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Trends of ozone and precursors in Europe
status report TOR-2, Task Group 1

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Summary

Tropospheric Ozone Research (TOR-2) is one of the projects in the second EUROTRAC framework. The overall aim of TOR-2 is to quantify crucial processes in the atmosphere in order to improve the scientific background for the development of effect-based control strategies for photochemical oxidants over Europe. The work is organised in three task groups with the following themes: 1) evaluation of trends in relation to emission changes; 2) investigation of the exchange of ozone between the atmospheric boundary layer and the free troposphere; and 3) analysis of the temporal and spatial scales of the processes underlying the seasonal cycles of ozone. The project started in 1998 and will end at the end of 2002.

This status report presents an overview of trends of ozone and precursors. The information is taken from data and results provided by TOR-2 participants, but also from sources outside TOR-2. The intention of this report is to describe the status of our knowledge on trends, to identify gaps and weaknesses, and to recommend activities for the last phase of TOR-2. It is hoped that this status report, which can be seen as preparatory to the final report next year, will help increasing the transfer of science results of the project into the larger EUROTRAC body and that it will stimulate discussions with the policy community in Europe.

In North-western Europe and Alpine Europe, long-term measurements of NO_x and VOC in urban areas, where traffic is the dominant source of emissions, demonstrate that a substantial reduction of traffic emissions has been achieved. The decline of traffic emissions has started already in the mid to late 1990s and became gradually apparent during the 1990s. The reductions over the last 10-13 years are in the order 40% for NO_x and somewhat more for VOCs. In the majority of cases the timing and rate of the decrease is in excellent agreement with the traffic emission inventory data.
Non-urban data and data from moderately polluted places, which can be regarded as more representative of all sources, show in majority substantial decreases and usually in good agreement with the national emission inventory data and the data from neighbouring countries. National reductions seem somewhat less than the traffic reductions, suggesting that the other main source category is trailing compared to traffic. However, there are a few sites where no significant trends are reported. The reasons for this seemingly discrepancy are not clear yet.
At remote sites in this part of the continent the trends in precursor emissions are very small. The meteorological variability and the background are potentially very important contributors to trends, and much work need to be done to separate the contributions.

In Eastern Europe several rural sites report a decrease in NO_2 concentrations, a decrease that usually started near 1991 and coincides with the economical crisis that

struck Eastern Europe at that time. A similar development is seen in the time series of hydrocarbons in K-Puszta, Hungary. The timing of the changes in the observations seems in good agreement with data from other sources.

In Southern Europe a downward trend of NO_x is reported from a site in central Athens over the 90-97 period. It is not known (yet) how this corresponds to emission developments in this area. There is a strong need of much more information on trends in Southern Europe. It is felt that much of this information is somewhere available but that it is not obvious how to retrieve it.

There is very strong evidence that the annual maximum concentrations and the 98th-percentiles of ozone have been reduced over the last ten years. Most of the sites report downward trends, and often at a substantial rate of 1-2% per year. Upward trends are not observed at none of the site. A few of the trends are based on regression methods that remove the meteorological variability from the records, which strongly suggest that the reduction of emissions is responsible for the downward tendencies.
The concurrence of the emission reductions with the reduction of maximum ozone concentrations, which are predominantly determined by European emissions, is a strong indication that there is an effect visible as a result of past ozone policy, at least qualitatively. Whether the effects are quantitatively in line with the expectations needs to be seen. It is advocated to perform model sensitivity runs with various model and following a set of prescribed emission reductions to see if the modelled responses are in line with the observed ozone reductions.

There are strong indications that the background concentrations of ozone have increased over the last 10-13 years. At sites in Ireland and Scotland ozone concentrations in wind sectors receiving air masses from the Atlantic and Polar areas show upward trends. Also in Sweden and Finland positive tendencies of ozone are seen in the least polluted classes and wind sectors. Elevated sites in the Alps (Jungfraujoch, Zugspitze) have upward trends, in particularly during the winter. In the winter period these sites are predominantly affected by the free troposphere, and because of the increased lifetime of ozone and the reduced intensity of the photochemistry, advection constitutes an important term in the ozone budget. However, upward trends are not confirmed by all sites. Downward trends in the Atlantic sector are seen at Yarner Wood in the United Kingdom. Annual mean ozone concentrations at 4-5 km altitude have stabilised during the 1990s in Uccle, Belgium. Strong downward trends are observed at Kislovodsk, in the Causussian mountains in Russia. It is not clear yet whether there is conflicting information on the background, whether background trends may differ for different parts of Europe, or that there are important local disturbances in the data selected as background. An evaluation by means of 10-days back trajectories which is foreseen for the last TOR-2 phase could shed more light on this matter. Furthermore, it is recommended to calculate the effects of emission changes on a worldwide scale by means of 3D global dispersion models.

Rural and non-urban sites in interior Europe report predominantly upward trends for the mean ozone concentrations, in particular during the winter, but also often in the summer. In polluted places there is certainly an important contribution from a reduced titration by NO as a result of decreasing NO_x levels. There is likely also a contribution from an increase of the background concentrations for which there is much evidence that this has occurred. Contributions from other mechanisms, such as changes in the chemistry (other than a shift in the photostationary state) cannot be excluded either.

High and consistent quality of data over a long period of time is essential in establishing proper and accurate trends. Despite quality checks performed at the institutes and despite various pan-European intercalibration efforts detailed evaluation show that part of the data is affected by artefacts, which in a few cases could be linked to changes in monitors and in calibration. Data from the regular monitoring networks can be useful for a trend study, but it requires a thorough and extensive screening. Comparison with data from other data-providers has proven to be very instrumental in detecting erroneous data. Furthermore, it has become clear that documentation of the history of the instruments, and of the calibration and sampling procedures is in general poor. The "human data discs" appeared to be an indispensable source of information, more than official institute documents.

Growing interest in trends and supported by the continuation of long-term measurements have resulted in a promising large number of trend studies, both for ozone and precursors. The available information for trend assessment is now much larger than 6 years ago at the end of TOR-1. The on-going activities in TOR-2 and elsewhere suggest that a thorough evaluation of past ozone policy is possible in the next 1-3 years. To be successful, it is necessary to make a stronger link to the model community and to specifically address regions (South and East) from which more information needs to be retrieved.

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1. Introduction

Our knowledge of the fate of atmospheric constituents is based on various sources of information. Process studies in the field and in the laboratory have increased our insight in the interactions that play a role in chemical transformation, dispersion, emissions and deposition under specified and sometimes controlled conditions. The regular monitoring networks provide a European broad picture of the spatial and temporal variations of ozone and precursors under a wide range of different climatological conditions. The collection of statistical data on land-use, population density, emission factors, industrial turnover is essential in determining the emissions in Europe and specified to sources, geographical areas and temporal variations.
The mainstream of information from these disciplines is taken together and incorporated into chemical dispersion models that aim primarily at linking the emissions to the observations. The increase in our knowledge of dispersion characteristics and the increase in computer resources have lead to a gradual but steady increase in the ability of models to resolve smaller scale processes, to address a more comprehensive set of chemistry, and to simulate longer time intervals following analysed meteorological fields. All this together suggests that our ability in terms of tools and perception has increased to a level that we may expose ourselves to intriguing challenges such as a quantitative evaluation of historical emission changes.
This is not only a scientific challenge, it also bears a relevant link to policy makers: looking back to evaluate the success (or lack of success) of past policy in order to learn for future policy. Since the start of the regular network monitoring of ozone in the mid-seventies Europe has faced various economical developments that were not the same for the different geographical and political entities in Europe. These developments offer a good opportunity to test our ability in linking and quantifying the changes in emissions to the changes in ozone concentrations.

This is the central theme in Task Group1. In order to answer the main question: "is there an effect on ozone concentrations visible due to European anthropogenic emission changes" various complex steps need to be taken. Figure 1 shows the four basic blocks of information: 1) observations of ozone and other secondary species, 2) observations of precursor concentrations, 3) emission information, and 4) chemical dispersion tools.

There are sources of "contamination" that might affect a possible trend in the data records. The data needs to be examined for being representative for a larger region, and the quality of the data should permit a trend analysis.
Ozone and precursors concentrations are subject to variations acting on different time scales ranging from diurnal variations to variations on a synoptical time scale to an annual variation. Beyond the annual time scale there are variations due to variations and long-term changes in the atmospheric circulation. The long-term and short-term meteorological variability should therefore be accounted for in any sort of ozone and precursor regression. The effect will be a reduction of the margins of

uncertainty in the trend estimate (removing the short-term variability) and a possible adjustment of the trend by removing the trend component caused by the long-term variation in the meteorological conditions.

Precursors time series, which have been subjected to data quality checks and for which regression studies have included meteorological variability, are fit for comparison with emission inventory time series. It requires that the inventory be compiled in consistent fashion over the years, that definitions of source categories, species (i.e.: NMVOC) have not changed. The main objective of the precursor comparison is to have an independent check on trends in the emissions. The findings in this line will help in the evaluation of ozone trends.

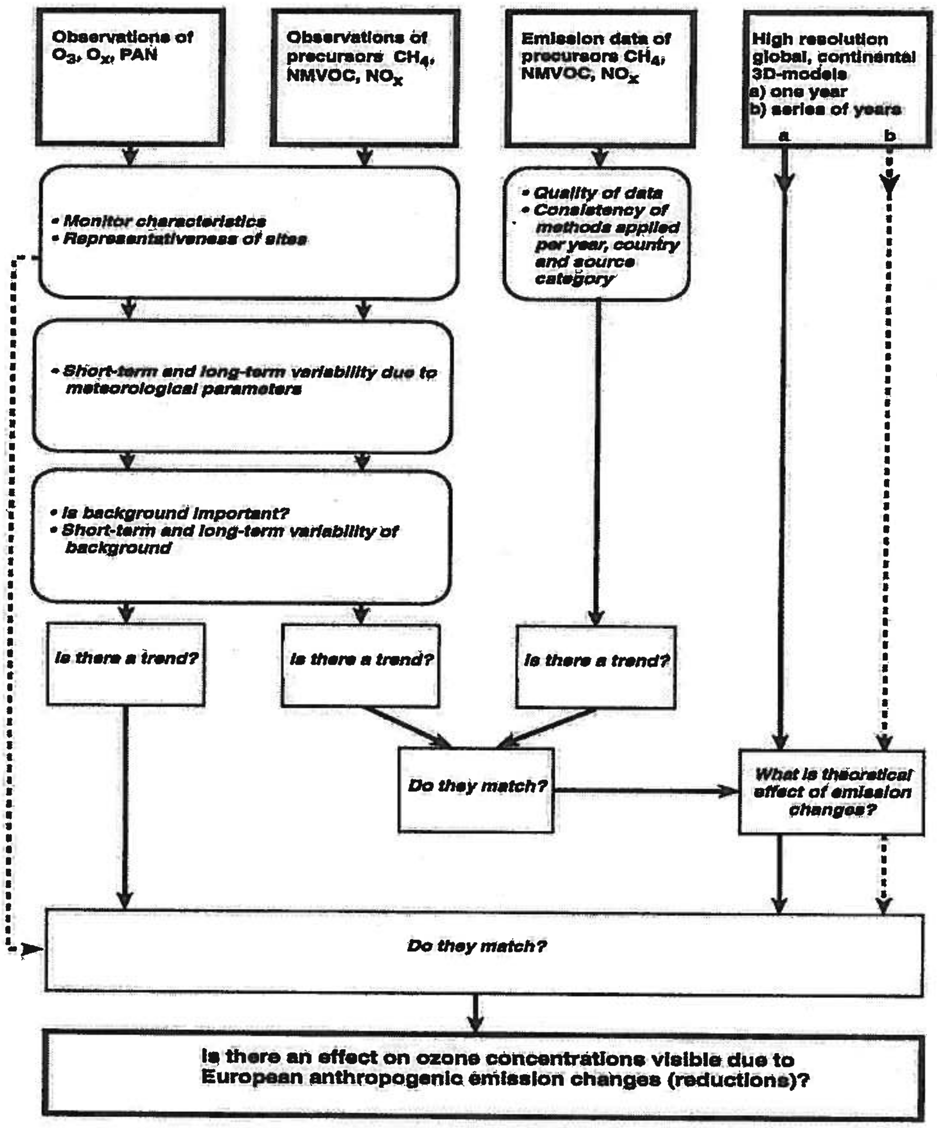

Figure 1 An overview of objective, data, tools and issues in relation to trend research

If emission trends are confirmed by trends in precursor concentrations, the emission trends can be fed into the chemical dispersion models. Objective of this step is to investigate what the effect of the observed historical emission developments is on ozone, at least according to our theoretical tools. The pre-final step is then to piece together the different streams of trend information. On one side is a set of ozone trends according to the observations, screened for data quality. By taking into account meteorological variability in the regression the remaining trends is void from possible long-term and short-term variations therein. By doing the analysis for a set of stations in different parts of Europe, by selecting subsets of data for background and polluted conditions, by selecting seasons and different segments of the ozone distribution (averages, high-percentiles) it is possible to distinguish between the European and non-European (background) contributions to a trend. The non-European trend could be confronted with modelled trends from global models. The European trend component in the observations should be confronted with trends based on the European scale models fed by the aforementioned analysed emission developments. The latter is the ultimate test of our ability to describe the processes determining the ozone budget. By using a set of models ranging in complexity and characteristics a picture will emerge as to what level of detail is required to properly describe the ozone budget over Europe. It is expected that such "confrontations" will address the sensitivity in trend description to spatial resolution, to NO_x/VOC ratios, to VOC speciation, to chemistry, to vertical exchange processes, and other elements.

The final objective will be to assess what we can expect from ozone policy ahead of us, whether the planned emission reduction will meet the targets, or that adjustments need to be considered.

2. Workplan in TOR-2 Task Group 1

The approach depicted in the Introduction is a very ambitious one, which requires combining resources from a large number of research groups for various years. The intention in task group 1 is to address all items as described in Figure 1 with the objective and ambition to:

1. present an overview of what we know on trends with specified degrees of confidence,
2. to identify gaps and deficiencies in our knowledge concerning trends,
3. to offer recommendations for future trend research.

From the start of the project it was clear that it was unlikely to answer the main question: "is there an effect on ozone concentrations visible due to European anthropogenic emission changes" within the time frame of TOR-2. However, it was felt essential to attempt an all-encompassing approach in order to see how much short we are of answering that question.

The main strength of a network of research groups such as in TOR-2 is to obtain insights in the developments of ozone and precursors over large areas in Europe by performing joint activities. Furthermore, it is clear that the sources of information would come not only from the TOR-2 community but from other groups and networks as well. An inherent weak element is the lack of central funding. Contributions from groups disappear or become less due to funding problems.

In the first place TOR-2 is a data oriented group. The majority of research groups run one or more sites in their countries. Continuation of measurements and analysis of data is an essential element in the research. There are also groups (and TOR-2 serving projects) that develop and apply tools for data analysis, as well as groups with dispersion models and modelling expertise. The role of models in the analysis (Fig. 1) is recognised as important. The aim is to address the model aspects in the last phase of TOR-2. The fact that this takes place in the final phase of the project is due mainly to funding opportunities for the groups involved.

During the evolution of TOR-2 the interpretation activities undertaken in task group 1 could be categorised in one of the following clusters:

1. data quality aspects
2. methods to account for (meteorological) variability
3. ozone trend analysis
4. precursor trends and the relation with emission trends
5. ozone trends in relation to emission trends and in relation to model results.

The logical order of activities would be from cluster one to five. In practice however, things work differently. With simple means ozone regression analyses can be performed, while the awareness concerning cluster 1 and 2 had to develop during the project. At the end of the year 2000 most of the PIs were involved in the clusters 1-3. Less has been done so far on cluster 4, and cluster 5 has still to begin.

This overview starts with a short section on data quality aspects emerging from TOR-2 studies. In the next two chapters an overview of recent studies on ozone and precursors trends will be presented. The status of our present trend knowledge will be given, together with suggestion for the years ahead. Annex A lists the data sets and tools accomplished in TOR-2 (trend section). The list of Principal Investigators in TOR-2 is given in Annex B. Annex C outlines the invitation to the model community.

3. Data quality

Trends, especially trends of ozone that are small, require high and consistent quality of data over a large span of time. In the regular networks as well as at dedicated scientific sites quality procedures are adopted to ensure that only technically valid data are stored in the database. The regular networks provide the core of ozone and precursor trend information.

A study by Low et al. (1992) indicated trends of opposite sign at coastal and near coastal sites in the UK, Netherlands and Germany. Large differences in trends, and even opposite trends were also found in a study comparing nearby (< 300 km) EMEP-sites in Scandinavia, Finland, Germany and The Netherlands (Roemer, 1997). The time series of differences of pairs revealed remarkable fluctuations that could not be ascribed to differences in advection or local emissions.

A similar study is started to compare three sites in the northern tip of Europe: Jergul/Krasnajok in Norway, Esrange in Sweden and Pallas in Finland, which are within a distance of about 200 km in an almost pristine environment. The average ozone concentrations of the three sites over the entire length (10 years) of the time series are very similar. Occasionally, differences in all three combinations of up to 10-20 ppb can be observed. These differences usually last for a few days or less, and are likely to be caused by meteorological conditions. Of more concern is the fact that the averaged ozone concentrations at two sites differ approximately 5 ppb over a period of three year, while during the other years the difference is negligible. The combination with the third site has not produced conclusive evidence of one site being flawed.

Ozone and NO_x data at 35 non-urban sites and from 5 data-providers in The Netherlands, Belgium and Germany were evaluated on data quality issues and on trends (Roemer, 2001). Comparing time series of data from different data-providers revealed various discontinuities and drifts. In one case it could be demonstrated that a change in monitors and in calibration had caused a serious discontinuity of the ozone time series. The information came from the data, the link with the history of the instrument required interviewing various technicians of the laboratory, and by confrontation with evidence from the data comparison. Much of the essential information on the history of the instruments and calibration was stored at "human data discs" and only accessible by employing Sherlock Holmes type of detective work. Other cases of suspect data await information from other laboratories.
In the case of NO and NO_2 the method of comparing nearby sites has not yet been applied. Linear regression applied on subsets of the data (low and high percentiles; winter and summer; daytime and night-time) revealed remarkable differences in trends under relatively clean conditions. A closer inspection of the data revealed that all five data-providers have other methods of the processing of near zero values of NO and NO_2, which will produce systematic differences between neighbour-

ing countries at the low tail of the concentration distribution. More importantly however, is that methods are not constant in time, and likely to be of effect when trends are calculated under relatively unpolluted conditions.

From these studies it is concluded that data provided by the regular monitoring networks can only be used for trend studies after a thorough and extensive screening. Despite quality checks performed at the institutes (and often also at EMEP-CCC) the data still contain discontinuities, drifts and offsets that cannot be tolerated in a trend analysis. This is not to say that they are useless, but that substantial effort is asked to create subsets that are reliable for a trend study.
Part of the suspicious data could have been detected by simple means such as visual inspection. The fact those data slipped through may point towards deficiencies in the quality control at the institutes. The other part is more difficult to detect. Only by working with the data, by comparing with data from other institutes, by performing more sophisticated analyses than calculating averages and percentiles, the true nature and status of the data becomes clear. At that point the institutes responsible for the networks often fail the necessary resources and commitment.

4. Trends of ozone concentrations

The literature gives a large number of studies concerning trends of ozone. The methods involved are not the same, which does not necessarily pose a problem in the comparison of trends, but of a more serious problem is that it is not always clear what the status of the quality of the data is. Furthermore, the time intervals are usually not identical which can be of importance, especially in cases where regressions without meteorological adjustment are used. It is known that especially then the choice of the interval can have a strong influence on the magnitude and even the sign of the trend. This section will start with a brief overview of recent trend studies, followed by a discussion of what can be regarded as emerging from these studies.

4.1 Rural west and north Europe

The Mace Head data is split into a polluted and unpolluted sector (Simmonds et al., 1996; Simmonds et al., 2001). In the unpolluted sector, which can be regarded as western background at mid-latitudes, an upward trend for the monthly means of +0.5 ppb/yr is reported over the 1987-2000 period, equivalent to about +1.6%/yr. In the polluted (European) sector the trend in the monthly means is +0.4 ppb/yr, equivalent to +1.3%/yr. The variation in the monthly means is larger in the polluted sector, but over a span of 2-3 years both sectors show common features. A rapid increase in the 87-90 period is followed by a standstill (unpolluted) or even decrease (polluted) in the period 91-95. A notable increase is seen in the 98-00 period, the increase being more pronounced in the unpolluted sector. From the data it is obvious that background concentrations of ozone are increasing. The interpretation of comparing the two sectors is not entirely clear yet, but it suggests that the large scale (hemispheric) background is important in driving the changes on a time scale of a few years and more. The effect of Europe is certainly seen in the variations on a shorter time scale, but whether the relatively small difference in trends between polluted and unpolluted sector is a manifestation of European emission policy, or must be regarded as negligible, needs to be sorted out yet.

Trends at Mace Head, Strath Vaich and Yarner Wood over the period 88-98 were determined for one to three clean sectors (South, Atlantic and North) depending on location, and one polluted European sector (Coyle, 2001). Ozone data were allocated to sectors by means of three-dimensional 5-day back trajectories from the ECMWF. Upward trends in the annual means are seen at all sectors at the two cleanest sites (Mace Head, Strath Vaich). In contrast, Yarned Wood at the south-western tip of Cornwall produces downward trends in both the Atlantic and the European sector.
All three sites have in common that trends of the annual maxima are downward in the European sector ranging from close to –0.6 ppb/year for Mace Head and Strath

Vaich to –2.0 ppb at Yarner Wood, the latter being significant at the 95% confidence interval. The annual maxima in the clean sectors show little trend or an upward trend (Mace Head).

At Lille Valby, Denmark the ozone and oxidant data are allocated to one out of three sectors (east, west, north) by means of local wind direction (Skov et al., 2001). The west and east sectors are obviously affected by NO_x emissions and oxidant seems a more appropriate parameter at this site. The annual oxidant means show a gradual decrease over the 92-98 period, especially in the eastern (40-160) sector. The year 1999 however, displays the highest annual means of the time series. For the 98-percentile a remarkable strong downward trend is observed at all sectors over the 92-98 period, but including the year 1999 renders trends still downward but less spectacular. For the 98-percentile the trends over the entire period are still in the order of –2%/yr.

Five background sites in Sweden were used for trend analysis over the last 10 years (Lindskog, 2001; Lindskog and Kindbom, 2001). The annual means are upward for nearly all sites most notably in the winter and early spring. For a few sites and shorter time intervals sectors were defined. The first indications confirm upward trends or no trends both in the polluted and the unpolluted sectors.
From the Mann-Kendall test over the 90-98 period significant (at 75% confidence level) upward trends were found for the means in the winter. In the summer period only one southern site (Norra Kvill) produced downward trends, the rest showed no significant change. For the 98-percentiles the prevailing picture is upward in the winter, and downward in the summer at two southern sites.

The same Mann Kendall test has been applied for a number of Norwegian sites over the period 88-97 (TOR-2, 2001a). For the means and 50-percentile the predominant picture is a significant upward trend in the winter. In the summer the tendencies are mixed with negative, positive as well as no trends (significant at the 75% confidence level). For the 98-percentile the winter shows upward trends all over. For the summer more downward trends are seen, although a few sites report upward trends.

At a few Finnish sites the ozone data in the May-July period was sorted according to total nitrate, which was measured simultaneously (Laurila et al., 2001). The average ozone concentrations in the 89-95 period were compared to those in the 96-99 period for the different NO_3 classes. An increase of average ozone concentrations, equivalent to about +1.0%/yr, is observed at the lowest NO_3 classes representing the unpolluted air masses. In the more polluted air masses a smaller increase is seen. This finding is supported by the notion that sites in southern and central Finland report increasing AOT40 values over the period 89-99, although the scatter from year to year is very large.

4.2 High altitude Europe

The homogenised time series of ozone sondes at Uccle in Belgium was subjected to the Kolmogorov-Zurbenko filter to separate different time scales (de Backer et al., 2001). Free tropospheric ozone concentrations at 4 and 5 km show a stabilisation since 1990.

Since 1978 continuous measured are performed at the Zugspitze (2962 m) and Wank (1780 m). The sites are located in the German Alps very close to each other. Since 1994 a divergence in trends is seen with continuing increasing mean values at the Zugspitze and a slight decrease in Wank (Scheel, 2001). At the Zugspitze the seasonal cycle remains virtually unchanged during the 1990s, but at Wank the amplitude of the seasonal cycle diminishes with decreasing summer means and increasing winter means. As a possible explanation for the latter a reduced advection of polluted air during the winter is brought forward. Other phenomena that need examination are a possible change in the vertical exchange between the two altitudes, a reduction of ozone precursors (which has occurred) or changes in the circulation.

The Alpine research site at Jungfraujoch is located at an altitude of 3580 m and continuous measurements are carried out since 1986 (Schuepbach et al., 1999). Application of various tests to detect shifts and discontinuities in trends revealed a discontinuity in 1989 to be associated with a change of monitor, and one in 1991. The latter anomaly is also seen at a neighbouring highly elevated site of Davos (but not at Zugspitze). It is suggested that at least part of the anomaly is related to natural variability. A linear regression on the adjusted data (for the 1989 shift) shows for the 88-96 period upward trends for all monthly means, except for May. Significant trends are seen in the winter period.
Application of general state space approach to the monthly means of May for the 88-97 period showed significant downward trends (Schuepbach et al., 2001).

Kislovodsk HMS is located at 2070 m altitude in the Caucasus in Russia. The Mann-Kendall test showed statistically significant downward trends of the monthly means at a rate of 1.75 ± 0.4 % per year over the 89-00 period (Elansky et al., 2001). The annual means continuously decrease from 1991 to 1997, increases are observed over the first two years and the last two years. The trends in January are much smaller than in July where after 1995 a rapid decrease of concentrations is noted.

4.3 Polluted west and alpine Europe

Brönniman and Wanner (2001) have investigated ozone trends at thirteen urban, rural and elevated sites in Switzerland. At some stations, data from before 1991 cannot be used due to data quality problems.

With a non-linear regression model with 10 independent variables to account for meteorological variability and the seasonal cycle they were able to explain about 65-75% of the variance in ozone concentrations (average of the four highest half hourly values between 11.00h and 18.30h) during the April-September period. The model consists of a baseline trend and a radiation interaction trend, which allowed for more than one tendency term. A second approach consisted of separating the different time scales in the time series (by application of the Kolmogorov-Zurbenko filter $KZ_{15,5}$), followed by application of a linear regression model on the high-frequency part of the time series for summer (April-September) and winter (October-March). This method resulted in explained variances of 70-85%.
Daily maximum values under "fair weather" situations have decreased since the start of the 1990s at rural sites, but increased at urban sites. These changes are not due to trends in meteorological conditions. The mean value has increased at all sites, most notably at urban and elevated rural sites. An increasing baseline trend seems eminent at all sites, to which adds a local trend, which could be due to local and regional emissions. Including the (decreasing) precursor concentrations in the regression approach resulted in an improved description of the local trends, while the baseline trend remained more or less intact.

A rather similar study was conducted by Kuebler et al. (2001) for Swiss sites over the 87/89-98 period. As in Brönniman and Wanner (2001) the ozone time series were separated into different temporal scales. The low frequency component was removed by application of a $KZ_{365,3}$ filter. A linear regression model with meteorological variables was used to account for the short-term variations. The method was applied on the daily maximum ozone concentrations (mean of the four highest half hour values) in the May-September period. The explained variance is 73% for Dübendorf. As a result of their method they find no significant trend in the summer season 90th percentile of ozone. Urban sites indicate positive tendencies; at the Jungfraujoch no statistically significant trend was found.
It is not clear yet how to interpret the no-trend of the 90th percentile for rural sites below 1000 m as reported by Kuebler et al. (2001) in relation to the downward trends of peak values in rural areas shown by Brönniman and Wanner (2001). It is possible that the 90th percentile is too far off from the peak values, and influenced too much by processes that underlie the increase of averages (see also the German study in the next paragraph). A more detailed inspection of results could disclose causes for this seemingly discrepancy. Both studies agree with respect to increasing mean values.

A few hundred German sites were examined on trends by Beilke and Wallasch (2000). Over the period 1990-1999 the number of days that at least at one site in Germany the threshold values of 180 $\mu g.m^{-3}$ is exceeded has been reduced by as much as 80%. The reduction is even stronger for the 240 $\mu g.m^{-3}$ threshold with virtually no exceedances at the end of the 1990s. By describing the distribution of the ozone maxima as convolution of three distribution functions for ozone, NO and a

measurement error, they were able to relate the reduction in peak values to the decrease in emission precursors that has occurred in Germany.
While the 99th percentile of daily maximum ozone concentrations show a reduction in the order of 2% per year, the reduction has almost disappeared at the 90th percentile. At lower percentile classes upward trends are seen, increasingly becoming stronger at the lowest percentiles. The 25th percentile shows an annual increase of about 2.5%. The increase is attributed to a reduction in titration by NO.

In a study to trends in the Netherlands, Belgium and Germany (Roemer, 2001) data quality problems shortened the available period to after 1992. A Dutch and a German site (50 km apart) showed consisted trends in ozone and oxidant over the 93-98 period. A linear multiple regression model which incorporated local meteorological parameters, a seasonal cycle and two trend terms explained about 80% of the variance of the daily maximum ozone concentration (highest hourly value between 12.00 and 20.00h) in the May-September period.
Significant increases were seen in the 25th and 50th percentiles, whereas the 90th percentile dropped by as much as –2.7%/yr in Eibergen and –1.4%/yr in Wesel. Virtually no trend difference was seen at the 90th percentile between ozone and oxidant. For the two lower percentiles the trends in oxidant were considerably less upward than for ozone, indicating that an important part of the upward trend can be attributed to reduced titration by NO. Nevertheless, the remaining positive tendency of oxidant indicates that other factors play a role as well. This can be an increase of background ozone, or otherwise a change in the chemical composition affecting the chemistry over this still polluted region.

By means of linear regression, trends were established for nineteen sites in the United Kingdom and Ireland over the 88-98 period (Coyle, 2001). Most sites showed little to no changes in their annual means. Two sites had significant upward trends; Lough Navar, which is located in a forest, had a significant downward trend. The annual maxima went down at all sites, four of them being statistically significant at the 95% confidence interval.

4.4 South Europe

Patission is located in downtown Athens (Varotsos et al., 2001). Monthly mean values in January over the 87-98 period are upward. In April, July and October there is little change of the monthly means or they are slightly downwards (Varotsos et al., 2001).

4.5 East Europe

Preila is a coastal site at the Baltic Sea in Lithuania where ozone is being measured since 1981. Over the entire 81-99 period as well as over the shorter 89-99 period upward tendencies are observed for both the summer as the winter season. Splitting up to monthly means and performing the Mann-Kendall test over the 89-99 period, it is seen that downward tendencies (at a confidence interval better than 90%) are seen in June and August, and that all months in the October-May period show upward trends (Girgzdiene, 2001).

A gradual increase of mean ozone values are reported for K-puszta in Hungary (Haszpra et al., 2001). The annual trend over the 90-99 period is 0.36 ppb, equivalent to about 1% per year.

Puntijarka is a site at 980 m altitude in Croatia not far from Zagreb, where since 1989 continuous ozone measurements are carried out. The data show increases in the 12-months running mean until 1993/94, followed by a stabilisation or even a small decline (Butkovic et al., 2001). The war in Croatia, which resulted in a drastic decrease in industry and transportation, might have been of influence to the stop of increasing levels.

4.6 Summary

The previous sections describe the results of recent trend studies in Europe. Does it produce a coherent picture of ozone developments? Are there regions where ozone developments have gone in another direction, or is the information not conclusive, or even contradictory? To order trend study results it is important to keep in mind the objectives. There are two objectives: 1) is there a trend as a result of changing emissions in Europe, and 2) is there a trend in the background?
The first question will be addressed by looking at trends of peak values and high percentiles, but also by looking at the means and lower percentiles (Table 1). For sites at the rim of Europe sector splitting is very informative, for sites located more inward in the continent all sectors are to be used. Background information comes in the first place from sites at the rim with sectors facing outwards, and from high altitude sites and balloon soundings (Table 2).

Tables 1 and 2 are constructed by the information provided in the reports and sometimes based upon interpretation of graphs. The interpretation of graphs creates an element of uncertainty; also the significance of reported trends is not always clear. In this evaluation only rural and non-urban sites are discussed. The majority of sites span a period of about 10 years covering a large part of the 1990s, with sometimes a few years at the end of the 1980s. Shorter intervals are used at Lille-Valby and the Netherlands-German study.

Table 1 *Overview of trend results for rural and non-urban sites in Europe; trends apply to ozone unless stated differently.*

Site	Type	Peaks, 98th-P, 95th-P	90th-P	Averages and 50th-P	Averages and 50th-P	Averages and 50th-P
		summer or annual	summer or annual	summer	annual	winter
Europe						
Mace Head (Ire)	rural	--			+	
Strath Vaich (Sco)	rural	--			+	
Yarned Wood (Eng)	rural	--			--	
Norway South 5x	rural	--/0		0/+		+
Sweden South 3x	rural	--/0		--/0		+
Sweden North 2x	rural	0		0		+
Finland	rural			+		+
Lille Valby (Den)	non-urb.	--			-- (ox)	
UK 19x	rural	--/0			0/+	
Neth-Germ. 2x	non-urb.		--	+		
Germany (many)	all	--	0		+	
Switzerland	rural	--	0	+		
Preila (Lith.)	rural	--/0		+	+	+
K-puszta (Hun)	rural				+	
Puntijarka (Cro)	rural				0/+	
Switzerland	>1000	--		+		
Wank (Ger)	1780			--	0	+
Zugspitze (Ger)	2962	--/0		0/+	+	+
Jungfraujoch (Swi)	3580		0			+
Uccle (Bel)	4-5 km				0	
Kislovodsk (Rus)	2070			--	--/0	--

-- : downward trend; +: upward trend; 0: no trend; blank fields: not applicable or no information

There is very strong evidence that the peak values of ozone are decreasing. All available information shows downward trends, a few report no significant trends. Upward trends are absent.
There is very strong evidence that wintertime averages are increasing. All sites, except one, report positive tendencies. The exception is the elevated site at Kislovodsk. More detailed analyses are required to investigate whether this is due to different developments in the background (the site is located far to the east compared to the others), to the collapse of the Russian economy after 1991, or to other things.

The increase in wintertime means is to a certain extent reflected in the increase of annual means. Annual means are predominantly increasing, but there are also sites with no trend or a downward trend. A mixed picture is produced when summertime means are considered. Tendencies of both signs are seen, although more sites with positive than with negative tendency.

In polluted places the increase of ozone averages is attributed to the reduced titration by NO, but apart from that, there are indications that there is also a contribution from other processes.

Concerning changes in the background, ground level sites in Ireland, Scotland, Sweden and Finland report increases. Elevated sites like Zugspitze and Jungfraujoch indicate predominantly upward tendencies, especially in the winter, when the background prevails over chemistry. But: the soundings in Belgium at 4-5 km do not show a trend, and on the other end, the trend at Yarner Wood is downward in the clean sector. The indications for an increase of background are very strong but not conclusive yet.

Table 2 *Overview of trend results for rural sites facing the background of Europe; trends apply to ozone.*

Site	**Type**	**Peaks, 98th-P, 95th-P**	**90th-P**	**Averages and 50th-P**	**Averages and 50th-P**	**Averages and 50th-P**
		summer or annual	**summer or annual**	**summer**	**annual**	**winter**
Background-Europe						
Mace Head	rural				+	
Strath Vaich	rural				+	
Yarner Wood	rural				–	
Sweden	rural				0/+	
Finland	rural			+		
Zugspitze (Ger)	2962			0/+	+	+
Jungfraujoch (Swi)	3580					+
Uccle (Bel)	4-5 km				0	
Kislovodsk (Rus)	2070			–	–/0	–

– : downward trend; +: upward trend; 0: no trend; blank fields: not applicable or no information

5. Trends of precursor concentrations

This chapter will give a brief overview of the results of trend studies based on TOR data and on other data. The studies are divided into the two main groups of ozone precursors (NO_x and VOC) and to the type of data (rural and urban).
Most of the rural nitrogen data is as NO_2, urban data is often both in NO_2 and in NO_x. The definition of urban and rural is not very strict in terms of concentration levels and depends also on the local perception. In densely populated and small countries like The Netherlands and Belgium, it would be misleading to define many of their sites as rural, only because they are located outside the population centres. The concentration levels are high enough to classify them as semi-urban or non-urban instead of rural.
It was not always possible to discriminate between the terms VOC and NMHC. In cases where it was not clear the data are referred to as VOC.

5.1 NO_x and NO_2

5.1.1 Urban and semi-urban

Daily averaged NO_2 concentrations are measured at a large number of urban and regional sites in Sweden since 1986. The national winter-time (October-March) NO_2 averages are calculated as the average over 15 urban areas in Sweden. Figure 2 shows the temporal development of the national averages of NO_2 and the emissions of passenger cars, total traffic and the emissions in urban areas.

The national Swedish NO_x emissions from traffic and passenger cars are estimated to have been reduced by respectively 40 and 52% over the measurement period, which is good agreement with the 39% reduction in national average NO_2 burden.

In the United Kingdom emissions from road traffic, which have the greatest influence on ground level concentrations, increased steadily prior to 1989. There has been a decrease of 22% between 1989 and 1994 (PORG, 1997). Significant downward trends of NO_x concentrations over the same period are reported from all five sites that had, in 1994, at least five years of measurements. The trends are consistent with emission inventory data.
Non-urban NO_2 data from Lullington Heath and Ladybower show moderate to substantial downward trends in the 88-98 period (Coyle, 2001).

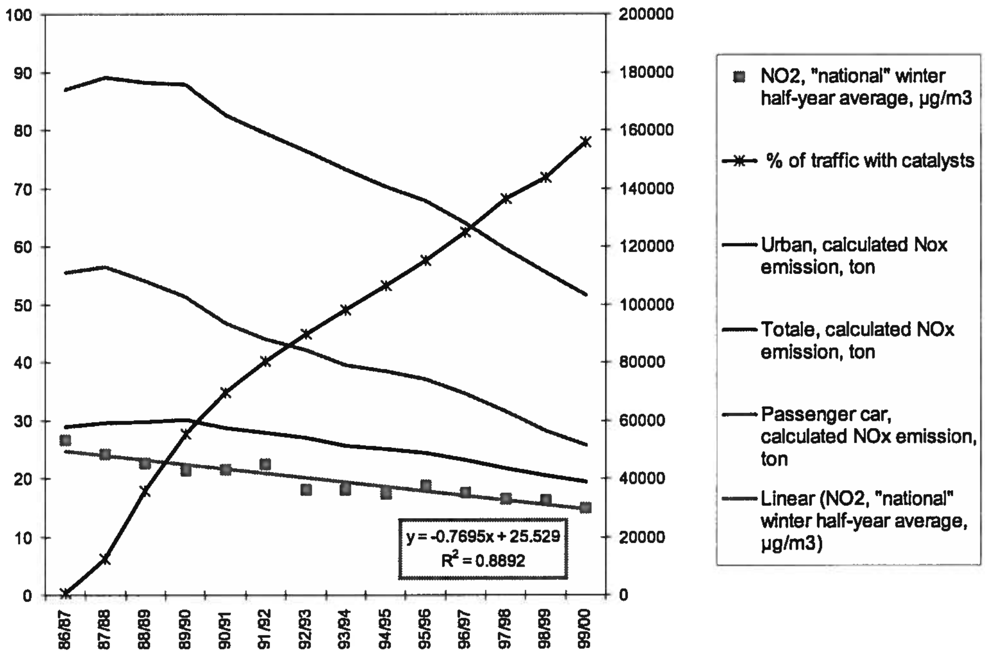

Figure 2 *Trends of Swedish national winter half-year averages of NO_2 ($\mu g.m^{-3}$), percentage of traffic with catalysts (left vertical axis), and urban emissions, national passenger car emissions and total traffic emissions (right axis).*

In The Netherlands 18 non-urban sites showed that in 1999 the average NO_x concentrations have dropped by about 30% compared to the early 1990s (Roemer, 2001). No trends were found during the 1980s. By comparing the data with model calculations which take into account the interannual variability in the meteorological conditions much of the variations from year to year could be ascribed to changes in the circulation. The timing and the rate of the NO_x concentration changes compared well with national emission inventory data. This study included also a few sites in the neighbouring German province Northrhine-Westphalia. It seems that at the German sites the reduction of NO_x started a few years earlier, thereby making the total reduction over the entire period somewhat larger than 30%.

The concentrations of NO_x measured at nine Netherlands urban sites, predominantly influenced by traffic, were adjusted for meteorological fluctuations and variations in background concentrations. Urban traffic emissions were calculated for the specific conditions (intensity, type of traffic) at these sites (Noordijk et al., 1999). There is an excellent agreement in trends between the concentrations and the urban traffic emissions (Figure 3). Both sources of information show a reduction in NO_x traffic emissions of 35-40% over the 88-97 period.

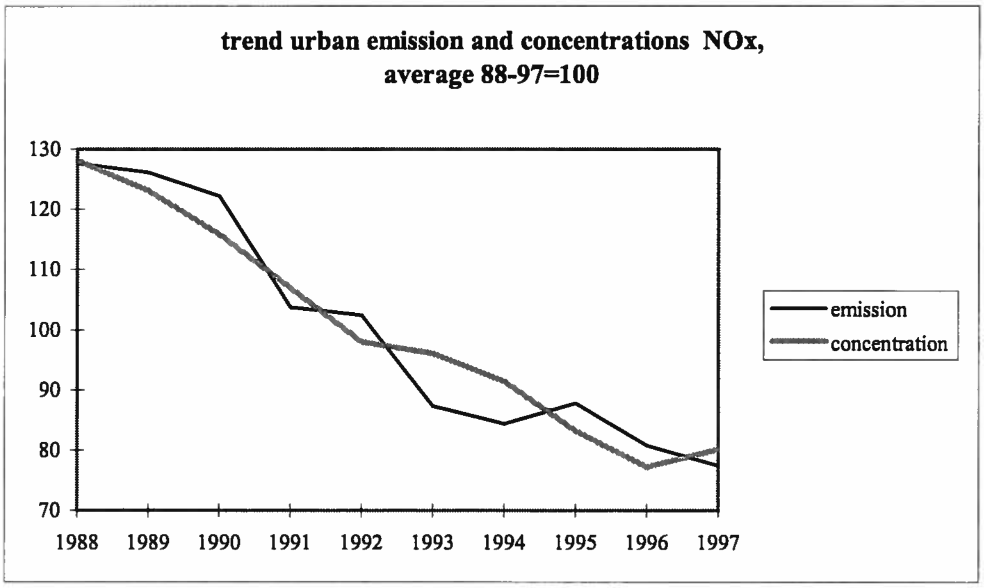

Figure 3 *Trends of urban traffic NO_x emissions and measured NO_x concentrations at nine urban sites in The Netherlands.*

The urban NO_x data in Switzerland report a clear downward trend of 50% in Zürich (86-98) and of 40% in Dübendorf (88-98) but a smaller downward trend of 13% in Lugano over the 89-98 period (Kuebler et al., 2001). According to the Swiss emission inventory (SAEFL, 1995) the national Swiss NO_x emission has dropped in the 85-98 period by –30%. For the two main source categories, traffic and industry the reductions are –39% and –14% respectively. The smaller source categories are residential (-23%) and agriculture (+19%). It is important to note that in these emission estimates the last few years are based on future projections.

At the Athenian site Patision a reduction of about 25% of NO concentrations is reported over the 90-97 period, and a smaller trend of NO_2 over the same period (Varotsos et al., 1999).

5.1.2 Rural

At Strath Vaich in the north of Scotland the annual NO_2 show little change over the 90-97 period, although a small downward tendency seems eminent (Coyle, 2001).

The five rural Swiss stations show little downward or no trend of NO_x over the period 88/91 to 98 (Kuebler et al., 2001).

An overview of long-term NO_2 measurements from the EMEP network is given in Barrett et al..(2000). A brief summary will be given below. NO_2 concentrations at the German north-sea coastal site Westerland show almost no change in the 85-98

period, in contrast to the Dutch coastal site Kollumerwaard which is located at a distance of about 300 km to the west. Other rural German NO_2 sites show a mixed picture with clear downward trends in Deuselbach and Waldhof, and no trends at the higher altitude sites Schauinsland and Brotjacklriegel in the southwest and southeast respectively. The national NO_x emission has been reduced substantially over the last 10-15 years, so it is not clear why the rural sites do not show a more coherent trend. Data quality issues are suggested by Barrett et al. (2000) as a possible explanation.
Another contrast of trends was seen in Scandinavia. Norwegian EMEP sites report a reduction in NO_2 concentration of about 35% (87-98), while the Swedish EMEP sites show no trends or even a slight upward trend over the same period. This is even more remarkable in view of the fact that the Norwegian NO_x emissions have hardly changed since the mid-1980s, but that the Swedish NO_x emissions dropped by almost 40%. Rural sites in both countries are strongly influenced by foreign emissions, most notably by German and UK emissions. Since EMEP sites are selected on the criterium of little local influence, it was unclear why the trends in the two countries diverge so much. Recent information, which was not incorporated in the Barret et.al. (2000) report, offers the explanation. In 1988-1989 a new method was introduced in Sweden. As a result of a re-examination of the two methods it became clear that the NO_2 measured by the former method was systematically too low compared to the new one The old data have been adjusted by multiplication with a factor of 1.44 (Sjöberg, 2001). As a result the NO_2 trends in the adjusted data in Sweden have become downwards with a decrease of more than 30% in the winter (86-99). In the summer the decrease is somewhat smaller. The developments are now in line with the emission trends. This example illustrates once more the importance of documentation and quantification of changes in the monitoring practices.
The Polish site Jarcew displays a decrease in NO_2 concentrations consistent with reported changes in national Polish emissions which have declined with about 30% since the mid-1980s. A substantial decrease in NO_2 concentrations is also seen at the Czech station Svratouch after 1990, which seems to be confirmed by similar developments at K-Puszta in Hungary, although their records in the 92-94 seem suspiciously low (Barrett et al., 2000).

A similar development is seen at Preila in Lithuania where after 90/91 a notable decrease in NO_2 concentrations is observed which continues until 1999 (Girgzdiene, 2000).

5.2 VOC

A general "complaint" is that there is a lack of observational data. It is certainly true that the size of the VOC databases is considerably smaller than of NO_x and O_3, but in the course of time interesting and useful time series of VOC have been constructed. Some of the data has already been used in trend studies, others await further evaluation.

5.2.1 Urban and semi-urban

In the United Kingdom 26 hourly speciated C_2-C_8 hydrocarbons are routinely measured at 11 urban and 1 rural site since 1994 (Derwent, 1999; Derwent et al, 2000). The majority of the hydrocarbon concentrations show good correlation with benzene indicating that motor vehicle exhaust and evaporative emissions are the major source of hydrocarbon emissions at the UK urban background sites. The concentrations of the majority of the hydrocarbons have shown significant decreases, typically in the order of 20-40% between 1996 and 1998. At sites heavily influenced by industrial emissions, some of the hydrocarbon concentrations have shown reduced trends, no trends or sometimes upward trends.

In Sweden an urban network of benzene and other aromatics was initiated in 1992 (Lindskog, 2001). During the winter half-year weekly averaged samples of 8 speciated VOCs are taken at 10 urban sites. In the course of time the number of urban sites has increased to 39 in 1999/2000. The average of the 10 urban sites show a 60% drop in winter half-year concentrations from the winter 92/93 to the winter 99/00 (Svanberg and Lindskog, 2001). The data of the aromatics indicate a much stronger reduction than the trend in the urban total VOC emissions, which are calculated at –38% over the same period. It is not clear yet whether the discrepancy is due to a reduced share of aromatics in traffic exhaust, or that the urban sites are substantially influenced by other source categories, which have undergone much more change.

In the Netherlands the longest records of continuous VOC measurements are at Moerdijk nearby an industrial plant for which the sites were designed (Thijsse, 1983). Hourly data of C_2-C_5 are collected for the 81-91 period, daily data for the 92-99 period. In the second period ethene, propene and acetylene are measured each day, C_6-C_{12} once every fourth day. A method to filter out the data contaminated by the refinery worked satisfactorily for the longer lived species, but seemed less appropriate for a very reactive component as propene (Roemer, 2001). Typical traffic components like acetylene and ethene showed a downward trend of about 50% over the 81-99 interval (Fig. 4), which is in excellent agreement with the trends according to the traffic emission inventory. The decrease starts in the mid 1980s and accelerates in the 1990s. For benzene and toluene downward trends of 3-4%/yr are found over the 92-99 interval.

For propene almost no trend was found which could be due to local changes that slipped through the filter. Also hexane didn't show much change over the 92-99 interval. It is not clear yet why these components differ so much in trends from the others.

In The Netherlands there are other continuous non-urban data available, such as Zegveld (C_6-C_{14} since 1992) and Kollumerwaard (C_2-C_8 since 1994) but evaluation of those has been so far very limited.

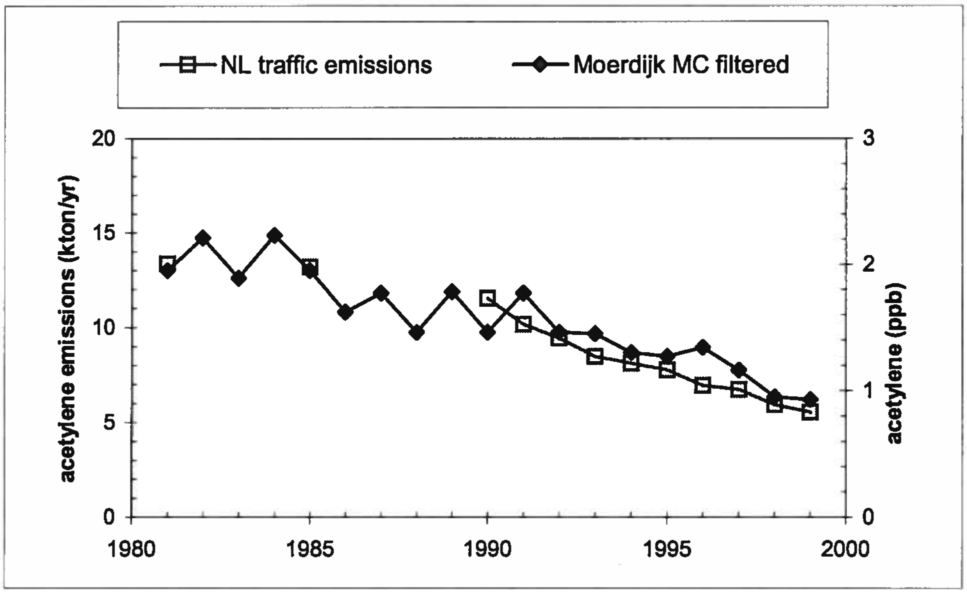

Figure 4 *Trend of filtered Moerdijk MC acetylene concentrations (ppb) and trend of Netherlands traffic acetylene emissions (kton/yr) over the 1980-1999 period.*

In Switzerland long-term continuous measurements of VOC are available for Zürich (urban) and Dübendorf (near a road in an industrial area). At both locations VOC concentrations decreased with about 50% in the period 86-98 (Kuebler et al., 2001). According to the Swiss emission inventory (SAEFL, 1995) the national Swiss VOC emission has dropped in the 85-98 period by –42%. For the two main source categories, traffic and industry the reductions are –70% and –29% respectively. It is important to note that in these emission estimates the last few years are based on future projections.

5.2.2 Rural

At the Wank mountain site in southern Germany of measurements of VOCs were carried out by grab samples during periods from March '87 to July '90 (part 1), and by means of an automatic device in the period January '96 to June '98 (part 2). In the first part samples were taken in the morning while in the second period the observations were done at night. Due to differences in sampling strategy and due to improvements in analytical techniques it is not possible to derive a trend from these two data sets.

The EMEP network started in 1992 with regular monitoring of light VOCs (C_2-C_7) and carbonyls at a few selected sites in Europe (Solberg et al., 1993). The programme for VOC monitoring was substantially revised in 1997 implying the closure of a few sites. There are five EMEP-VOC sites (Waldhof, Kosetice, Starina, Donon and Taenikon) that report data since 1993. In addition data from Birkenes (since 1987), Pallas and Utö are included in the annual EMEP reports (Solberg et al., 1998). The intensity of data collection varies from 70 to 140 samples per year. Trends at Birkenes are currently being sorted out.
The two Finnish sites Pallas and Utö showed no significant difference in concentrations between the 95/97 period and the 98/00 period (Laurila et al., 2001). For the winter season (October-March) no changes were found, for the summer periods (June-August) there are indications of a decrease in concentrations, but the scatter in the data is too high to establish a trend.

The rest of the sites reporting to EMEP have not yet been subjected to a through trend evaluation. It would be interesting to investigate whether the apparent VOC emission trends in various parts of Europe over the last ten years will show up in the data when the observations are assigned to the corresponding wind sectors.

At the site of K-puszta in Hungary VOCs are measured since 1988. Total NMVOC concentrations stayed fairly constant in the 88-91 period and then dropped steeply to much lower values in 1992 which is attributed to the economic crises that occurred in Eastern Europe (Haszpra et al., 2001). Comparing the 88-90 period with 92-93 the decrease in concentrations is in the order of 50-70% in the NE-SE-SW sectors and much smaller in the NW sector facing western Europe (and a part of Hungary). Since 1993 the concentrations stayed at a fairly low level, but since 1997 an increase seems eminent.

5.3 Summary

The urban data, and the non-urban data in The Netherlands and Germany have in common that the trends are downward, and very substantially, and that there is a good agreement with the emission inventory data. This hold for NO_x and for VOC. Urban data sites are predominantly traffic sites, and the agreement shows that the traffic emission information is consistent (in relative terms) with the observations.

In cities in Sweden, Switzerland and The Netherlands the reduction of NO_x emissions by traffic seems to have set in already in 1987 while for the United Kingdom it is 1989. In these countries the traffic emission reduction of NO_x over the last 10-13 years is 35-50%. Non-urban sites in The Netherlands show a 30% in reduction in NO_x concentrations, in agreement with the trend of national NO_x emissions. The onset is not before the early 1990s, so a few later than in the cities.
The traffic related reduction of VOC is about 50-60% in The Netherlands (85-99), 50% in two Swiss cities (86-98) and 60% (aromatics only) for Swedish cities (92-

99). In Sweden the reduction is much stronger than the reduction in VOC emission according to the Inventories, but this might be due to a shift in the partitioning of aromatics to the total.

Sites in downtown Athens show a downward trend in NO_x, it is not clear yet whether this is in line with information from the emission inventory.

The situation for the rural sites is less clear. Clean sites in Scotland and Germany (Westerland, Schauinsland, Brotjacklriegel) show no trend or little trend in NO_2 and NO_x concentrations, despite the reported emission reduction in these and other countries. In contrast, Deuselbach and Waldhof two rural sites closer to the main source areas show a significant downward trend. Agreement between NO_2 reductions in concentrations and emissions of NO_x are reported for a Polish and a Czech site, whereas the Norwegian and Swedish sites showed tendencies opposite to national emission developments.

Rural VOC measurements are scarcely investigated to trends. The two Finnish sites Pallas and Utö showed no trend over the last five years. At K-puszta in Hungary the sharp drop in NMHC concentrations seems to be linked to rapid decline of the eastern economies after 1990.

It is clear that an urban network offers excellent opportunities to track down changes in traffic emissions. It is also clear that traffic emissions in NW-Europe and Alpine Europe are reduced by as much as 35-50% for NO_x and even somewhat more for VOC. The start of the VOC emission reduction is a few years ahead of NO_x. Non-urban sites in polluted and moderately polluted areas also show clear downward tendencies of emissions, and in some cases in good agreement with trends of national emissions. The indication is that the traffic emission reduction is stronger than the national emission reduction, which suggest that the main other source (industry) is trailing compared to traffic.

However, the cleanest sites and a few of the moderately polluted sites show little trend, or trends opposite to the national emission changes. These sites, which are remote from the source areas, may have been influenced by opposing trends from different geographical areas, or by changes in the chemistry that could have affected the NO_2 share in NO_x and NO_y. At these very low concentration regimes data quality problems cannot be excluded either.

6. Discussion and future perspective in TOR-2

Returning to Figure 1 it becomes clear that a few of the questions (“Is there a trend”, and “do they match”) can be answered with more confidence than a few years ago.

Emissions

There is conclusive evidence that reduction of NO_x and VOC emissions by traffic is confirmed by long-time series of observations in urban and polluted areas in NW-Europe and in Alpine Europe. The only south-European location for which information is available, Athens, shows also a decrease in NO_x. Information from other urban areas in Southern and Eastern Europe is heavily needed to assess the developments in these areas. It will be investigated whether this information, if available, can be channelled through by means of GENEMIS, SATURN, or ETC-AQ.

Emissions of industrial sources are more difficult to capture. Without having to measure at individual plants, rural and non-urban sites have the potential to assess concentration and emission changes on a larger than urban scale. There are indications that for NO_x downward trends are observed which relate to emission changes, in NW-Europe but also in a few East-European countries.
However, the situation in the rural department is far from conclusive, since a number of sites might have suffered from data quality problems. For remote sites the meteorological variability and the background are potentially very important contributors to trends, and much work need to be done to separate the contributions.

Ozone observations

There is very strong evidence that over the last ten years annual maximum concentrations have dropped. At a number of sites data quality problems were identified and suspicious data removed. In nearly all studies downward trends emerged, and often at a substantial rate of 1-2% per year.

There is substantial evidence that background ozone concentrations at ground-level in the sectors North and West have increased over the last 10-13 years. For the sectors South and East potential information is available (Izana, Kislovodsk) but not processed yet in the proper format. The developments at (primarily) free tropospheric sites suggest increases of ozone concentrations during the winter and less increase or no increase in the summer. Also at the lower rural sites the dominant tendency in the winter is upward, whereas in the summer upward tendencies still prevail, but with more sites reporting no trend or a downward trend. Advection of ozone from elsewhere is much more of importance to the ozone budget in the winter than in the summer. The fact that trends are almost exclusively upward in the winter, is another indication of increasing background levels. The summer produces a more mixed picture of tendencies of mean concentrations, suggesting that at some sites changes in the chemistry have been of influence.

Relation with models
Annual peak values of ozone are known to be produced by European emissions under the proper meteorological conditions. In a qualitative way, the conclusion that emission reductions are visible in the reduction of peak values is inevitable. However, do they match the expectations? That question can only be answered by feeding chemical dispersion models with the emission changes as have occurred over the last 10-13 years. A first and preliminary comparison suggested that the scavenging of peaks runs more efficient than expected by a model (Roemer, 2001). An important task ahead in TOR-2 and in GLOREAM is to perform a number of sensitivity studies with a set of models to identify the response of models under a specified regime of emission reductions. Candidate models are: LOTOS, EMEP (3D and Lagrangian), CHIMERE, EURAD, REM3, Danish Eulerian, and others. The fact that the models differ in complexity, in vertical and horizontal resolution, in chemistry, in parametrisation of sub-grid processes, in deposition, etc .. offers a good opportunity to testing critical factors determining the sensitivity of ozone trends.

A similar model approach is advocated to address questions concerning the background. There are qualified models available that can simulate changes in the chemistry on a global scale due to worldwide emission, and due to changes in the stratosphere. A pre-requisite is of course that the worldwide emissions are covered well. This approach might identify key contributors to an increase in the background, and whether the changes in background are regionally dependent. Concerning the background much more observational information is required to establish increases in the northern hemisphere. Such information could come from other continents, but also from European sites located south and east. Essential is the availability of back-trajectories to sort out the origin of air parcels.

The workplan for trends in the next year of TOR-2 consists of the following elements:

1. Sorting and filtering
Sorting the 50-70 ozone sites in Europe by means of 3D and 2D back-trajectories (Annex A). The 10-day back-trajectories are calculated on the basis of the reanalysis data, and will be available for the 79-00 period in the autumn of 2001. The 2D back-trajectories are on the 925 mbar level and go 4 days back in time. They cover the 88-96 period and are primarily meant for flat low level areas. An important objective of using trajectories is to sort to geographical areas. Are developments in the south or east different from those in the west? Keeping this in mind it is up to the PIs, who know the local circulation patterns and the local pollution situation, to define the most appropriate sectors at their sites.

Other filtering methods that work on local parameters (NO_3, Be^7, meteorology) and have been applied in the past will be pursued as well.

The sites listed in Annex A are not all TOR sites. It is important that sites lacking human-power to process the data by means of the trajectories will be "hosted" by others in order to prevent holes on the European map. Resources will be sought for to get maximum coverage.

2. Model comparison

The model community will be invited to engage in model sensitivity study directed to trends (see above, and Annex C).

3. Data quality

Data quality aspects remain essential in trend studies. A few sites have been subjected to certain degrees of testing. Others have yet to start. Pairing of nearby sites (especially across borders) is encouraged as a cheap and fast method to identify possible problems. The general state space approach is offered to process a number of sites in order to check for discontinuities.

7. Conclusions

Based on studies conducted and collected in TOR-2 the following conclusions are drawn:

- There is conclusive evidence of a very substantial reduction of NO_x and VOC emissions by traffic in NW-Europe and in Alpine Europe.
- There is very strong evidence that traffic emission trends in the inventories are reflected by the urban observations.
- There is very strong evidence that over the last ten years annual maximum concentrations of ozone are reduced.
- There are strong indications that background ozone concentrations have increased over the last 10-13 years.
- There are indications of emission reductions since the early 1990s in Eastern Europe.
- Data from the monitoring networks needs thorough and extensive screening before it can be used in a trend analysis.

The following aspects need attention in order to provide a pan-European picture:

- There is insufficient information available to draw firm conclusions for the regions East and South.
- It is not known what underlies the increase of background concentrations.
- It is not known whether the changes in ozone (peak and mean) concentrations are *quantitatively* in line with the modelled projections.

It is recommended to work on the following in the last phase of TOR-2:

- Use the trajectory sites to establish a pan-European view of ozone trends, including background trends in all directions.
- Perform model runs with different models to examine the sensitivity of ozone trends to process descriptions.
- Conduct studies dedicated to obtain information from the South and East.

8. Acknowledgements

The convener is grateful to his colleagues in TOR-2 and in Europe for the provision of data and results, for the support in scientific discussions, and for the pleasant human interactions.

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10. Authentication

Name and address of the principal:

TOR-2

Names and functions of the cooperators:

Dr. M.G.M. Roemer

Names and establishments to which part of the research was put out to contract:

-

Date upon which, or period in which, the research took place:

May 2001

Signature:

18/06/01

Dr. M.G.M. Roemer
Project Leader

Approved by:

19/06/01

Dr. M.P. Keuken
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Annex A Data and tools in TOR-2

Since TOR-2 is a project without funding it cannot claim products and data that were developed in projects funded by national and international agencies. However, by combining and comparing the work of a large number of European research groups, by defining a set of joint activities to support a pan-European approach, TOR-2 can claim that it has played an important and stimulating role in creating data sets and tools that are useful and will be useful in the future for the analysis of trends and spatial variation of ozone and precursors. The following data sets and tools can be mentioned:

Data quality aspects
With respect to data quality (not only ozone but also precursors) two approaches are developed. The first approach screens the data to detect irregularities, breaks, drifts by means of statistical methods. Several methods were tested in their ability to detect irregularities. Evi Schuepbach (University of Berne) has offered to run her general state space method for TOR2 ozone and oxidant data. The second approach compares data from neighbouring sites to look for irregularities. Several PIs have volunteered in comparing their ozone with those from neighbouring countries.

Methods to account for (meteorological) variability
There are various methods to account for (meteorological) variability in the data. An overview of methods was circulated after the Dubrovnik workshop (see also: Roemer, 2001). Some of the methods use local meteorology, others use filters by means of chemical tracers (^{7}Be, NO_3). Also large scale parameters (NAO-index, GrossWetterLagen, ..) are used. Back-trajectories are useful in defining the the origin of air. In TOR-2 activities are started to create a large dataset of ozone and trajectory sites.

segregation and classification by means of 3D trajectories.
A few PIs have performed analysis by means of trajectories from NOAA-CMDL and ECMWF. In order to harmonise the analysis for Europe, and thanks to an offer by Andreas Stohl of the University of München it was decided to prepare a set of 3D back trajectories for about 50 sites, covering the period 1979-2000. The 10-day back trajectories are based on the ECMWF re-analysis data. The trajectories have a resolution of 3 hours and provide data of potential vorticity, potential temperature, pressure, and other parameters at a 3h interval. Figure 5 and Table 3 give an overview of sites including the length of the ozone records. It includes three sounding sites, 13 ground level sites above 1 km altitude, and 36 ground level sites below 1 km. The set covers almost the entire European area, with a large range in pollution levels and climatological conditions. A few years are available (May 2001) and it is expected that the entire series will be completed in the autumn of 2001.

segregation and classification by means of 2D trajectories

Sverre Solberg of NILU has calculated and made available 1988-1996 trajectories for 70 sites in Europe, including parameters such as integrated NO_x and VOC. About 40 sites have both the 2D and the 3D trajectories available. Mhairi Coyle and Jana Moldanova have written a programme to visualise the 2D trajectories in excel.

Table 3 *Overview of sites for which 3D trajectories are calculated. Sites have continuous measurements, except the sounding sites. (Coordinates expressed in degrees and minutes)*

Country	Site	Northern Latitude	Eastern Longitude	Altitude m.a.s.l.	Period
NO	Ny Ålesund	78°54'	11°53'	474	89-00
NO	Kårvatn	62°47'	08°53'	210	88-00
NO	Birkenes	58°23'	08°15'	190	85-00
NO	Jeløya	59°26'	10°36'	3	80-00
SE	Vindeln	64°15'	19°46'	271	86-00
SE	Rörvik	57°25'	11°56'	2	87-00
SE	Esrange	67°53'	21°04'	524	91-00
FI	Utö	59°47'	21°23'	7	89-00
FI	Pallas	67°58'	24°07'	566	91-00
DK	Lille Valby	55°42'	12°07'	15	91-00
IE	Mace Head	53°20'	-09°54'	15	87-00
UK	Strath Vaich	57°44'	-04°47'	270	87-00
UK	Aston Hill	52°30'	-03°20'	370	86-00
UK	Eskdalemuir	55°19'	-03°12'	269	86-00
UK	Harwell	51°34'	-01°19'	137	83-00
UK	High Muffles	54°20'	-00°48'	267	87-00
UK	Yarner Wood	50°36'	-03°42'	119	87-00
UK	Sibton	52°11'	01°17'	46	73-00
NL	Kollumerwaard	53°20'	06°17'	0	78-00
NL	Vredepeel	51°32'	05°51'	28	86-00
NL	Bilthoven	52°07'	05°12'	5	94-00 [1]
DE	Waldhof	52°48'	10°46'	73	81-00
DE	Deuselbach	49°46'	07°03'	480	81-00
DE	Zugspitze	47°25'	10°59'	2962	78-00
DE	Brotjacklriegel	48°49'	13°13'	1016	81-00
DE	Wank	47°31'	11°09'	1780	90-00
DE	Schauinsland	47°55'	07°55'	1205	81-00
CH	Jungfraujoch	46°33'	07°59'	3580	87-00
CH	Payerne	46°49'	06°57'	500	92-00
AT	Sonnblick	47°03'	12°58'	3106	88-00
AT	Illmitz	47°46'	16°46'	117	92-00
SL	Krvavec	46°18'	14°32'	1720	89-00
HR	Puntijarka	45°49'	15°59'	980	89-00
FR	Puy de Dome	45°46'	02°57'	1465	95-00
FR	Pic du Midi	42°56'	00°09'	2877	90-93; 96-98
PT	Funchal	32°38'	-16°53'	58	89-99
ES	Noia	42 °44'	-08°55'	685	95-00
ES	San Pablo	39°33'	-04°21'	917	95-00
ES	Logrono	42°27'	-02°30'	445	95-00

Country	Site	Northern Latitude	Eastern Longitude	Altitude m.a.s.l.	Period
ES	Izana	28°18'	-16°29'	2370	84-00
IT	Monte Cimone	44°11'	10°42'	2165	96-00
TU	Uludag	40°17'	28°13'	1685	96-00
BU	Sofia	42°39'	23°23'	588	94-00
HU	K-Puszta	46°58'	19°33'	125	89-00
CS	Kosetice	49°35'	15°05'	633	93-00
SK	Starina	49°03'	22°16'	345	94-00
PL	Puszcza Borecka	54°09'	22°04'	157	96-00
LT	Preila	55°20'	21°00'	5	82-00
LV	Rucava	56°10'	21°11'	18	94-00
RU	Kislovodsk	43°44'	42°40'	2070	89-00
BE	Ukkel	50°48'	04°21'	1000[2], 5000[2]	69-00
GR	Athens	37°59'	23°44'	1000[2], 5000[2]	91-99
FR	OHP	43°54'	05°48'	1000[2], 5000[2]	84-00

1) LIDAR measurements

2) For sounding sites two altitudes (above ground level) are used.

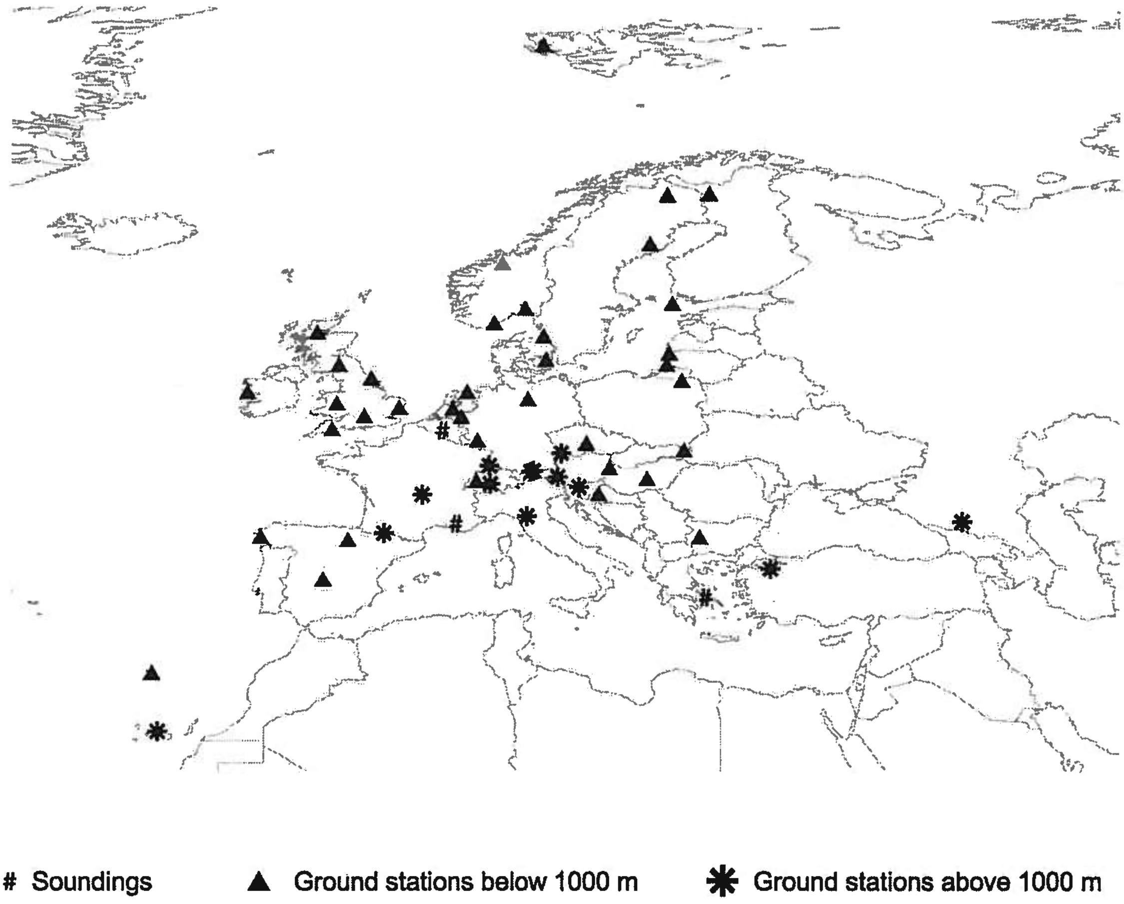

Figure 5 *Location of sites for which 3D-trajectories (10-day back) will be available over the 1979-2000 period.*

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Annex C Invitation to model community

Title: Ozone response to emission changes as a function of model description

Phase 1
An initiative in TOR-2, Michiel Roemer

Questions to be answered in phase 1:

1. what is the model's response on one or more predefined emission changes?
2. Does the model's response depends on the process description?
3. What must be the scope and set-up of a large project (phase 2) to more thoroughly address the first two questions?

The model should be run for a base case and for an emission case. As first emission case it is suggested to reduce anthropogenic national totals for all European countries with 30% (NO_x) and 50% (NMVOC). In phase 2 (or perhaps later in phase 1) more sophisticated cases could be considered, for instances cases for specified regions and source categories (traffic in particular).

A base case consists of one meteorological year, including a shorter episode in that year (see also output).

<u>Question for modelers</u>: which years (episodes) are available?? Preferably we should compare as much as possible model runs based on the same meteorological year.

Suggestion for output, base case and emission case:

A) Ground-level (1-5 m above surface) calculated concentrations for all grid cells. Species: O_3, NO, NO_2, OH, CO, and 1-2 selected VOCs.
B) Aggregated output for ozone: B1) annual mean, winter mean (Nov.-Feb.), summer mean (May-Aug), B2) maximum, P98, P95, P90, P50, P25 of daily maximum concentration in the summer.
C) Aggregated output for episode: as under B, but then for episode period only.

Sensitivity runs are performed by rerunning the base case and the emission case under different conditions. Examples of different conditions are:

1) changing the horizontal or vertical resolution (switch zooming option on or off)
2) changing the VOC/NO_x ratio in the anthropogenic emissions (an example has been illustrated recently at the EGS meeting in Nice by Sandy Sillmann with the CHIMERE model)
3) changing the reactivity of the VOC emissions.

These changes need to be within reasonable margins and need to be specified later in the project.

The requested information to answer the first two questions will come from the sensitivity runs and from comparing the results of the different models. A description of processes in the different models is essential to evaluate the results.

Schedule

June-July	response of model groups (yes or no; available meteo, ..)
August	definition of model runs, definition of output
Sept.-Jan.	model runs
Feb.-Mar.	collecting results, writing summary
March.	Presentation results Eurotrac 2002
April	Definition of phase 2