EMEP/CCC-Report 10/2002

EMEP Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe

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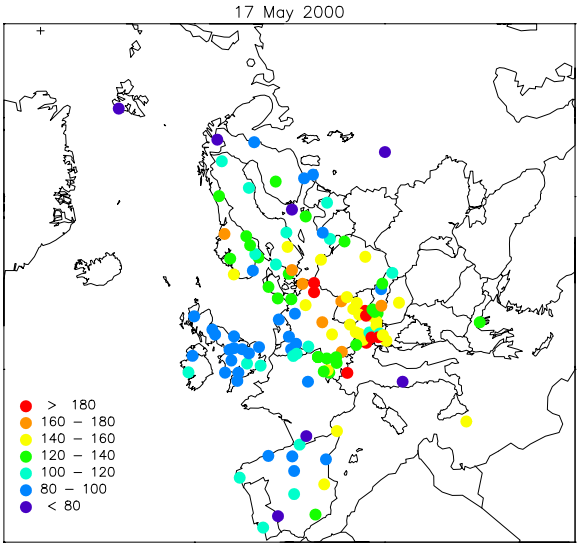

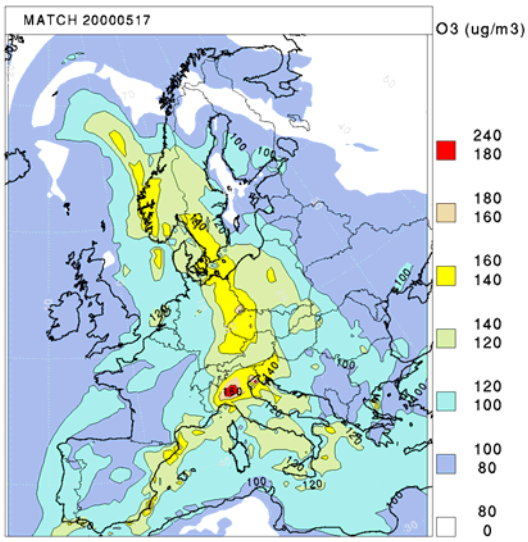

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A Nordic study

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Summary

This report presents the results from the project "Changes in Nordic ozone episodes due to emission reductions" carried out for the Nordic Council of Ministers. The aim of the project was to evaluate to what extent there has been a reduction in concentration level and extent of peak ozone episodes in Norway, Sweden and Finland as a result of European emission reductions during the last 10-15 years. EMEP and national ozone monitoring data were used in the project together with the MATCH model from SMHI.

When taken together the combined results indicate that it's very likely that European emission reductions have lead to a reduction in the extent and severity of surface ozone episodes in S-Norway and S-Sweden in the 1990-ies. Conservative estimates indicate a reduction of the order of 30 $\mu g/m^3$ ozone for the highest peak values and less for the less pronounced episodes. It is also likely that the number of episodes of exceedance of the EU threshold value of 180 $\mu g/m^3$, and the 160 $\mu g/m^3$ applied by the Norwegian Pollution Control Authority, has been reduced in the same period, although the few number of these episodes make this statement uncertain. Although the model indicates similar results for S-Finland, the agreement with the measurements were somewhat poorer, possibly reflecting that ozone episodes in Finland in general are more linked to transport from the east than in Sweden/Norway and that the emission data in Russia/E-Europe is less established.

These conclusions are based on a number of analyses carried out in the project. The evaluation of the model performance for six selected ozone episodes during 1999-2000 as compared with observed data was a main focus in the work. Additionally, long-term measurements were analysed separately, and, to a small extent, ozone episodes of similar characteristics in recent and previous years were compared with each other.

Inspection of the 99-percentiles of hourly measured ozone concentrations from the summer half years, during a 10-15 years' period indicated that in the southern part of the Nordic area episodic ozone concentrations are indeed decreasing. These trends are apparently less pronounced at the more eastern (Finnish) sites and in the northern parts of the region. A reduction in the 99-percentiles of the order of 10-20 $\mu g/m^3$ per ten years in southern and central parts of Sweden and Norway is indicated by the model as well as by the measurements.

In general, we found a good agreement between the observed and modelled time series of ozone. For the 2000 data the correlation coefficient based on daily maximum values during the summer half year was between 0.6 and 0.8 for each station separately and the correlation for 1999 was slightly lower. This gives a strong confidence to the model performance in general. Furthermore, the modelled regional (European) distribution of ozone in general also agreed well with the patterns evident from the European maps of daily maximum concentrations for the selected episodes. However, the comparison also revealed that the model had a tendency to underestimate the ozone concentrations during

the episodes. In some episodes fairly large underestimations were found. Although a reason for concern and for initiating further model development, this underestimation should indicate that the predicted change in ozone due to the emission changes from 1990 to 1999 could be regarded as a conservative estimate, i.e. that it's likely that the effects on ozone in reality may even have been larger than indicated by the model results presented in this report.

Changes in ozone episodes due to emission reductions
A Nordic study

1. Introduction

This report presents results from a collaborative project by the Nordic Council of Ministers (Hav- och luftgruppen) in 2001. As the project was closely linked to essential EMEP questions, both thematically and with respect to the monitoring data used, the project is reported as an official EMEP report. The thematic link to EMEP and the reporting to EMEP was a condition stated already in the project proposal.

The aim of the project was to evaluate to what extent there has been a reduction in concentration level and extent of peak ozone episodes in the Nordic countries as a result of emission reductions in Europe the last 10-15 years.

According to official, national data there has been a substantial reduction in anthropogenic emissions of NO_x and VOC the last 10-15 years in Europe (Mylona, 1999). For Europe as a whole the reduction was approx. 17% for both NO_x and VOC in the period 1987-1997 according to the reported data. There are, however, large differences between the countries. In countries that probably have the largest influence for generation of ozone episodes affecting the Nordic countries, such as Poland, Germany and United Kingdom, there have, according to the official numbers, been emission reductions of more than 40% in the same period. Thus, it's natural to ask if the effect of this reduction is observable as reduced ozone concentrations in the Nordic countries.

Based on studies conducted and collected in the EUROTRAC project TOR-2 (Roemer, 2001, and references therein), it was concluded that there is strong evidence that annual maximum concentrations of ozone have been reduced during a ten years period (from the last part of the 1980-ies). Furthermore, based on a study of some hundred German ozone monitoring sites, Beilke and Wallasch (2000) found a strong reduction in the exceedances of the EU threshold limits 180 $\mu g/m^3$ and 240 $\mu g/m^3$ during the period 1990-1999. At the same time, several authors report indications of an increasing trend in the background ozone concentration. Monitoring data from the United States from the period 1980 to 1998 show that ozone concentrations have decreased at the high end of the probability distribution and have increased at the low end (Lin et al., 2000). Similar results are also reported for sites in the UK (NEGTAP, 2001). For high-altitude monitoring sites in Germany, Scheel (2001) found a decreasing ozone seasonal amplitude with reduced summer concentrations and increased winter concentrations.

For several reasons it is worthwhile to focus on the Nordic countries in particular. Firstly, Norway, Sweden and Finland, as studied here, are in some respects located at the "fringe" of the European continental air mass and their monitoring

networks are well suited to separate occasional European pollution episodes from the dominant cleaner, background air masses. Secondly, as mentioned, Europe's largest national emission reductions in ozone precursors have apparently been experienced in the main source areas for the Nordic countries.

The project work was based on a combined study of monitoring data and modelled concentrations. EMEP ozone monitoring data were used in the project together with the MATCH model from SMHI. The idea was to select a number of marked ozone episodes affecting one, or preferably several, of the Nordic countries and to study these in more detail by the model. Official EMEP emission data for 1990 and 1999 were used in the study to calculate the temporal development of the selected episodes that were compared with measurements. In such a study the natural question is how one could trust that the modelled response actually have taken place. Whereas this is impossible to answer directly, the idea was that episodes with sufficiently good spatial and temporal fit between the modelled and observed data would give confidence in the model results, and could be used to make statements about the likely change in Nordic ozone episodes that have taken place during the 1990-ies.

2. Measurement data and model description

2.1 Measurement data

The study was based on hourly surface ozone measurement data from the EMEP network from 1999 (Hjellbrekke and Solberg, 2001) and 2000 (Hjellbrekke and Solberg, 2002). For comparison with the modelled results we mostly used daily maximum values, referring to UT+1, both for the measurements and for the model data. Whereas the Norwegian and Swedish data were already given at the time UT+1, the Finnish data were given in UT+2 that were taken account for when comparing with the model. Details regarding the exact definition of the hourly values, i.e. whether centred at the whole hour, starting at the whole hour, etc. was, however, not considered, as such differences would have negligible influence in this study.

All ozone monitoring data using the standard UV monitor, give the concentration values in mixing ratio, i.e. ppbv, directly. In this report all data are presented as $\mu g/m^3$, which was calculated simply as the ppbv value multiplied by a factor of 2. This was done in the same way for the modelled values as for the observed values, thus giving a coherent data set. The true atmospheric concentration values in $\mu g/m^3$ will, however, depend on the atmospheric pressure and temperature, but that was not considered here.

During the project work all ozone monitoring sites from Finland (Laurila, 1999), Norway (Solberg et al., 1997) and Sweden (Lindskog and Kindbom, 2001) were used, including national sites not reported to EMEP. Only a representative selection of the sites, shown in Figure 1, is presented in this report in order to keep the presentation clear and not too voluminous. Additionally, ozone monitoring data from the complete European EMEP network were used in the regional comparisons with the model to study the model's spatial performance vs. the measured regional distribution.

Figure 1: Map of the Nordic ozone monitoring sites used in this study. Time series are only shown for the underlined stations.

2.2 Model description

The MATCH (Multi-scale Atmospheric Transport and Chemistry) photochemistry model for Europe was used in this study. Details about the model can be found in Appendix A and references therein.

2.2.1 Model domain

The MATCH photochemistry model domain, shown in Figure 2, covers most of Europe, with a horizontal resolution of ca 44×44 km^2.

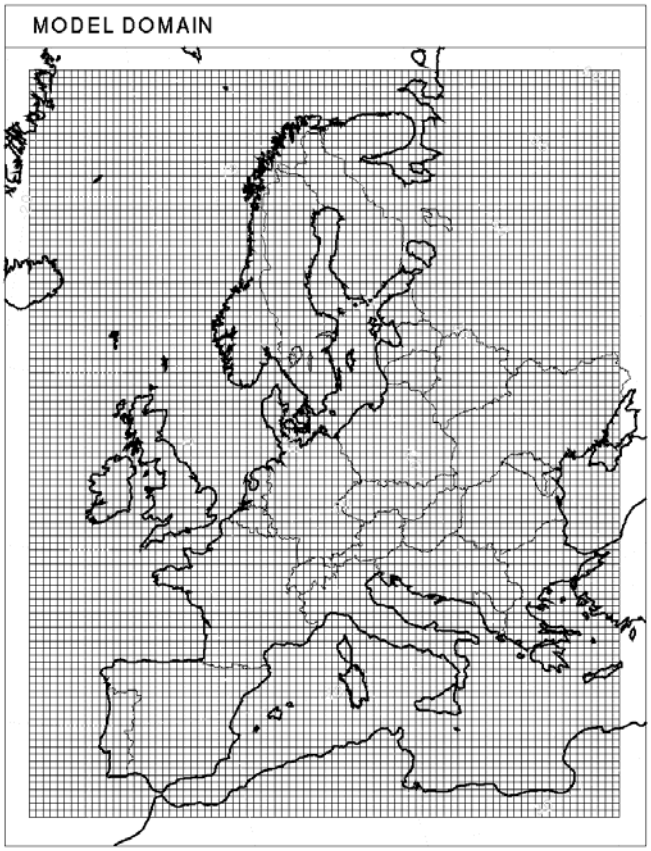


Figure 2: The MATCH photochemistry model domain used in this study.

2.2.2 Model emissions

Anthropogenic emissions for the simulations carried out within this project were derived from the 50×50 km^2 emission data provided by EMEP MSC-W at the Norwegian Meteorological Institute. The emissions for 1999 and 1990 for NO_x, SO_2, nonmethane volatile organic compounds (NMVOC), CO and NH_3 were used in the model calculations. Biogenic emissions of isoprene were also included (see Appendix A).

Maps showing the emissions of NO_x, NMVOC and CO are given in Figure 3 and Figure 4. In Figure 5 the relative changes in emissions from 1990 to 1999 are shown.

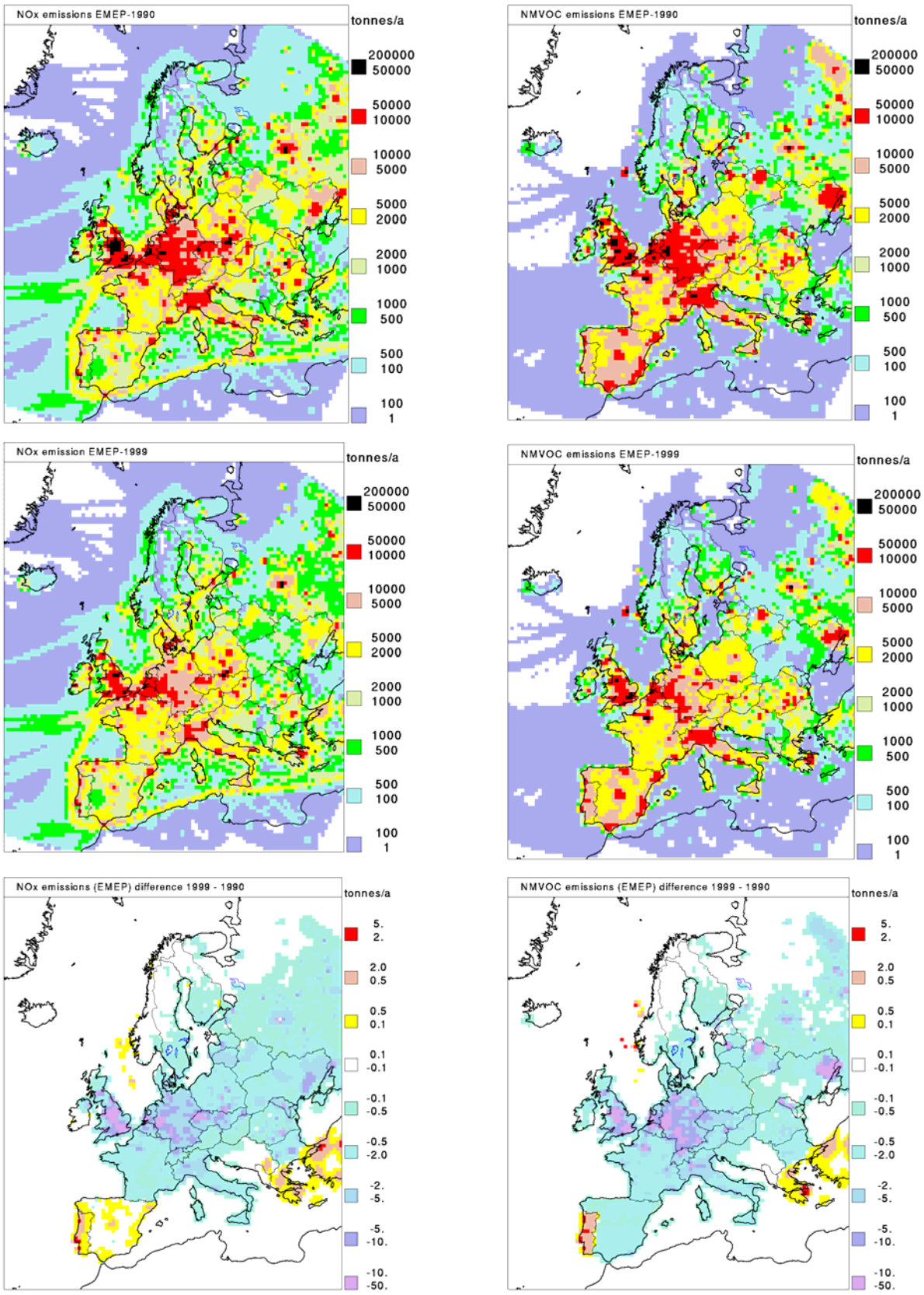


Figure 3: Anthropogenic emissions of NO_x (left) and NMVOC (right) for the years 1990 (top) and 1999 (centre); the change in emissions from 1990 to 1999 is given in the lowest panels. Unit tonnes/year.

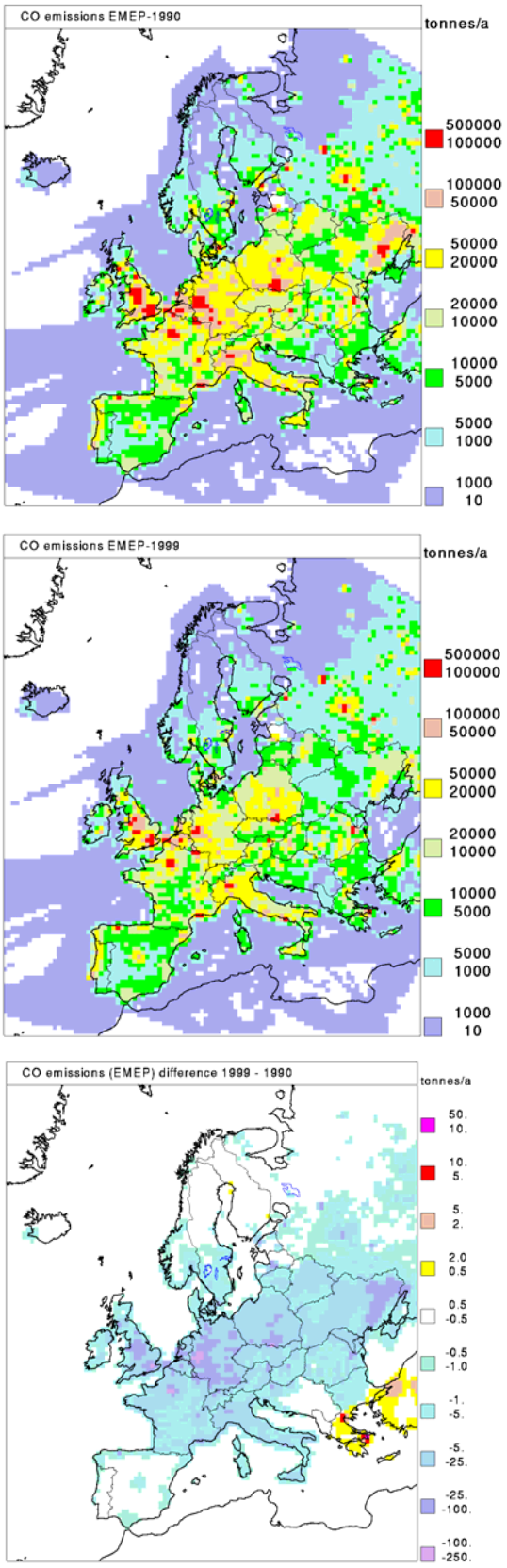


Figure 4: Anthropogenic emissions of CO for 1990 (top) and 1999 (centre); the change in emissions from 1990 to 1999 is given in the lowest panel. Unit tonnes/year.

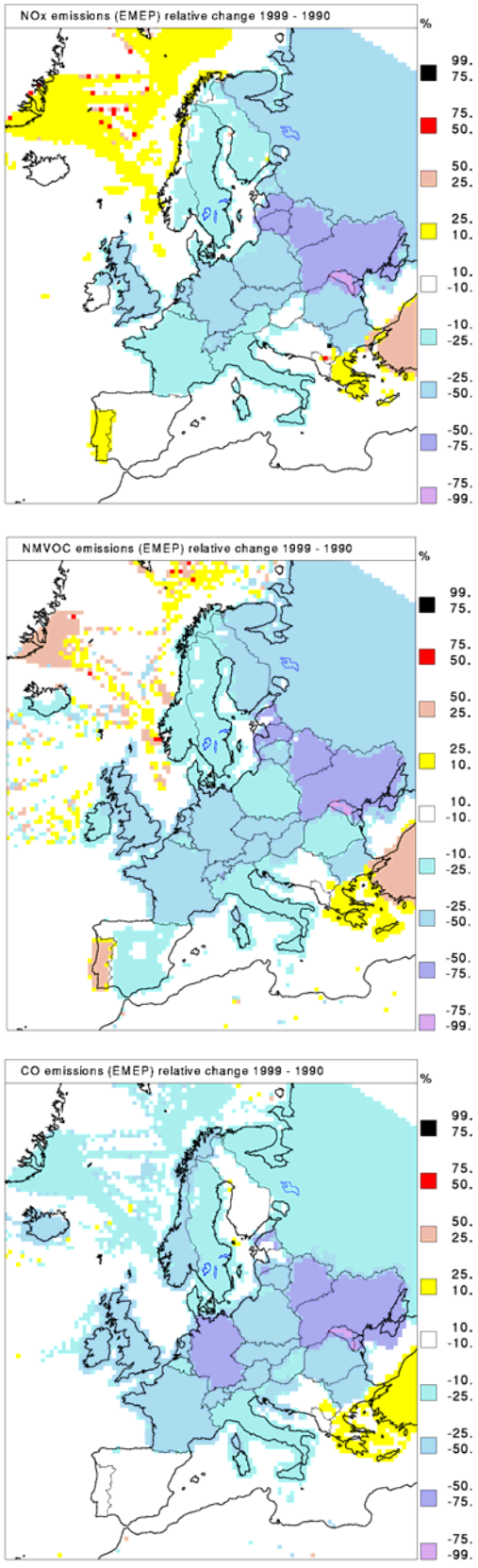


Figure 5: The relative change in anthropogenic emissions from 1990 to 1999 for NO_x (top), NMVOC (centre) and CO (lower panel). Unit: % of the 1990 emissions.

3. Ozone in Northern Europe

3.1 Ozone climatology

There is a clear difference between southern and northern parts of the Nordic region in terms of ozone levels and variation over the year. At the more remote sites up north, e.g. Esrange and Vindeln, the highest concentrations are normally obtained in April and May and the lowest in August-September (Esrange) or in October-November (Vindeln) when daytime observations are considered. Those sites are also less affected by photochemical episodes, as compared with sites in southern parts closer to the main source areas. The southern sites have similar seasonal cycles characterised by a peak in May and a winter minimum. The sites in the south are exposed to polluted air masses from Continental Europe and southern UK quite frequently, even if the interannual variation is large. In summer this transport often results in enhanced ozone concentrations. Any tendency in observed peak concentrations as a result of reduced precursor emission is thus expected to be traceable mainly at sites in the southern parts of the Nordic region.

3.2 Model evaluation for 1999–2000

The original data consisted of hourly concentration values of ozone expressed as $\mu g/m^3$ (or rather ppbv·2) for the 6-months period April-September. A first screening of the data revealed that using daily maxima gave a better agreement between measured and modelled data (measured by the correlation coefficient) than daily average or daytime (11-18) average. The daily maxima will not necessarily occur at the same hour in the measured and the modelled time series. This difference is, however, acceptable, as there are always uncertainties in the exact timing of the episodes and in the model transport. In principle, the daily maxima from the model and from the measurements may be up to 23 h apart, and thus belong to separate episodes. We don't believe this to be a large problem, but it should be kept in mind when analysing individual short-term episodes. Furthermore, when an episode occurs during night, the allocation of the max values to the day before or after midnight may become somewhat arbitrary.

The time series of observed and modelled daily maxima (with the linear regression coefficients, r) as well as the scatter plots of the observed data vs. the difference in modelled values, using 1990 and 1999 emissions, are shown in Figure 6 and Figure 7.

First of all, the time series show fairly good agreement between modelled and measured values. The model generally overestimates the concentrations for August and September 2000, except for the sites furthest north. A similar but weaker tendency is seen for September 1999. The temporal fluctuations in the time series of the modelled and measured data, i.e. the peaks and dips, show a good agreement through all six months for both years in spite of the shift in level in the last period. For the 2000 data the correlation coefficient, r, varies between about 0.6 and 0.8 (except for Utö), which gives a highly significant correlation for n=183 daily values. The correlation for the 1999 results is generally slightly lower. The episodes (which are the focus in this project) are fairly well reflected in the model, although at somewhat lower values.

Compared to 2000 the results for 1999 differ in several ways. First of all, the measurements show that the ozone concentrations at the last part of the 6-months period were considerably higher in 1999 compared to 2000. A few very marked episodes occurred in 2000, all in May-June, followed by particularly low concentrations (also compared with the model) in July-September. In 1999, however, elevated ozone episodes were observed through the whole 6-months period April-September, although the peak values were lower in 1999 compared to 2000. This apparently also affects the model predicted drop in daily max ozone (using the two emission years).

3.3 Model sensitivity to emission changes 1990–1999

The scatter plots in Figure 6 and Figure 7 confirm the hypothesis that the effect of the reduced emissions (from 1990 to 1999) is most evident in the peak ozone episodes. There is indeed a general tendency that the predicted drop in peak ozone values due to the reduced European emission increases with increasing observed ozone values. This is natural as these are the situations when the photo-chemistry has the strongest impact, whereas in other situations, other parameters as wind speed, vertical mixing, dry deposition etc is controlling ozone to a larger extent.

For the Norwegian data in 2000 the results indicate a difference for days below and above 100 $\mu g/m^3$. For days with maximum ozone values less than 100 $\mu g/m^3$, this result indicate a 0-10 $\mu g/m^3$ drop in modelled concentration due to the reduced emissions, and no clear correlation between the observed ozone and the change in modelled ozone. On a few occasions (August-September only) a small increase is predicted. For days exceeding 100 $\mu g/m^3$ ozone, the results indicate a clear relationship between observed max ozone and modelled change, and predict a reduction of up to 30 $\mu g/m^3$ (changing from 1990 to 1999 emissions) for the observed peak episodes of 150 $\mu g/m^3$ or more. This distinction is less evident for 1999 and for the other stations although the general pattern with increasing ozone reduction predicted for the days with the higher observed ozone maxima is seen all over.

This result is important for abatement strategies as these, at least for human health effects, have traditionally been linked to peak ozone episodes, as e.g. the 180 $\mu g/m^3$ threshold value in the EU ozone directive. In Norway the Norwegian Pollution Control Authority uses a threshold of 160 $\mu g/m^3$ for sending out information to the public about ozone episodes. As the annual peak values in the Nordic countries the last years tend to be in the range of 150-170 $\mu g/m^3$, a drop of the order of 30 $\mu g/m^3$ in these episodes would indicate that a marked improvement in the number and extent of episodes related to threshold values for health may have taken place in these countries during the 1990-ies. The relationship between predicted drop in peak ozone and observed ozone concentrations is somewhat weaker when looking at 1999-data compared to 2000.

3.4 Ozone in Finland

The results also indicate marked differences between the Nordic countries, or rather between Norway/Sweden on one hand and Finland on the other. Whereas 2000 was a year with particularly high ozone peak values in S-Norway and S-Sweden, ozone concentrations were generally low in Finland. There were,

however, some interesting episodes also in Finland in 2000. During those episodes concentrations were usually higher at the inland stations compared to concentrations at Utö that is located on the Baltic Sea and reflects concentrations in the air masses transported from the European continent. This may indicate the importance of domestic precursor emissions for episodic ozone concentrations in Finland.

The systematic difference between episodes in Finland compared to Norway and Sweden presumably reflects the difference in meteorological situations for these two regions. As will be shown in the following, Sweden and Norway are in general more exposed to episodes with transport from S or SW, whereas Finland is more influenced by the episodes with transport from E and SE. This is linked to the synoptic situations typical of ozone transport from the European continent. These situations are commonly characterized by a blocking high situated over central Europe for several days, leading to warm, stable, slowly descending and slowly moving air masses over the continent, accumulating emissions and accelerating in photochemical activity and gradually increasing the ozone concentration. In the Nordic countries, however, ozone peak values during such situations are typically observed only when the high-pressure system is breaking up due to an approaching frontal system leading to a short-term pulse of continental polluted air masses ahead of the front. This is a more common situation for Norway and Sweden, as the cyclones tend to move northwards when approaching the blocking high or when encountering the Norwegian/Swedish mainland. Thus, the tongue of continental, photochemical pollution may reach far north in Norway/Sweden while only parts of the more dissolved air masses reach as far as Finland.

Similarly, as seen in the following discussions regarding individual episodes, S-Finland may be affected by episodes and transport from E and SE that are not reaching as far west as Sweden/Norway. Thus, to some extent it's fair to regard the ozone climatology and the associated meteorological situations as rather different in Finland compared to Norway and Sweden. Furthermore, as mentioned above, the climatology for the Nordic countries also differs from that observed in central Europe. Knowledge of these systematic differences is valuable information in a political and abatement perspective.

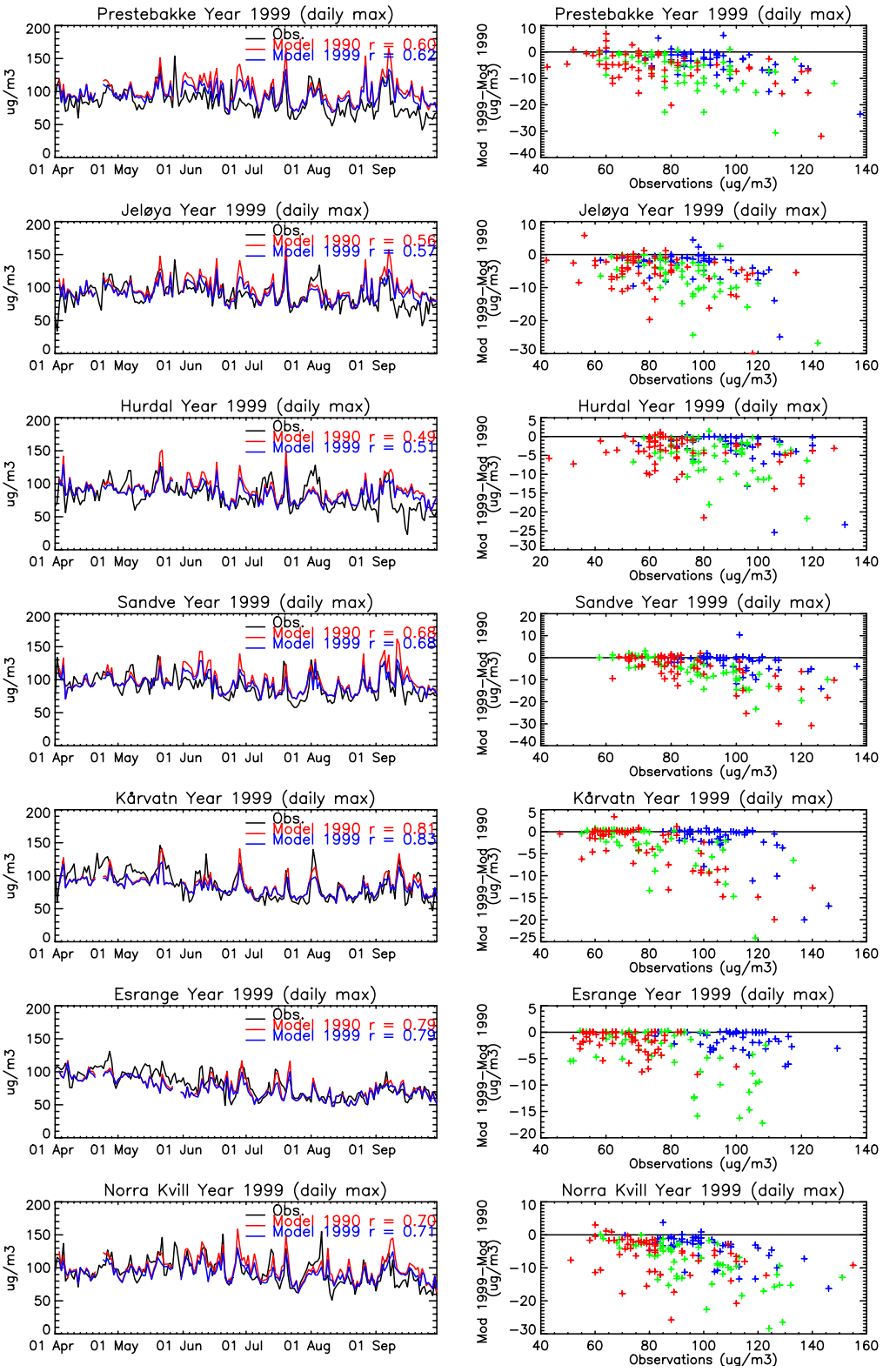


Figure 6: The left panels show the observed daily ozone maximum during the summer half year of 1999 together with the modelled values using 1990 and 1999 emissions. The right panels show the predicted change in daily max ozone (due to emission changes 1999-1990) as a function of observed ozone. The colour of the crosses indicates the time of year. Blue: April-May; Green: June-July; Red: August-September.

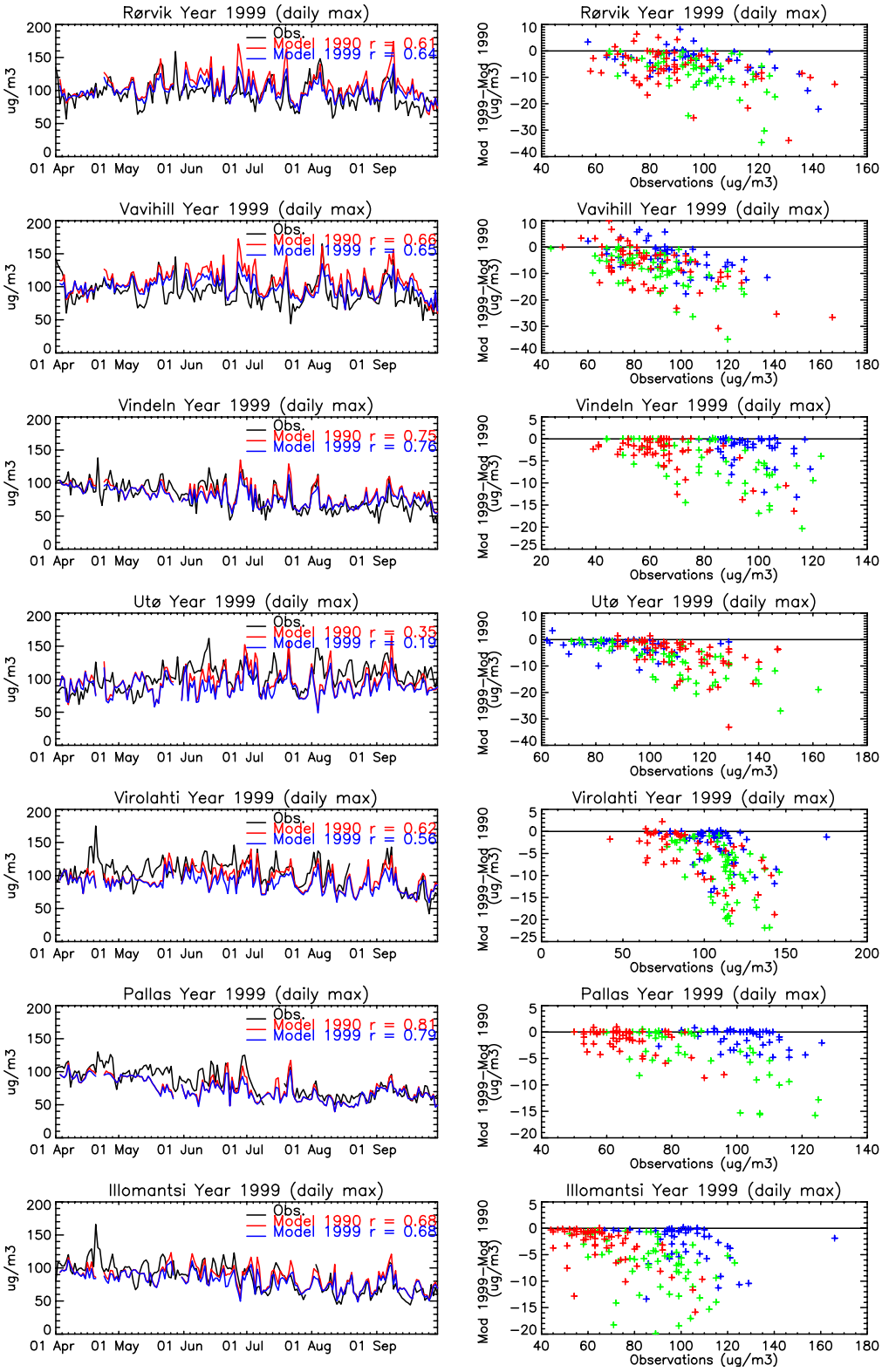


Figure 6, cont.

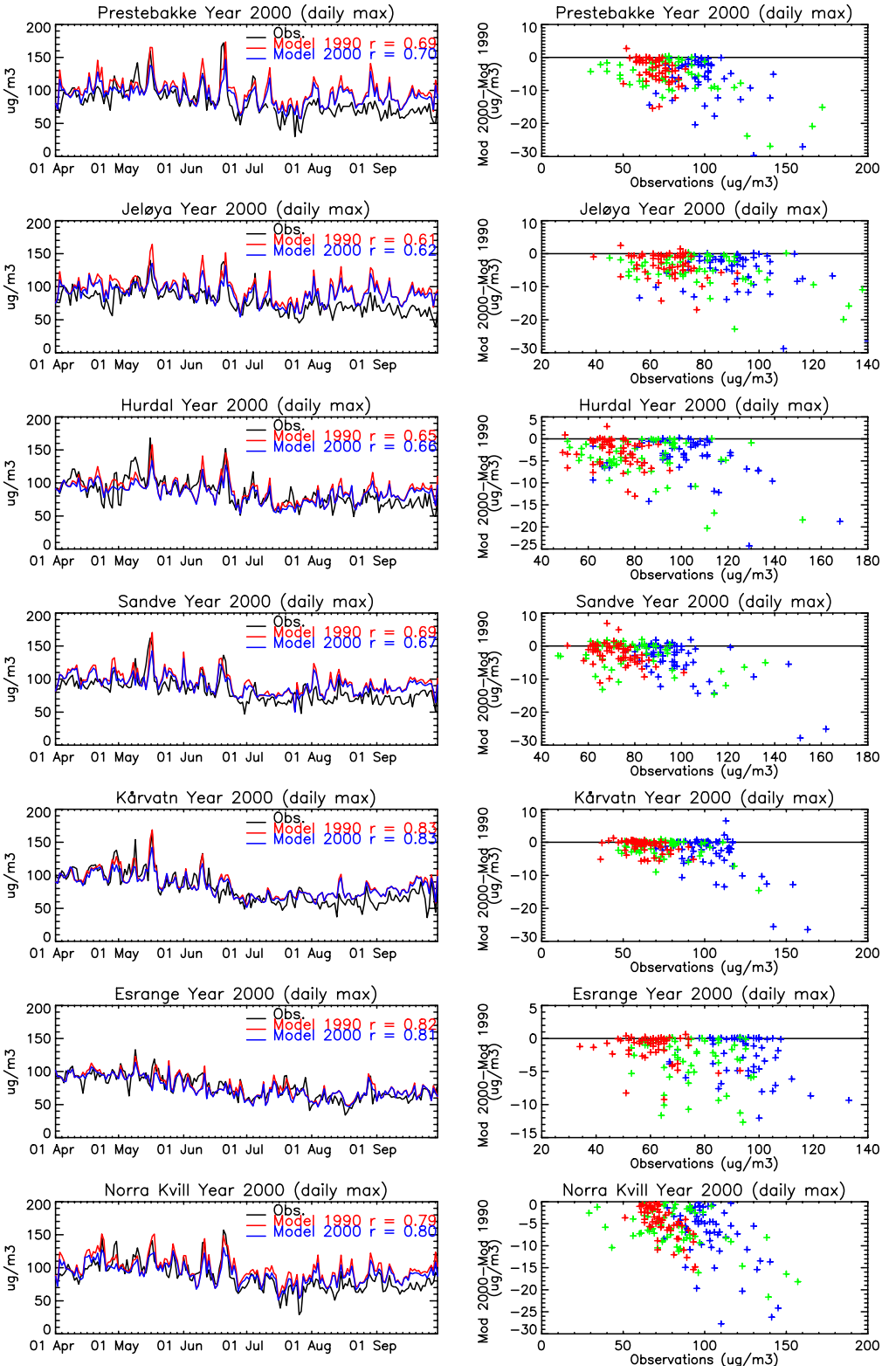


Figure 7: The left panels show the observed daily ozone maximum during the summer half year of 2000 together with the modelled values using 1990 and 1999 emissions. The right panels show the predicted change in daily max ozone (due to emission changes 1999-1990) as a function of observed ozone. The colour of the crosses indicates the time of year. Blue: April-May; Green: June-July; Red: August-September.

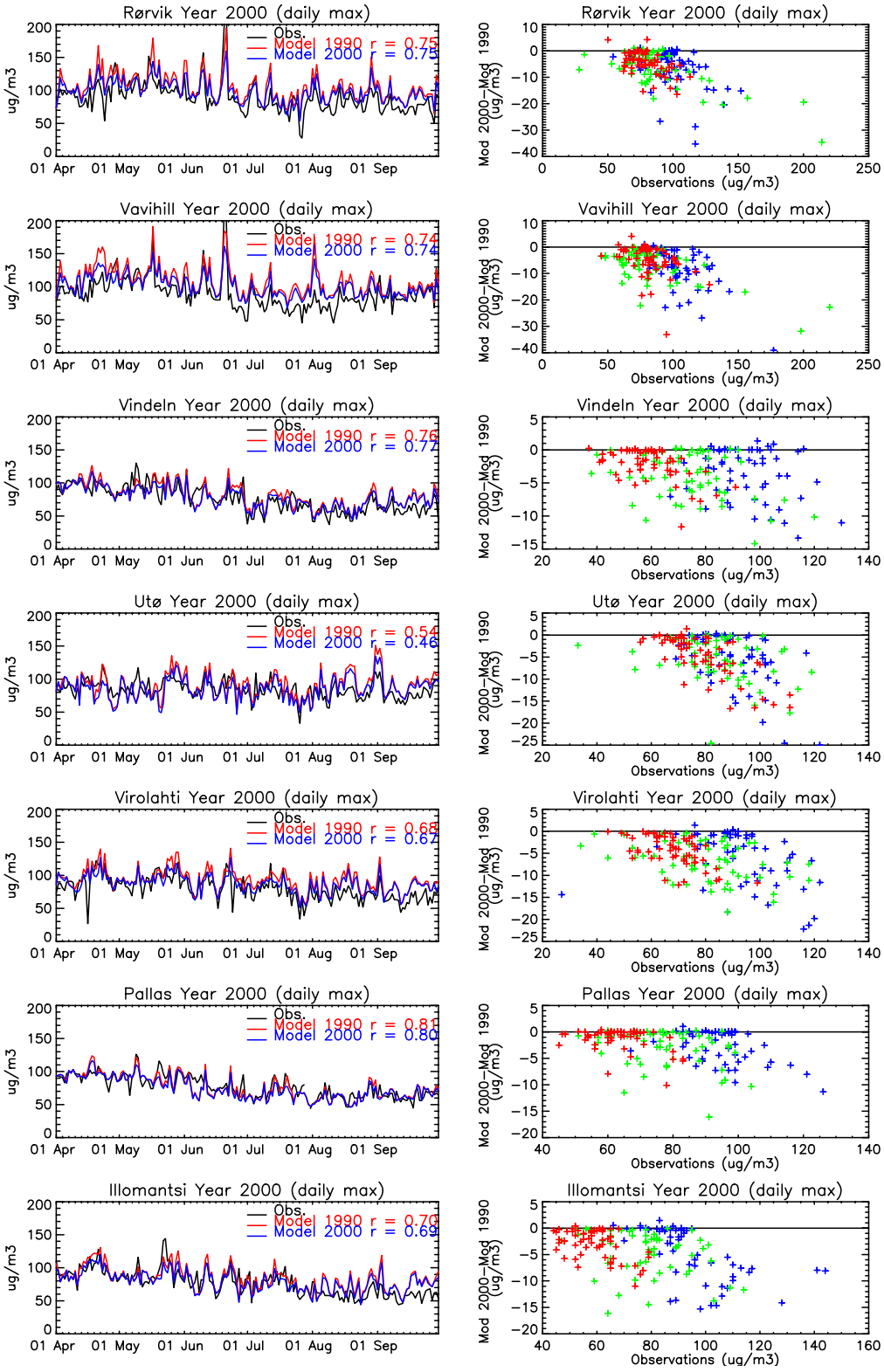


Figure 7, cont.

3.5 Long-term trends in measured ozone-percentile values

Long-term trends of observed episodic ozone concentrations in April–September were calculated for 7 Norwegian, 6 Swedish and 5 Finnish sites. First we calculated 99-percentiles of hourly observed ozone concentrations in April–September for each year separately. Linear regression of the 99-percentile concentrations together with 95% confidence intervals are shown in Figure 8. Trend estimates per 10 years are shown by bold numbers. We show also 99-percentile concentrations simulated by the MATCH-model using 1999 and 2000 weather fields and reported 1999 emissions. These 99-percentiles are added to the figures as two-letter codes. The MATCH model was also used to estimate the effect of emission changes on ozone concentrations during the 1990's by repeating the calculations using 1990 emissions. From these sets of ozone concentrations we calculated 99-percentiles and subtracted those calculated using 1999 emissions from those calculated using 1990 emissions. The two italic numbers are the trends per 10 years simulated using 1999 and 2000 weather fields, respectively.

The observed trends of 99-percentiles at 7 sites in the southern and central parts of Sweden and Norway, south of 61°N, are between -10 and -22 $\mu g/m^3$ per 10 years. Prestebakke in Norway is an exception showing increasing, though not significant, episodic ozone concentrations +11 $\mu g/m^3$ per 10 years. At the Norwegian sites north of 61°N trends are decreasing (Osen), increasing (Tustervatn and Kårvatn) or not changing (Spitsbergen). However, these trends are not significant. At the Swedish site Vindeln (64°N) 99-percentile ozone concentrations are decreasing by -4 $\mu g/m^3$ per 10 years. At the more northern site Esrange (68°N), trends are also negative. At the Finnish sites trends are insignificant. However, it seems that in the southeastern part of the country, which is mostly influenced by the air masses originating from Eastern Europe ozone concentrations are increasing.

As a conclusion, in the areas close to the European areas where the emission density is high, and where 99-percentiles have been mostly above 120 $\mu g/m^3$, episodic ozone concentrations are decreasing. It seems that these trends are less pronounced at the more eastern (Finnish) sites. Longer time series are needed to reach significant trends in the northern parts of Scandinavia.

The comparison between observed and calculated 99-percentile concentrations in 1999 and 2000 shows that the MATCH-model simulates episodic ozone concentrations well. The model results indicate downward trends between 1990 and 1999 typically in the range 15 to 20 $\mu g/m^3$ in the southern Norway and Sweden, around 10 $\mu g/m^3$ in the southern and central parts of Finland and about 5 $\mu g/m^3$ in the northern parts of Scandinavia. These trends are relatively good estimates of the observed trends. One of the notable differences is the observed increasing trend at the easternmost site, Virolahti, which is not seen in the model calculations. Generally, however, the time series are too short to get significant trends.

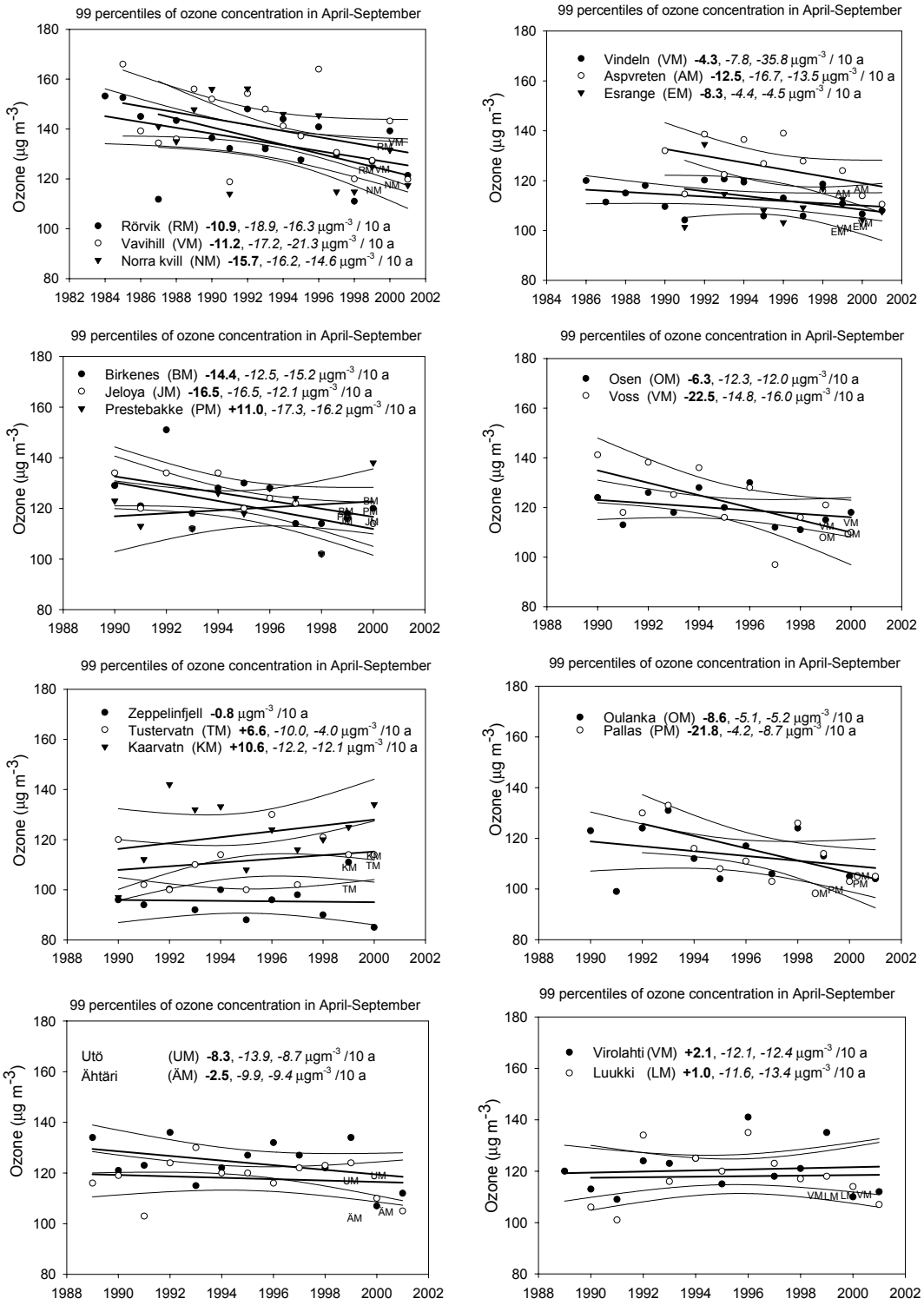


Figure 8: 99-percentiles of ozone concentration in April–September. Bold numbers indicate the change in 10 years according to a linear fit to the observations. Thin lines indicate 95 percent confidence limits. The two-letter code shows the 99 percentile ozone concentrations simulated by the MATCH-model for 1999 and 2000. Italic numbers after the bold trend numbers show trend estimates from MATCH-model simulations as a difference of 99 percentiles when 1999 and 1990 emissions where used. The first italic number shows the simulation results for 1999 and the second for 2000 meteorology.

4. Description of selected episodes

A total of six episodes were selected based on the monitoring data, three in 1999 and three in 2000. As mentioned above, the ozone climatology in 2000 was rather different from 1999, and whereas very marked episodes were experienced in Sweden and Norway in 2000, 1999 was a year with low ozone concentrations in all the countries. Thus, the episodes differ substantially in concentration level observed as well as in spatial and temporal extent. The episodes were subjectively chosen based on the deviation from the seasonal background concentration. Thus, as the background level of ozone is much lower in September compared to May in the Nordic countries, the criteria for selecting an episode in terms of absolute concentration level is also lower.

A presentation of each of the episodes is given in separate chapters below. The presentation is accompanied with daily meteorological maps showing the surface pressure fields, wind speeds and directions, 2-m temperatures as well as precipitation areas (e.g. Figure 9). All the meteorological data were taken from the HIRLAM model run by SMHI. Furthermore, maps showing the regional distribution of daily maximum ozone, measured and modelled, are also given (e.g. Figure 10). The measurement maps show the daily maximum values as coloured dots at each EMEP station, whereas the modelled fields are given as interpolated contour fields covering the model domain. To ease the interpretation and the application of the maps the same colour scale was used for the measured and modelled fields for all the days and episodes presented. Finally, time series of observed and modelled *hourly* ozone values around the episodes are also included.

The diurnal cycle and in particular the night time ozone concentrations are difficult to model properly in a regional scale model. This is due to the fact that night-time inversions and ozone surface deposition leading to lowered concentrations are determined by local conditions as surrounding vegetation and topography, which could not easily be incorporated into a regional scale model. Thus, discrepancies between the modelled and measured night-time values does not necessarily indicate an erroneous model, rather that the measured values during night are not regionally representative or that sub-grid processes are determining the concentrations.

For each episode concentration maps for a six days' period are shown. The May 2000 episode is an exception, for which maps for an extended period is shown. This was due to the fact that an episode was experienced in the first part of the period in Norway and Sweden, while elevated concentrations were seen several days later in Finland.

4.1 17-22 July 1999

This episode was characterized by an extended high pressure area over all of central Europe moving eastwards and dissolving due to a low and a frontal system approaching from the N Atlantic. The front passed N-Europe and Scandinavia 20-21 July. High temperatures (above 30°C) and humid air were observed in central Europe and UK, and thunder occurred in association with the approaching frontal system.

In terms of ozone concentrations this was a minor episode in the Nordic countries, peaking at 142 μg/m^3 at Jeløya in SE-Norway. The European monitoring data show ozone concentrations gradually building up during 17-19 July, exceeding the EU threshold value of 180 μg/m^3 at a few sites. At 19-20 July the monitoring data indicate that the area with elevated ozone concentrations were extended to the north, and concentrations exceeding 140 μg/m^3 (and also 160 μg/m^3) were observed in Norway and Denmark. Elevated concentrations of ozone were observed at Rörvik and Vavihill on 19th and 20th, and at Norra Kvill and Vindeln on 21st. At 21st July, values exceeding 140 μg/m^3 were also observed in Finland, whereas the concentrations in Norway and Sweden had dropped to background levels due to the frontal passage.

The MATCH model simulated the geographical distribution and temporal development of the episode very closely, as seen from Figure 10. The stretching of the European ozone plume and the filament of it crossing the Nordic countries 20-21 July matched very well the observational field, indicated by the EMEP network data. For Central Europe the peak concentrations given by the model was, however, clearly lower than observed.

The time series of hourly ozone concentrations at the Nordic sites (Figure 11) also indicate a generally good agreement between the modelled and measured data. The model predicted reduction in peak ozone due to the emission changes from 1990 to 1999 is fairly large, of the order of 20-30 μg/m^3. For the Norwegian sites, the model predicts that several of the monitoring sites would have crossed the 160 μg/m^3 threshold if assuming 1990 emissions data while not when applying the 1999 emission. Thus, based on the general model fit it is likely that this is an example of a situation where the European emission reductions changed this from an ozone episode to a non-episode in Norway in terms of press release and public information, referring to the 160 μg/m^3 threshold applied by the State Pollution Authority.

At Rörvik the highest concentration was observed during the night when the model indicated a minimum. This may be due to a local sea-land breeze situation. At Vindeln the model indicated a second maximum in the evening not present in the observations.

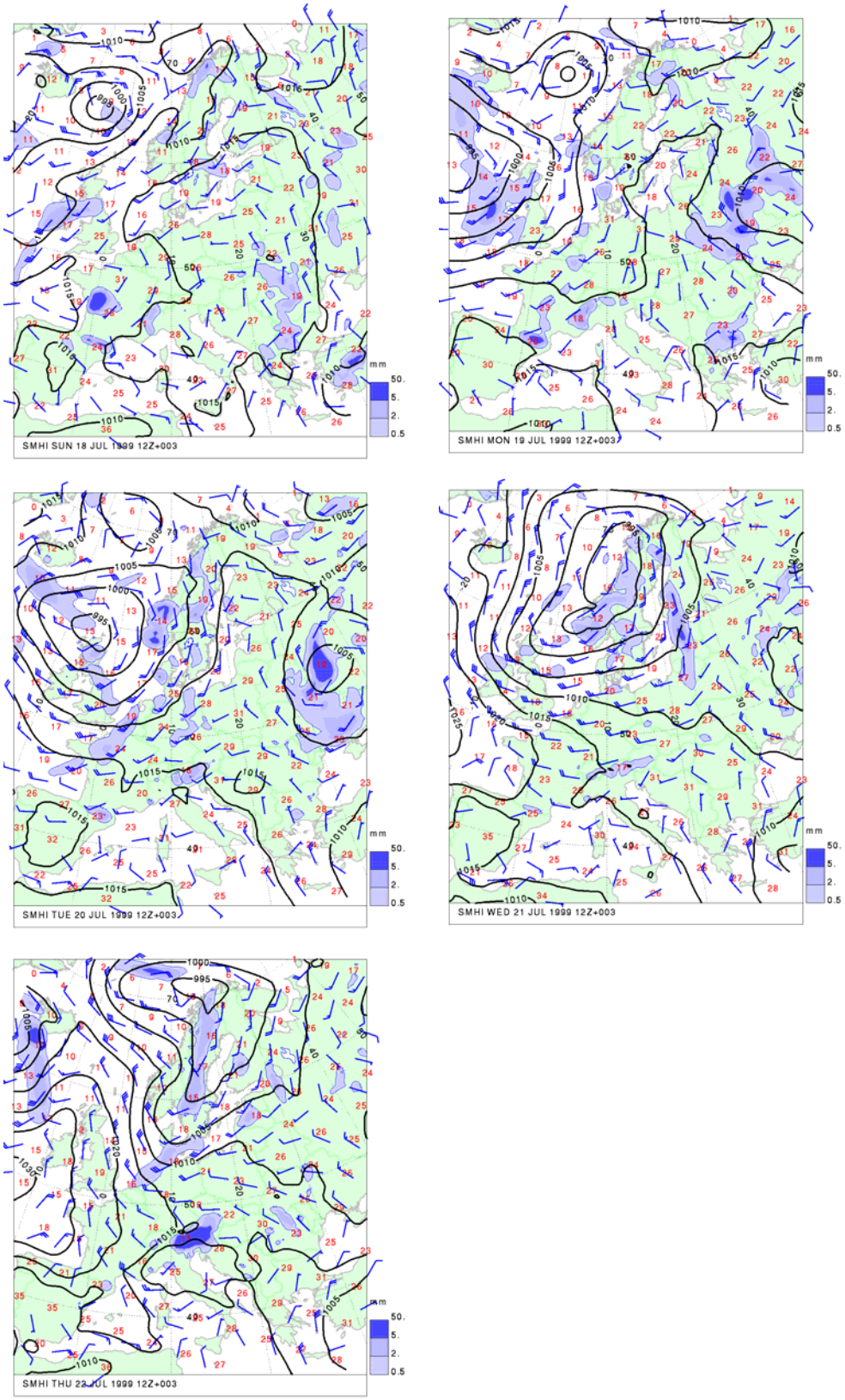


Figure 9: HIRLAM meteorology 18–22 July 1999. Surface pressure (black isobars, hPa), horizontal wind vectors at model level 4 [ca. 600 m height] (blue WMO-arrows), 2-m temperatures (red numbers, °C), and precipitation areas (blue).

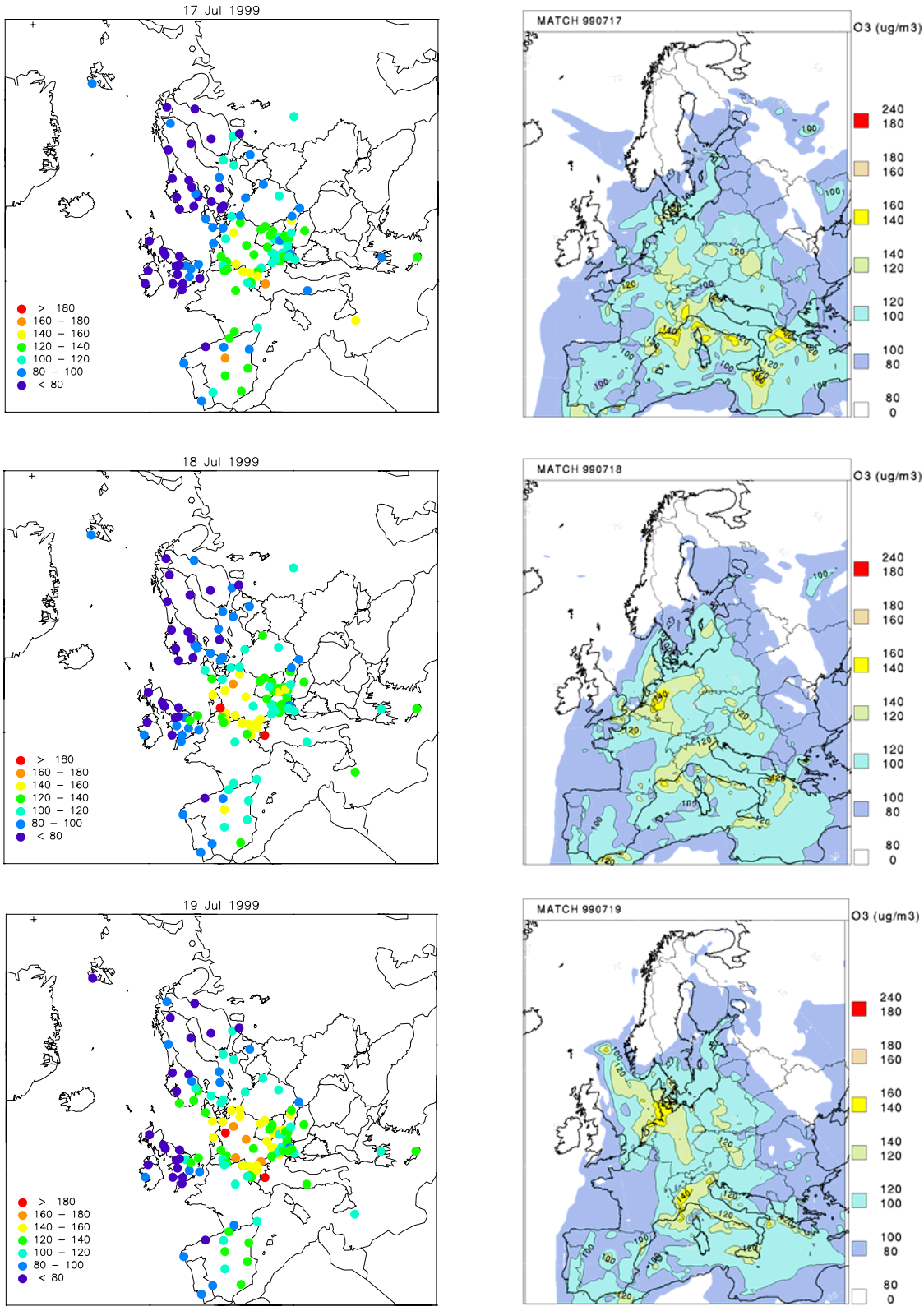


Figure 10: The episode 17–22 July 1999. Daily maximum ozone concentrations as observed at the EMEP network stations (left panels) and as modelled by the MATCH model (right panels). All values are given in $\mu g/m^3$.

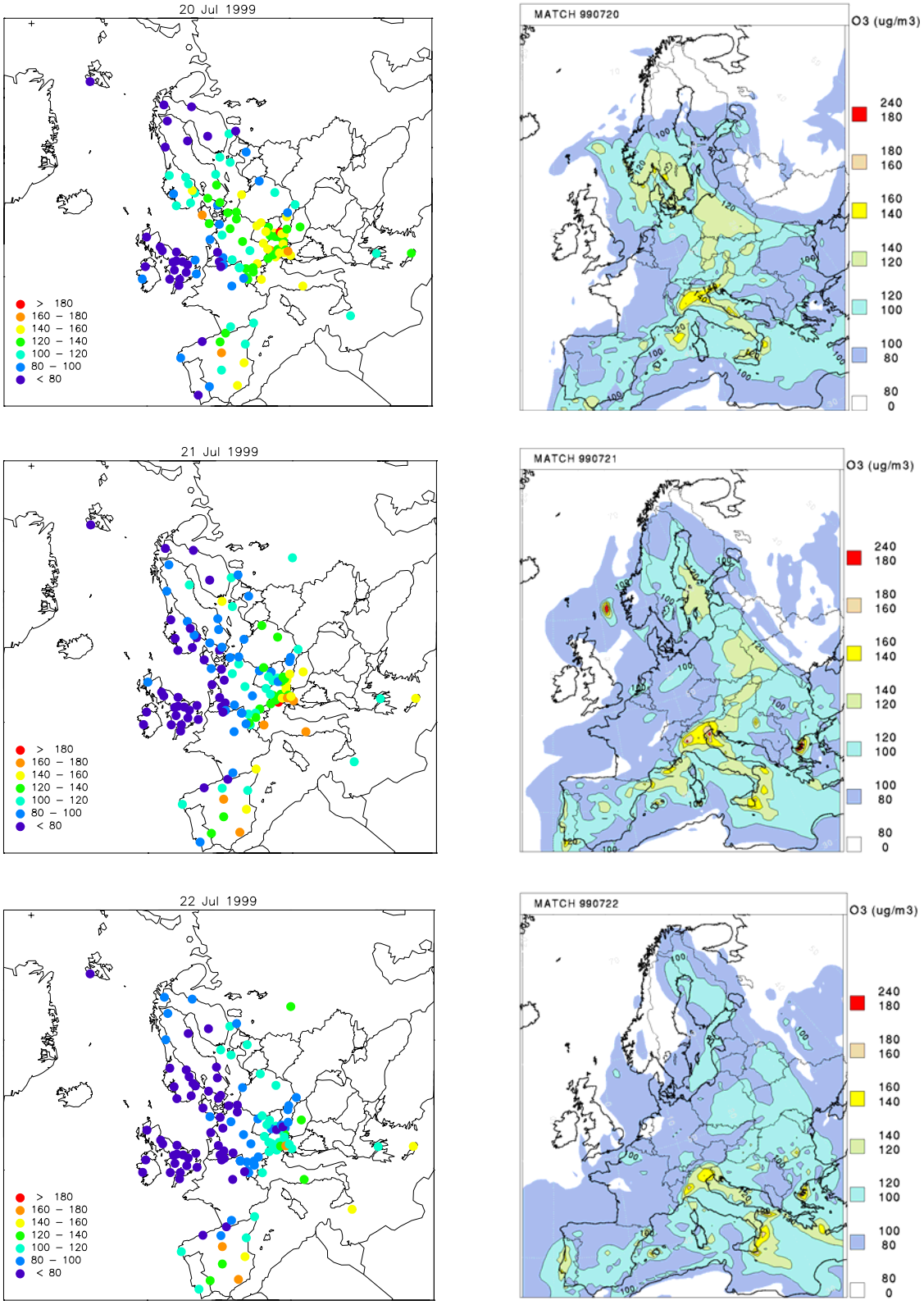


Figure 10, cont.

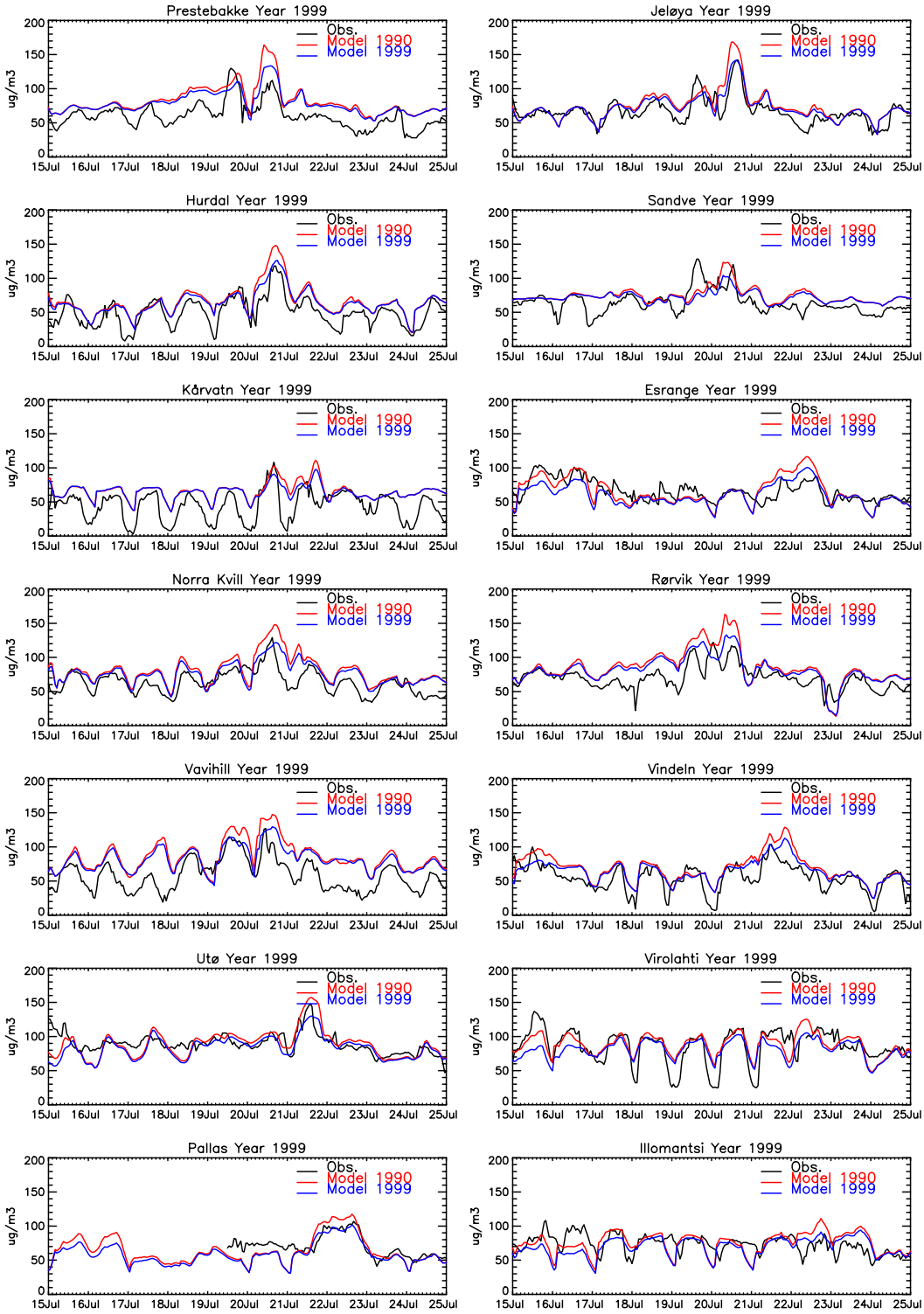


Figure 11: Observed hourly ozone concentrations together with the modelled values using 1990 and 1999 emissions.

4.2 4-9 August 1999

During this period a low pressure system was gradually moving into Biscay, replacing an area with very small pressure gradients over central Europe. Gentle winds from S-SW were observed, and a trough located NE-SW from UK to E-Europe was associated with a marked shift in wind direction and air masses. Fronts associated with the Biscay low brought rain and thunder from 7 August in western parts. Warm and humid continental air was gradually replaced by colder air masses from west.

The ozone monitoring data indicate an area with elevated ozone concentrations extending from northern UK to SE Germany, apparently coinciding with the location of the through on 4 August. Concentrations above 160 $\mu g/m^3$ were seen at several sites in Germany and the Netherlands, also exceeding 180 $\mu g/m^3$ at a few locations. On the following two days the measurements indicate that this air mass moved slowly towards northeast, giving rise to concentrations exceeding 140 $\mu g/m^3$ at several sites in southern Norway and Sweden, also exceeding 160 $\mu g/m^3$ at Vavihill in Sweden. In the following days the whole episode was gradually disappearing and dissolving while the air mass was apparently transported slowly to SE.

The model predicted the NW-SE location of the plume, but the modelled concentrations are significantly lower than the measurements. The ozone plume just "touching" the southern parts of Norway and Sweden, then dissolving, is reproduced by the model, although the measurements indicate that the air mass was reaching further to the north than given by the model.

In general, this kind of transport situation is a challenge to the model due to the gentle winds and the very slow atmospheric transport. Uncertainties regarding transport direction etc become important during such episodes. Thus, reproduction of details in the ozone concentration fields at the outskirts of the large-scale plume is difficult. The general underestimation of the ozone concentrations is, on the other hand, of more concern, and points to deficiencies in e.g. the emission data, the vertical exchange or the dry deposition.

Furthermore, on the 4th, the model predicted an area of very high ozone concentrations in the North Sea, presumably caused by a transport of the already polluted air masses from N-Scotland to the areas of very high emissions of NO_x and particularly VOC from the petroleum activity in the North Sea (Figure 3). However, whereas the VOC emission in this area is indeed very high, nearly all of it is slow-reacting light alkanes (mostly ethane and to a less extent n-butane) from loading buoys filling the gas tankers offshore. In the model, all VOC emissions were given the same VOC source split (Appendix A) with significant portions of alkenes and xylenes which obviously will exaggerate the reactivity of the emissions. It's still an interesting question to what extent ozone is formed in this area. However, neither the model nor the measurements actually indicated elevated concentrations over land.

From the time series plots (Figure 14) it is clear that Vavihill is the only site where the episode was seen both in the measurements and in the model. For this

site the model underestimated the concentrations considerably, and, interestingly, the model predictions using 1990 emissions matched closely the observations.

To summarize, the model performance for this August 1999 episode is not particularly good, which probably could partly be explained by a weak pressure field, increasing the uncertainties and representing a general modelling challenge. Additionally, systematic underestimation of the ozone concentrations may indicate other modelling problems as well, e.g. related to emission data or dry deposition.

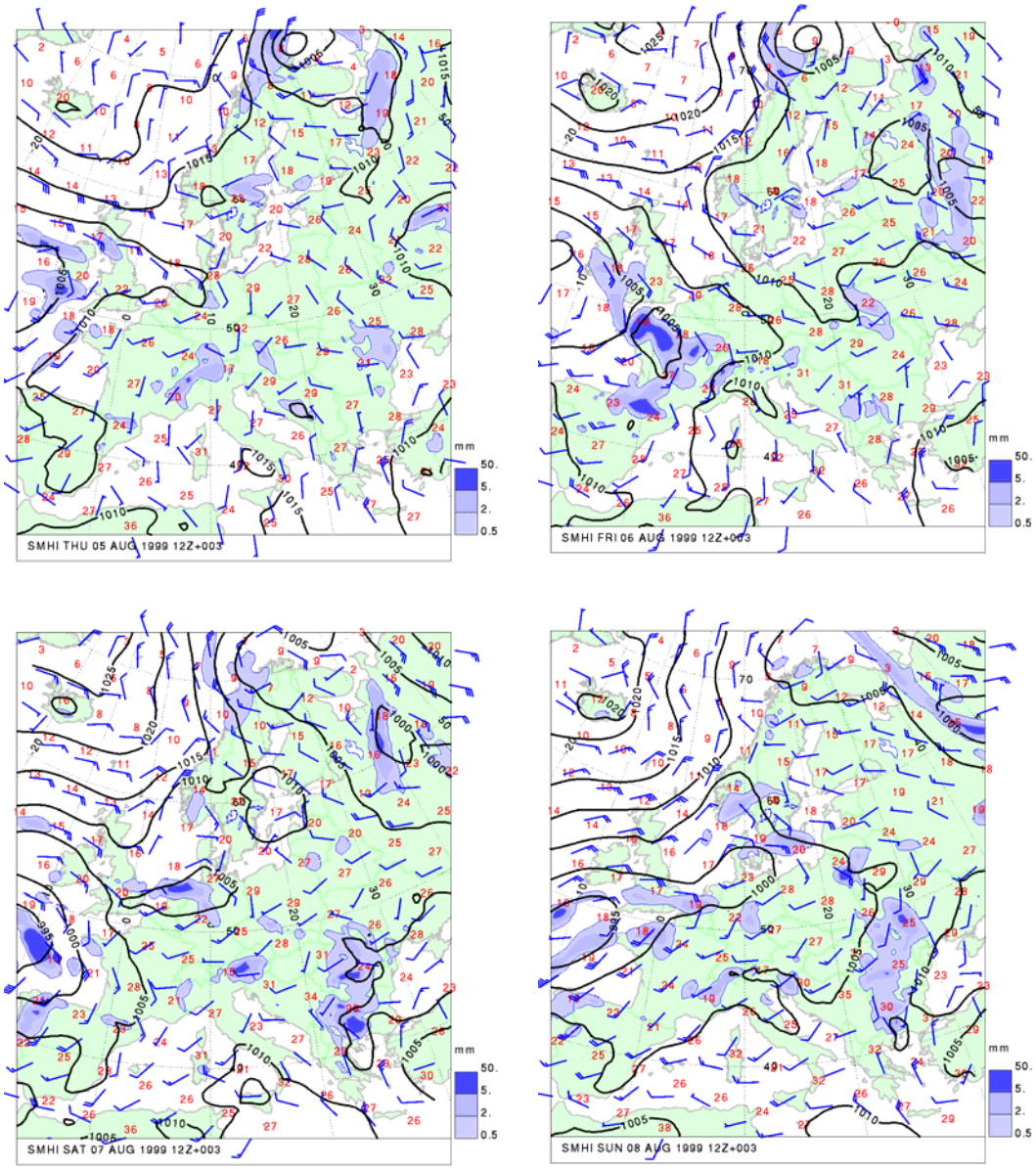


Figure 12: HIRLAM meteorology 5–8 August 1999. Symbols as in Figure 9.

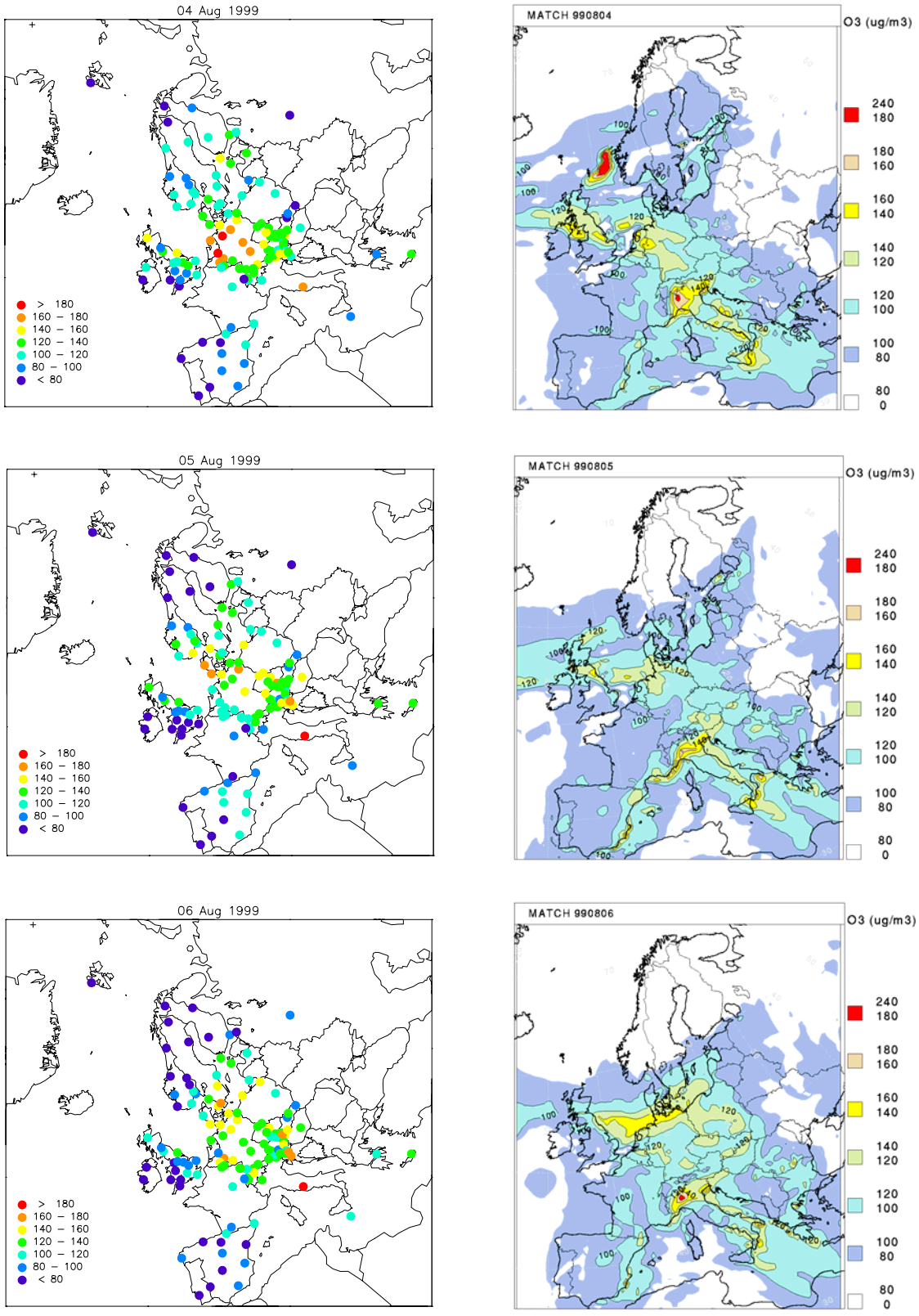


Figure 13: The episode 4–9 August 1999. Daily maximum ozone concentrations as observed at the EMEP network stations (left panels) and as modelled by the MATCH model (right panels). All values are given in $\mu g/m^3$.

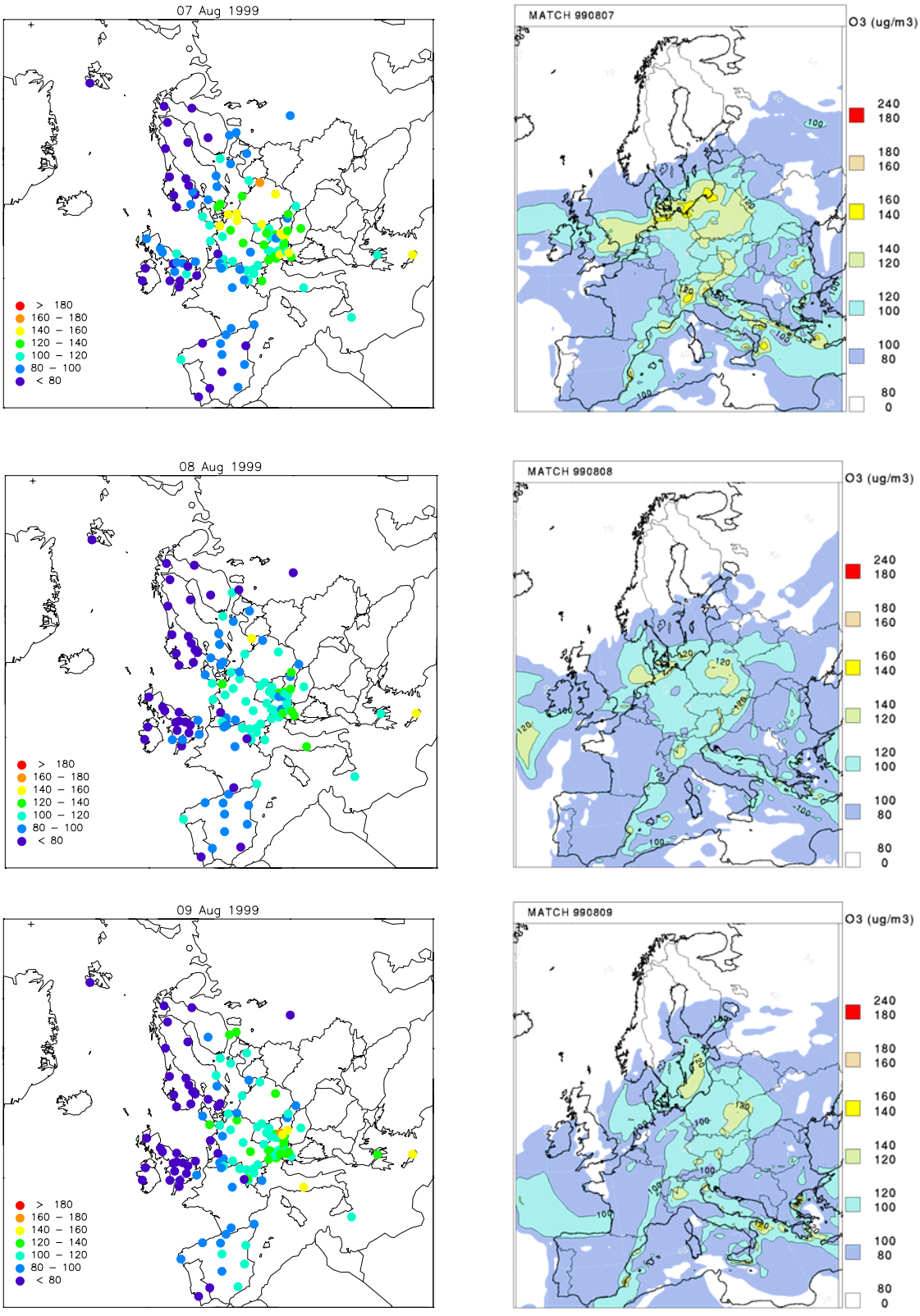


Figure 13, cont.

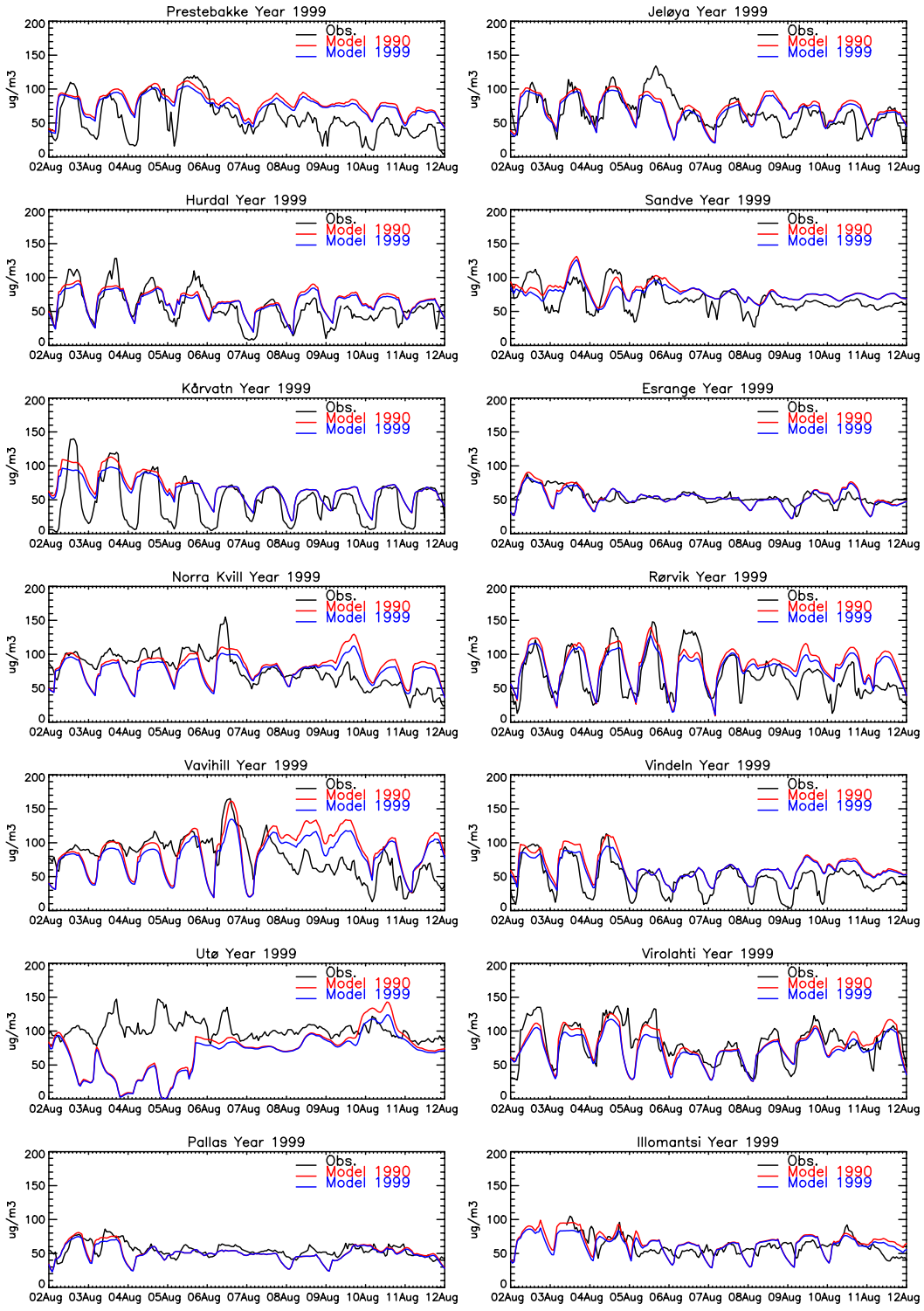


Figure 14: Observed hourly ozone concentrations together with the modelled values using 1990 and 1999 emissions.

4.3 5-10 September 1999

An extensive high pressure system located over the Baltic States at the start of this period gradually developed and was established over S-Scandinavia by the 10 September. Exceptionally high temperatures for the season were recorded in UK, and also elsewhere in N-Europe (including Scandinavia) temperatures of 20°C and higher were observed. The wind fields were generally southerly in the first part of the period, turning more westerly in the last part as an intense low pressure system was passing in the North Atlantic.

In the Nordic countries ozone episodes in September are very rare, partly due to the low background concentrations of ozone at this time of year. In terms of ozone concentrations recorded, this episode was thus not very pronounced with a peak value of above 140 $\mu g/m^3$ at Virolahti in E-Finland. The maximum value in Norway was 126 $\mu g/m^3$ at Prestebakke. The regional distribution of measured ozone indicates an episode originating in SW England, then gradually moving NE with a band of elevated ozone stretching SW to NE across Europe to southern Finland on the 8th September then weakening in the north while increasing in ozone in central Europe.

This development of the episode was well reproduced by the model. The model also indicates an "initial" region of elevated ozone on the 5th September in SW England and the Biscay, which could be interesting to trace further back. The model and, to a small extent the measurements, indicate that the ozone plume just barely reached the southern tip of Norway. According to the model, the ozone plume passed southern Norway the night between 7 and 8 September, which in fact could explain why no pronounced episode was observed in that region. During calm anticyclonic conditions in September the nighttime inversion may become very strong, separating the shallow boundary layer from the air aloft very efficiently. Thus, it is not unlikely that the ozone plume, evident from the model and from the rest of the EMEP network passed over the nighttime inversion without being mixed down and made visible at the surface stations. At Rörvik and Vavihill the episode was seen on the 9th September and the model agreement was fairly good. The predicted change in peak ozone from the emission change 1990-1999 was rather large, particularly at Rörvik, indicating a drop in maximum ozone of more than 30 $\mu g/m^3$ that day and indicating an episode close to the EU threshold of 180 $\mu g/m^3$ applying 1990 emissions.

Although very late in the ozone season in the Nordic countries, the general model agreement during this episode gives confidence in the model predictions. The prediction of an actual episode during this situation is rather uncertain due to several causes: Firstly, the ozone plume was apparently just reaching the southern part of the Nordic countries. Secondly, low wind speeds as well as presumably well developed night-time inversions at this time of year make comparisons with surface measurements particularly difficult. On this background the model agreement is satisfactory.

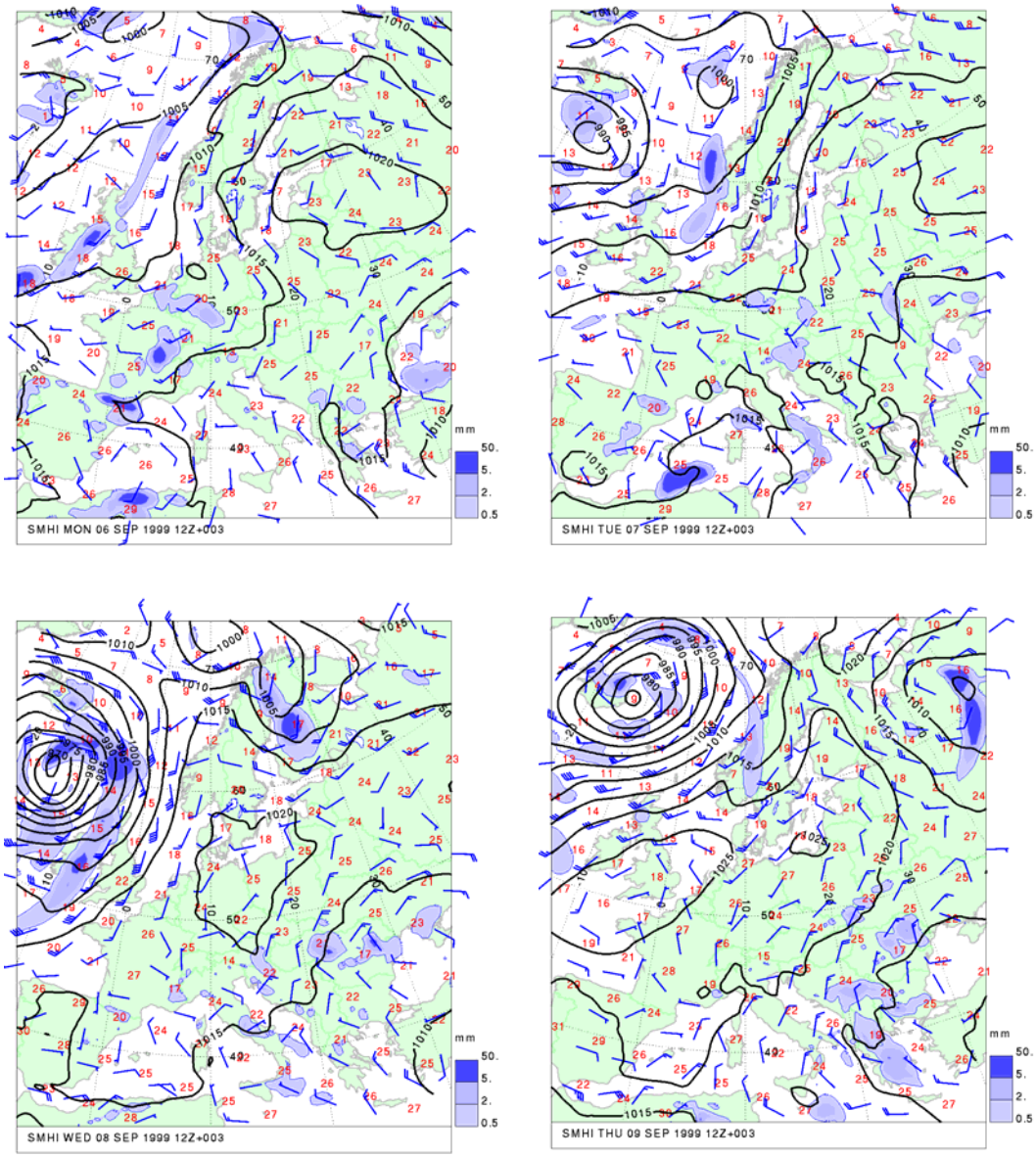


Figure 15: HIRLAM meteorology 6– 9 September 1999. Symbols as in Figure 9.

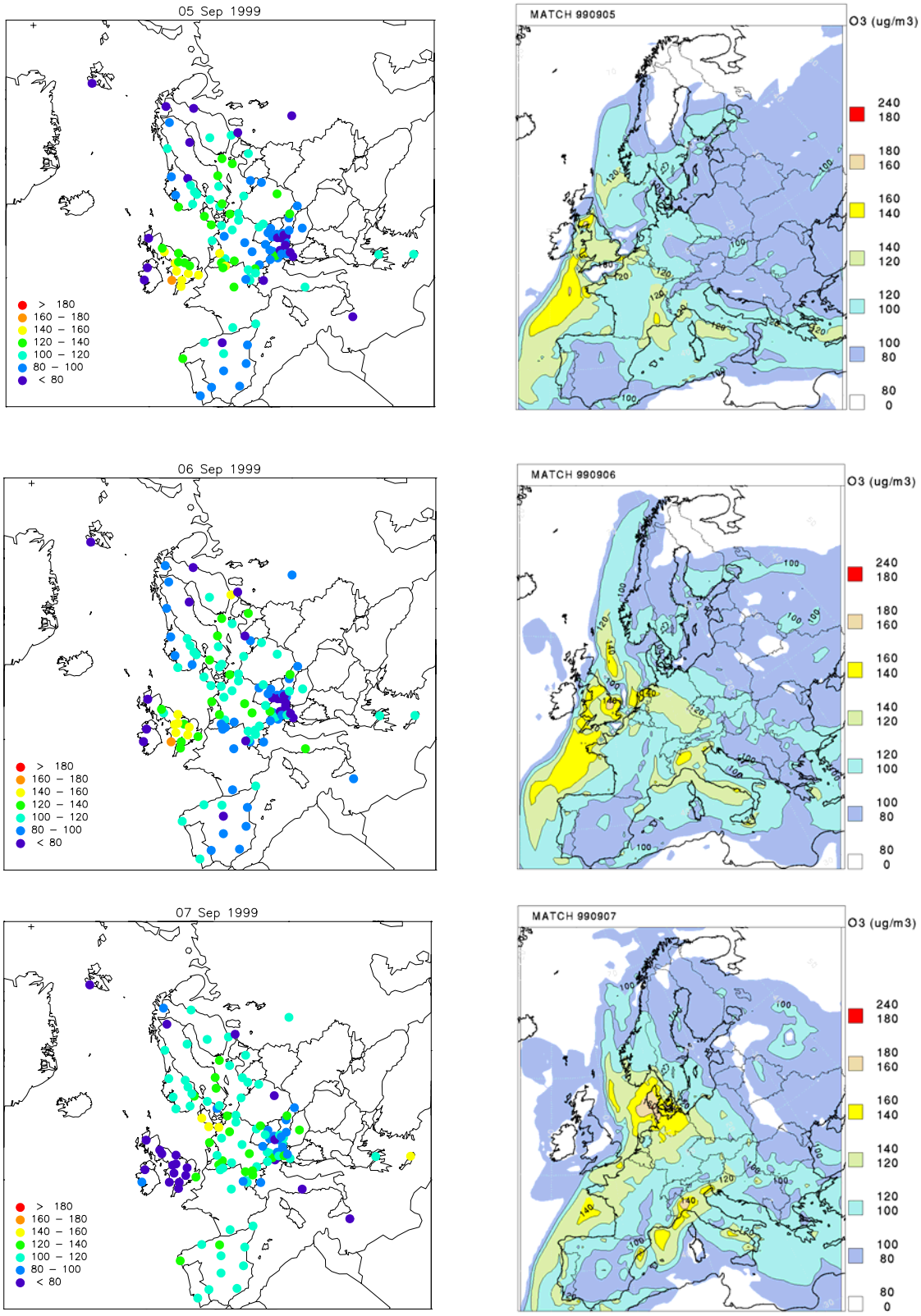
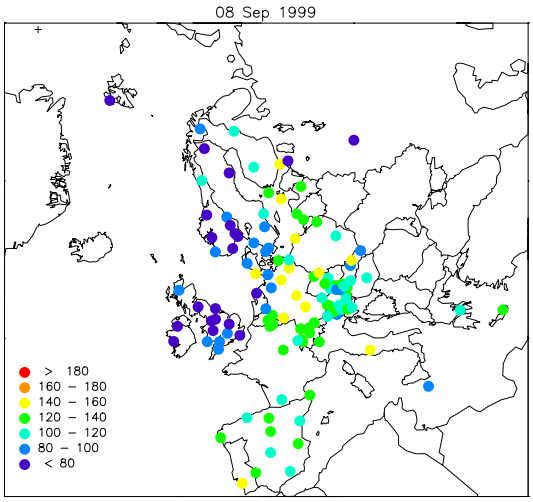
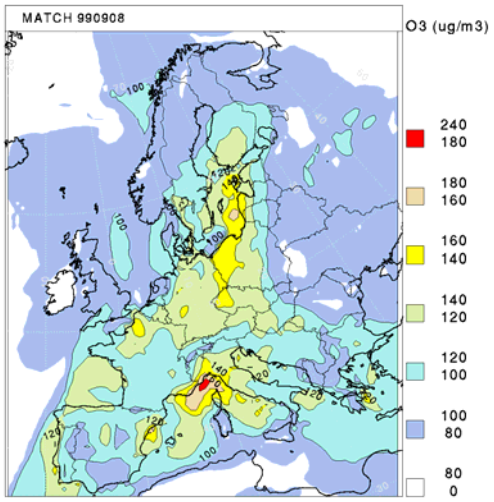
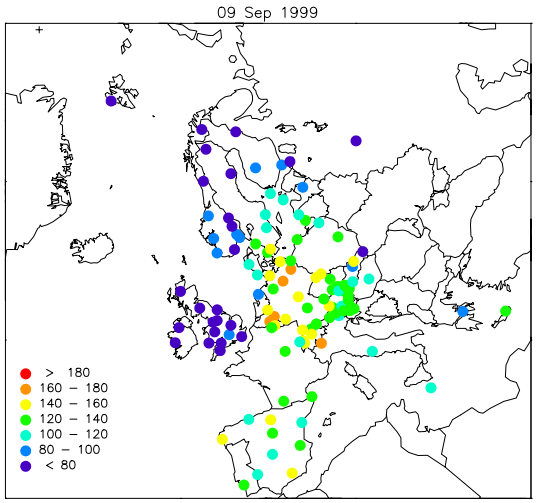
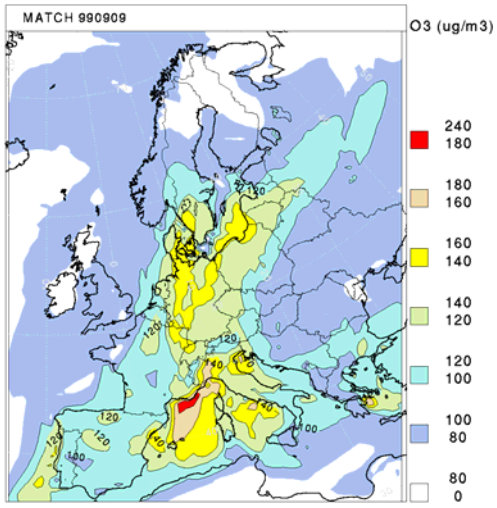


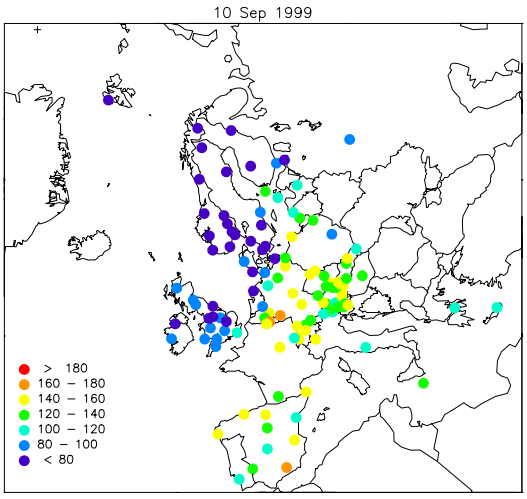
Figure 16: The episode 5–10 September 1999. Daily maximum ozone concentrations as observed at the EMEP network stations (left panels) and as modelled by the MATCH model (right panels). All values are given in µg/m^3.











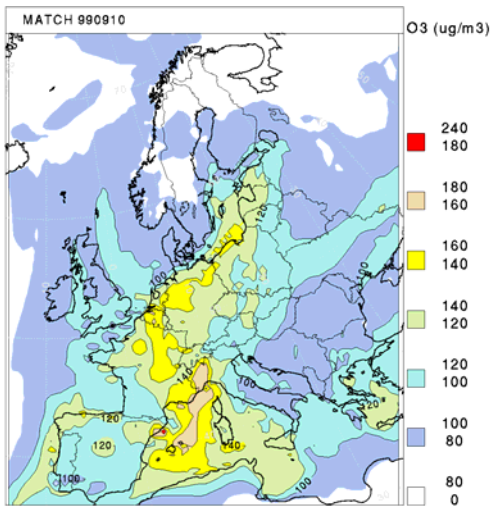


Figure 16, cont.

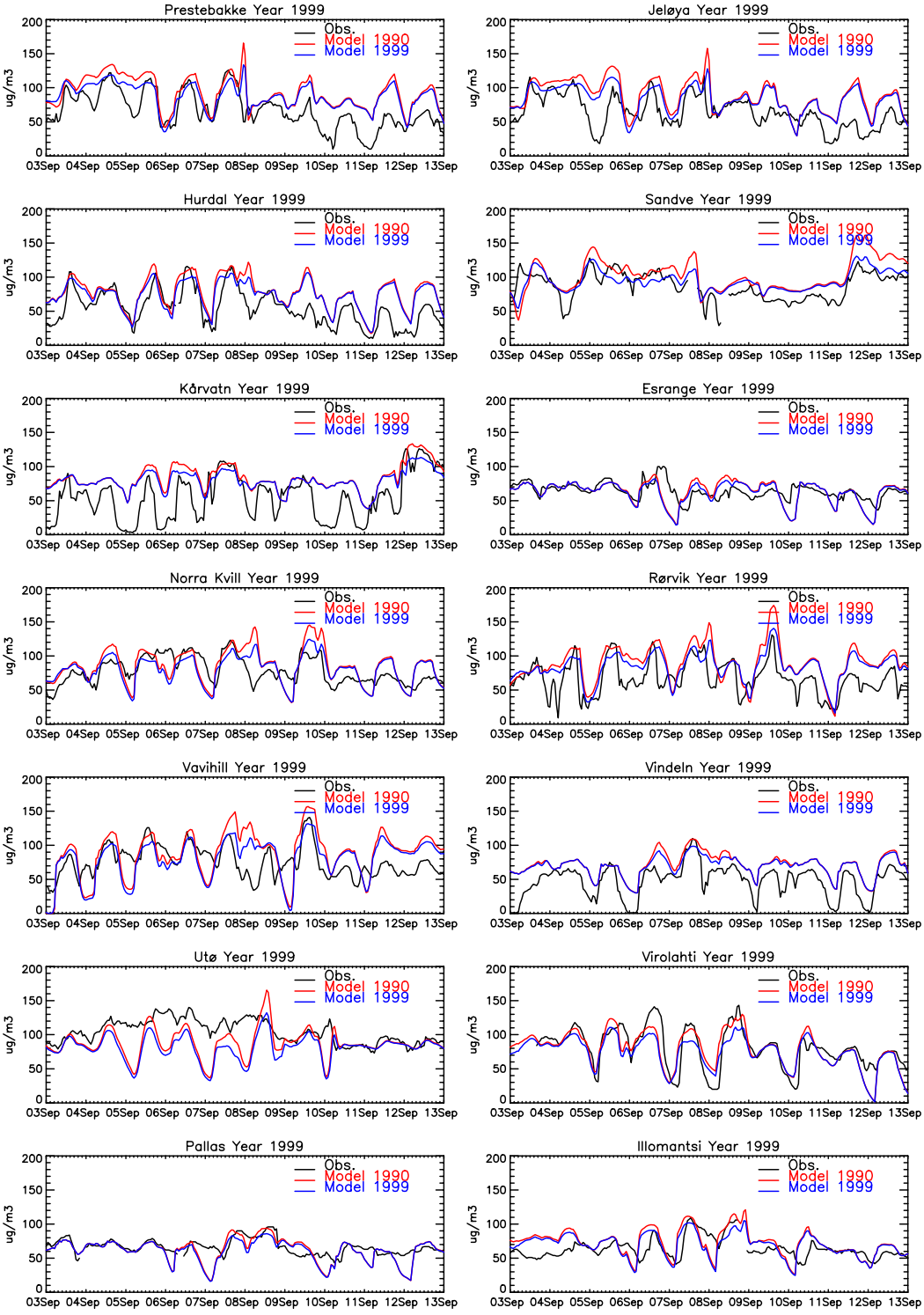


Figure 17: Observed hourly ozone concentrations together with the modelled values using 1990 and 1999 emissions.

4.4 14–25 May 2000

An extensive high pressure system developed already around 10 May over central Scandinavia and moved gradually towards central Europe during a week's time. The period was accompanied by dry and warm weather over large regions of northern Europe. On the 16th and 17th a very marked cold front swept across the continent, including Scandinavia, bringing thunder and hail all along the frontal zone and was followed by cold, unstable marine air masses and a north-westerly flow. Southerly winds increased prior to the frontal passage, bringing continental air masses up to Norway and Sweden and eventually to southern Finland. The episode could be regarded as the "school-book" situation of an ozone episode in the Nordic countries.

With this in mind and also the fact that background concentrations are at the highest in May, it may be somewhat surprising that ozone concentrations didn't reach even higher values than observed. In Norway the ozone concentrations peaked at 168 μg/m^3 at Kårvatn and 163 μg/m^3 at Hurdal, and in Sweden values above 170 μg/m^3 were observed. At least for Norwegian conditions, these are indeed high values, actually then the highest observed since 1994, but still far from the annual peak values observed in the early 1990-ies in Norway which were around 200 μg/m^3 and higher. In May 1992 there was an interesting ozone episode affecting all of southern Norway, that was also caused by a high pressure system over central Europe and southerly transport from the European continent. In that episode the peak values observed in Norway were around 200 μg/m^3 at two sites. It could be tempting to relate the difference in ozone peak values these two years to the emission reductions that apparently have taken place in Europe in the mean time. The difference of 30-35 μg/m^3 corresponds well to the drop in peak ozone generally predicted for such episodes for the Nordic sites by the MATCH model as shown in this work. On the other hand, two episodes are never equal and thus in principle couldn't be compared in this way. The 1992 episode was for instance not associated by a strong cold front passage as in 2000, thus more intense vertical mixing in the 2000 episode could also be an explanation for the lower ozone concentrations observed.

The modelled regional ozone distribution in general agreed fairly well with the measured distribution. However, the results also show that the model under-estimated significantly the ozone concentrations in central Europe particularly 17 May.

The hourly ozone time-series (Figure 20) indicate that the model underestimated somewhat the maximum ozone concentration seen at the Norwegian sites although the timing of the episode was fairly well described. The predicted reduction of peak ozone due to the 1990-1999 emission reductions was of the order of 20-30 μg/m^3.

For the Swedish sites, high concentrations were first observed at Rörvik on the 15th. On this and the following day the model values were lower than observed. During the rest of the period the model overestimated the Rörvik concentrations, especially on the 17th. The development at Vavihill was similar. The best agreement between daytime observations and the model was obtained for Norra

Kvill, with slightly underestimated values in the first part of the period followed by an overestimation in the end.

In the last part of the period the high pressure area was situated over northern Russia and an occluded front was located over Finland. There was advection of warm air (25°C) in sunny weather from the southeast to the most eastern parts of Finland. On May 22 the evening ozone concentrations at Illomantsi exceeded 140 $\mu g/m^3$. According to the meteorology the air mass had passed the Moscow region two days earlier where pollutants had accumulated in nearly calm conditions.

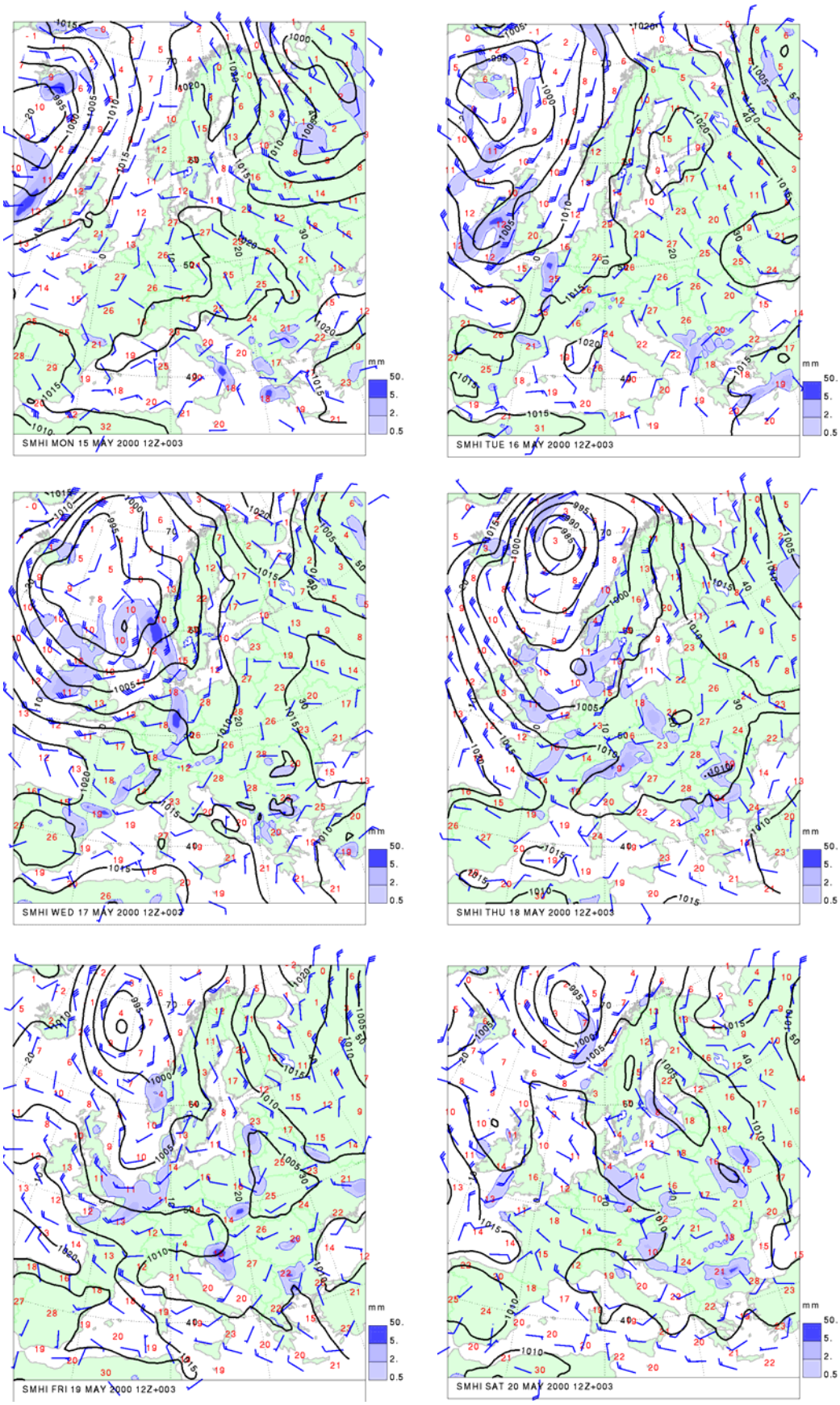


Figure 18: HIRLAM meteorology15–24 May 2000. Symbols as in Figure 9.

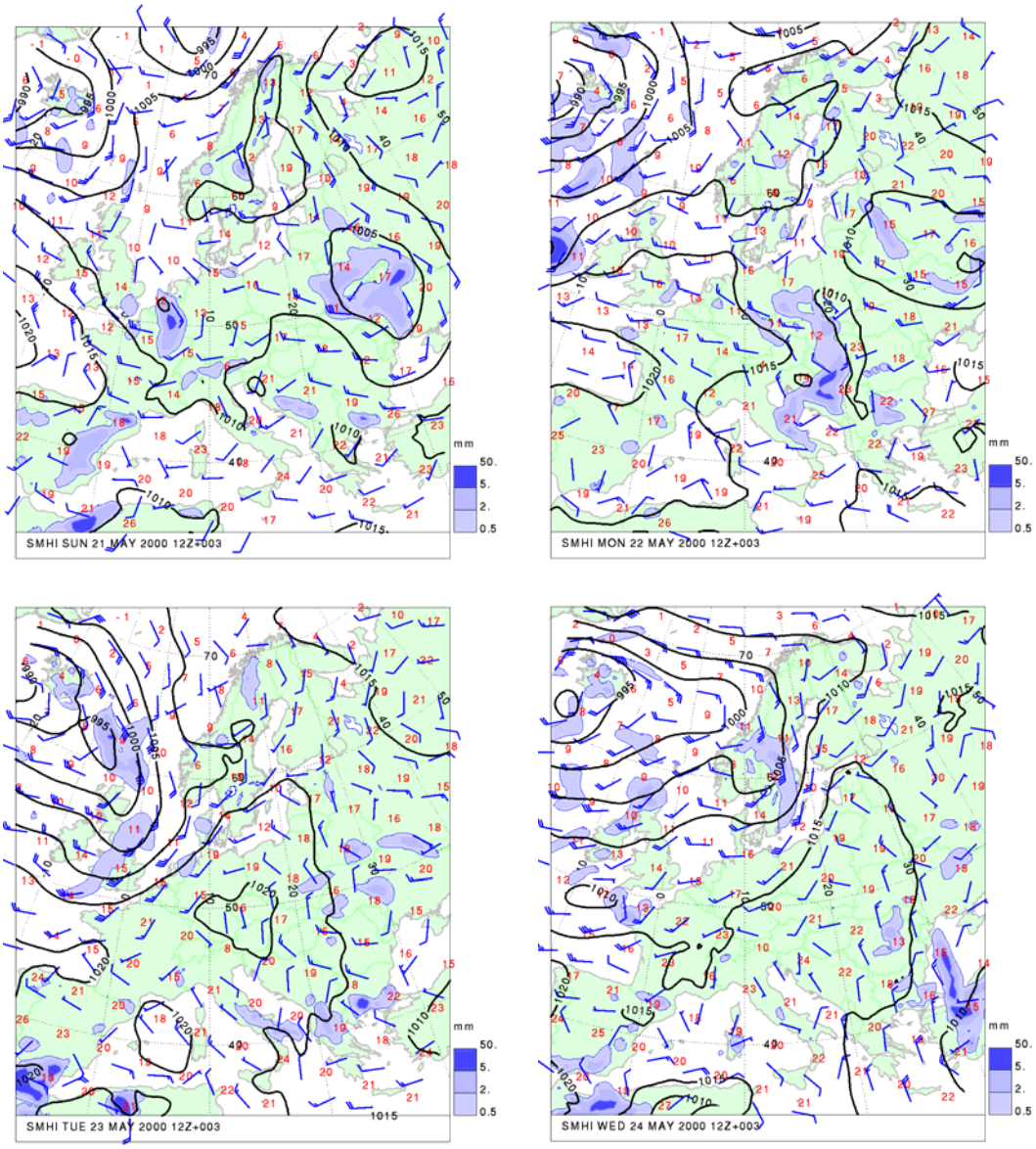


Figure 18, cont.

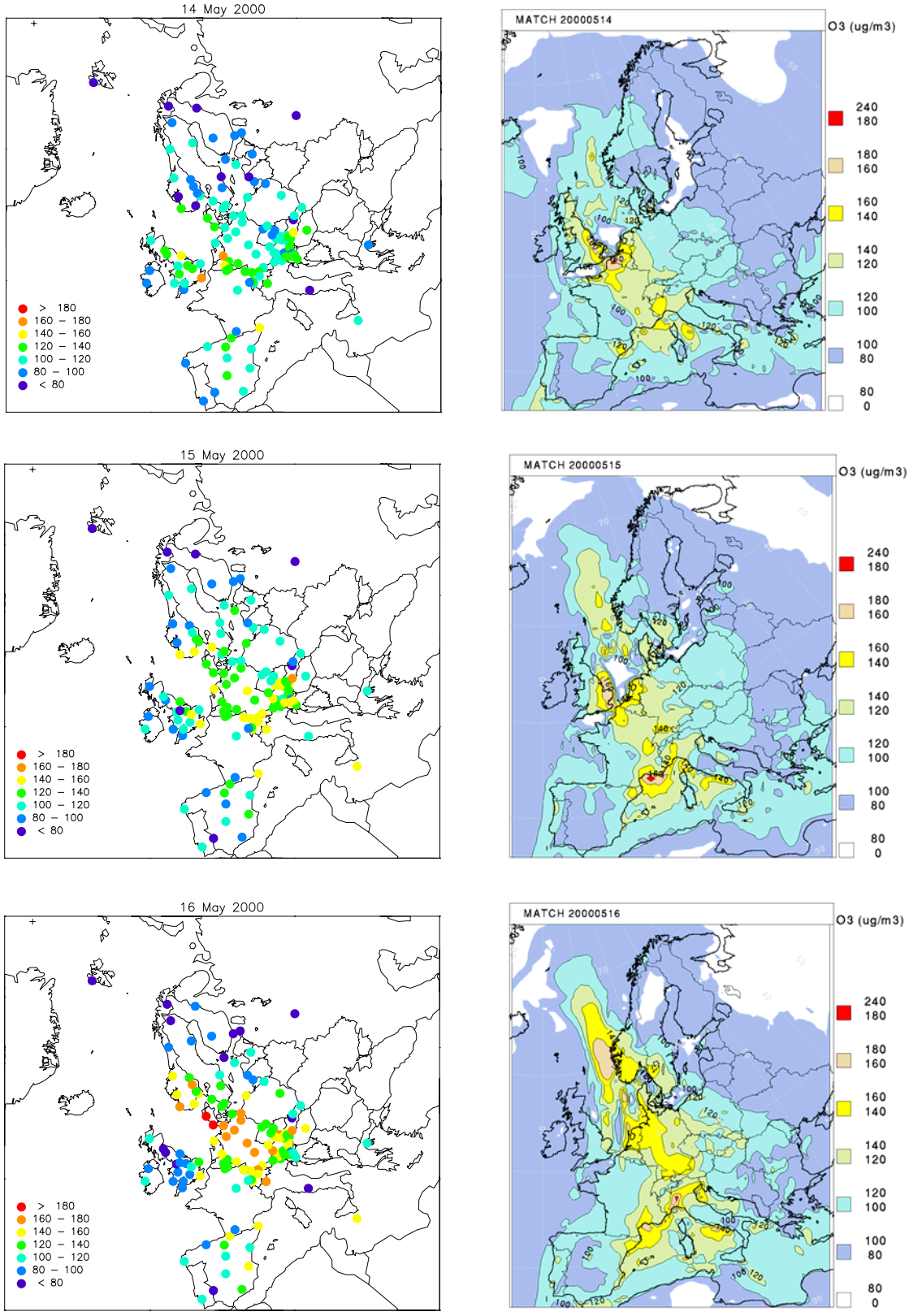
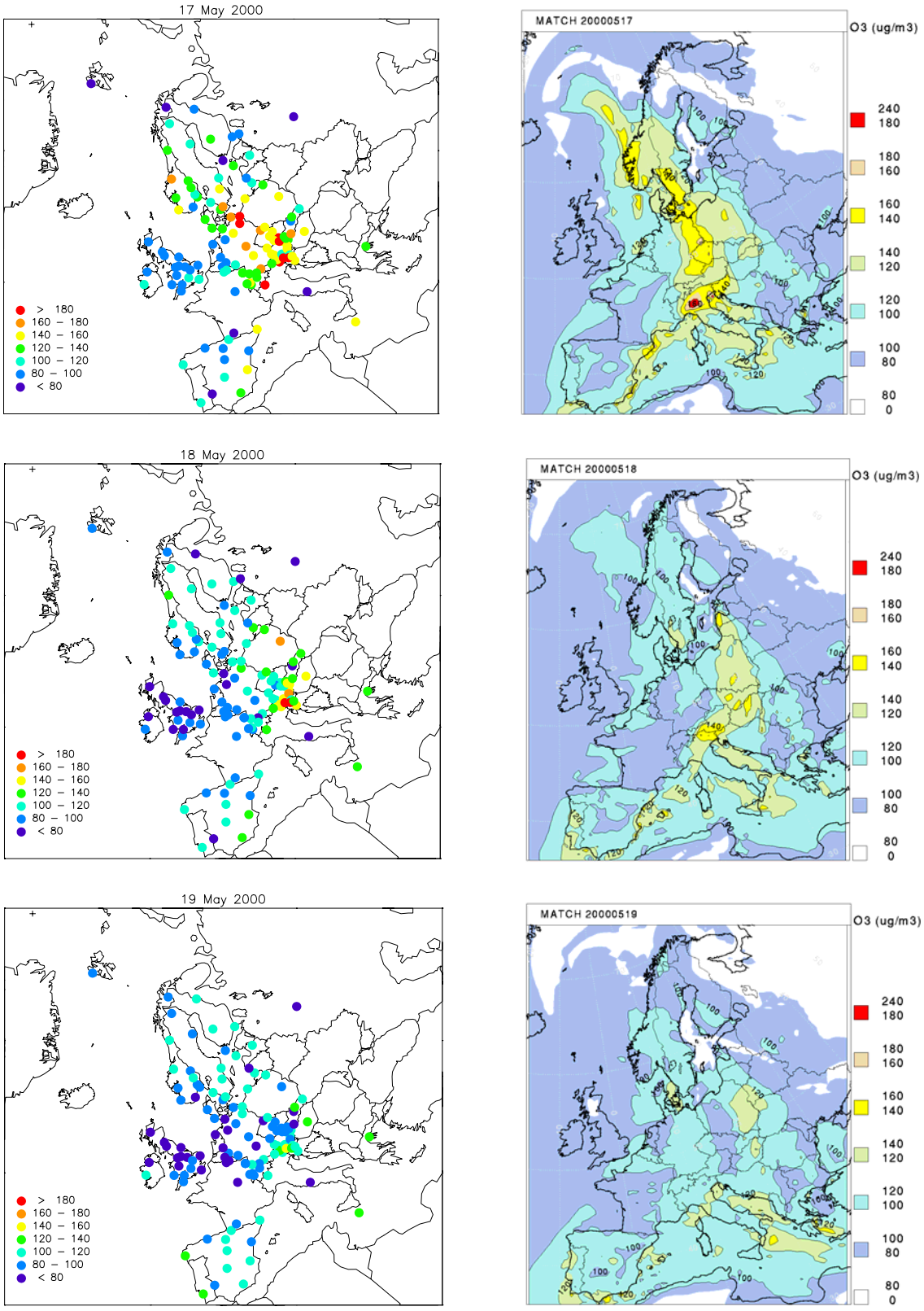


Figure 19: The episode 14–25 May 2000. Daily maximum ozone concentrations as observed at the EMEP network stations (left panels) and as modelled by the MATCH model (right panels). All values are given in $\mu g/m^3$.



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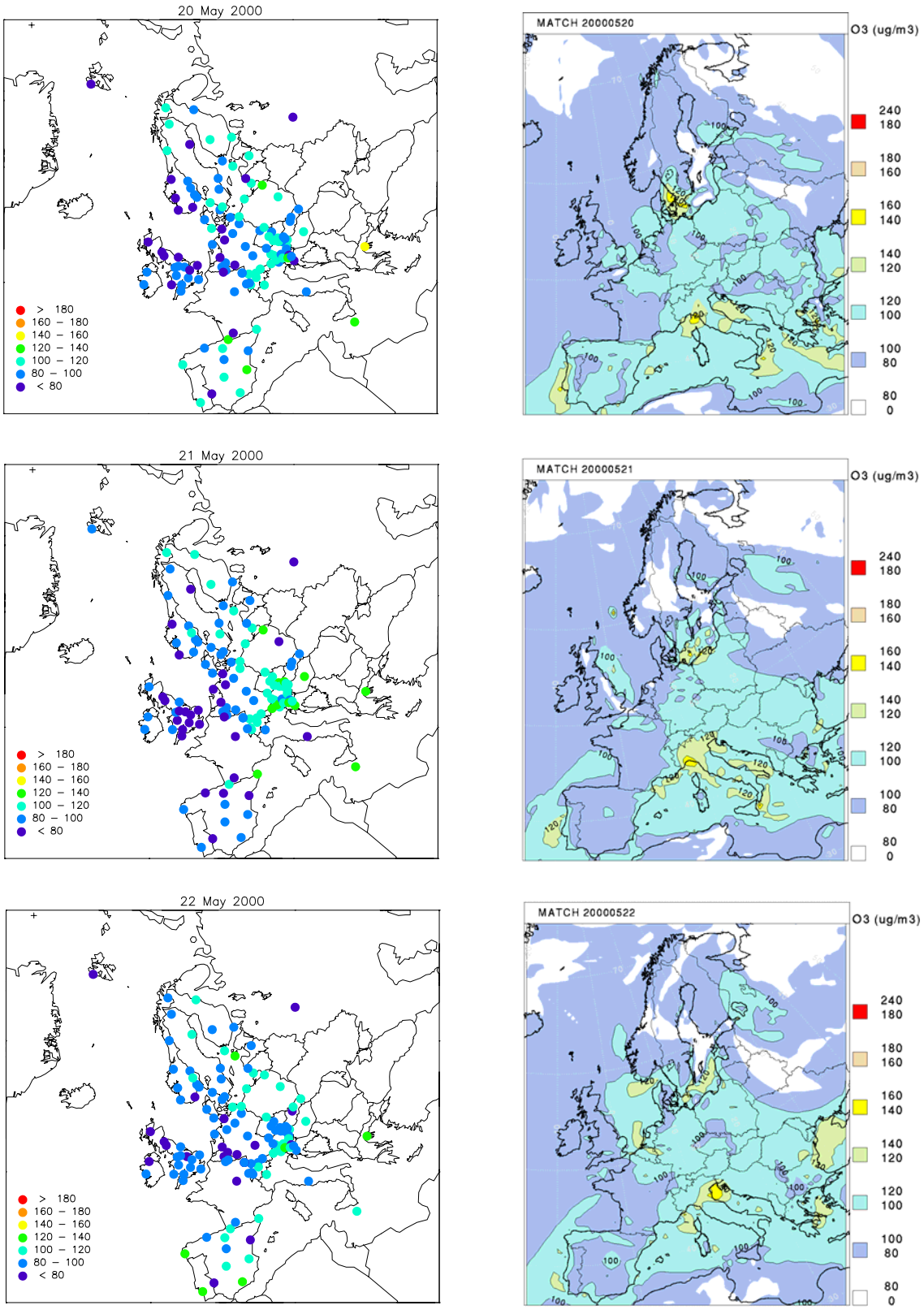

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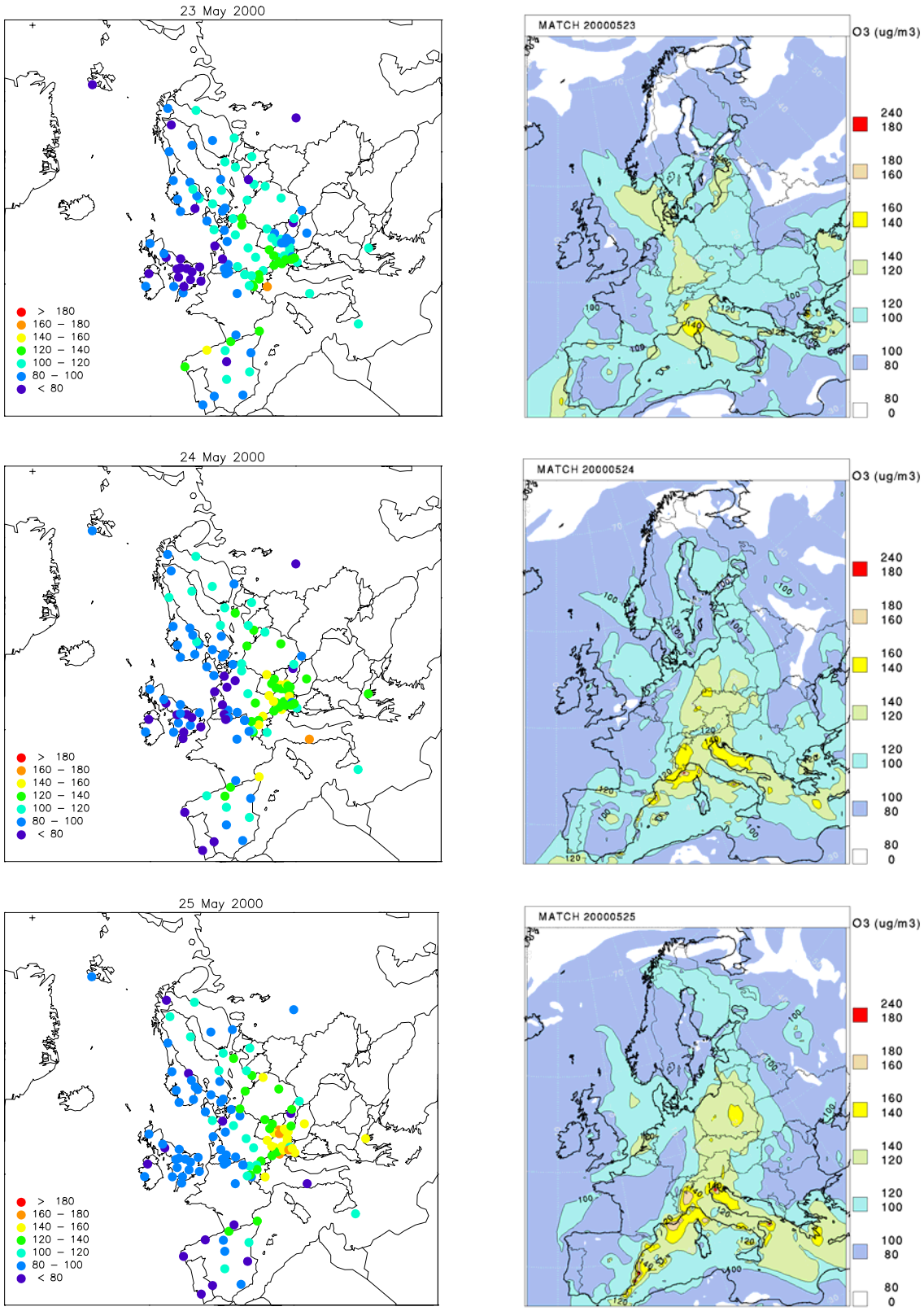

Figure 19, cont.

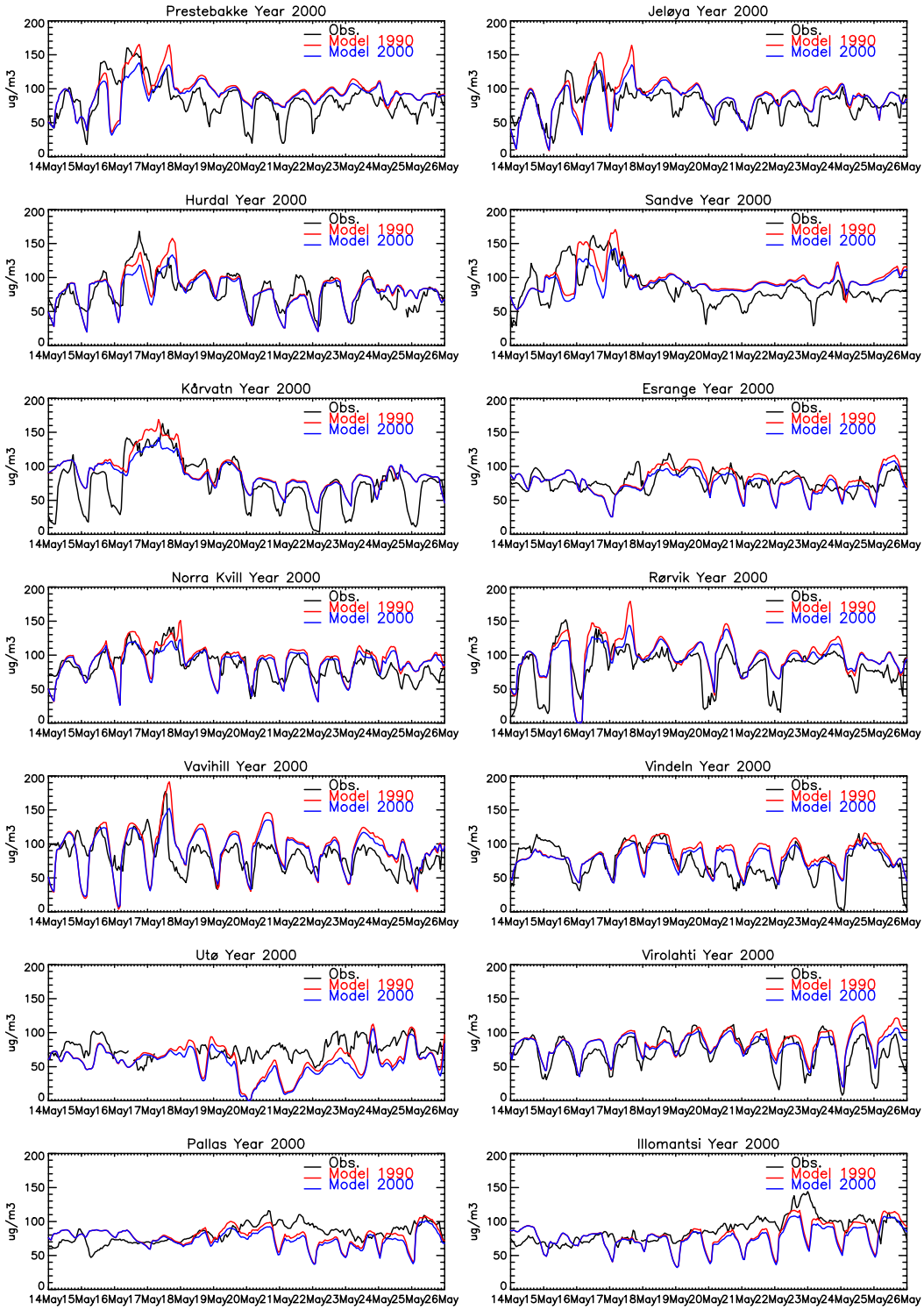

Figure 20: Observed hourly ozone concentrations together with the modelled values using 1990 and 1999 emissions.

4.5 8–13 June 2000

A high pressure region that built up over Poland/E-Europe during the 8-9 June gave SE winds in southern Norway/Sweden and north-westerly over Finland. In the following days the anticyclone weakened and moved to the east and was replaced by SW-W winds over the Nordic countries.

This was a minor episode in terms of ozone concentration. Peak values in Finland and Sweden exceeded 140 μg/m^3 while in Norway the highest observed value was 133 μg/m^3. For the episode days, the weather maps indicate advection to Finland across the Baltic Sea. At Utö the ozone concentration was about 120 μg/m^3 while in the inland areas in the southern parts of Finland (Evo and Jokioinen, around 100 km from the coast) the ozone concentrations increased to about 130 μg/m^3. At Ähtäri in central Finland (about 350 km from the coast) the ozone concentration exceeded 140 μg/m^3. This may indicate photochemical ozone formation over the Baltic Sea from precursor emissions in Poland followed by further ozone production over southern Finland. The ozone concentrations decreased on June 12 when an occluded front passed bringing air masses from the west.

Enhanced ozone levels in connection with transport from SE were observed on the 9th at both Rörvik and Vavihill; at Rörvik with a small delay compared to the model. The following day the observed concentrations were even higher, about 150 μg/m^3, which is higher than indicated by the model. For the following days however, the model overestimated the levels. A third episode is indicated by the model on the 13th, and a corresponding increase is also found in the observations for Vavihill and Norra Kvill.

Compared to the European measurements the modelled regional ozone distribution was fairly well reproduced, but at too low concentrations. Both the measured and modelled fields indicate an area of elevated ozone concentrations in central Europe being transported to the east and north during the episode. The model predicted drop in peak ozone values is around 15 μg/m^3.

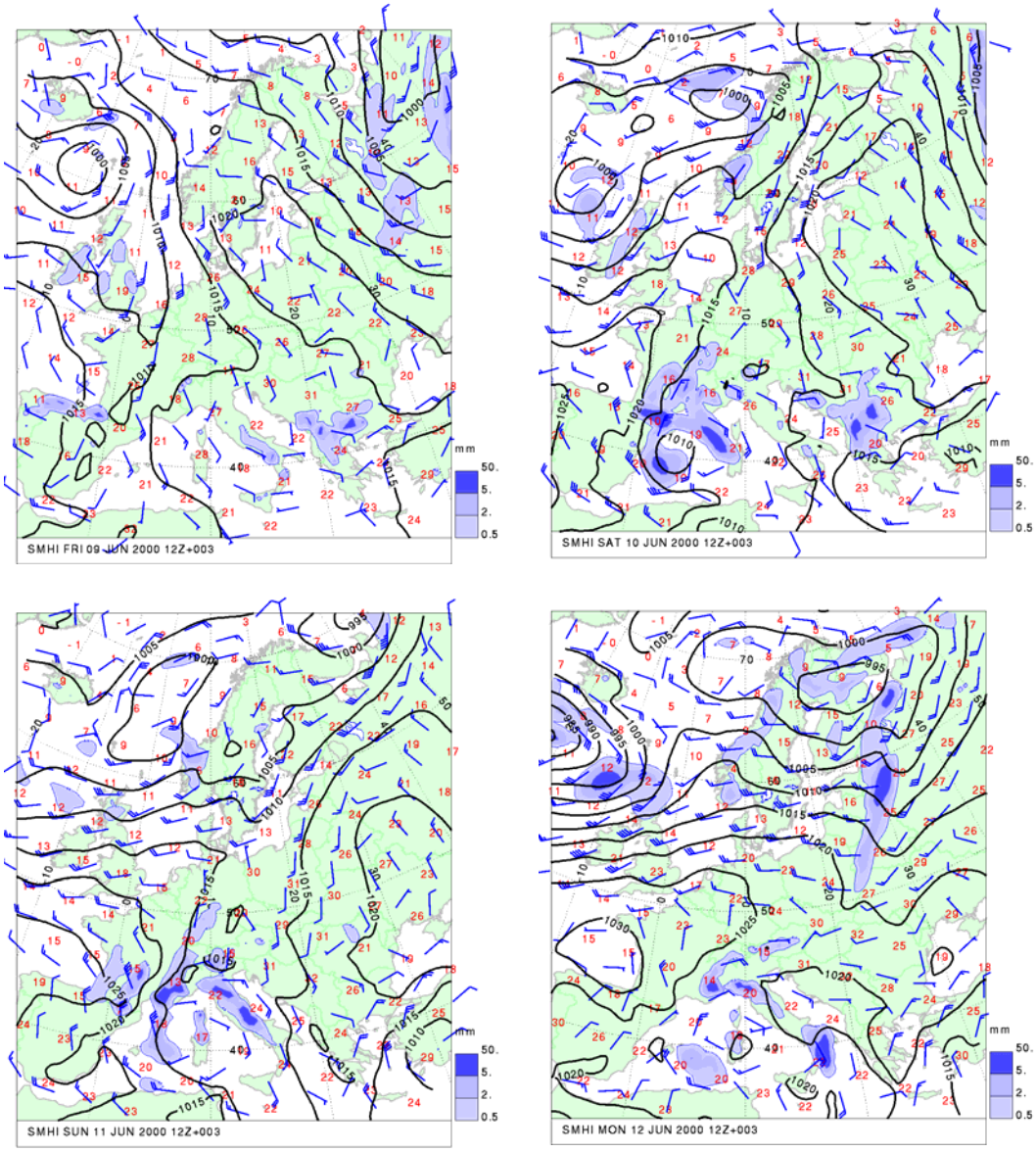

Figure 21: HIRLAM meteorology 9–12 June 2000. Symbols as in Figure 9.

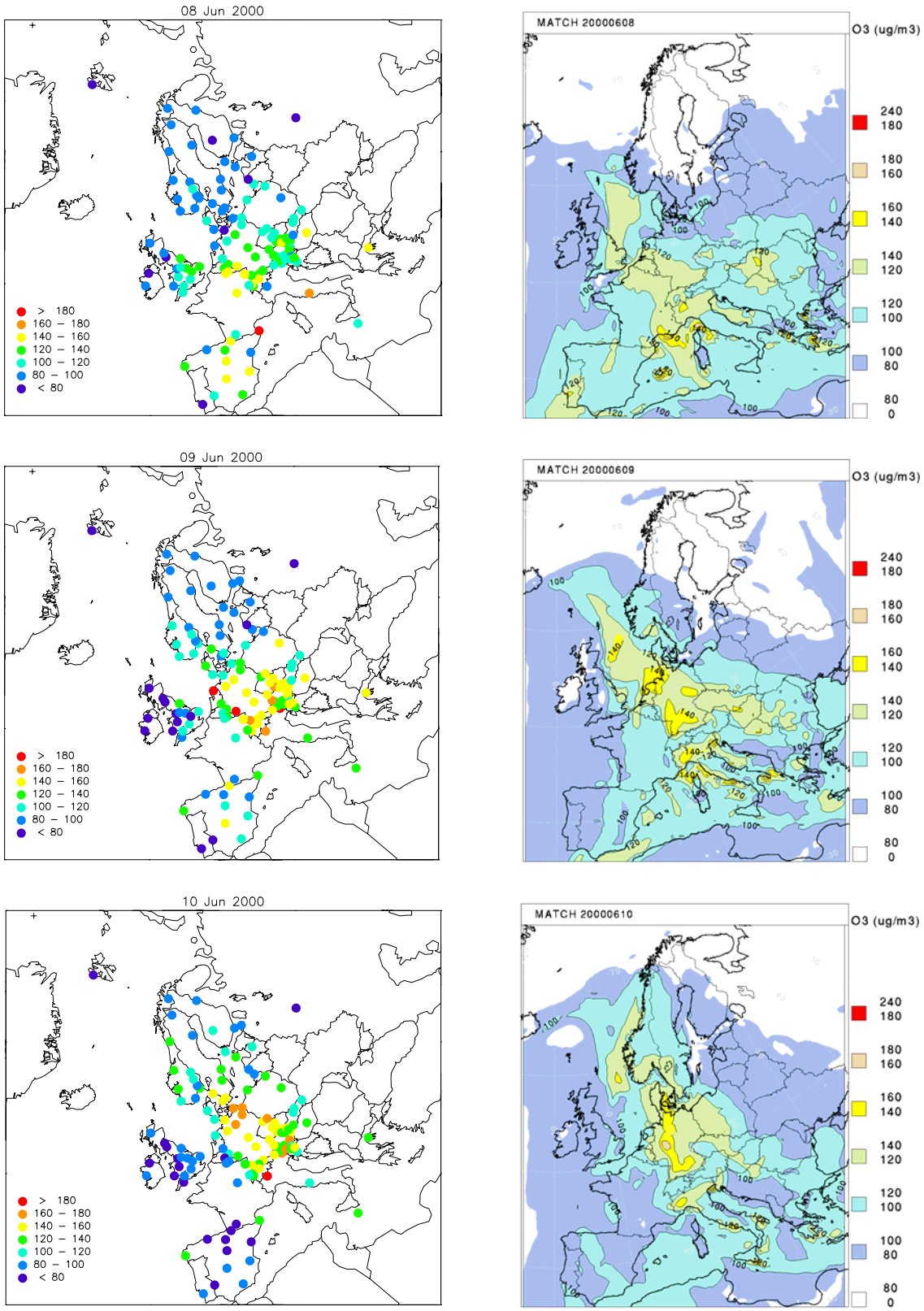

Figure 22: The episode 8–13 June 2000. Daily maximum ozone concentrations as observed at the EMEP network stations (left panels) and as modelled by the MATCH model (right panels). All values are given in µg/m^3.

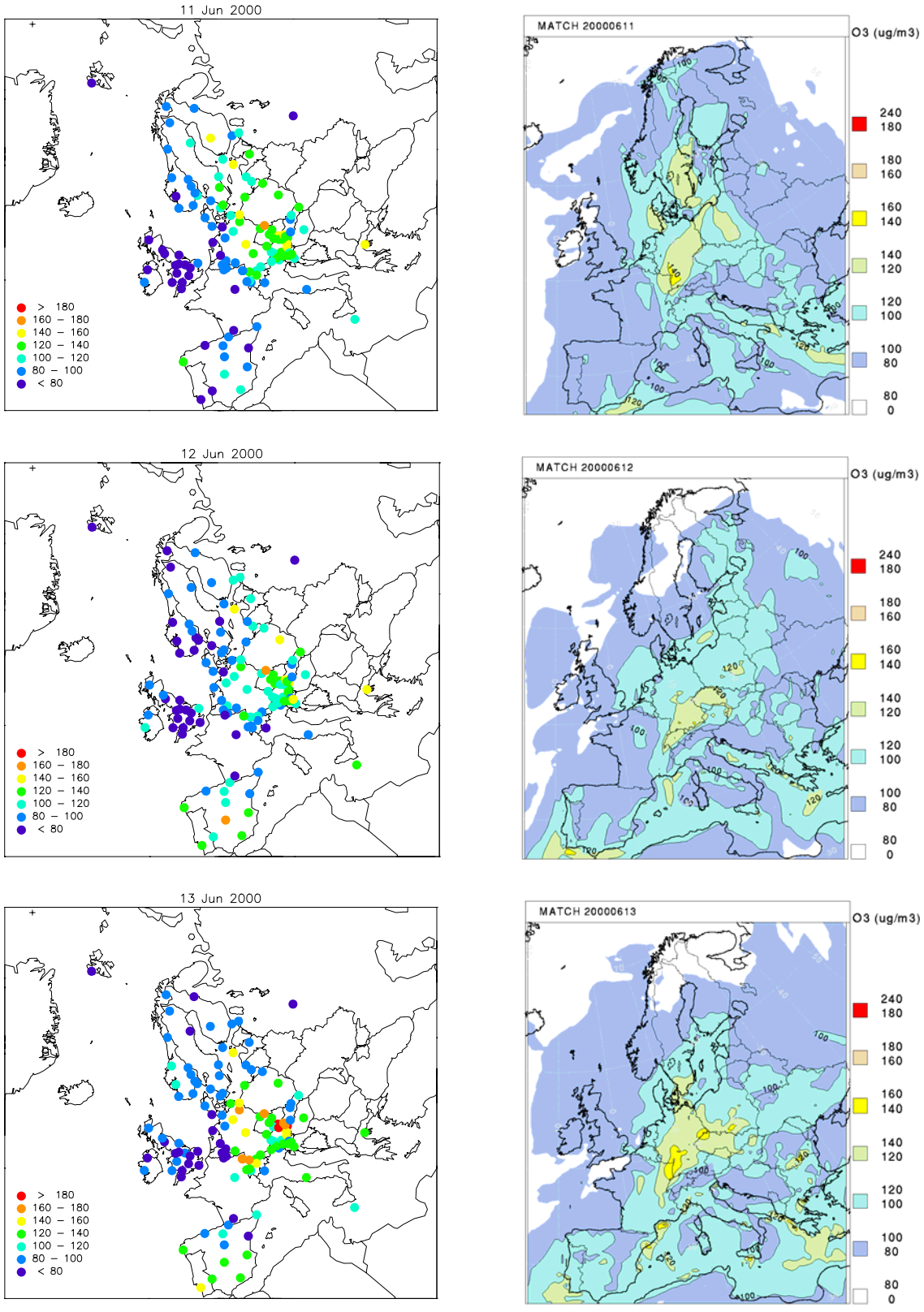

Figure 22, cont.

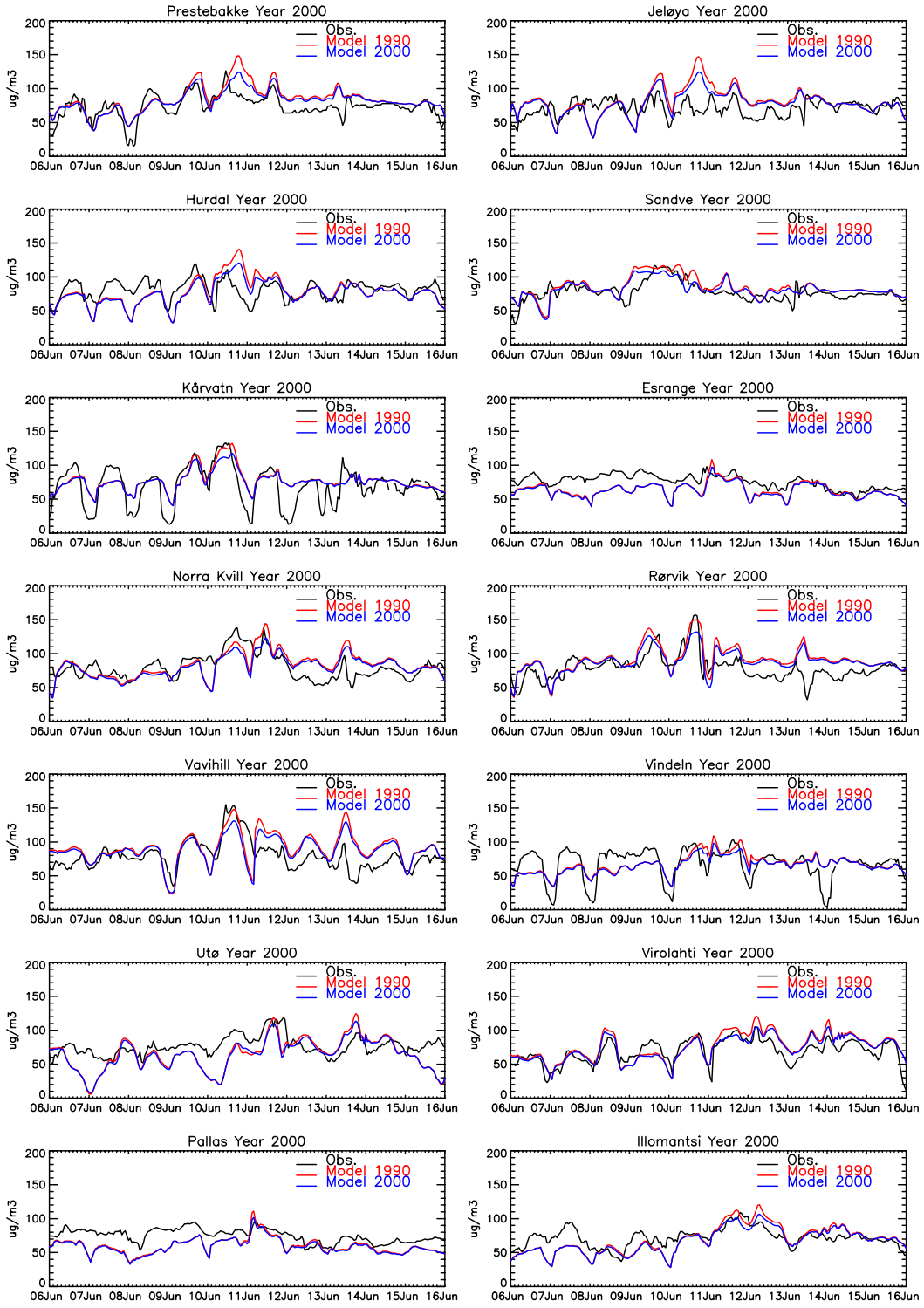

Figure 23: Observed hourly ozone concentrations together with the modelled values using 1990 and 1999 emissions.

4.6 18–23 June 2000

In the days prior to this episode a high pressure system developed over NW-Europe and moved slowly towards southeast. On 18-19 June the extensive high pressure was centred over E-Europe, giving weak southerly winds and high temperatures (above 30°C) over large regions. In the following days the high weakened and gave way for a cyclone and fronts approaching from the Atlantic, crossing the British Isles on 20-21, increasing the SW winds over the Nordic countries.

This was a very marked ozone episode, affecting most of Europe, particularly the central parts for several days. The ozone concentrations exceeded the EU threshold of 180 $\mu g/m^3$ at many sites on the continent, including in South Sweden. Peak values in Norway were 172 $\mu g/m^3$ (at Prestebakke). The meteorological data showed that the air mass reaching Evo and Ähtäri (in Finland) had passed the Helsinki area earlier. Interestingly, the air mass had apparently circulated over the southern and central parts of Finland for two days, and the ozone concentration before the episode was only 80-100 $\mu g/m^3$.

The model simulated the regional ozone distribution fairly well, but at much too low concentrations. This is a situation when one would expect very high ozone concentrations in southern Sweden and very high concentrations, >200 $\mu g/m^3$ at Vavihill and Rörvik and about 150 $\mu g/m^3$ at Norra Kvill, were also observed. The calculated values were much lower and, as was the case also during some of the previous episodes, the model overestimated the concentration on the subsequent days. The model values using the higher 1990 emissions produced higher ozone peak values, but still significantly lower than the measured values. The predicted drop in peak ozone due to the emission reductions from 1990 to 1999 was of the order of 15-30 $\mu g/m^3$ for the Nordic stations.

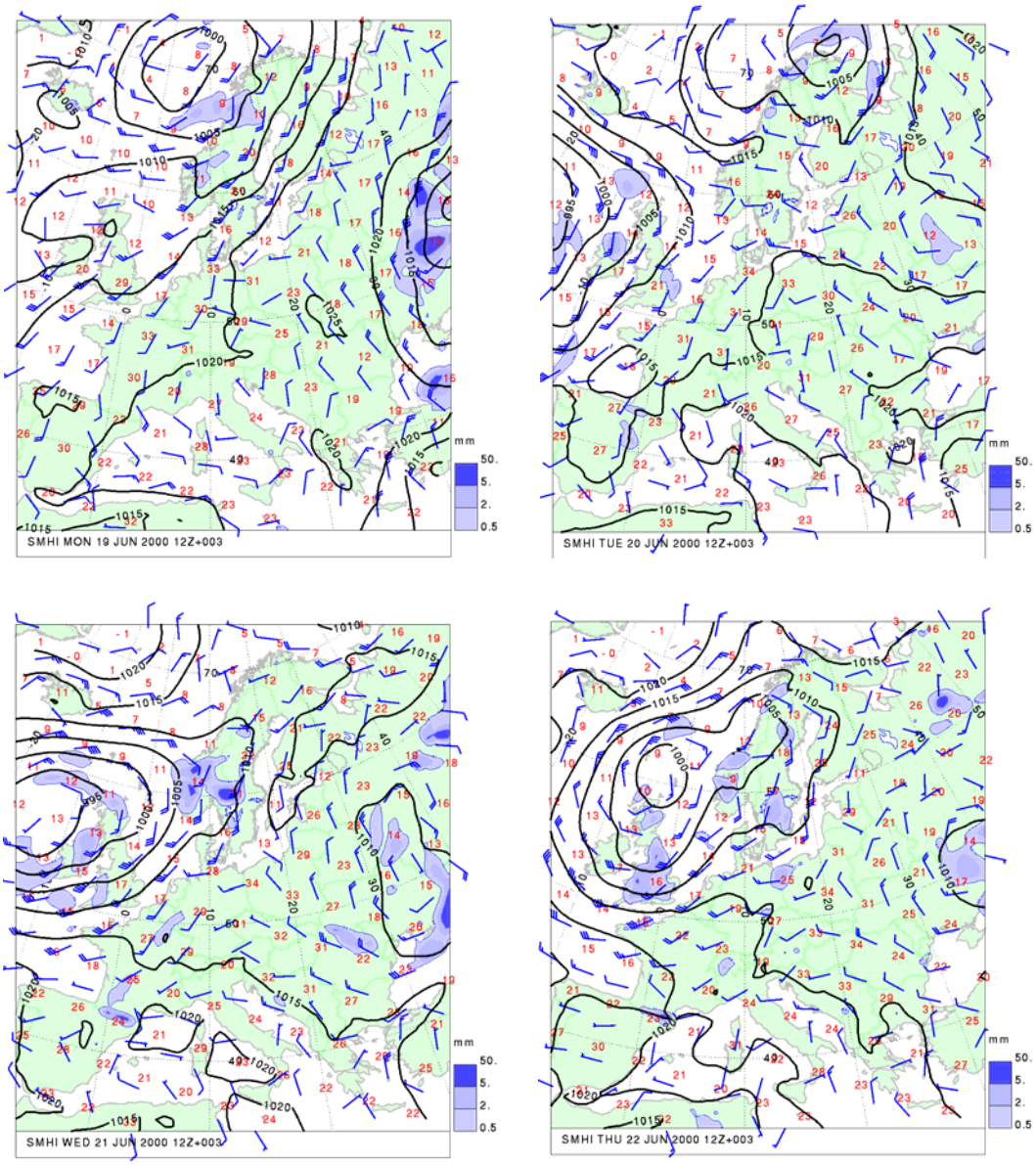

Figure 24: HIRLAM meteorology 19–22 June 2000. Symbols as in Figure 9.

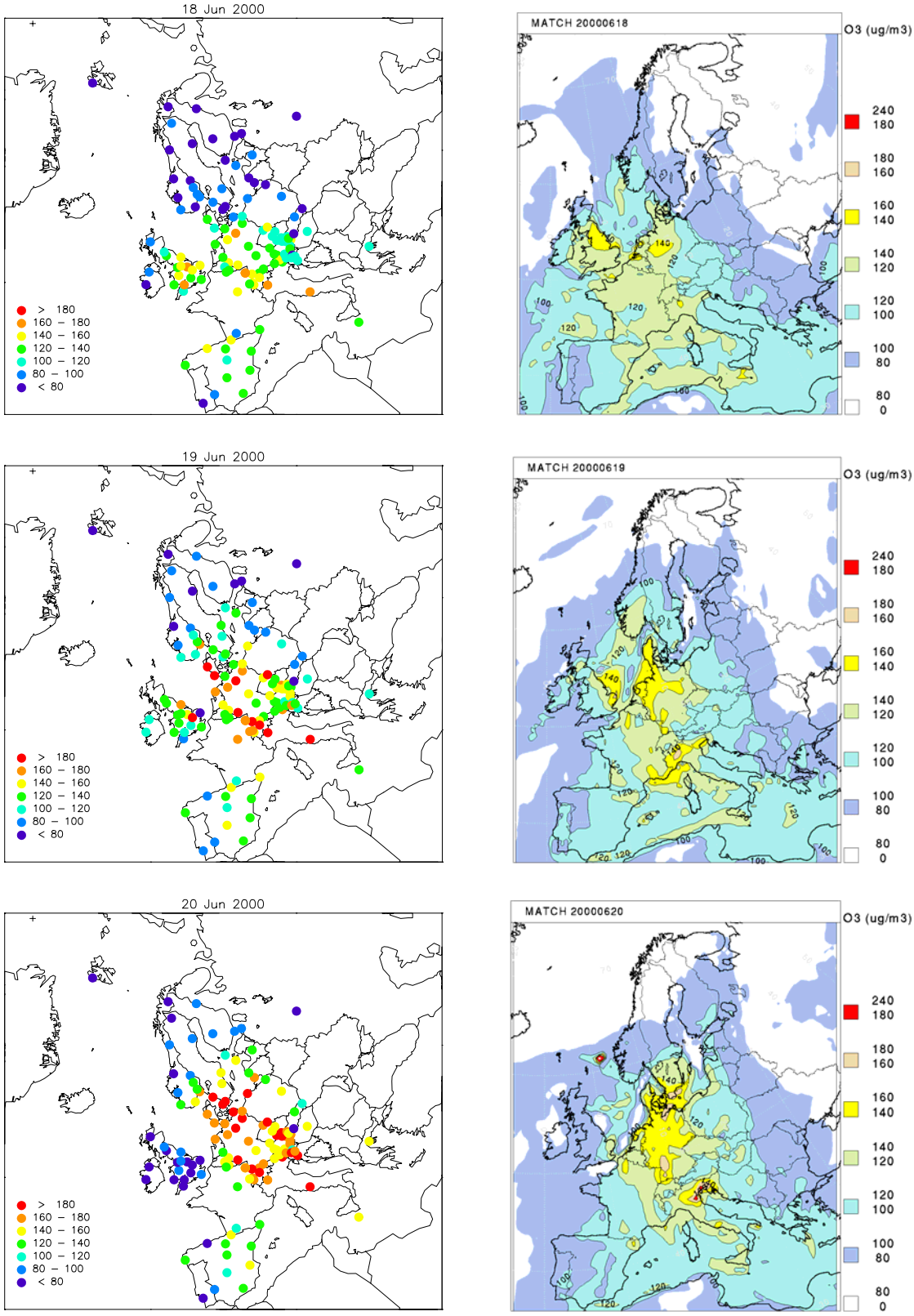

Figure 25: The episode 18–23 June 2000. Daily maximum ozone concentrations as observed at the EMEP network stations (left panels) and as modelled by the MATCH model (right panels). All values are given in $\mu g/m^3$.

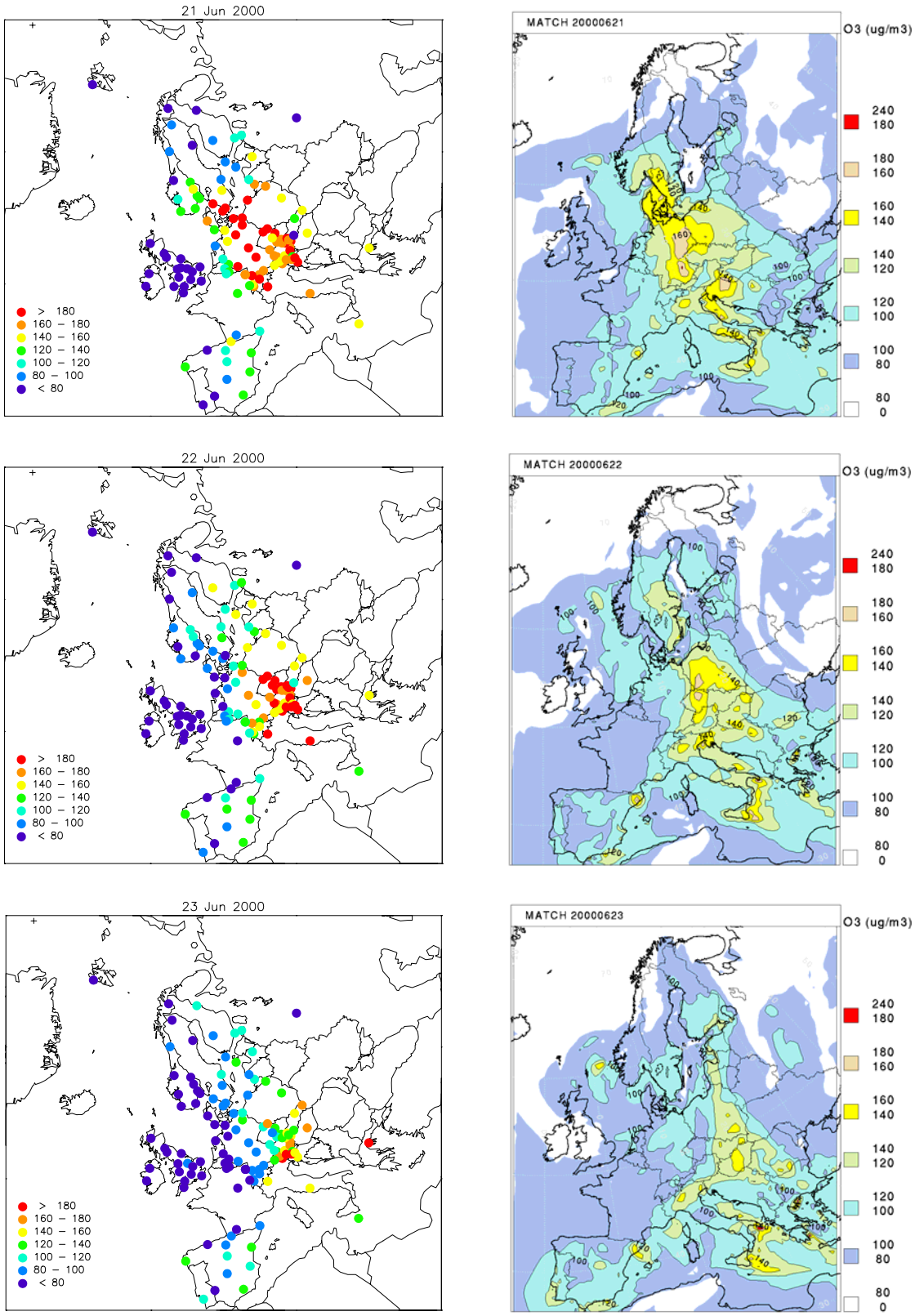

Figure 25, cont.

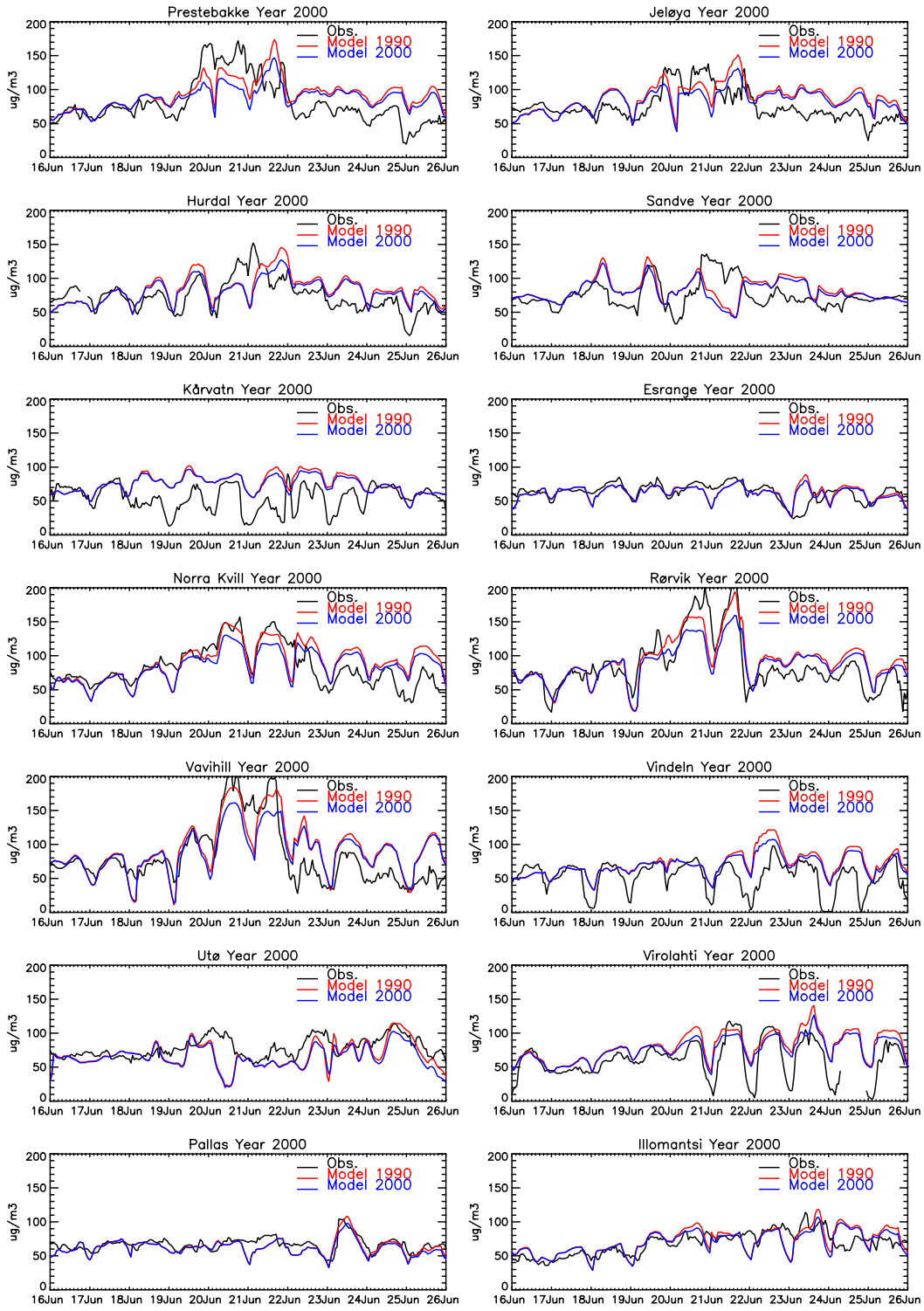

Figure 26: Observed hourly ozone concentrations together with the modelled values using 1990 and 1999 emissions.

5. Discussion and conclusions

Results from a collaborative project by the Nordic Council of Ministers (Hav- och luftgruppen) in 2001 have been presented. The aim of the project was to evaluate to what extent there has been a reduction in concentration level and extent of peak ozone episodes in the Nordic countries as a result of emission reductions in Europe the last 10-15 years. EMEP ozone monitoring data were used in the project together with the MATCH model from SMHI.

Six episodes from 1999 and 2000 were selected and compared with the model results using EMEP emission data for 1990 and 1999, respectively. The idea was that episodes with sufficiently good spatial and temporal fit between the modelled and observed data would give confidence in the model results, and could be used to make statements about the likely change in Nordic ozone episodes that have taken place during the 1990-ies.

We found several independent indications that high ozone values have decreased in the 1990-ies. Firstly, inspection of the 99-percentiles of hourly measured ozone concentrations from the summer half year indicates that in the Nordic areas close to the European areas where emission density is high and where 99-percentiles have been mostly above 120 $\mu g/m^3$, episodic ozone concentrations are indeed decreasing. It seems that these trends are less at the more eastern (Finnish) sites. Both the observations and the model indicate reductions in the 99-percentiles of the order of 10-20 $\mu g/m^3$ per ten years in southern and central parts of Sweden and Norway. Time series should, however, be longer to reach significant trends in the northern parts of Scandinavia.

Next, the comparison between the modelled and observed ozone time series gave a somewhat mixed picture. In general, we found a good agreement between the observed and modelled time series. For the 2000 data the correlation coefficient based on daily maximum values during the summer half year was between 0.6 and 0.8 for each station separately which gives a highly significant correlation. The correlation for 1999 was slightly lower. In our view, this gives a strong confidence to the model performance in general.

Furthermore, the modelled regional (European) distribution of ozone in general also agreed well with the patterns evident from the European maps of daily maximum concentrations for the selected episodes. This is a further support for the model, indicating that the transport description is reasonable. However, the comparison also revealed that the model had a tendency to underestimate the ozone concentrations during the episodes. A closer look at the performance of the model showed that the model results in general tended to be more "smoothed out" than the measurements, i.e. that the highest observed values were underestimated and the low observations were overestimated in the model. This kind of result is probably rather common when comparing models with monitoring data, and to some extent reflects spatial and temporal smoothing in e.g. the meteorological fields, the emissions etc. The underestimation of ozone were however during some of the episodes (notably the marked episode 18-23 June 2000) too large to be explained by this alone, and points to other causes as well, such as e.g. the temporal variation in the emission data, vertical exchange processes etc.

Anyway, this underestimation and the general tendency of the model to reduce the amplitudes compared to the measured values, should indicate that the predicted change in ozone due to the emission changes 1990-1999 from the model could be regarded as a conservative estimate, i.e. that it's likely that the effects on ozone in reality were larger than indicated by the model results presented here.

Thus, to conclude, we have found several results that taken together indicate that it's very likely that there has been an improvement in ozone episodes (i.e. that peak values have been reduced) in S-Norway and S-Sweden due to European emission reductions during the 1990-ies. The results, which could be regarded as a conservative estimate, indicate a reduction of the order of 30 $\mu g/m^3$ ozone for the highest peak values and less for the less pronounced episodes. It is furthermore likely that the number of episodes of exceedance of the EU threshold value of 180 $\mu g/m^3$ or at least the 160 $\mu g/m^3$ applied by the State Pollution Agency in Norway, has also been reduced in the same period, although the few number of these episodes make the statements uncertain. The 99-percentiles of summer six-months hourly data (approx the 40th highest value) have probably been reduced by the order of 10-20 $\mu g/m^3$ in the same region. For Finland and for the northern part of the region, the conclusions are more difficult to assess and become more uncertain. Although the model indicates similar results for S-Finland, the agreement with the measurements are somewhat poorer, possibly reflecting that ozone episodes in Finland in general are more linked to transport from east than Sweden/Norway and that the emission data in Russia/E-Europe is less established.

These conclusions are based on fairly simple analyses with indeed a high degree of uncertainty. The length of the time period studied is by itself a reason for being careful with making strong statements. However, the fact the results from separate analyses (the measurements by themselves, the comparison with model predictions, and the comparison of similar episodes many years apart) are fairly coherent and not conflicting, should be given weight, i.e. that all applied analyses tend to go in the same direction.

Finally, we certainly admit that the present analyses could easily be extended in all directions. More years could be included (particularly in the modelling), more detailed studies of trends for the separate percentiles of measured values could be carried out, the analyses of *reasons* for lack of model agreement could be widely expanded etc. However, the present work was simply limited by the resources and time available during this rather small one-year project.

6. Acknowledgement

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Appendix A

MATCH model description

A1 Introduction

The MATCH (Multi-scale Atmospheric Transport and Chemistry) model (Robertson et al., 1999) has been developed as a flexible transport/chemistry/ deposition model for atmospheric pollutants. It is used in a range of applications from urban scale studies on ~5 km or higher horizontal resolutions to continental scale studies on acid deposition and photochemistry (see www.smhi.se/sgn0106/ if/meteorologi/match.htm). MATCH is also used operationally to provide forecasts of radioactivity in case of nuclear emergencies in Europe (Langner et al., 1998a). Here we report the model setup (v.3.9.1d) used for the studies of ozone episodes in the Nordic countries. For details on the basic transport model the reader is referred to Robertson et al. (1999). More details about the photo-chemistry version of the model can be found in Langner et al. (1998b) and in Tilmes et al. (2002).

A2 Model

The MATCH model solves the advection diffusion equation for atmospheric tracers in a three-dimensional, Eulerian framework:

$$\frac{\partial c_i}{\partial t} = -\nabla(\mathbf{v}c_i) + \nabla(\mathbf{K}\nabla c_i) + Q_i + S_i \qquad (1)$$

where c_i represents the mass mixing ratio of the trace species of interest, **v** is the three-dimensional wind, **K** is the turbulent diffusion tensor and Q_i and S_i represents internal sources and sinks.

The basic transport model includes modules describing emissions, advection, turbulent diffusion and dry and wet deposition. Depending on the application, specific modules describing, e.g., chemistry can be added to the basic transport model. MATCH is an ”off-line” model. This means that atmospheric weather data are taken from some external source, usually a numerical weather prediction (NWP) model, and fed into the model at regular time intervals, currently every three or six hours. Such data are then interpolated in time to yield hourly data. Special attention is given to interpolation of the horizontal wind where vector increments are applied. The vertical wind is calculated internally to assure mass consistency of the atmospheric motion after the time interpolation of the horizontal winds.

The model design is flexible with regard to the horizontal and vertical resolution, principally defined by the input weather data, and allows for an arbitrary number of chemical compounds. The model is written in η (or hybrid) vertical co-ordinates which is a linear combination of pressure and σ vertical co-ordinates.

A2.1 Advection

Advection is modeled using a positive definite flux form advection scheme (Bott, 1989, 1992), utilizing polynomials fitted to the concentration distribution in order to reduce numerical diffusion. The scheme has been rewritten using primitive functions to be applicable in situations with variable grid distances (Robertson et al. (1999)). The scheme is mass conservative. For the calculations presented here,

fifth order integral functions were used in the horizontal and an upstream scheme in the vertical direction.

A2.2 Boundary layer parameterization

Boundary layer processes, such as turbulent vertical mixing in the boundary layer and dry deposition, are parameterized using three primary parameters: the surface friction velocity (u_*), the surface sensible heat flux (H_0) and the boundary layer height (z_{PBL}). The friction velocity is calculated for neutral stratification in order to avoid unrealistic values of numerical origin for strongly stable and unstable conditions.

The sensible heat flux is given by the surface energy balance equation, utilizing different formulations for land and ice covered sea and for open sea. For land and ice covered sea H_0 is defined from similarity theory, using the surface friction velocity, u_*, and the temperature scale, θ_* (van Ulden and Holtslag, 1985).

The calculation of the boundary layer height for unstable conditions is based on a bulk Richardson number approach (Holtslag et al., 1995), where the boundary layer height is defined as the height where the bulk Richardson number, *Ri*, reaches a critical value of 0.25. For neutral and stable conditions a formulation proposed by Zilitinkevich and Mironov (1996) for the equilibrium stable boundary layer is used. The formulation accounts for the combined effects of rotation, surface momentum flux and static stability in the free flow and remains applicable in the limits of a rotation-free stable layer and a perfect neutral layer subject to rotation.

The horizontal diffusive fluxes are assumed to be small compared to the advection along the direction of the horizontal wind. Therefore only the vertical turbulent mixing is taken into account. Two different formulations of the vertical turbulent exchange coefficient, K_z, are applied. The exchange coefficient within the boundary layer for neutral and stable conditions follows Holtslag et al. (1995). For unstable conditions the convective turn-over time, z_{PBL}/w_*, is used directly to determine K_z. Above the boundary layer K_z is set to zero. A scheme for transport by deep convection is available but was not used for the simulations presented here.

A2.3 Dry deposition

Dry deposition is modeled using a resistance approach (Chamberlain and Chadwick, 1965), where the component dry deposition flux is proportional to the concentration of each component and the inverse of the sum of the aerodynamic resistance and a species specific surface resistance. For simplicity we use the same aerodynamic resistance for all surfaces in a grid square.

Variations in the surface resistance are calculated using the methodology of Noilhan and Planton (1989). The surface resistance is dependent on soil moisture, soil type, vegetation type, leaf area index, photosynthetically active radiation and temperature. For some components a lower deposition velocity is used for snow covered surfaces and/or at temperatures below 273K. The maximum surface deposition velocities are given in Table A1.

The surface characteristics are important in determining the turbulence in the atmospheric surface layer and the surface resistances for different compounds. In this study we have used the land-use information available in the HIRLAM model (Bringfelt, 1996). Currently the dry deposition model differentiates between water surfaces, forested surfaces, low-vegetation land and no-vegetation land. Information about the fraction of each of these surface types is available for each grid square. The forest cover is taken from The Remote Sensing Forest Map of Europe (ESA, 1992). Information about the dominating types of forests, low vegetation and soil is derived from the land-use data set of Henderson-Sellers et al. (1986).

Table A1: Removal parameters employed in the model. Maximum 1-m dry deposition velocities to different surfaces (cm s^{-1}) and wet scavenging coefficients (s^{-1} mm^{-1} hour)

Component	Dry deposition						Wet deposition
	low vegetation day	low vegetation night	sea	forest day	forest night	snow	scavenging coefficient
O_3 (Apr)	0.7	0.3	0.05	0.6	0.3	0.05	*
O_3 (May)	0.7	0.3	0.05	0.7	0.3	0.05	*
O_3 (Jun)	0.7	0.3	0.05	0.8	0.3	0.05	*
O_3 (Jul)	0.8	0.3	0.05	0.8	0.3	0.05	*
O_3 (Aug-Sep)	0.7	0.3	0.05	0.8	0.3	0.05	*
H_2O_2	1.0	1.0	0.6	1.0	1.0		*
NO_2	0.4	0.1	0	0.6	0.2	0.1	0
HNO_3	4.0	4.0	1.0	5.0	5.0		3.89e-4
NO_3	2.0	2.0	0.5	2.0	2.0		3.89e-4
N_2O_5	2.0	2.0	0.5	2.0	2.0		3.89e-4
NITRATE	0.1	0.1	0.05	0.5	0.5		2.78e-4
PAN	0.25	0.05	0	0.25	0.05		0
ONIT	0.1	0.1	0.1	0.1	0.1		0
HCHO	0.2	0.2	0.2	0.2	0.2		1.4e-5
CH_3CHO	0.1	0.1	0.1	0.1	0.1		0
$CH_3COC_2H_5$	0.1	0.1	0.1	0.1	0.1		0
METHYLGLYOXAL	0.05	0.05	0	0.05	0.05		0
GLYOXAL	0.1	0.1	0	0.1	0.1		0
CH_3OH	0.6	0.1	0.5	0.6	0.1		3.89e-4
C_2H_5OH	0.6	0.1	0.5	0.6	0.1		3.89e-4
CH_3OOH	0.1	0.1	0.1	0.1	0.1		1.e-5
C_2H_5OOH	0.1	0.1	0.1	0.1	0.1		0
OXYO2H	0.1	0.1	0.1	0.1	0.1		0
MALO2H	0.1	0.1	0.1	0.1	0.1		0
SO_2 (Apr-May)	0.8	0.3	0.5	1.3	0.6	0.06	*
SO_2 (Jun)	0.8	0.3	0.5	1.5	0.7	0.06	*
SO_2 (Jul)	0.8	0.3	0.5	1.7	0.7	0.06	*
SO_2 (Aug-Sep)	0.8	0.3	0.5	1.8	0.8	0.06	*
SULFATE	0.1	0.1	0.05	0.5	0.5		*
NH3 (Apr-May)	1.5	1.5	0.6	1.5	1.5	0.4	3.89e-4
NH3 (Jun-Aug)	1.0	1.0	0.6	1.0	1.0	0.4	3.89e-4
NH3 (Sep)	1.2	1.2	0.6	1.2	1.2	0.4	3.89e-4

A2.4 Wet scavenging

For ozone, H_2O_2 and SO_2 in-cloud scavenging is calculated by assuming Henry's law equilibrium in the clouds; sub-cloud scavenging is neglected. For sulfate particles, in-cloud scavenging is assumed to be 100% effective, i.e., all sulfate particles within the precipitating clouds are deposited. Sub-cloud scavenging for sulfate particles is calculated as in Berge (1993).

Wet scavenging for all *other* species is assumed to be proportional to the precipitation intensity and a species-specific scavenging coefficient:

$$\frac{dc_i}{dt} = -c_i \Lambda_i P \qquad (2)$$

where c_i is the concentration of species i, Λ_i is the scavenging coefficient given in s^{-1} mm^{-1} hour and P is the precipitation rate in mm $hour^{-1}$. The scavenging coefficients employed in this study are given in Table A1.

A2.5 Radiation

Estimates of radiation are needed in the calculation of photolysis rates and in the calculation of biogenic emissions. A relatively simple model has been used to estimate global radiation and photosynthetically active radiation (PAR), using model calculated total cloud cover from HIRLAM as the main input.

The rates for photolytical reactions in the lower troposphere depends on a number of factors, the most important being the solar elevation, the presence of clouds, the surface albedo and the vertical distribution of gases absorbing at the wave lengths for which the photolytic reaction in question can take place. On-line calculation of the photolysis rates is rather computationally demanding and for the present study a simplified approach has been used.

Expressions for the photolysis rates depending on solar elevation derived for clear sky situations were taken from Derwent and Jenkin (1990). To account for the effect of clouds the photolysis rates given by Derwent and Jenkin were scaled by the ratio of the actual global radiation (corrected for clouds) to the clear sky global radiation. This ratio was estimated using a simple analytical expression for the global radiation, based on measurements in Denmark, (Nielsen et al., 1981). The expressions for the photolysis are calculated for a given ozone column, but we have used them for all levels and independent of the actual ozone column.

A2.6 Emissions and boundary conditions

The basic version of the MATCH transport model includes modules for inclusion of area emissions of the simulated species. Emissions can be introduced at any height in the model and at different heights simultaneously. Emissions are initially distributed in the vertical based on a Gaussian plume formulation (Berkowicz et al., 1986), evaluated at a downwind distance of $x=u_h \Delta t$, where u_h is the wind speed at the effective plume height.

If desired, standard plume-rise calculations can be performed (Berkowicz et al., 1986), based on stack parameters (stack diameter, effluent temperature and

volume flux) that are given as input to the model. It is also possible to specify temporal variations in the emissions over the diurnal time scale as well as variations between days. The emissions that enter the model calculations are updated every hour to account for temporal variations and the influence of the stability on the plume rise and initial vertical spread calculations.

A2.6.1 Anthropogenic emissions

Anthropogenic emissions for the simulations carried out within this project were derived from the 50 × 50 km^2 emission data provided by EMEP MSC-W at the Norwegian Meteorological Institute. The EMEP emission data are divided into emissions below and above 100m. The emissions for 1999 and 1990 for NO_x, SO_2, nonmethane volatile organic compounds (NMVOC), CO and NH_3 were used in the model calculations.

Simple variations of the emissions with the time of day and with the day of the week were used for NO_x, NMVOC, CO and SO_2. 75% of the low-level emissions were assumed to occur during the hours 0600–1800 (UTC). For the high-level (>100m) emissions the variation between daytime and night was smaller, 60% during 0600-1800. The weekly low-level emissions were divided between days as: Monday–Friday 15%/day, Saturday 13% and Sunday 12%. The corresponding figures for high-level emissions were: Monday–Friday 14.4%/day, Saturday 14.1% and Sunday 13.8%.

The emission of anthropogenic hydrocarbons was split on the components used in the chemical scheme, using data from the UK (Derwent and Jenkin, 1991). The resulting split is given in Table A2. The same VOC emission profile was used in all grid points.

Table A2: Model split of hydrocarbon emissions.

Component	Mass%
C_2H_4	3.6
C_2H_6	6.7
C_3H_6	3.8
n-C_4H_{10}	37.1
o-Xylene	24.5
CH_3OH	1.0
C_2H_5OH	14.0
HCHO	0.9
CH_3CHO	0.1
$CH_3COC_2H_5$	3.3
unreactive	4.9

A2.6.2 Biogenic emissions

Biogenic emissions of isoprene (C_5H_8) were estimated using the E-94 isoprene emission methodology proposed by Simpson et al. (1995). The emission rate, *ER*, is given as,

$$ER = \sum_{j=1}^{m} \left[A_j \cdot AEF_j \cdot ECF\left(PAR, T\right) \right] \quad (3)$$

where m is the number of vegetation categories, A_j is the area of vegetation category j, AEF_j is the area-based emission factor for vegetation category j and *ECF(PAR,T)* is a unitless environmental correction factor representing the effects of temperature and solar radiation on emissions. Following Simpson et al. (1995) five vegetation categories are used: Oak, Other broadleaf, Spruce, Other coniferous and Crop. The distribution of these five categories over Europe is derived by combining the information about coverage of forest and low-vegetation land from HIRLAM and the information on a national basis given by Simpson et al. (1995). The emission calculation was included in the model using the two-meter temperature available from HIRLAM and *PAR* calculated as described in section A2.5. The emissions were updated hourly based on current values of *T* and *PAR*.

A2.6.3 Initial and boundary concentrations

For some components in the chemical mechanism it is necessary to specify mixing ratios on the boundaries. In the present study the boundary conditions were treated in a rather simplified way. For each boundary (the four sides and the top of the model domain) a concentration (c_{north}, c_{east}, c_{south}, c_{west} and c_{top}) was assigned for each of the components. c_{top} represents the concentration at the top *surface* boundary, while the four lateral boundary concentrations represent the ground level concentrations at the *midpoints* of the four sides. Linear interpolation is used to get the boundary values between these points.

The boundary concentrations were as far as possible based on measurements of the various components at sites which were considered representative for the model boundaries. Due to lack of observational data for many of the components the boundary values are in many cases fairly crude estimates. The situation is especially uncertain for the eastern and southern boundaries, where suitable measurements are scarce. This is also true for the top boundary.

Different boundary values were used for different months for some components, due to seasonal variability in the background concentration. In Table A3 a few of the boundary concentrations used for the year 2000 are given.

Table A3. Some boundary concentrations used in the model runs for the year 2000. Unit ppb(v).

Component	ctop	cwest	ceast	csouth	cnorth
O_3 (Apr)	64	43	40	40	31
O_3 (May)	64	41	40	40	29
O_3 (Jun)	64	35	40	40	25
O_3 (Jul)	62	31	40	40	24
O_3 (Aug)	62	31	40	40	24
O_3 (Sep)	62	34	40	40	29
NO_y(N) (Apr)	0.4	0.9	1.6	0.6	0.5
NO_y(N) (May)	0.4	0.8	1.6	0.6	0.4
NO_y(N) (Jun)	0.3	0.7	1.5	0.6	0.3
NO_y(N) (Jul)	0.3	0.6	1.5	0.6	0.2
NO_y(N) (Aug)	0.3	0.6	1.5	0.6	0.3
NO_y(N) (Sep)	0.3	0.7	1.6	0.6	0.3
NMVOC(C) (Apr)	17	19	20	6	19
NMVOC(C) (May)	10	12	15	6	11
NMVOC(C) (Jun)	7	9	15	5	8
NMVOC(C) (Jul)	6	6	15	4	7
NMVOC(C) (Aug)	7	7	15	4	8
NMVOC(C) (Sep)	8	9	15	4	9
CO (Apr)	130	160	190	130	170
CO (May)	110	140	160	110	140
CO (Jun)	91	110	110	91	110
CO (Jul)	80	95	85	80	87
CO (Aug)	82	100	92	82	89
CO (Sep)	85	110	110	85	100

A2.7 Chemistry

The gas-phase chemical mechanism used is mainly based on the EMEP MSC-W model chemistry (Simpson et al., 1993). The main difference is that for the isoprene chemistry an adapted version of the so-called Carter 1-product mechanism (Carter, 1996) has been used instead of the EMEP mechanism (Langner et al., 1998b). A key feature of the chemical scheme is that a simplified mixture of a dozen *representative compounds* is used to model the many different organic molecules emitted to the atmosphere. The model compounds are chosen to span the normal range of ozone creation potentials for the most important organic pollutants (Pleijel et al., 1996). The photochemical model includes ca. 130 thermal and photochemical reactions between 59 chemical components and it is designed to provide a good description of the chemistry for both high and low NO_x conditions.

In addition to the gas-phase photochemistry, the reduced nitrogen chemistry from the EMEP acid deposition model is included, as well as aqueous-phase oxidation of SO_2 by ozone and H_2O_2 in cloud water.

Standard numerical integration techniques following the work by Verver et al. (1996) are used to integrate the chemical mechanism. This leads to stable integrations, where the accuracy of the calculations can be controlled. We have used the Kinetics Pre-Processor (KPP) developed at the University of Iowa. The use of a standard solver coupled with KPP makes it easy to change the chemical mechanism without having to recode the solver.

A2.8 Meteorological data

Meteorological data were taken from archived output of the operational HIRLAM model at SMHI. A selection of fields for a sub-area of the operational grid was archived specially for the purpose of dispersion modelling. The fields available are listed in Table A4. Initialized analysis, three- and six-hour forecasts for every six hours were archived. Precipitation and cloud cover were taken from the three- and six-hour forecasts. The three-hour forecast was also used to get wind fields with three-hourly resolution.

The horizontal resolution was approximately 44 km on a rotated latitude longitude grid. The vertical resolution was 31 levels. The wind field at all levels was used together with the tendency of the surface pressure to achieve a balanced wind field as described in Robertson et al. (1996). In the transport calculation, however, fewer levels were used, in order to reduce computing and storage requirements. 16 levels were used for 1999 and 12 levels for 2000. The average heights of the 16 lowest model levels are given in Table A5.

Table A4: Meteorological fields available from HIRLAM.

Field	Note
Temperature	31 model levels
Specific humidity	31 model levels
Horizontal wind components	31 model levels
Cloud water content	31 model levels
Cloud cover	31 model levels
Mean sea level pressure	
Surface pressure	
Two-meter temperature	
Surface temperature	
Large scale precipitation	Accumulated for three- and six-hour forecasts
Convective precipitation	Accumulated for three- and six-hour forecasts
Sensible heat flux	
Latent heat flux	
Soil moisture	
Ice concentration	
Albedo	
Total cloud cover	
Snow depth	

Table A5: Average height of the model levels and thickness of the corresponding model layers for HIRLAM in 31 level configuration as used in the model calculations. Only the 16 lowest levels are shown for brevity. Unit: m.

Level/Layer	height (m)	thickness (m)
1	30	60
2	150	160
3	350	240
4	620	300
5	950	360
6	1330	400
7	1750	440
8	2200	470
9	2690	500
10	3210	530
11	3750	550
12	4310	580
13	4900	600
14	5520	630
15	6150	650
16	6810	670

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