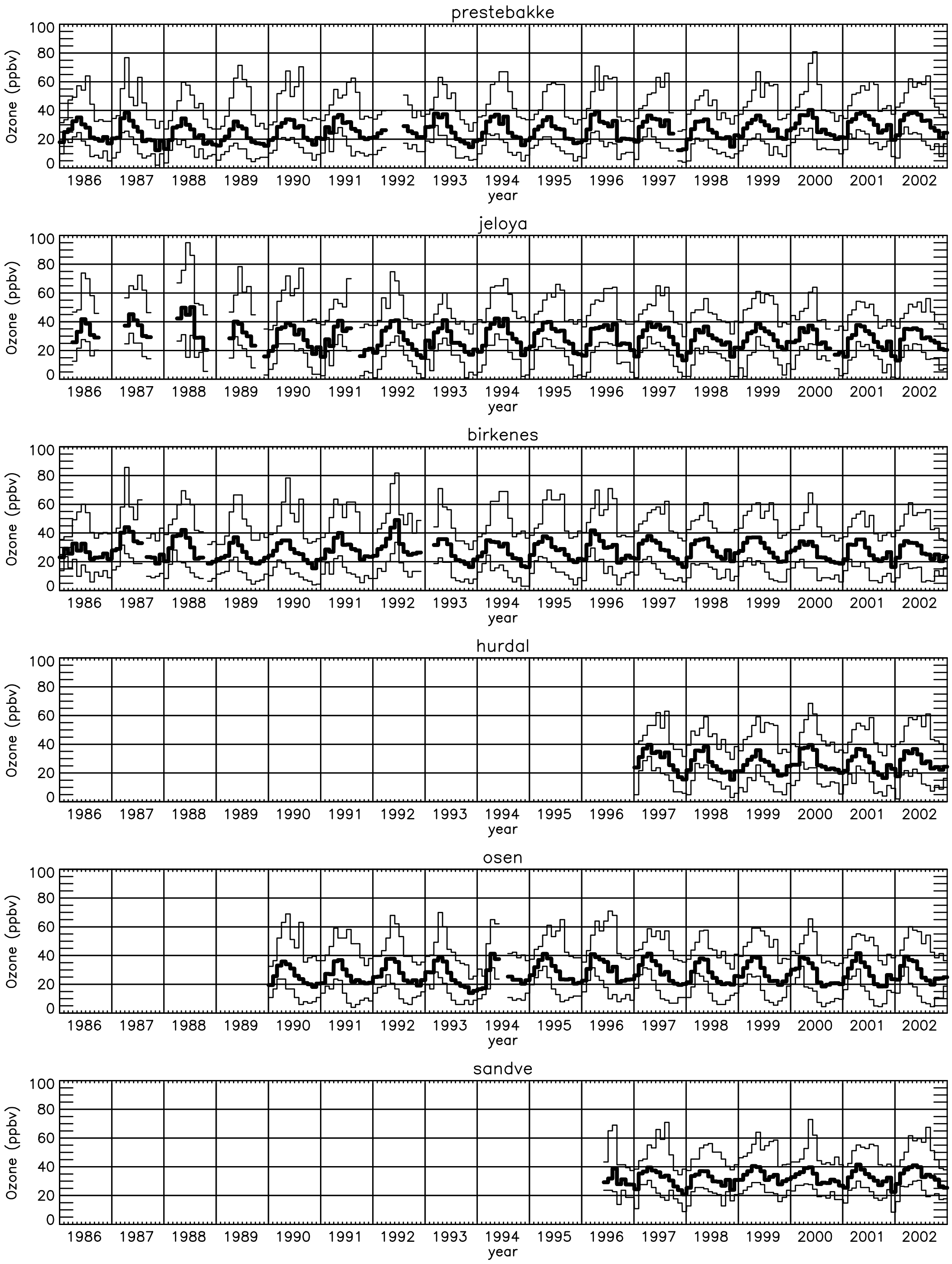


Figure 2: Monthly medians, 25- and 75-percentiles of ozone based on daytime (10:00-18:00) hourly values during 1986-2002.

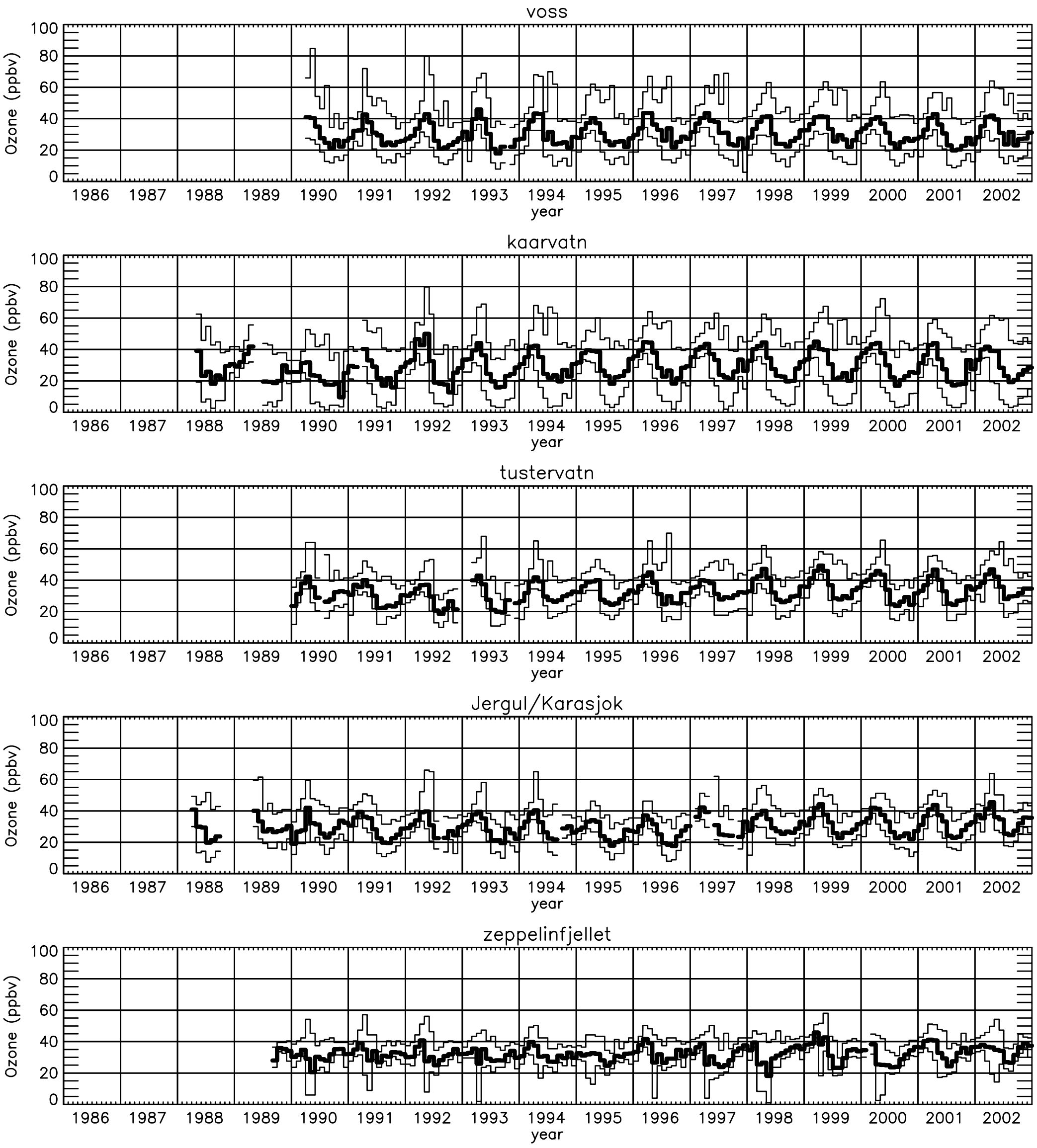


Figure 2, contd.

3.1 Screening of monitoring data according to the monitoring history

It turned out that a detailed verification of the monitoring history at each individual site was needed to evaluate the quality of the historical monitoring data, as pointed out by Roemer (1998). The main reason for this is that the quality assurance procedures for the monitoring have changed significantly during the history of ozone monitoring in Norway. While the original aim of the monitoring was to evaluate harmful effects from ozone pollution on human health, crops and other vegetation, the more recent focus on long-term trends in ozone has increased the demands for the accuracy of the ozone data substantially. A 10% error in the estimated ozone exposure (e.g. AOT40 value) is small compared to the uncertainty in the estimated damage to the crops. However, a 10% error in the ozone concentration values could render a long-term trend analysis completely, as the trends looked for are believed to be small compared to the background ozone level. Thus, without an elaborate screening of the monitoring history, any long-term trend study would be futile.

An investigation of the history of ozone monitoring at Birkenes, Jeløya and Prestebakke until 1995 has already been made (Roemer, 1998), and the present study is in some sense a continuation and extension of this.

Details of the instrumentation and monitoring for each individual site are given in the Appendix. The main conclusions from this could be summarized in the following: A strict procedure for regular calibrations of the ozone monitors was not followed until 1997 when a transfer standard, used for regular field calibrations, was held at NILU's lab and calibrated against the NIST standard at ITM, Stockholm, once a year. Before 1997 the monitors were never calibrated in the field, only at NILU's lab during maintenance. As the stability of the monitors varied so did the frequency of calibration at the various stations. Furthermore, with this practice the instruments were typically run at the sites until they eventually broke down in the field, making a post-calibration impossible. Unfortunately, this turns out to be a major problem with the historical data. In addition, the general quality and stability of the UV monitors has clearly improved during the last 15 years. Not only is the QA procedures improved the last 5-7 years, so are also the monitors.

A detailed investigation of log-books and other documentation was carried out. Discussion with the responsible technical personnel was essential in this work. Based on this, the dates for field inspections, technical service, malfunctioning, calibrations etc. were determined, which in turn was used to make an evaluation of the quality of the historical data. The results of this are visualised in Figure 3. The time series show the daily ozone values normalized according to the following expression (removing the seasonal amplitude):

$$z_i = (x_i - \mu_i)/\sigma_i \qquad (1)$$

where

- z_i = the daily normalized ozone concentration at day i
- x_i = the daytime (10:00-18:00) average ozone concentration at day i
- μ_i = the 30 days' running average concentration (based on the daytime data) at day i
- σ_i = the corresponding 30 days' running standard deviation at day i

Figure 3 shows the normalized daily values as well as the 90 days' running median, 25- and 75-percentile of these values. The dates for calibrations as well as periods with uncertain data quality are also indicated in the figures.

The first choice was to restrict the trend evaluation to the period after 1990. The procedures of the ozone monitoring in the 1980-ies turned out to be poorly documented, and few calibrations were carried out with the UV-monitors applied at that time, making the precision of these data questionable.

Secondly, certain periods with data of unknown quality were identified (and marked as red in Figure 3). There have been regular field inspections of the ozone monitors approximately every third month (sometimes much longer) since the

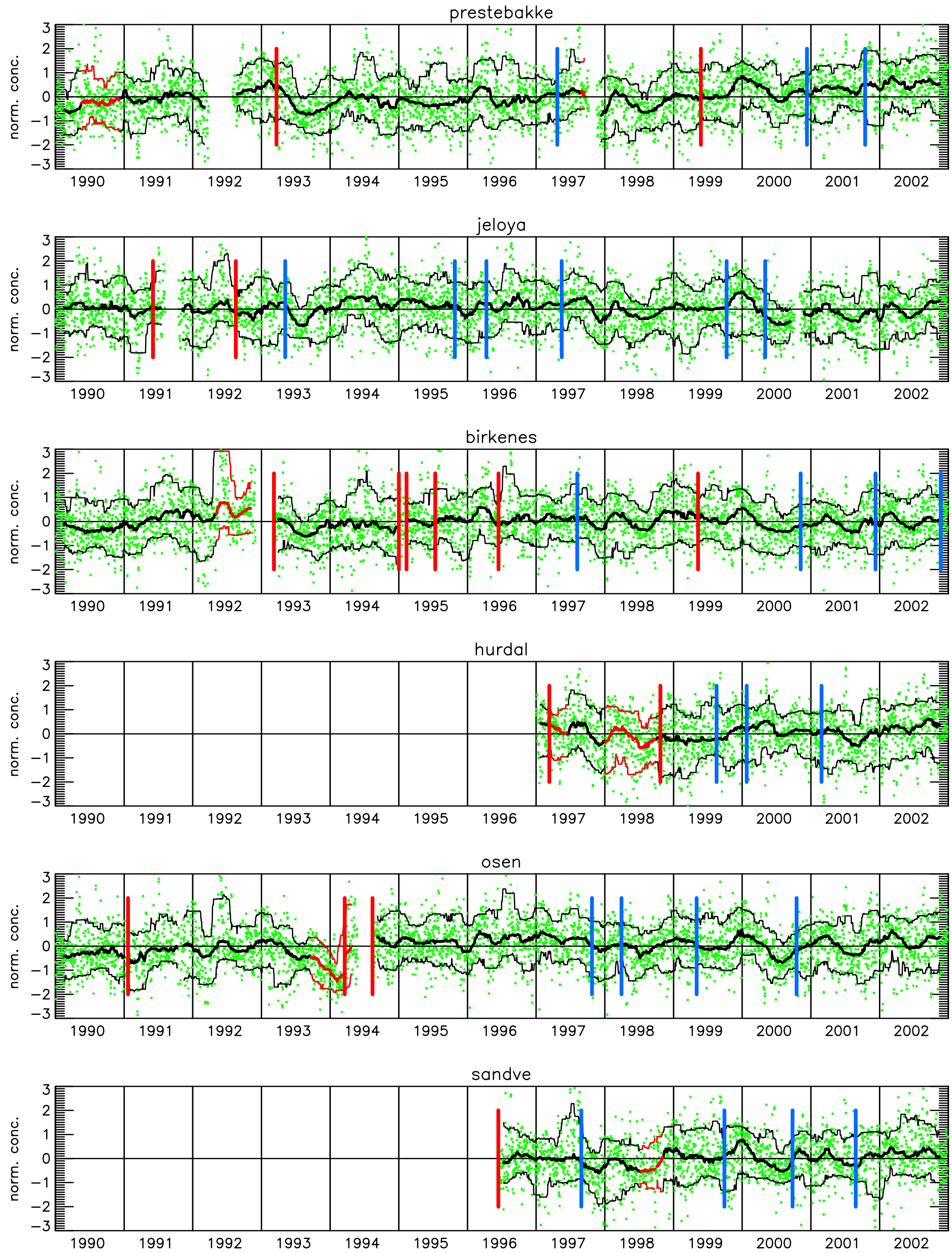


Figure 3: Time series of normalized ozone data during 1990-2002 at Norwegian sites. See text for details of the normalisation. Green dots mark normalized daytime average (10:00-18:00) concentrations of ozone. Red vertical lines mark "forward" calibrations, i.e. calibrations of new monitors at the site, or calibrations after a repair, but without any calibration of the previous monitor at the site. Blue vertical lines mark "backward and forward" calibrations, i.e. either by a field calibration using a transfer standard, or by a calibration of the old and new instrument when a monitor was replaced. Red curves mark periods with uncertain data quality.

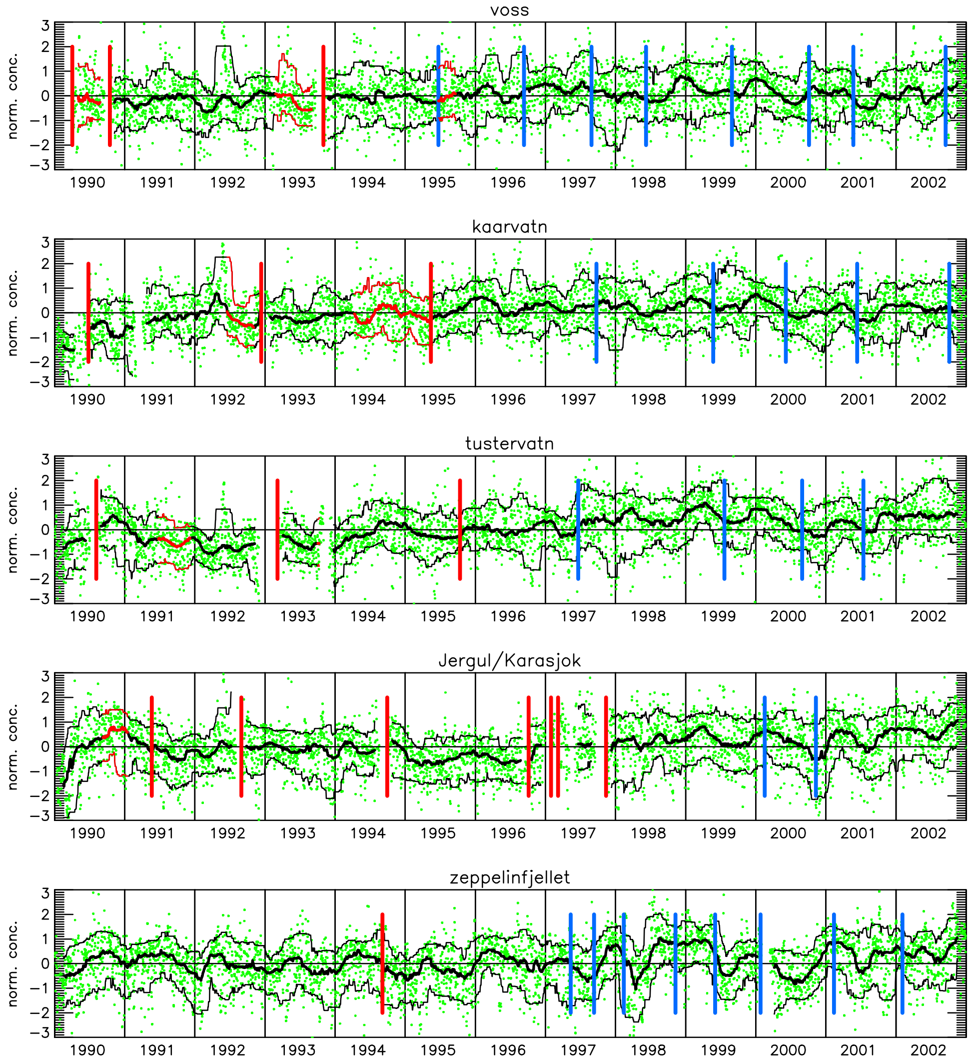


Figure 3, contd.

monitoring started. At a number of these occasions serious faults with the monitors were discovered, which may have lead to erroneous data values. Leakage in the magnetic valve has been the most common problem. Blocking of the inlet filter or pump failure are other potential sources of error. Normally, data values would then have been rejected for a certain time period, but this has not always been done. For the present study we have marked all data before such events and after the last preceding field inspection when the monitor behaved satisfactory.

One example of this is the monitoring at Osen in the last part of 1993 and the beginning of 1994 (shown in Figure 3). At a routine inspection a leak in the valve was discovered in March 1994. The last previous inspection of the monitor reported in the log-books were in September 1993, and thus a six-months period of data is marked as having a questionable quality. The data values themselves clearly indicate a drop in the average concentration level which could be caused by a gradually reduced sensitivity of the monitor during this period. Without screening of the data in this way, such periods of doubtful data values could weaken and even corrupt long-term trend analyses.

The dates of calibrations are indicated by coloured vertical lines in Figure 3. From 1997, when a transferable standard was started to be used, standard procedures for field calibrations and for replacement of monitors were designed. The dates of these field calibrations, or of any other parallel monitoring in the field, are marked with blue vertical lines in Figure 3. According to these standard procedures, a monitor replacement should be accompanied by pre- and post-calibrations of the new and old monitors in the lab and in the field, as well as parallel monitoring in the field. Since 1997 this practice has been followed when replacing an ozone monitor. Furthermore, the running monitors should be calibrated in the field by the transfer standard once a year, in addition to regular field inspections every third month. The practice of yearly field calibrations has been carried out for a number of the sites but not for all since 1997. However, according to the technical personnel and the log-books the newer instruments are very stable, so that drift in the monitors from one year to the next is almost negligible.

Before 1997 calibrations were only applied in the lab, prior to a field installation or after a repair of a malfunctioning monitor. The dates of these calibrations are marked by red vertical lines in Figure 3. Given that the transportation and field installation itself didn't influence the instrument performance, the monitoring data for a certain time period following these dates should be regarded valid, but the calibrations are not "backward compatible", i.e. these dates mark a shift in the data series and the agreement between the monitoring data before and after the calibrations is unknown.

A filtered data set, consisting of a conservative selection of the original data based on the monitoring history, was then prepared from the following criteria:

- Data are valid 1.5 year from a "forward calibration" (red bar).
- When a successful standard procedure for calibration or replacement is followed (blue bar) the data are valid backward and forward to the next calibration.
- All periods with doubtful data (marked red) are rejected.

By adopting these rather strict criteria the time series of the normalized data become as shown in Figure 4. Due to the infrequent instrument calibrations in the early 1990-ies, the time series become substantially shortened at several of the sites. It should be stressed that this screening of data are made for the purpose of trend analyses which set particular demands for the data quality, and does not apply to the ozone dataset in general.

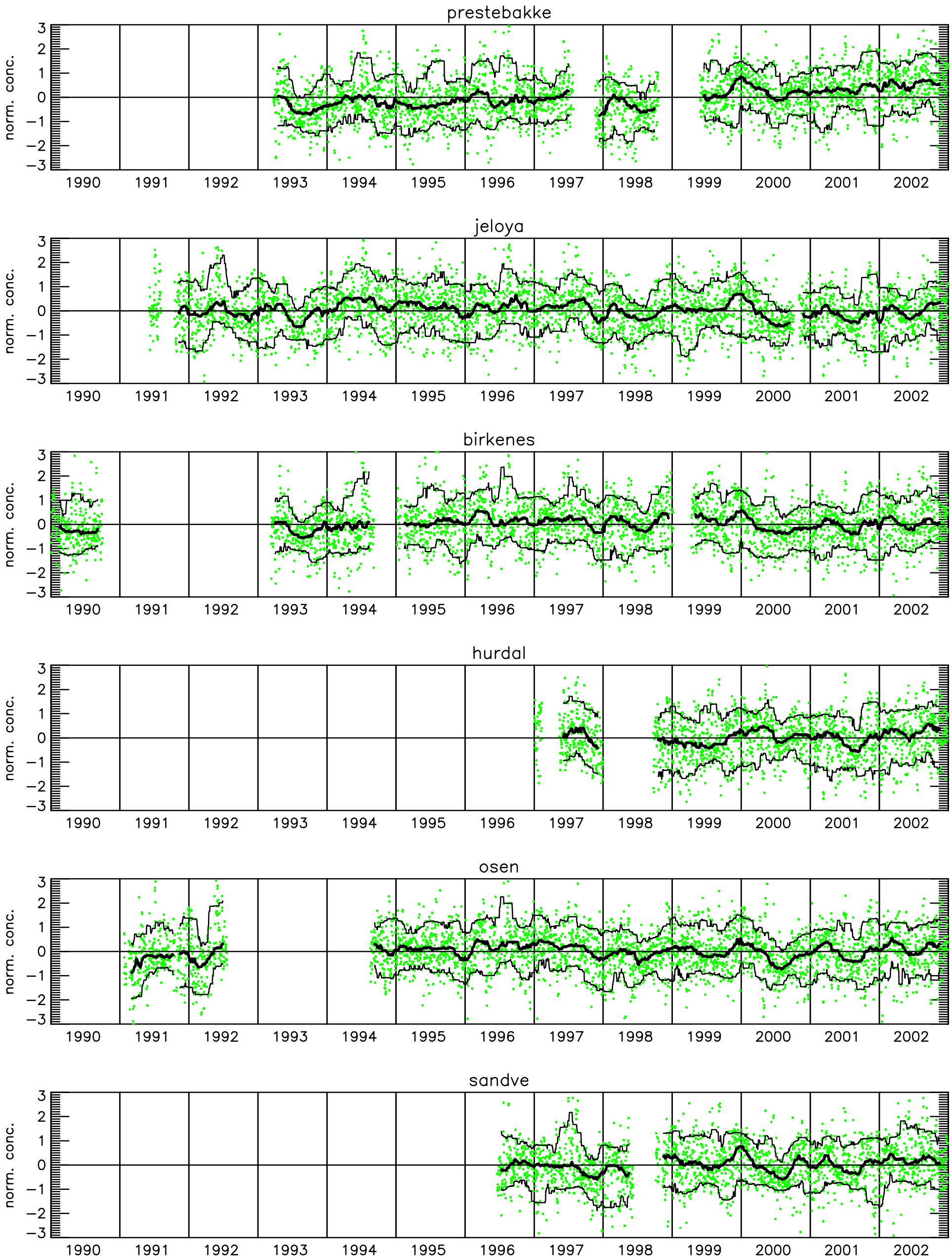


Figure 4: The time series of normalized ozone data during 1990-2002 after filtering based on the monitoring history. See text for details of the normalisation. Green dots mark normalized daytime average (10:00-18:00) concentrations of ozone. Black lines mark the 90 days' running median, 25- and 75-percentiles.

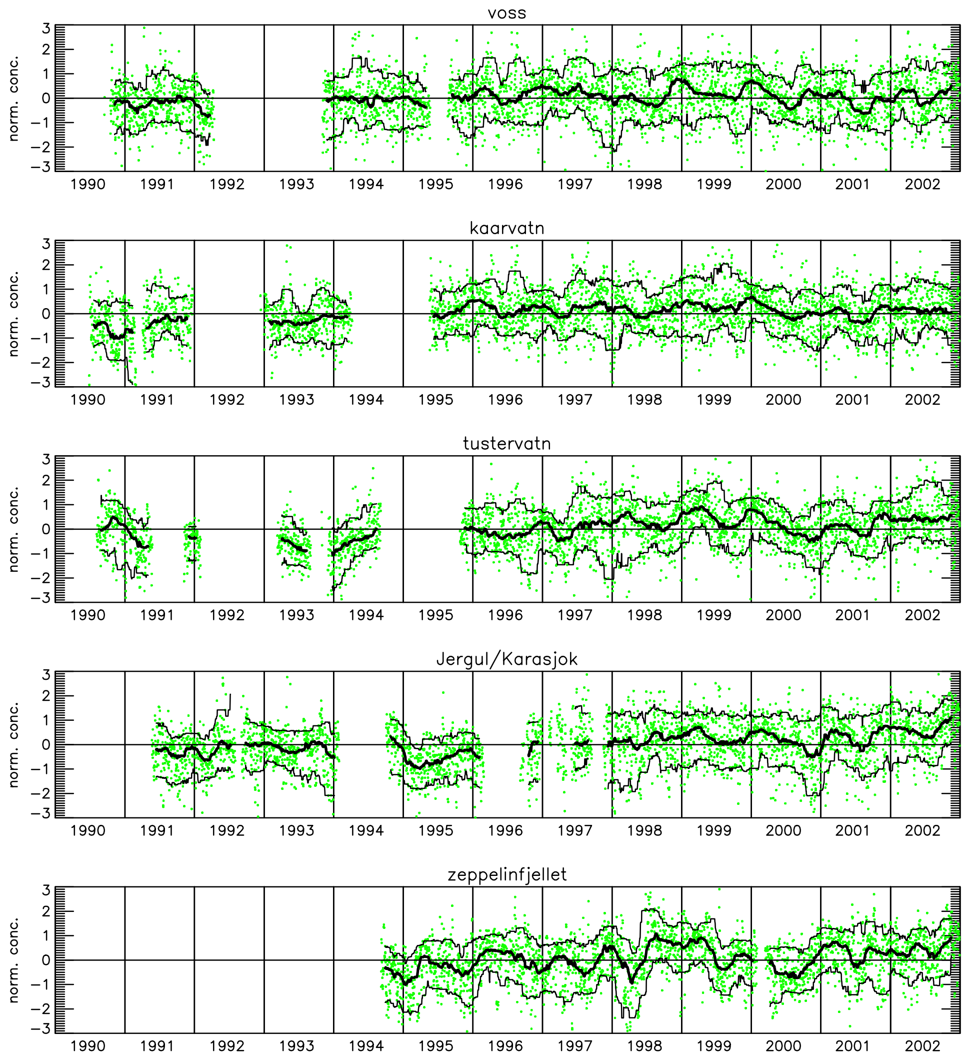


Figure 4, contd.

3.2 Ozone monitoring before 1990

In the preceding chapters the emphasis has been on ozone data after 1990. However, as mentioned in the introduction, surface ozone has been measured at various sites in Norway since the mid 1970-ies, and thus time series for several decades exist although that includes all kinds of problems with respect to changing procedures and instrumentations.

A new, improved method for calibration of the ozone monitors was introduced in 1981, and applied to NILU's monitoring data from 1980 at Maridalen and Jeløya. The other sites, run by SFT (the State Pollution Authority), were still calibrated with the old method. An empirical correction factor of 0.76 (to be multiplied with the old data) could be used to harmonize the old and new data (Schjoldager et al., 1981). However, as the old calibration method had a poor reproducability, correction of old data was not really recommended by the US EPA (Schjoldager et al., 1981).

According to the annual reports, all monitors were calibrated annually with the reference method used at that time. In 1981 and 1982 NILU used the improved "KIBRT" method and in 1983 the use of an UV monitor was introduced at NILU as a reference method.

The annual hourly maximum concentrations during the years 1977-1984 are given in Table 3. These data include data from the station at Jeløya established in 1979, at Maridalen, approx. 8 km north of the centre of Oslo, and at three sites around the industrial area of Telemark in south Norway. During 1980-1983 NILU used its own air plane to make a number of flights with ozone measurements in South Norway, mostly following the southwest coast or the southeast coast crossing the border to Sweden. The peak concentrations from the various flights in these years are given in the Table as well. In addition to these data, there were ozone measurements at other sites in Telemark back to 1975 with annual hourly maximum concentration up to approx. 200 µg m^{-3}. These sites were generally located in more urban areas.

Table 3: Annual maximum hourly concentrations of surface ozone measured in Norway from the start in 1977 to 1984. Unit: µg m^{-3}.

	1977	1978	1979	1980	1981	1982	1983	1984
Maridalen	218	184	142	307	182	167	122	143
Jeløya	-	-	185	257	209	190	157	198
Langesund	-	-	198	148	154	183	150	133
Klyve	-	-	183	-	-	195	153	129
Haukenes	-	-	398	146	160	225	148	145
Air plane	-	-	-	180	189	290	162	-

There are considerable variations in the data shown in Table 3, both from one year to anther and among the different sites. Furthermore, very high peak values, even above 300 µg m^{-3} are reported. The stations at Maridalen and at Klyve and Haukenes were all presumably influenced by local emissions from Oslo and from the heavy petrochemical industry (at that time) in Telemark, respectively.

It is an open question to what extent the measured peak values from these years are real and to what extent they are locally influenced. When compared to the annual maxima at the other sites the extreme value of 398 µg m^{-3} at Haukenes in 1979 seems unrealistic, and is certainly not regionally representative. The original report (Schjoldager and Stige, 1980) suggests local formation of ozone from emissions from the petrochemical industry in Bamble as an explanation.

The few number of sites and the differing procedures for calibration and instrumentation makes a more detailed evaluation of these earliest data difficult. An annual maximum value of above 300 µg m^{-3} at the station in Maridalen (in 1980) seems unlikely compared to the annual max of only 148 µg m^{-3} at Langesund some 200 km to the southeast. We note that in 1983-84 the peak values were closer to the ones observed in later years and lack the extremes seen during the years 1977-1982.

3.3 Comparison of ozone monitoring data from neighbouring stations

Comparison of data from neighbouring monitoring sites has previously been used to validate ozone data (Lindskog, 2003). For the present study triplets of sites were compared and the monthly medians of the differences based on daytime averages of the filtered data are shown in Figure 5.

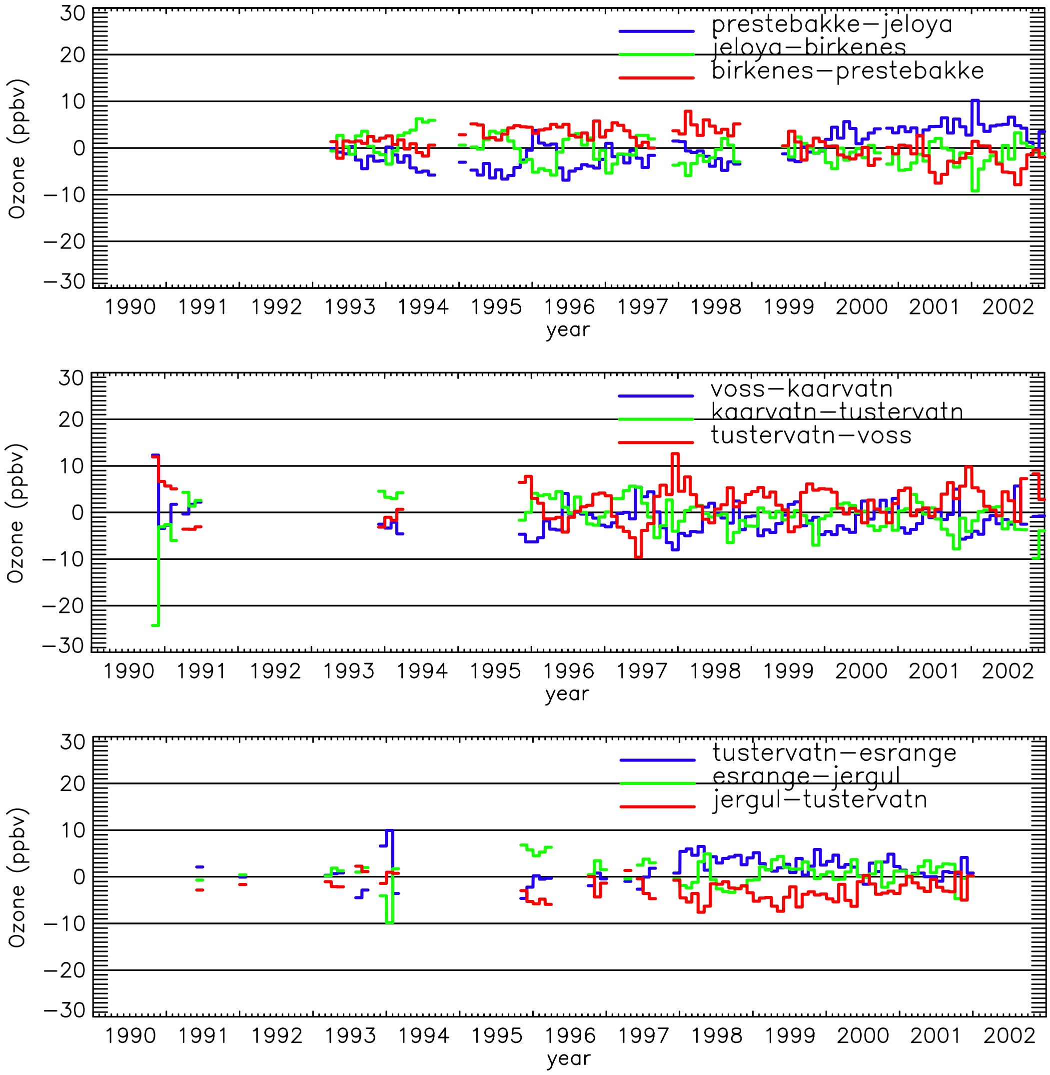


Figure 5: Monthly medians of the differences in observed daytime mean ozone concentrations at various sites. The filtered ozone data (described above) were used in the calculations.

The upper panel of Figure 5 clearly indicates that the long-term trend at Prestebakke differs from that at Jeløya and Birkenes. Relative to the other two sites the ozone values at Prestebakke increase with time. A similar trend was found when comparing Prestebakke with the Swedish site Rörvik located some 150 km to the south. This behaviour is most apparent for the last four years i.e. from 1999. As regular calibrations have been carried out during this period, there is no reason to suspect a drift in the monitor at Prestebakke, thus other local effects should be looked for, like the possibility of trends in the land use, deposition, nearby emissions etc. At present no obvious explanation for the growing trend observed at Prestebakke is given, except that it most likely is caused by local scale effects, not regionally representative.

The middle and lower panels in Figure 5 indicate a fairly good agreement between the neighbouring sites presented with a few exceptions. The station triplet of Voss, Kårvatn and Tustervatn (middle panel) reflects the varying seasonal cycle at the sites. The data may indicate a slow increase in the ozone values at Tustervatn relative to the two other sites, but this signal is not very obvious. The scattered data from the first years, 1990-1991, do, however, show larger differences probably reflecting less reliable data quality at that time.

In the lower panel of Figure 5 the data from Jergul/Karasjok and Tustervatn is compared to the Swedish site Esrange, located in northern Sweden (Lindskog, 2003). This triplet of sites show a fairly good agreement for the sites during the whole period, except for a short period by the end of 1993 and start of 1994. Apart from this, the monthly medians of the differences are of the order of 7-8 ppb or less, giving confidence to these data.

4 Trends in monitored ozone in Norway

Based on the filtered ozone data described above, simple linear trend calculations were made and the results for means and percentiles for each of two six-months seasons (Oct.-Mar. and Apr.-Sep.) are shown in Figure 6. The lines mark linear trends that were found to be significant on a 1-σ and 2-σ level as explained in the figure. These plots take all data together, thus the varying meteorology from one year to another is inherent in these results as is any possible changes induced by trends in anthropogenic emissions of NO_x, VOC and other trace gases.

As the time series become rather short and fragmentary due to the filtering of the data, no robust conclusions should be stated. However, a pattern of decreasing 99-percentiles at the southern stations in summer and an increase in the mean concentrations in the north in both seasons, most dominant in winter, is seen from Figure 6. All stations from Voss and northwards indicate significantly increasing mean concentrations in winter, of the order of 0.34 to 0.56 ppbv/year. Many of these sites also show increasing low or high percentiles in the same season, indicating a general rise in the ozone level. The estimated rise in the mean concentration in summer is of the same order at these sites, but less significant, and for some of the sites (Zeppelin Mountain and Voss) no trend is found in summer.

At Jeløya and Birkenes a decline of about 1 ppbv/year in the 99-percentile is found for the summer season. The data from Prestebakke show the opposite trends with increasing values both in the summer and the winter seasons. As discussed above there are no obvious reasons why the data from Prestebakke show these differing trends.

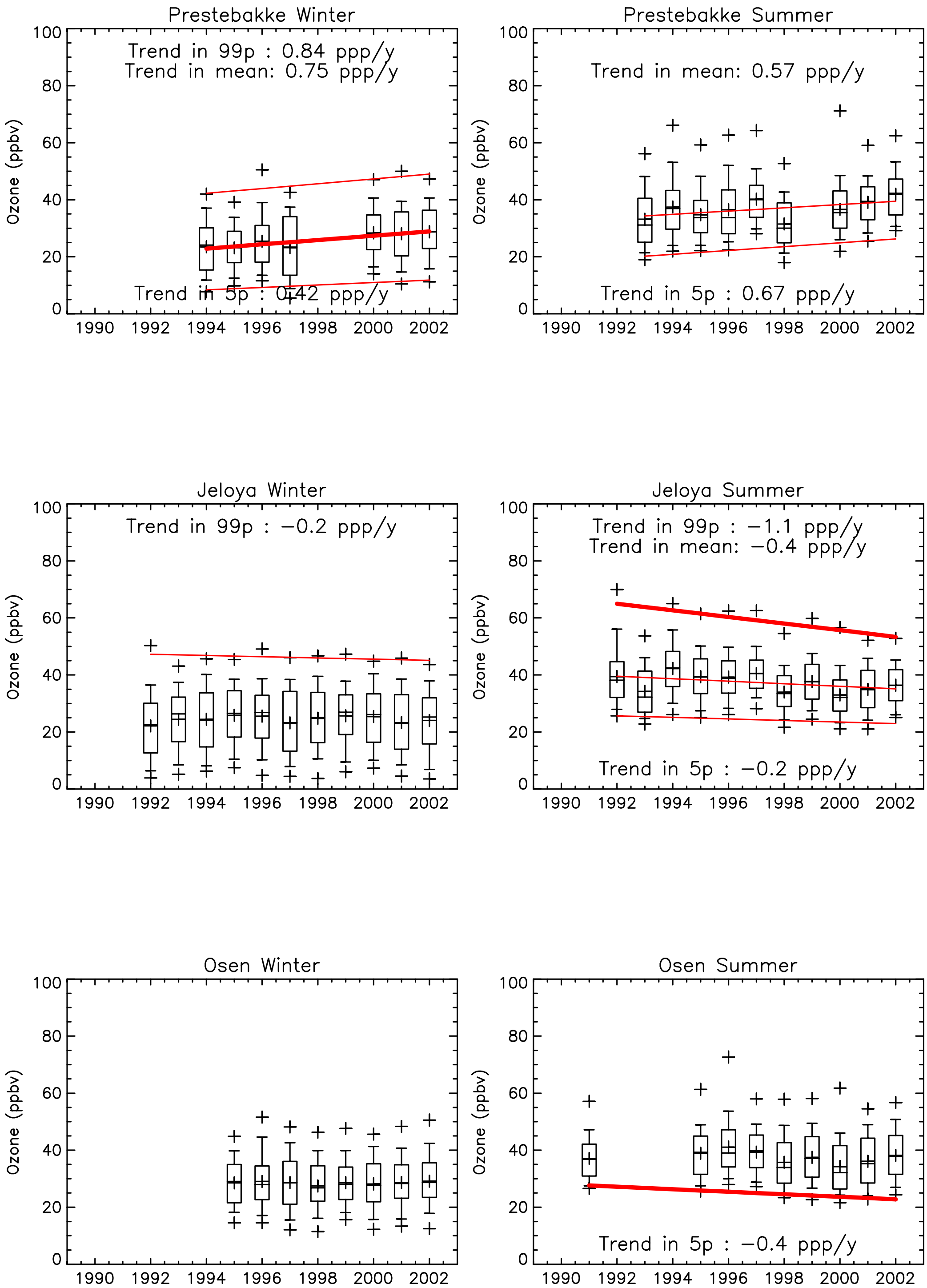
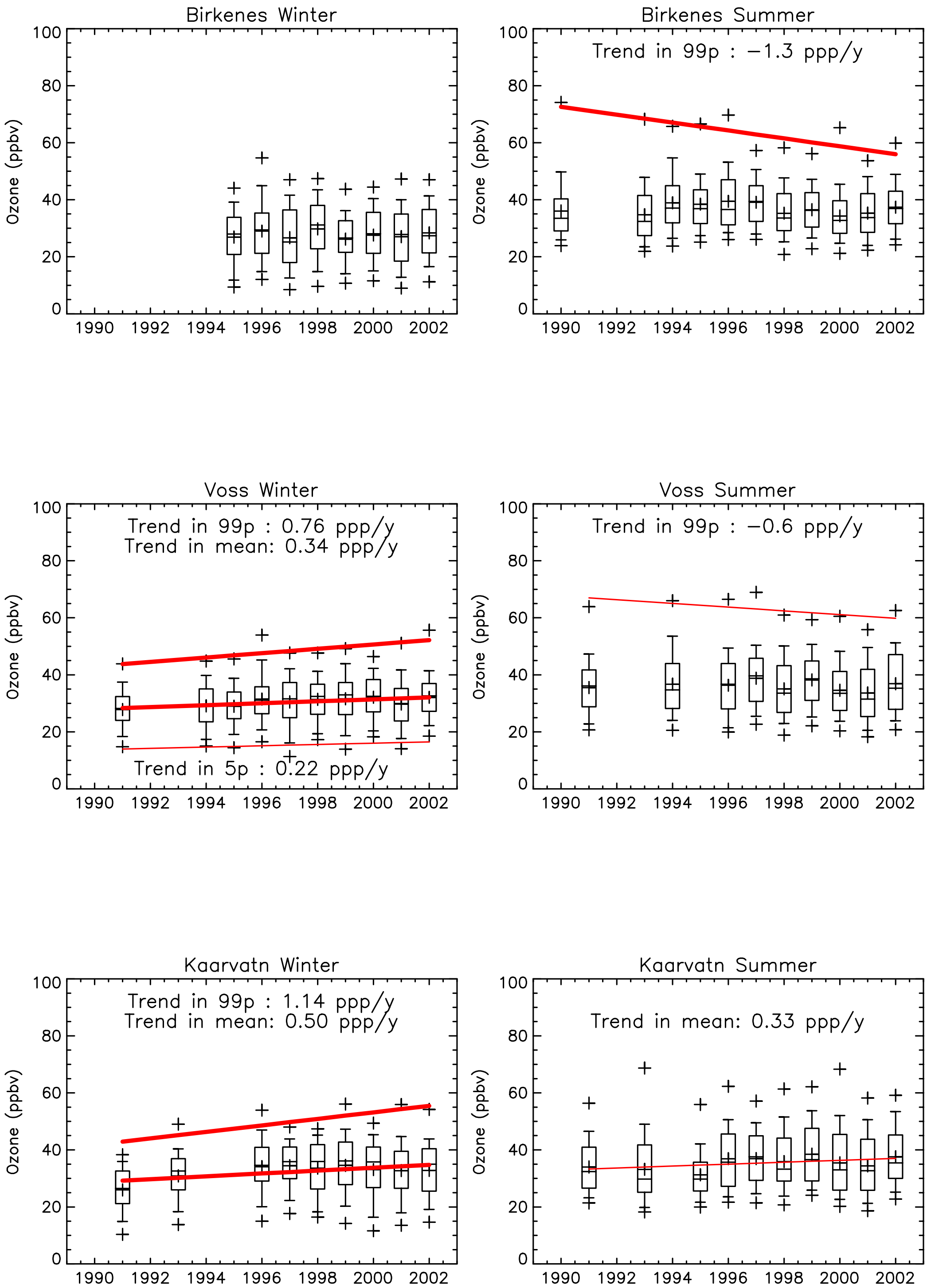


Figure 6: Trends in six months' means and percentiles of filtered daytime average ozone data 1990-2002. The bars give the 10-, 25-, 50-, 75- and 90-percentiles, and the crosses mark the 5- and 99-percentiles and the means. A 70% data coverage was required for each season. Lines mark the linear trends in the 5- or 99-percentiles or the mean if significant on a 1-σ level (thin line) and a 2-σ level (thick line). Winter = Oct.-Mar; Summer = April-Sept.



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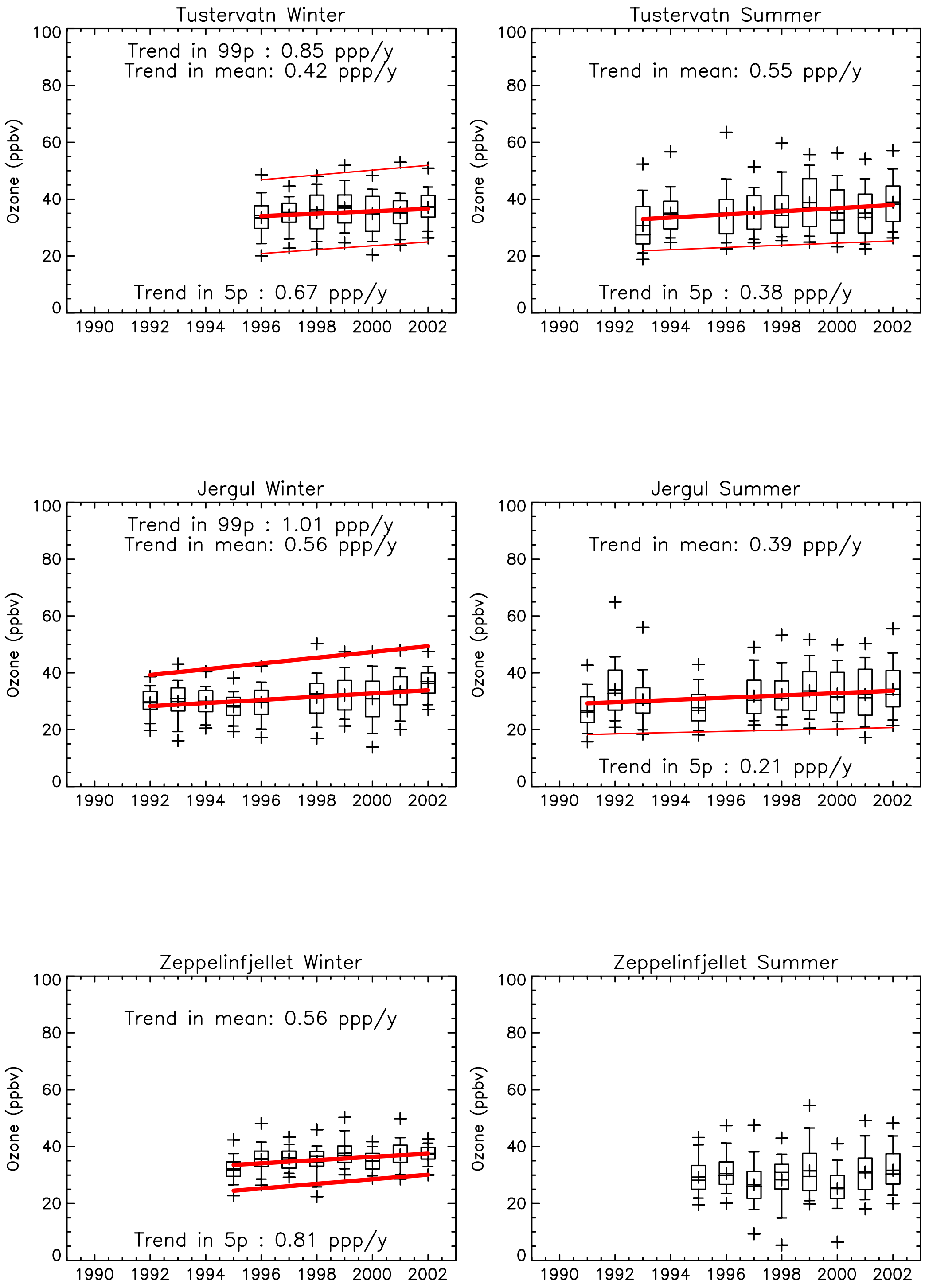

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4.1 Trends in relation to atmospheric long-range transport

To look more closely into the link between variations in long-range transport and observed ozone concentrations, the monitoring data were coupled with daily transport sector values based on 96h trajectories for the planetary boundary layer calculated by EMEP-MSC/W at the Norwegian Meteorological Institute (available at www.emep.int).

The daily sector values are based on 96 h backwards trajectories. The area around the arrival point has been divided into 8 equal sectors. If the origin of the circle points straight upwards, Sector 1 (North) begins from -22.5 degrees to 22.5 degrees. The next sectors are counted clockwise. The area around the arrival point extends from a radius of 150 km to a radius of 1500 km. The criteria for allocation of trajectories to one sector is that at least 50% of its given positions are found within that sector. Otherwise, sector 9 (inattributable) is allocated. Figure 7 shows the sectors for the site at Kårvatn. These daily sector indices were available for the years 1985-2001.

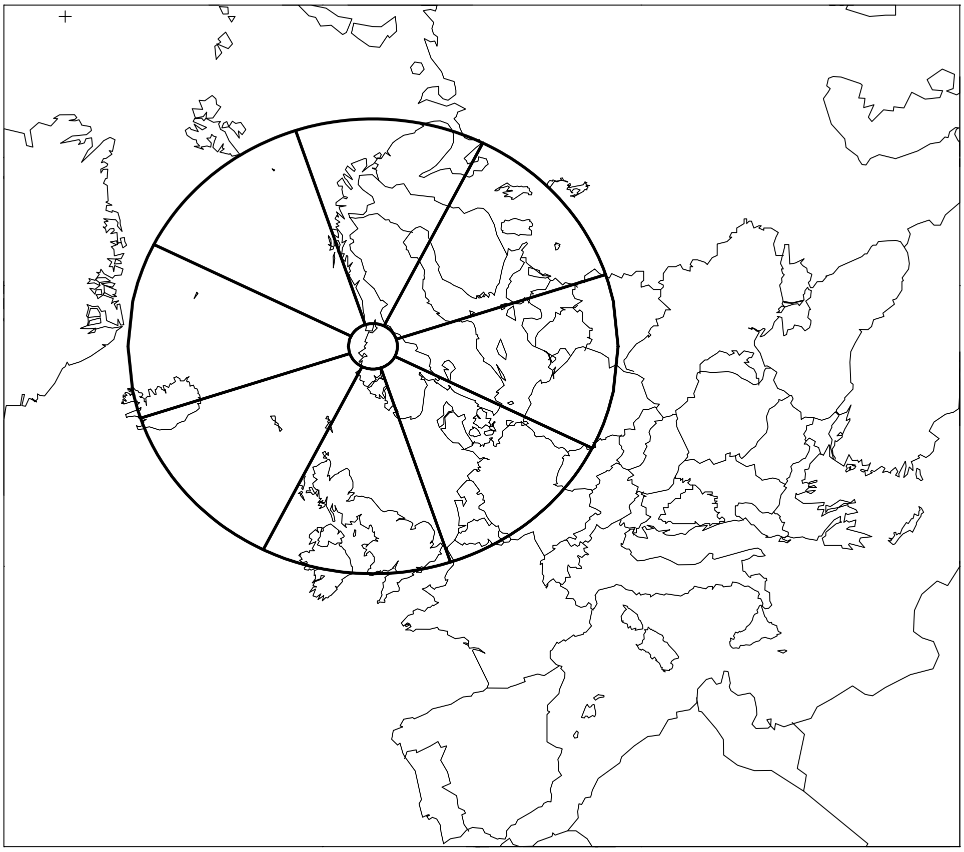

Figure 7: Map showing the transport sectors calculated by EMEP MSC-W for the station at Kårvatn. The North Pole is indicated by a cross in the upper left corner.

Background air was defined as either of sector 1, 2, 7 and 8, i.e. N, NE, W and NW for all the Norwegian sites. Although this procedure mixes together Atlantic and Arctic air masses, we wanted the selection to capture as many days as possible in order to get a statistically more robust selection of data. Additionally, the data were split into two six-months seasons of winter (Oct.-Mar.) and summer (Apr.-Sep.) as above.

Figure 8 shows the annual medians (and 25- and 75-percentiles) of the normalized daytime ozone concentrations for the background sectors and for the two seasons separately. First of all, the results indicate substantial variations in the seasonal median background concentration from one year to another for several of the sites. Secondly, the results indicate a significant increase in the general background ozone concentration in both seasons at the northern sites (Kårvatn and northwards), and with the clearest trend in winter. Due to the strong fluctuations from one year to another the results indicated by Figure 8, it could not be concluded if the rise in background ozone concentration is due to a period of several years in the mid 1990-ies with low background values followed by another period with generally high concentrations, or if a more steady growth is going on.

At the stations further south the results are more mixed. The data from Prestebakke give a strong increase in the last 4-5 years contrary to the nearest station Jeløya which doesn't show any trend in the data, and also contrary to the other stations in the southeast (Osen and Birkenes). The reason for this discrepancy is not clear. The data for these years are believed to be of high quality at all sites, and differing long-term trends in the background ozone level at these neighbouring sites are unexpected. Indications of an increase in the background ozone are to some extent seen also at the southern sites, but less significant.

Another interesting question is whether there has been any trend in the atmospheric transport of air masses into the monitoring sites. Based on the daily sector indexes described above, the frequency of various transport regimes during the years was calculated. To take into account that the filtered data are scattered in time, only those days with valid (filtered) ozone measurements were used in these calculations. Furthermore, to simplify the evaluation, the sectors were combined into the "background" as explained above (sector N, NE, W and NW), E+SE, S+SW and the undefined situations. The results for the different sites and for the two six-months seasons, separately, are shown in Figure 9.

No overall clear trend in transport sectors could be drawn from these results. The data for Jergul/Karasjok indicate that the E-SE sector was more common in summer in the period 1997-2001, compared to the previous years. Also for the Zeppelin Mountain a peak in the E+SE sector in the period 1998-2000 is found, but with lower frequencies in 1997 and 2001.

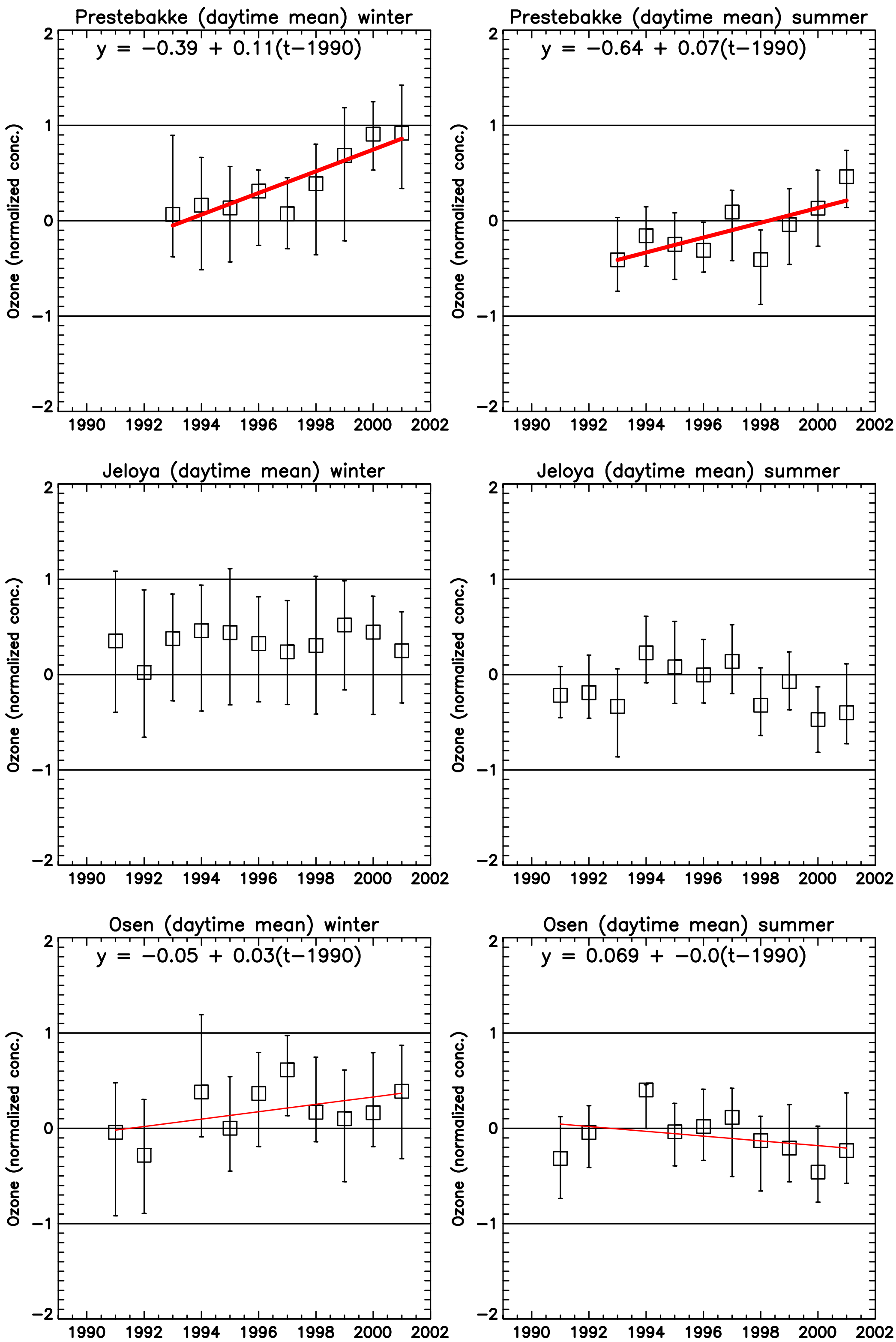

Figure 8: Trends in normalized ozone concentrations based on filtered data and only for background transport sectors (based on trajectory filtering). The boxes mark the annual medians, and the error bars the 25- and 75-percentiles. Lines mark the linear trends in the medians if significant on a 1-σ level (thin line) and a 2-σ level (thick line).

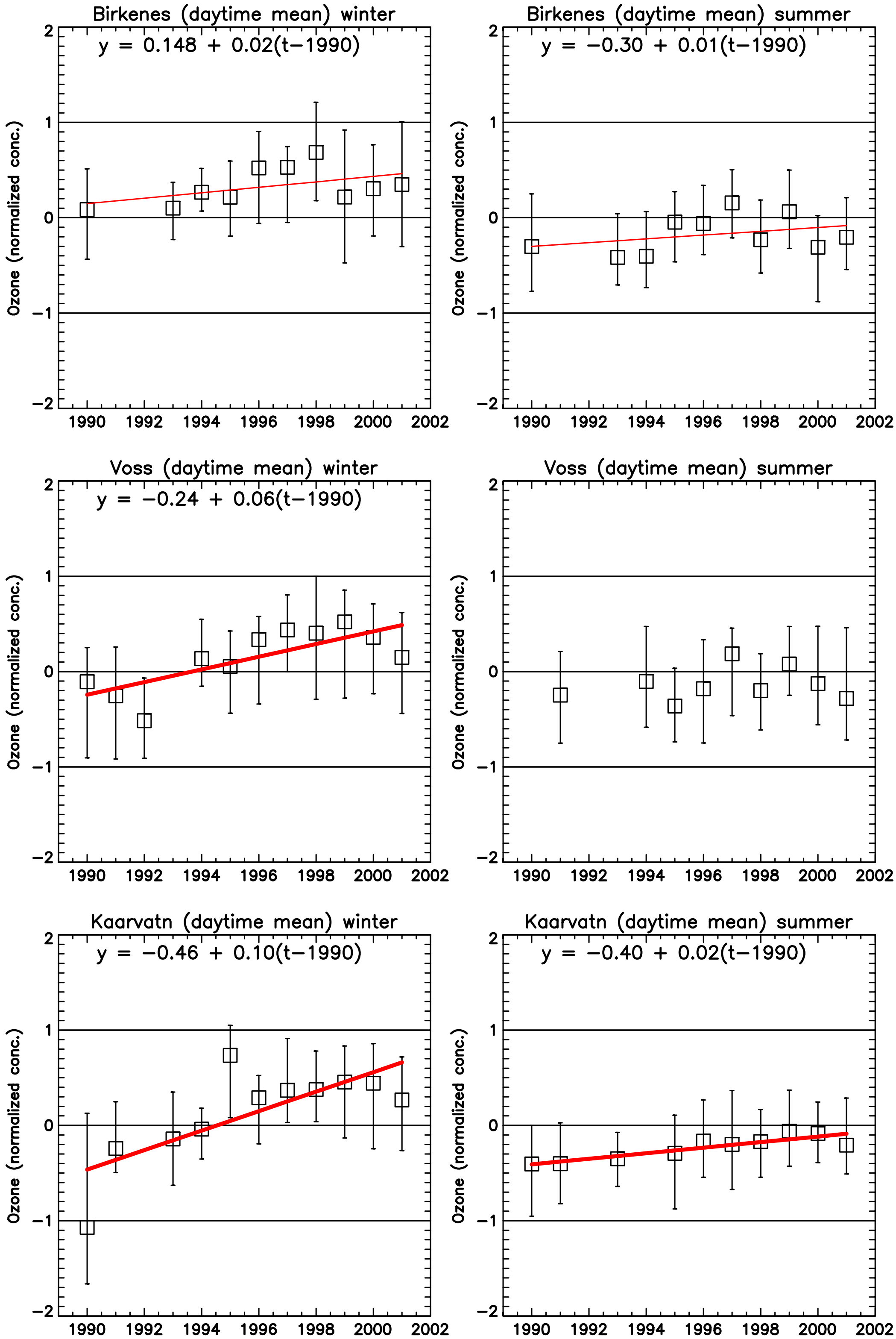

Figure 8, contd.

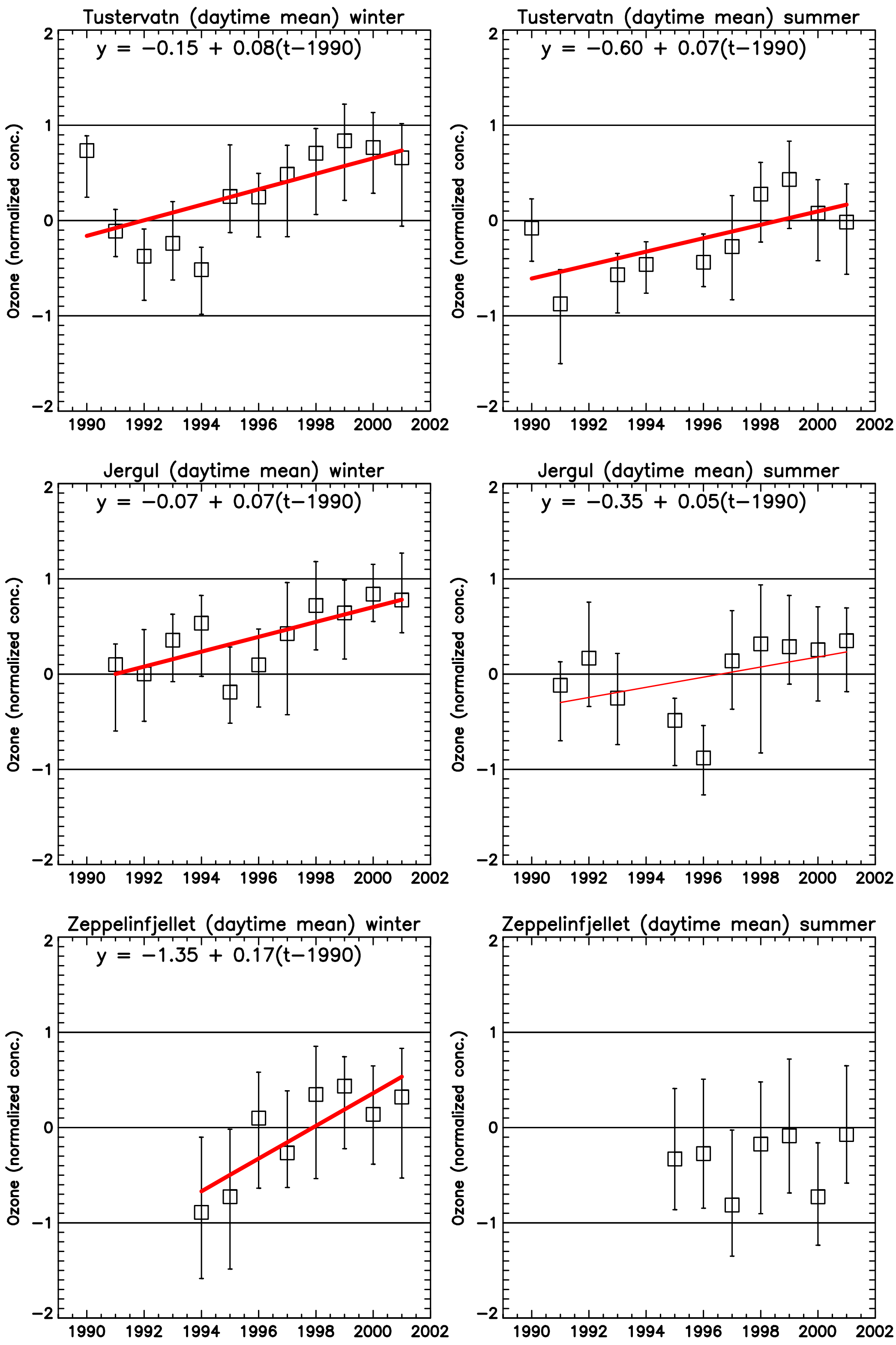

Figure 8, contd.

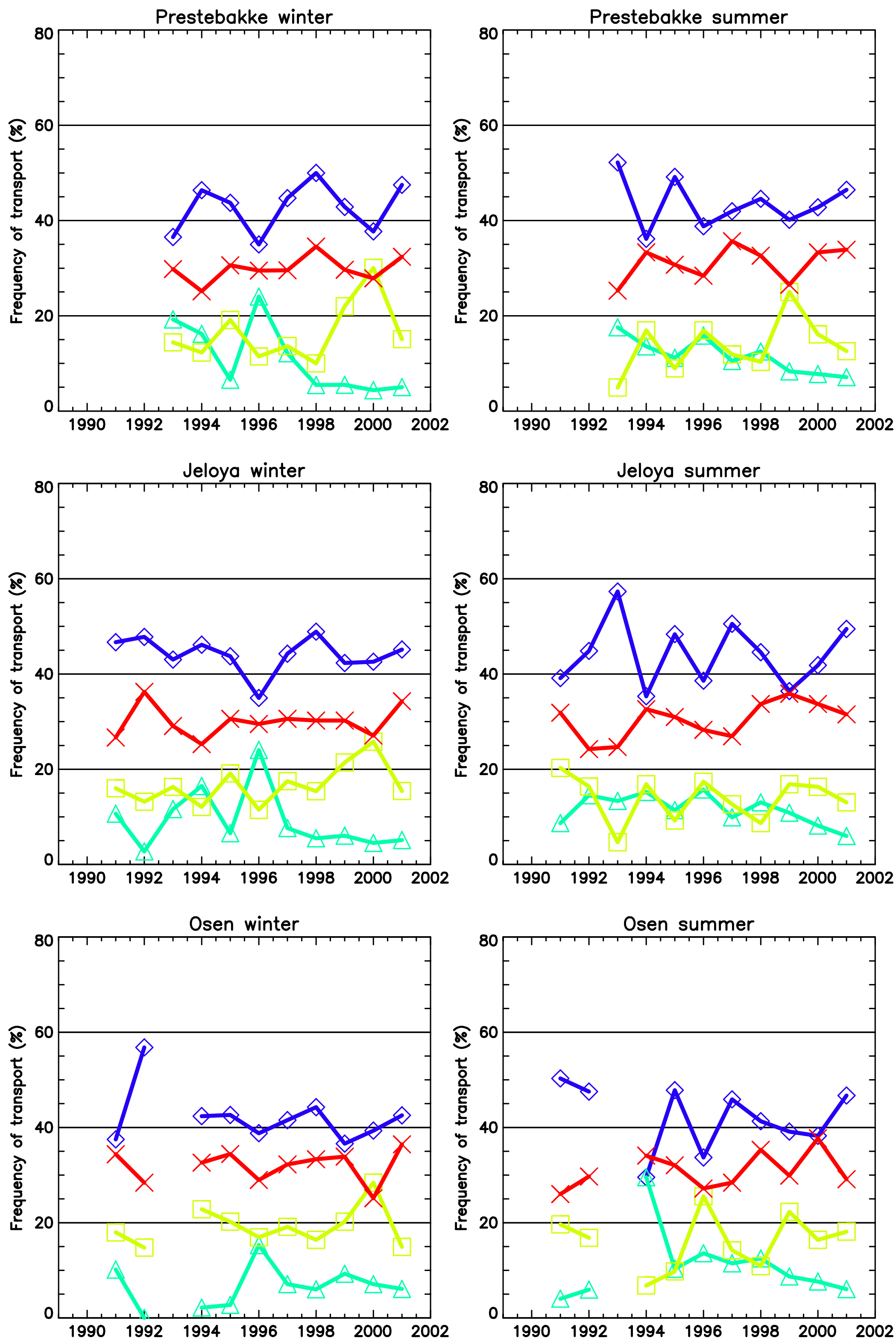

Figure 9: Frequency of various transport types based on trajectory sectors. Blue diamonds: Clean background; blue-green triangles: E-SE; Yellow squares: S-SW; red crosses: Undefined.

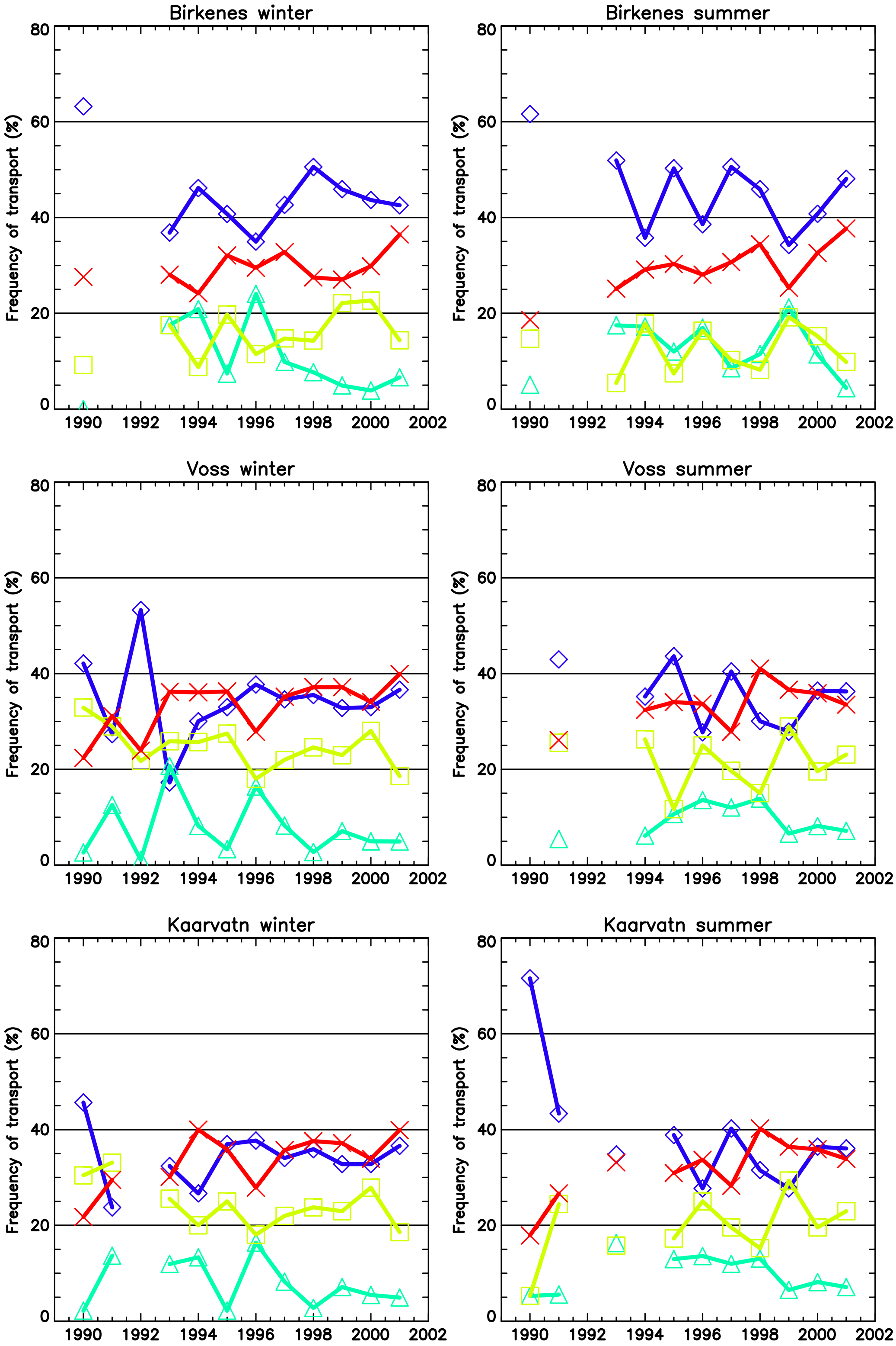

Figure 9, contd.

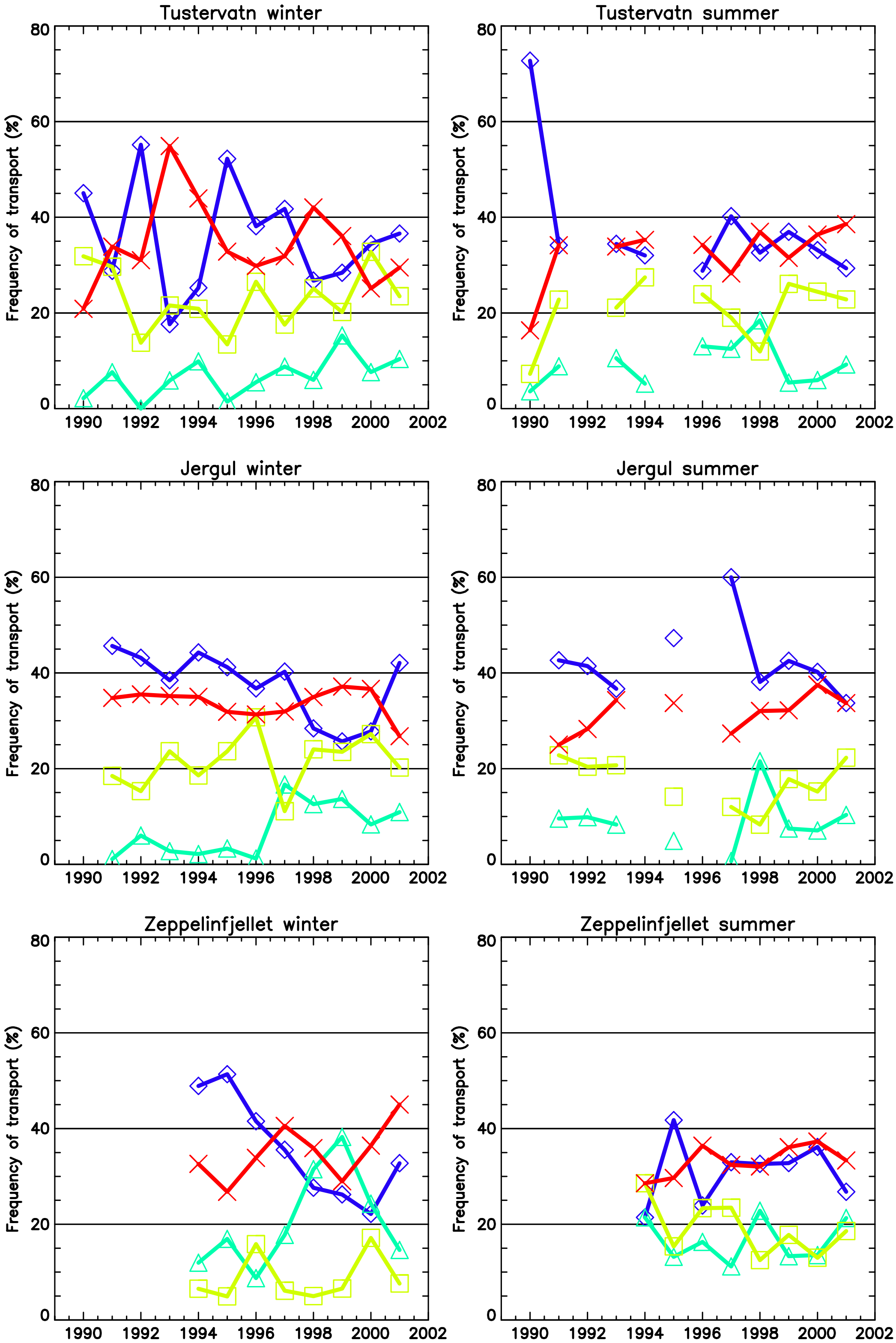

Figure 9, contd.

5 Conclusions

An evaluation of the Norwegian ozone monitoring data and the long-term trends in these has been presented. Due to varying procedures for the ozone monitoring back in time a detailed investigation of the monitoring history of each of the stations was required. A particular problem was the infrequent calibrations of the various instruments until approximately 1997 when a more rigorous procedure for quality assurance, field inspections and field calibrations was started.

Based on the revealed monitoring history periods of the ozone data were rejected for the purpose of long-term trend studies, while regarded valid for other applications with less strict requirements for accuracy. This reduced the amount of data and the length of the time series considerably, decreasing the relevance of the trend study, but increasing the accuracy and reliability of the underlying data. All further analyses were based on this filtered data set.

These data indicated a reduction in the 99-percentile of the daily (daytime) ozone data in the southern part of the country in the summer half year, although not seen at all sites, of the order of 1 ppbv/year during the 1990-ies. It should be stressed that the uncertainty in the trend rates is high due to the variability from year to year and due to the fragmentary data set. The station Prestebakke was in contradiction to this showing significantly increasing ozone concentrations, particularly marked since 1999. The reason for this is unclear, but must probably be explained by changes in local conditions (land use, nearby emissions etc).

For all sites from Voss and northwards a statistically significant growth in the mean ozone concentration was found for the winter half year with rates of the order of 0.3-0.5 ppbv/year. An increasing trend in the mean ozone concentration was found also in the summer season for a number of the same sites, but not for all, and generally with less significant trends.

An analysis of the ozone data with respect to atmospheric air mass transport was carried out to separate cleaner background conditions from air masses more influenced by European anthropogenic air masses. This indicated a significant increase in the ozone concentration in the background air masses at the northern sites (north of mid-Norway) both in summer and winter. Due to the large variations from year to year it is not clear if this could be explained by a steady growing ozone concentration in the background or due to shifts from one 3-4 years period with low background ozone in the mid 1990-ies to another period of high background ozone values at the end of the 1990-ies. At the stations in the south no clear trend in the background air masses was possible to identify. Whether this is due to problems of separating the types of air masses or reflects that an increase in background ozone is confined to the northern region is not clear.

Based on the trajectory data used (4-days trajectories for the planetary boundary layer) no clear trends could be seen in the transport pattern itself. Trajectories for Jergul/Karasjok in the far north of the Norwegian mainland indicated higher frequency of transport from E-SE during the last part of the 1990-ies and less periods with background air masses. For the other sites there was no corresponding trends in the frequency of transport sectors.

6 References

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Schjoldager, J. and Stige, L. (1980) Ozone measurents in Telemark, Oslofjorden and Oslo during the summer of 1979. Lillestrøm, Norwegian Institute for Air Research (NILU OR 5/80) (in Norwegian).

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7 Acknowledgements

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Appendix A

Monitoring history (1990–2002)

NO-01 Birkenes

A DASIBI-SN4275 was in operation since October 1987. The last calibration (in the lab) was in April 1989. This instrument was running at the site until replaced by a ML8810-SN1134 11th March 1993. The ML8810 was calibrated at the lab before installed in the field, while the DASIBI was not calibrated. The reason for the shift of monitor was that the old DASIBI showed signs of being unstable, implying that spikes could occur in the data. The ML8810-SN1134 was running at the site without calibration until late December 1994 when it was destroyed in a fire at the station caused by a lightning stroke. A new monitor API400-SN214 was installed 5th January 1995 after calibration in the lab. In February 1995 another lightning strike caused a new fire and the API monitor was also destroyed. It was replaced by a API400-SN213 15th February 1995 (after calibration in the lab). In July the trafo of the new API was burnt and 18th July 1995 the monitor was replaced by an API400-SN401 (calibrated in the lab before installation). The SN401 was repaired and calibrated (after another lightning strike) in June 1996 in the lab and reinstalled 19th June 1996. Following this, the monitor was replaced by an API400-SN911, with both monitors calibrated, 13th August 1997, which in turn was replaced by an API400-SN981 (both monitors calibrated) 19th May 1999. The SN-981 was calibrated with a transfer standard 16th November 2000, 19th December 2001 and 2nd December 2002.

NO-41 Osen

Ozone has been monitored at Osen since 1990. In the beginning a DASIBI instrument was used, and no calibrations of this monitor are given. The monitor was replaced by a ML8810-SN836, calibrated in the lab, 23rd January 1991. A leak was revealed during a routine check in the field in March 1994. The last previous field inspection was 22nd September 1993. The monitor was taken to the lab for repair, calibrated and then reinstalled at the station 23rd March 1994. Following problems with the monitor in 1994, it was replaced 18th August 1994 by an API400-SN289 after calibration in the lab. This monitor ran without calibration until replaced by an API400-SN341 31st October 1997. The new monitor was calibrated in the lab, and the two monitors were run in parallel at the station for a few days with satisfactory results. The SN341 started showing suspiciously low values in January 1998, was taken to the lab and replaced by a (calibrated) API400-SN93 6th April 1998. Parallel measurements in the field revealed a reduced sensitivity of the old SN341. The data between January and March 1998 have been corrected. The SN93 was subsequently calibrated in the field 11th May 1999. On 25th October 2000 the instrument was replaced by an API400-SN376 following the standard procedures for replacement. This monitor was running at the station until replaced in June 2003. The instrument was not calibrated during this period.

NO-15 Tustervatn

A Dasibi instrument was running from 1989 until replaced by a ML8810-SN662 6th August 1990. No calibrations are reported for the Dasibi. During a routine inspection a leak in the valve was found and fixed 9th December 1991. The last previous field inspection was 26th June that year. The SN662 monitor was running until it broke down by the end of 1992. The instrument was replaced by a ML8810-SN663 9th March 1993 (calibrated in the lab prior to this). At a routine inspection 1st December 1993 a leak in the valve was discovered and repaired.

The last previous field inspection was 6th October that year. The SN663 stopped in October 1995, probably due to lightning and was replaced 18th October by a ML8810-SN662 (calibrated in the lab). The old SN663 could not be calibrated. The SN662 instrument was replaced by an API400-SN376 26th June 1997 following the standard procedure for replacement (both monitors calibrated). The monitor was calibrated with a transfer standard in the field 29th July 1999 and was replaced by another API-SN213 at 7th September 2000, following the standard procedures for replacement. The SN213 was calibrated in the field 24th July 2001.

NO-42 Zeppelin Mtn (Ny-Ålesund)
A ML8810-SN659 was running at the site from September 1989. Five years later the instrument was brought to NILU's lab because the monitor started to behave unstable and was replaced 8th September 1994 by a ML8810-SN611. No calibrations of the SN659 monitor are reported. The SN611 monitor was running until replaced by a ML8810 SN1150 17th May 1997 in accordance with a standard replacement program. Thus, both monitors were calibrated before and after replacement and in the field. Good agreement was found. GAW representatives visited and inspected the site and calibrated the SN1150 monitor against the GAW standard 17th September 1997 with good results. A separate note was prepared by GAW. The SN1150 was replaced by an API400-SN980 19th February 1998, following the standard program for replacement (both monitors calibrated). The next field calibration was done 16th November 1998. The monitor was calibrated 11th June 1999 prior to moving to the temporary site down by the sea ("Gruvebadet") while a new monitoring station was built on the Zeppelin Mountain. The SN980 monitor was moved back to the new station 3rd February 2000. It was replaced by an API400-SN534 20th February 2001 which in turn was replaced by an API400-SN612 13th February 2002. The standard replacement procedure was followed during all these occasions (old and new monitors calibrated before, in and after replacement)

NO-43 Prestebakke
A DASIBI-SN4229 was running at the site since 1987. No calibration of this instrument is given. On a routine inspection 6th December 1990, it was discovered that the filter was blocked (packed). On replacement of the filter it was noted that the ozone concentration increased from 33 to 39 ppbv. The last previous field inspection was 18th May 1990. On 24th March 1993 the DASIBI instrument was replaced by a ML8810-SN1150 (calibrated on the lab prior to field installation). The SN1150 was running at the site until replaced 29th April 1997 by a ML8810-SN836 following the standard procedure for replacement. Very good agreement was found between the two monitors. On a routine inspection 20th November 1997 a leak in the valve was detected, and the last previous field inspection was 2nd September that year. The SN836 was then replaced by an API400-SN911 following the standard procedures for replacement 3rd June 1999. This revealed that the old SN836 gave values of the order of 5 ppbv too low compared with the calibrator and the new API-monitor. The API-monitor was calibrated in the field 19th December 2000 and replaced by an API400-SN910 (following the standard procedures) 25th October 2001.

NO-39 Kårvatn
A DASIBI instrument was running at the site since 1988. No calibrations are reported. 26th June 1990 the instrument was replaced by a DASIBI 1008-RS SN4703 which was calibrated in the lab before field installation. On a routine inspection 5th November 1992 a pump failure is discovered, giving a sensitivity probably of the order of only 70%. The last previous inspection was 19th June that year. The pump was replaced in the field and the monitor was running until replaced by a ML8810-SN1228 14th December 1992 (calibrated in the lab). No calibration of the old DASIBI monitor is given. The new instrument was running until replaced by an API400-SN341 19th May 1995 after the old instrument have shown problems with "stabilizing" during a long period (since April 1994). When brought to the lab, it turned out that there was an electronic fault with the old SN1228 (the capacitor had dried out). The effect of this electronic fault is probably an increase in the number of missing monitoring values, while the 5-min and 1-h averages will presumably be correct although based on less values and thus becoming more uncertain. The API monitor was replaced by another API400-SN910 29th September 1997 following the standard procedure for replacement. This monitor was calibrated in the field on 1st June 1999 and 14th June 2000. On 21st June 2001 the instrument was replaced by an API400A-SN535 following the standard procedures for replacement. This monitor was calibrated in the field 15th October 2002.

NO-48 Voss
Monitoring of ozone at Voss started in April 1990. The instrument, a ML8810-SN836, was calibrated in the lab before installation in the field. At a later field inspection it was discovered that the valve didn't work, and the instrument was replaced by a ML8810-SN662 16th October 1990. This instrument was running until it stopped in September 1993. It was brought to NILU, repaired, calibrated in the lab and reinstalled at the site 4th November 1993. There it was running until replaced 27th June 1995 by a ML8810-SN1228. The old SN662 was not calibrated directly after this, but in October the same year, and before any maintenance of the instrument. This calibration showed that the monitor was very accurate. The SN1228 was running until 16th September 1996 when replaced by a ML8810-SN659. Once again the old SN1228 monitor was not calibrated directly after the shift, but a lab calibration in December 1996 showed a very good agreement. In September 1997 the SN659 monitor was run in parallel with a ML8810-SN663 with very good results. On 15th June 1998 the SN659 instrument was replaced by an API400-SN341, following the standard procedure for replacement. The API monitor was calibrated in the field 6th September 1999 and 13th October 2000. On 1st June 2001 the monitor was replaced by an API400-SN1077, following the standard procedure, and it was calibrated in the field 26th September 2002.

NO-52 Sandve
Monitoring of ozone at Sandve started 18th June 1996, using a ML8810-SN663 which was calibrated in the lab before installation. The instrument was replaced by a ML8810-SN611 4th September 1997, following the standard procedure for replacement. At a routine field inspection 26th August 1998 an error in the valve was discovered and repaired. The last previous inspection was 16th July the same year. At the next field inspection 12th November 1998, the monitor had stopped

and the monitor was replaced by an API400-SN912. This monitor was calibrated in the field 6th October 1999 and was replaced by an API400-SN913 3rd October 2000, following the standard procedure. This monitor was in turn calibrated in the field 5th September 2001.

NO-56 Hurdal

The monitoring station at Hurdal was opened in the beginning of 1997 to replace the more locally polluted site at Nordmoen. On 17th March 1997 ozone monitoring started with a ML8810-SN1228. At a field inspection 18th June the same year a small leak was detected in the valve which was then repaired. At a routine field inspection 21st April 1998 a leak in the valve was discovered and fixed. The last previous field inspection was 8th January the same year. At the next inspection 2nd October the same year a leak was detected once more and repaired once again. On 30th October 1998 the monitor was replaced by a ML8810-SN663 and the monitor site was moved to a forested area nearby and the air intake lifted to 15 m above ground. This monitor was calibrated in the field 25th August 1999, and it was replaced by an API400-SN1077 2nd February 2000, following the standard procedure for replacement .This monitor was in turn replaced (following the standard procedure) by an API400-SN980 7th March 2001.

NO-30 Jergul / NO-55 Karasjok

Ozone was monitored at Jergul since 1988, in the beginning by a DASIBI instrument. A DAS-SN4228 is reported to be at the site at the beginning of 1990. No calibrations of this instrument are given. At a field inspection in January 1991 the instrument was reported to be highly unstable. The last previous inspection was 9th September 1990. The monitor was replaced by a ML8810-SN874 on 23rd May 1991. In Autumn 1992 the monitor was brought to NILU for repair, was calibrated in the lab and reinstalled at Jergul 1st September 1992. The instrument was then running until it broke down 20th August 1994. It was brought back to NILU for repair and was calibrated in the lab and then reinstalled at Jergul 3rd September 1994. The instrument was then running until another breakdown in October 1996, when it was once again repaired and reinstalled at the site 10th October 1996. The monitor was then moved to the new station at Karasjok 4th February 1997. The instrument was experiencing several periods with failure during the following months and was finally replaced by an API400-SN289 18th November 1997. The old ML8810 was not working and was thus not possible to calibrate when replaced. The API monitor was calibrated in the field by a transfer standard 25th February and 18 November 2000, and was then replaced and brought to NILU in 2003 following the standard procedures for replacement. Thus, the monitor was running without calibrations for longer periods than the normal 1-year period at other stations. However, as these calibration results were very satisfactory, the data could still be regarded valid.

NO-45 Jeløya

Ozone has been monitored at Jeløya since the start of the 1980-ies. A DASIBI instrument was running by the beginning of 1990. No calibrations of this instrument are given. The DASIBI monitor was replaced by a ML8810-SN836 (calibrated at the lab prior to installation) 5th June 1991. The ML8810 was repaired and calibrated in the lab 19th August 1992. The instrument was replaced

26th April 1993 by an new API400-SN93. The old ML8810 was stopped and brought to NILU for calibration in the lab with very good results. According to the logg book, the new API400 was tested at the site for a period of several months, probably by running in parallel with NILU's ozone standard, although this is not explicitly documented. On 11th October the API400 was calibrated in the lab and was from that time running in a standard way at Jeløya. The monitor was running until replaced by an API400-SN213 31th October 1995. Both monitors were calibrated with good results. On 16th April 1996 the SN213 was replaced back again with the SN93 and both instruments calibrated. On 22nd May 1997 the SN93 was replaced back with the SN213 following the standard procedure for replacement. The SN213 was calibrated in the field 14th October 1999. The monitor was then replaced 11th May 2000 with an API400A-SN185 as part of the standard replacement program. This monitor was running at the site until the monitoring station was closed down in 2003. There were no calibrations of the monitor during this period.

Appendix B

List of reports

The national monitoring programme:

Bamble-undersøkelsen. Måling av luftforurensning i Grenland 1975/1976.
NILU OR 2/77 by J.E. Hanssen and B. Sivertsen.
Lillestrøm, Norwegian Institute for Air Research, 1977.

Målinger av ozon i nedre Telemark sommeren 1977.
NILU OR 23/78 by J. Schjoldager and O. Thorstad.
Lillestrøm, Norwegian Institute for Air Research, 1978.

Målinger av ozon i Oslo og nedre Telemark sommeren 1978.
NILU OR 10/79 by J. Schjoldager and O. Thorstad.
Lillestrøm, Norwegian Institute for Air Research, 1979.

Målinger av ozon i nedre Telemark, Oslo og Oslofjorden sommeren 1979.
NILU OR 5/80 by J. Schjoldager and L. Stige.
Lillestrøm, Norwegian Institute for Air Research, 1980.

Målinger av ozon i nedre Telemark, Oslo og Oslofjorden sommeren 1980.
NILU OR 42/81 by J. Schjoldager, R. Dreiem, G. Gundersen, L. Stige and B. Tveita.
Lillestrøm, Norwegian Institute for Air Research, 1981.

Målinger av ozon, Østlandet – Telemark – Sørlandet, 1981-1983. Målinger av PAN, Telemark, 1983.
NILU OR 34/84 by J. Schjoldager, R. Dreiem, B.M. Wathne, T. Johannessen, L. Stige and B. Tveita.
Lillestrøm, Norwegian Institute for Air Research, 1984.

Målinger av ozon, Østlandet – Telemark – Sørlandet. Målinger av PAN, Telemark, 1984-1985.
NILU OR 64/87 by J. Schjoldager, R. Dreiem, T. Krognes, T. Johannessen, L. Stige and B. Tveita.
Lillestrøm, Norwegian Institute for Air Research, 1987.

Målinger av ozon i Norge 1986.
NILU OR 35/88 by K. Hoem, R. Dreiem, T. Krognes, J. Schjoldager, L. Stige and B. Tveita.
Lillestrøm, Norwegian Institute for Air Research, 1988.

Målinger av ozon i Norge 1987.
NILU OR 53/89 by K. Hoem, R. Dreiem, J. Schjoldager, L. Stige and B. Tveita.
Lillestrøm, Norwegian Institute for Air Research, 1989.

Tidsrekker av ozondata og EUs ozondirektiv.
NILU OR 2/95 by J. Schjoldager.
Kjeller, Norwegian Institute for Air Research, 1995.

Statens forurensningstilsyn (1989) Overvåking av langtransportert forurenset luft og nedbør. Årsrapport 1988. Oslo (Statlig program for forurensningsovervåking. Rapport 375/89).

Statens forurensningstilsyn (1991a) Overvåking av langtransportert forurenset luft og nedbør. Årsrapport 1989. Oslo (Statlig program for forurensningsovervåking. Rapport 437/91).

Statens forurensningstilsyn (1991b) Overvåking av langtransportert forurenset luft og nedbør. Årsrapport 1990. Oslo (Statlig program for forurensningsovervåkning. Rapport 466/91).

Statens forurensningstilsyn (1992a) Overvåking av langtransportert forurenset luft og nedbør. Årsrapport 1991. Oslo (Statlig program for forurensningsovervåkning. Rapport 506/92).

Statens forurensningstilsyn (1992b) Virkninger av luftforurensning på helse og miljø: Anbefalte luftkvalitetskriterier. Oslo (SFT-rapport 92:16).

Statens forurensningstilsyn (1993) Overvåking av langtransportert forurenset luft og nedbør. Årsrapport 1992. Oslo (Statlig program for forurensningsovervåkning. Rapport 533/93).

Statens forurensningstilsyn (1994) Overvåking av langtransportert forurenset luft og nedbør. Årsrapport 1993. Oslo (Statlig program for forurensningsovervåkning. Rapport 583/94).

Statens forurensningstilsyn (1995) Overvåking av langtransportert forurenset luft og nedbør. Årsrapport 1994. Oslo (Statlig program for forurensningsovervåkning. Rapport 628/95).

Statens forurensningstilsyn (1996) Overvåking av langtransportert forurenset luft og nedbør. Årsrapport 1995. Oslo (Statlig program for forurensningsovervåkning. Rapport 663/96).

Statens forurensningstilsyn (1997) Overvåking av langtransportert forurenset luft og nedbør. Årsrapport 1996. Oslo (Statlig program for forurensningsovervåkning. Rapport 703/97).

Statens forurensningstilsyn (1998) Overvåking av langtransportert forurenset luft og nedbør. Årsrapport 1997. Oslo (Statlig program for forurensningsovervåkning. Rapport 736/98).

Statens forurensningstilsyn (1999) Overvåking av langtransportert forurenset luft og nedbør. Årsrapport 1998. Oslo (Statlig program for forurensningsovervåkning. Rapport 768/99).

Statens forurensningstilsyn (2000) Overvåking av langtransportert forurenset luft og nedbør. Årsrapport 1999. Oslo (Statlig program for forurensningsovervåkning. Rapport 797/00).

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Norwegian Institute for Air Research (NILU)

P.O. Box 100, N-2027 Kjeller, Norway

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ABSTRACT

An evaluation of the Norwegian ozone monitoring data is presented. A detailed investigation of the technical monitoring history was required to prepare a subset of data with acceptable data quality for long-term trend studies. These data indicated a reduction in the 99-percentile of the daily (daytime) ozone data in the southern part of the country in the summer half year of the order of 1 ppbv/year during the 1990-ies for some sites. For sites further north a statistically significant increase in the mean ozone concentration of 0.3-0.5 ppbv/year was found for the winter half year. An increasing trend in the mean ozone concentration was found also in summer but less clear. Based on trajectory analyses a significantly increasing ozone trend in background air masses was found.

NORWEGIAN TITLE

Overvåking av bakkenært ozon i Norge fra 1977 til 2002

KEYWORDS		
Ozone	Assessment	EMEP

ABSTRACT (in Norwegian)

En gjennomgang av norske ozonmålinger siden starten i 1977 er foretatt. Dokumentasjon av målehistorikken var viktig for å sortere ut data med tvilsom eller ukjent kvalitet. Resultatene indikerer en nedgang i 99-prosentilen i Sør-Norge og en økning i bakgrunnskonsentrasjonen i nord på 1990-tallet.

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