

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

The development of European surface ozone. Implications for a revised abatement policy

A contribution from the EU research project NEPAP









Norwegian Institute for Air Research PO Box 100, NO-2027 Kjeller, Norway **Chemical Co-ordinating Centre of EMEP (CCC)**

NILU	:	EMEP/CCC-Report 1/2005
REFERENCE	:	U-103003
DATE	:	MARCH 2005

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Norwegian Institute for Air Research P.O. Box 100, N-2027 Kjeller, Norway

EVK2-CT-2002-80019 NEPAP

Network for the support of European Policies on Air pollution

IVL Swedish Environmental Research Institute Ltd., P.O. Box 5302, SE-400 14 Gothenburg, Sweden

Swedish Meteorological and Hydrological Institute, SE-601 76 Norrköping, Sweden



Norwegian Meteorological Institute P.O. Box 43 Blindern, NO-0313 Oslo, Norway

Edited by Sverre Solberg¹⁾ and Anne Lindskog²⁾

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List of Contributors:

Richard G. Derwent³, Peringe Grennfelt², Øystein Hov⁴, Joakim Langner⁵, Anne Lindskog², Sverre Solberg^{1*}

- Norwegian Institute for Air Research, P.O. Box 100, NO-2027 Kjeller, Norway
- 2) IVL Swedish Environmental Research Institute Ltd, P.O. Box 5302, SE-400 14 Gothenburg, Sweden
- 3) rdscientific, Newbury, Berkshire RG14 6LH, United Kingdom
- 4) Norwegian Meteorological Institute, P.O. Box 43 Blindern, NO-0313 Oslo, Norway
- 5) Swedish Meteorological and Hydrological Institute, SE-601 76 Norrköping, Sweden

*Corresponding author

Email:	<u>sso@nilu.no</u>
Fax:	+47 63 89 80 50
Tel:	+47 63 89 80 00

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1. Summary

This report was made within the EU DG Research Project NEPAP (Network for the support of European Policies on Air Pollution) as a scientific support to the present revision of the air pollution abatement strategies within EU and the Convention on long-range transboundary air pollution (CLRTAP). The focus is on emissions and processes not controlled under the present policy for the reduction of surface ozone in Europe. This includes the influence of emissions from other continents, ship emissions and the increasing ozone background concentration (Chapter 3); the future global emission scenarios of ozone precursors and the link between climate change and surface ozone (Chapter 4); the importance of CO and methane for surface ozone (Chapter 5). Chapter 6 gives an overview of monitoring strategies in various organisations. Each chapter is preceded with a summary or a list of key points. These points are also given below. A list of abbreviations and acronyms used in the report is given in Chapter 7.

- European measures to reduce the emissions of ozone precursors have been effective in reducing peak ozone levels.
 - Ozone peak values have been decreasing significantly over the last 10-15 years in many parts of Europe.
- Surface ozone continues to pose a threat to human health as well as vegetation and ecosystems in Europe.
 - The exceedances of the critical levels are still considerable.
 - There is no clear trend in the medium scale ozone concentrations.
 - Without further actions the progress made in Europe to reduce peak ozone levels may, at some point in the future, be off-set by increased hemispheric background due to non-regulated processes on a global scale.
- The European measures to reduce ozone is presently counteracted by an increase in the hemispherical background ozone concentration
 - Surface monitoring sites indicate a steady increase in the ozone background. Some data from soundings and ships are, however, not in agreement with this.
 - Transport of pollutants from other continents and from shipping can contribute significantly to the European background ozone levels. However, large gaps still remain in our understanding of intercontinental transport and the impact for surface ozone in Europe.

- The future ozone level in Europe is closely linked to the future global emissions
 - Modelling studies within IPCC estimate a near-surface ozone increase of about 5 ppb by 2030 and a range from -4 ppb to more than +20 ppb by the end of the century.
 - Intercontinental transport is seen to have a key role in the future trend of surface ozone in Europe.
- The future ozone level in Europe is closely linked to global warming
 - Factors related to global warming such as reduced atmospheric mixing, reduced cloud cover, increased temperature and reduced dry deposition favour increased surface ozone concentrations.
 - o Global warming may lead to increased biogenic emissions of VOC.
 - By 2020 the man-made climate change signal will remain of the same magnitude as the inherent variability in the climate system, while towards the end of the 21st century the anthropogenic signal is expected to be ubiquitous.
 - The extensive episodes of high ozone concentrations observed during the 2003 heat wave in Europe could be seen as an example of the link between global warming and surface ozone.
- *Methane (and CO) emission control is an effective way of simultaneously meeting air quality standards and abating global warming*
 - \circ Methane and CO are, together with NO_x, controlling the hemispheric background level of ozone.
 - The effect of methane control will be global and independent on the location of emission reductions. Thus, reduction of methane emissions in Europe is as good as anywhere else. A reduction of European methane emissions to pre-industrial levels would reduce the background ozone concentration by up to 2 ppb and would at the same time reduce the contribution to global warming
- Long-term environmental measurements of known quality are crucial for scientific understanding and policy development.
 - A number of studies have revealed that data provided by the regular monitoring networks can only be used for trend studies after a thorough and extensive screening.
 - Research Infrastructures as a tool to ensure long-term high quality data may fit the requirements from CAFE.
- It is strongly recommended to strengthen the links between the various monitoring networks (EMEP, EuroAirnet, GAW, national networks) and between the various environmental issues (climate change, surface ozone etc).
 - A proper reference to the EMEP monitoring strategy should be made in the CAFE strategy, which, when implemented, will provide the information required also for these frameworks in background areas.

- A limited number of urban and suburban supersites should be established
 - Urban and suburban measurements are not covered by the EMEP monitoring strategy. However, such data are fundamental for process understanding and for evaluating the effects on human health.

2. Introduction

The 6^{th} Environment Action Programme (EAP) sets out the key environmental objectives to be attained in the European Community in the coming years, and the activities to implement the 6^{th} EAP are taking place within the Clean Air for Europe programme (CAFE). To support CAFE in this procedure, the WHO/ECEH project "Systematic Review of Health Aspects of Air Quality in Europe" was implemented from late 2001 until mid 2004. The project revealed that further actions are needed to reduce ozone levels in Europe. The decrease of ozone peak values achieved through the reduction of ozone precursor emission is important, but also mildly elevated concentrations lasting for longer periods need to be reduced. This calls for a revision of the present policy on air quality. The revision is in the hands of CAFE and will lead to the adoption of a thematic strategy on air pollution under the 6^{th} EAP by mid 2005. This report aims to support CAFE in this work.

The EU Framework 5 project NEPAP (<u>http://nepap.ivl.se/</u>) has been directed to evaluation and assessment of already existing knowledge and data in order to improve the scientific basis for CAFE and CLRTAP strategies. NEPAP's work package on ozone has been focused on the contribution from sources not presently controlled by the EU directives or the emission protocols within the CLRTAP. It is becoming evident that processes outside European control will be crucial for meeting long-term aims and air quality guidelines in Europe in the future.

Measurements and modelling results indicate that there is a strong link between climate change and surface ozone. A warmer and dryer European climate is very likely to lead to increased ozone concentrations. Furthermore, increased anthropogenic emissions in developing economies in Asia are likely to raise the hemispheric background level of ozone. A significant increase in the background concentration of ozone has been observed at several sites in North Europe although the underlying causes are not settled. The photochemical formation of tropospheric ozone from increased concentrations of methane and CO may also lead to a higher ozone level on a global scale. Gradually, these effects may outweigh the effect of the reduced European ozone precursor emissions. This calls for a global or hemispheric perspective in the revision of the European air quality policy for ozone.

3. Historical trends and present state of ozone in Europe

The effect of the decline in European ozone precursor emissions is evident: Ozone peak values have been decreasing significantly over the last 10-15 years.

There is strong evidence that the lower percentiles in the ozone values in polluted areas of Europe have increased, in particular during winter. An important contribution to this upward trend comes from a reduced titration through the O_3 +NO reaction, in response to the reduction of European NO_x emissions.

There is strong evidence that European background ozone has increased.

Current levels of ozone need to be reduced further, since the exceedances of the critical levels are considerable.

It has been suggested that intercontinental transport contribute significantly to the European background ozone levels. However, large gaps still remain in our understanding of intercontinental transport and the possible impact on the air quality at the ground level. Most of the evidence published concerns pollutant plumes in the upper troposphere and there are relatively few conclusive observations from surface stations.

3.1 Measurement data, data quality and representativeness in relation to trend assessments

Data quality and representativity are always important in the evaluation of observations, but are crucial in the search for relatively small changes in time series.

The issue of representativeness is linked to the objective of the interpretation. When assessing 'unpolluted' global or hemispheric background conditions and long-term baseline trend analysis, observations from a remote site would be representative, provided that the measurements are performed in a proper way. When the objective is to assess compliance with critical loads and levels for vegetation, human exposure etc. observations on a finer scale are required.

There are more than 1000 ozone monitoring sites in operation in Europe today in both rural and urban areas, with the majority of sites near local precursor emission sources. Still, the spatial resolution of the monitoring networks is in general insufficient, and the documentation of the representativeness of the individual sites is poor. Very few sites have a monitoring history of several decades and with a detailed knowledge of the historical data quality. From work performed within the EU projects TROTREP and NEPAP it was concluded that data provided by the regular monitoring networks can only be used for trend studies after a thorough and extensive screening (Monks et al., 2003; Solberg, 2003).

3.2 Observed trends in ozone levels in Europe

Comparisons of European ozone measurements made before 1960 and the early 1990s confirm a major increase by a factor of more than 2 in surface ozone in the polluted boundary layer over Europe (Volz and Kley, 1988; Staehelin et al., 2001).

Scheel (2002) has presented the time series of ozone measured at the mountain atmospheric monitoring station at Zugspitze in South Germany (Figure 1) since the 1970s. These data show a clearly increasing trend until the mid 1980s followed by a period with more steady seasonal cycles. Indications of slightly increasing minimum concentrations and reduced seasonal amplitude are seen.

In TROTREP, downward trends of high ozone concentrations (annual peaks or 98th or 95th percentiles) over the last 10-15 years were reported for various European areas (United Kingdom, Denmark, Norway, Sweden, Netherlands, Germany, Switzerland, and Lithuania). When applying methods to minimise the meteorological inter-annual variability, the results become more robust and suggest more strongly that a reduction in the emissions is responsible for the reduction in peak ozone (Monks et al., 2003).



Figure 1: Monthly mean ozone concentrations measured at Zugspitze, Germany since 1978 (from Scheel et al., 2002).

In the UK it has been estimated that peak ozone concentrations at the EMEP stations have declined about 30% in the period 1986-1999 (NEGTAP, 2001). At the same time a slight increase in the annual average concentration is seen. Statistically significant downward trends in the annual maximum concentrations of the order of 3 ppb yr⁻¹ were found for 8 monitoring sites. Downward trends were also found for the 8 h running mean concentration (exceeding 50 ppb) and AOT40 (AOT40 = Accumulated Ozone exposure over the Threshold of 40 ppb) but these trends are not statistically significant.

Results from more than 300 German ozone sites between 1990 and 2000 including urban locations show a pronounced downward trend of the higher percentiles while upward trend was indicated for low and medium percentiles (Beilke and Wallasch, 2000). During 1990-1997 a drop in the 99 percentile of $3.3 \,\mu g \, m^{-3} \, yr^{-1}$ was found. (2 $\mu g \, m^{-3}$ corresponds to about 1 ppb of ozone at the surface). In the same study no trend in AOT40 during 1990-1999 was found,

reflecting that for medium ozone concentrations the various trends may cancel each other.

Albeit the inter-annual variability in peak values of ozone in Europe is large, the trends in the extreme values are sufficiently consistent and in line with numerical model calculations, thus indicating that an effect of the decline in European ozone precursor emissions is evident (Solberg et al., 2005a; Derwent et al., 2003; Monks et al., 2003).

There is also strong evidence that the lower percentiles in the ozone values in polluted areas of Europe have increased, in particular during winter (Monks et al., 2003). An important contribution to this upward trend comes from a reduced titration by the O_3 +NO reaction in response to the reduction of NO_X emissions. Trends of oxidant ($O_X = O_3$ +NO₂) instead of ozone show much less positive trends or even no trend at all.

Observations in remote areas provide strong evidence that background ozone has increased (Monks et al., 2003). Increases of background ozone are reported from Ireland, Scotland, Norway, Sweden, Finland and mountain top sites such as Zugspitze and Jungfraujoch. This is particular true for the winter season. How much of this comes from a possible increase in the hemispheric ozone burden, or from recirculation of European polluted air is not yet known.

A statistically significant increase of 0.5 ppb yr⁻¹ in background air masses has been reported from Mace Head on the west coast of Ireland (Figure 2) based on measurements since 1987 (Simmonds et al., 2004). Furthermore, a major anomaly is evident in these data in 1998-1999, likely caused by large-scale biomass burning events in tropical and boreal regions coupled with an intense El Nino event.

For Esrange, in northern Sweden (Lindskog, 2003) observations from 12 years are available for trend calculations. A simple linear regression analysis applied on daily averages of daytime observations revealed an increase in concentration (Figure 3). The annual increase is about 0.34 ppb (p = 0.05). Also the higher percentiles are increasing. It is likely that the enhanced concentrations of ozone are the result of a general enhancement in the Northern Hemispheric background.

At Rörvik in Southern Sweden the winter average of daytime ozone has increased with 0.3 ppb yr⁻¹ (p=0.001) since 1988 (Figure 3). Also the daily winter maximum has increased. The decrease in summer maximum, as indicated in model calculations, is not observed. Rörvik is occasionally exposed to episodes of polluted air from the European source areas and also from regional sources. It is likely that the reaction with NO_x will act as a temporary sink for ozone during winter, and, consequently, the reduction in NO_x emission that has taken place, could partly explain the enhancement of ozone. However, since also O_x (NO₂+O₃) is increasing (0.1 ppb yr⁻¹; p=0.1), part of the observed change in ozone must be attributed to a genuine increase in ozone, supporting the assumption that the Northern Hemispheric ozone is increasing.



Figure 2: Ozone baseline monthly means and 12-month moving average measured at Mace Head, Ireland (from Simmonds et al. 2004).



Figure 3: Winter averages of O_3 daily means and O_X daily means at Rörvik, Sweden (left panel) and summer and winter averages of daily daytime means of O_3 at Esrange, Sweden (right panel). Winter = October - March; summer = April -September.

Within NEPAP an evaluation of trends in the Norwegian ozone monitoring data has been carried out (Solberg, 2003). A detailed investigation of the technical monitoring history was required to prepare a subset of data with acceptable data quality for long-term trend studies. These data indicated a reduction in the 99-percentile of the daily (daytime) ozone data in the southern part of the country in the summer half year of the order of 1 ppb yr⁻¹ during the 1990s for some sites. For sites further north a statistically significant increase in the mean ozone concentration of 0.3-0.5 ppb yr⁻¹ was found for the winter half year (Figure 4). An increasing trend in the mean ozone concentration was found also in summer but less clear.



Figure 4: Trends in six months' means and percentiles of daytime average ozone data 1990-2002 for Birkenes in South Norway and Voss at the West coast of Norway. The bars give the 10-, 25-, 50-, 75- and 90-percentiles, and the crosses mark the 5- and 99-percentiles and the means. A 70% data coverage was required for each season. Lines mark the linear trends in the 5- or 99-percentiles or the mean if significant on a 1- σ level (thin line) and a 2- σ level (thick line). Winter = Oct.-Mar; Summer = April-Sept.

An analysis of the ozone data with respect to atmospheric air mass transport was carried out to separate cleaner background conditions from air masses more influenced by European anthropogenic air masses. This indicated a significant increase in the ozone concentration in the background air masses at the northern sites (north of mid-Norway) both in summer and winter (Figure 5). Due to the large variations from year to year, it is not clear if this could be explained by a steady growing ozone concentration in the background, or due to shifts from one 3-4 years period with low background ozone in the mid 1990s to another period of high background ozone values at the end of the 1990s. At the stations in the south no clear trend in the background air masses was possible to identify. Whether this is due to problems of separating the types of air masses or reflects that an increase in background ozone is confined to the northern region is not clear.

In southern and central parts of Finland ozone concentrations in unpolluted air masses have been increasing by 1 μ g m⁻³ yr⁻¹ (= 0.5 ppb yr⁻¹) during summer (May-July) (Laurila et al., 2004).

As pointed out by Coyle et al. (2003) increasing tropospheric background ozone concentrations have considerable significance for future assessments of the risks of ozone impacts on vegetation in Europe. Based on ozone monitoring data for 1996-2000 combined with model calculations for future emission scenarios they calculated large increases in AOT40s, with values predicted to approximately double by 2030. Furthermore, this change is accompanied by an increase in early season exposure of vegetation to concentrations in excess of 40 ppb, which is not detected if the current fixed seasonal time-frames (May-July) are used.





Outside Europe increasing background ozone concentrations have been reported in US and were found to be largest in spring with values of the order of 3-5 ppb when comparing the periods 1980-1984 with 1994-1998 (Lin et al., 2000).

The situation with respect to the background is not entirely undisputed since two sites report negative trends over the last 13 years: Uccle, Belgium (at 5 km altitude) and Kislovodsk in the Caucasian mountains of Russia (2070 m altitude). At the Russian site part of the downward trend is due to a change in circulation favouring wind sectors with lower ozone. The trend in Uccle is more difficult to reconcile with other information. The apparent decrease in free tropospheric ozone in the Uccle soundings is in disagreement with the increase of O_3 in the upper troposphere observed in the data between 1994 and 2001from the EU FP5 project MOZAIC.

Lelieveld et al. (2004) have evaluated ship-borne measurements over the Atlantic Ocean during the period 1977-2002. They found that the median ozone concentrations in the northern mid-latitudes (40°N to 60°N) has shifted to higher mixing ratios, indicating an increase of background ozone, whereas the frequency of high-O₃ events has hardly increased. However, based on the monthly mean concentrations for the same region they didn't find any significant trend. In contrast, remarkably large trends occur at low latitudes and in the Southern Hemisphere, where near-surface ozone has increased up to a factor of two.

3.3 The present European ozone distribution

In 2002 the EU threshold value for informing the public of 180 μ g m⁻³ was exceeded in 17 out of the 27 countries reporting and was not exceeded in 9 countries (Scandinavia, Ireland, the Baltic and Romania). One third of the

stations (568 stations) reported one or more exceedances of the EU threshold for public information (Fiala et al., 2002).

The highest peak hourly ozone concentrations reported throughout Europe during 2002 were 185, 196 and 192 ppb for three sites in Spain, 186 ppb at a site in France and 189 ppb at a site in Italy. These levels are similar to the maximum levels reported during 2001.

Peak ozone concentrations tend to be highest in central and southern Europe and lowest in eastern and northern Europe. So, for example, over the period 1997-2002, Finland has reported no exceedances of the 180 μ g m⁻³ (90 ppb) information threshold and France, Spain and Italy regularly reported hourly peak concentrations in excess of 120 ppb (240 μ g m⁻³). At these concentration levels, ozone can cause serious health problems and damage to ecosystems, agricultural crops and materials. There is a general northwest-southeast gradient in the surface ozone distribution in Europe that is also reflected in the seasonal ozone cycles.

Based on rural, background monitoring sites EMEP calculates and reports the AOT40 values in Europe on an annual basis. The map of the 3-months AOT40 for crops and semi-natural vegetation in 2001 (Figure 6) show a general increasing gradient from northwest to south and southeast. The lowest values are found in Scandinavia, in the Baltic region and in the northern parts of Ireland and the United Kingdom, while the highest values are found in Austria, Hungary, Slovenia and on Malta.



Figure 6: AOT40 (ppbh) May, June and July 2001, daylight hours measured by the EMEP network (from Hjellbrekke and Solberg, 2003).

The map shows that the exceedances of the critical levels are considerable. The critical level for agricultural crops, 3000 ppbh, was in 2001 exceeded at most stations in central Europe. The critical level for forests (10 000 ppbh) is also exceeded in larger parts of central and Eastern Europe.

However, several important limitations and uncertainties have been recognised for using AOT40. In particular, the real impacts of ozone depend on the amount of ozone that reaches the sites of damage within the leaf, whereas AOT40-based critical levels only consider the ozone concentration at the top of the canopy. It was agreed at the Gothenburg Workshop in November 2002 (Karlsson et al., 2003) that ozone flux-effect models were sufficiently robust for the derivation of flux-based critical levels, and such critical levels should be included in the UNECE/ICP Mapping Manual for wheat, potato and provisionally for beech.

The new procedures will give very different levels of exceedance for AOT40 than the old. For crops the ozone concentration at the canopy height (about 1 m above ground level) will be significantly less than the ozone concentrations previously used to assess the AOT40. Thus the new AOT40 will be significantly lower than previously estimated values. Conversely, ozone concentration at the top of forest canopies are often 5-10% higher than those modelled or measured, so the new estimates of AOT40 for forests will be greater than previous estimates.

3.4 Import of ozone into Europe

3.4.1 The effect of intercontinental transport

European surface ozone is controlled by internal formation and loss processes and by changes in the concentration of ozone background, here referring to ozone imported into Europe unaffected by recent European influence. Background ozone concentrations are strongly controlled by global NO_x emissions and hence an increase in the hemispheric background concentration may be linked to the enhanced man-made NO_x emissions following growing industrialisation and population in Asia (Ma et al., 2002; Akimoto, 2003). The development in the anthropogenic NO_x emissions from Asia, Europe and North America is shown in Figure 7. Whereas the NO_x emissions in North America have levelled off the last years and have decreased in Europe since the mid 1980's the Asian NO_x emissions have nearly doubled between 1985 and 1998 according to Akimoto (2003). An increase rate of 1.5% per year in observed background ozone has been estimated in Southeast Asia related to the increase in Asian emissions (Chan et al., 2003).

With a photochemical lifetime of 1-4 weeks above the boundary layer, ozone has the potential for being transported and mixed on a hemispheric scale. Precursor emission sources in North America or Asia and stratosphere/troposphere exchange processes will both contribute to the ozone measured at European monitoring sites.



Figure 7: The development in man-made NO_x emissions from North America, Europe and Asia during 1970-2000.

The main characteristics of intercontinental transport between North America, Europe and Asia are shown in Figure 8 (adopted from Stohl and Eckhardt, 2004). Emissions from North America and Asia are mostly transported in the upper troposphere, following upward transport with so-called warm conveyor belts (WCB). This has been confirmed by aircraft measurements of distinct plumes over North America and Europe. Whereas the transport in the upper troposphere is very fast, low-level transport is much slower.

Emissions from Europe behave differently. In wintertime, practically no European pollutants are transported to the upper troposphere and even in summer low-level transport predominates. Furthermore, there is much more meridional transport than over the Asia and North America. In winter, most of the European pollutant outflow is directed towards the Arctic. A significant pathway, especially in summer, also leads into the Mediterranean and towards Africa.



Figure 8. Sketch of the global pathways of intercontinental pollution transport from the various continents. Transparent arrows show transport in the lower troposphere (below 3 km), whereas coloured arrows indicate transport in the middle and upper troposphere (above 3 km). The upper panel shows transport pathways in summer (June, July, August), the lower panel shows transport pathways in winter (December, January, February). Based on a 15 year model climatology and adopted from Stohl and Eckhardt (2004).

With a three-dimensional Lagrangian chemistry-transport model Derwent et al. (2004) have estimated the individual contributions from inter-continental transport and stratosphere/troposphere exchange to the annual mean ozone mixing ratios at a number of sites in Europe in 1998 and the results are summarized in Figure 9. They conclude that intercontinental transport from North America and Asia combined was a significantly more important source of surface ozone in Europe compared with the stratosphere in all months except December and February. They estimated that European ozone production contributed about 50% of the surface ozone mixing ratio in Europe.



Figure 9: Spatial patterns of the annual average contributions in ppb to surface ozone from internal production within Europe, intercontinental transport from Asia and North America and from stratosphere-troposphere exchange calculated with a global Lagrangian transport model (Derwent et al. 2004).

Furthermore, Derwent et al. (2004) estimated that a global 50% reduction of the NO_x emissions had an effect 2-5 times or more the effect of a similar reduction of CO on European ozone. For the calculated NO_x emissions reduction, the contribution from the North American source region varied from 10% to 25% depending on the time of year, and from 6% to 20% for the Asian region. Large regional differences within Europe was found, with the generally highest influence from intercontinental transport and from the stratosphere at the west coast of Ireland and the least in the southeast part of Europe.

In the same study, based on an empirical relationship between AOT40 and the mean monthly daily maximum ozone mixing ratio Derwent et al. (2004) estimated that a 50% reduction in North American NO_x emissions would lead to a 13% reduction in AOT40 in Europe on average. Correspondingly, a 50% reduction of Asian NO_x emissions was estimated to lead to an average reduction of 5% in European AOT40, whereas a similar emission reduction in Europe would lead to 43% reduction in European AOT40 on average.

Boundary layer air over North America is frequently lifted into the upper troposphere with the WCB over the Atlantic Ocean ahead of a frontal system. (Moody et al., 1996; Stohl and Trickl, 1999; Parrish et al., 2000; Cooper et al., 2001; Stohl, 2001; Eckhardt et al., 2004) and the transport is rapid. However, the

main part of the polluted air mass is left above the European boundary layer (Trickl et al., 2003).

Polluted air masses can also be uplifted due to a sea breeze front, become stratified and spread out 1-2 km above the surface in a thin layer, which can be transported all the way to Europe (Angevine et al., 1996; Li et al., 2002). Although the uplifted plume contains ozone precursors, it is suggested that the ozone reaching Europe through these transport processes is mainly formed in the North American boundary layer (Schultz et al., 1998; Flatøy et al., 1996; Stohl et al., 2002; Stohl and Trickl, 2001). It has been estimated that North American anthropogenic emissions enhance surface ozone in continental Europe by 2-4 ppb in summer in average and by 5-10 ppb during transatlantic events (Li et al., 2002). The influence was found particularly strong at the threshold of the European council ozone standard. It was estimated that 20% of the violations of this standard in the summer of 1997 were due to fossil fuel emissions in North America.

During the Indian monsoon the atmospheric transport from emission areas over Asia into the European region (Figure 8) may be very efficient and lead to strongly enhanced ozone concentrations, but mostly in the upper troposphere over Southeast Europe (the Mediterranean) as identified during the MINOS measurement programme (Scheeren et al., 2003). To what extent these fast transport mechanisms also affect the surface concentrations of ozone in Europe depends on the vertical exchange processes.

In general East Asia has a larger global impact on tropospheric ozone than North American or European sources due to its more southerly location, favouring ozone formation, and more efficient lifting of pollutants (Wild and Akimoto, 2001). Both observations and model calculations have verified trans-Pacific transport of Northeast Asian pollution to the Pacific and North America. The outflow of Asian emissions to the Pacific is strongest during spring. The increase of springtime background ozone mixing ratios observed along the US west coast over the past 18 years, amounting to about 10 ppb is most likely due to changes in ozone precursor emissions was discussed by Derwent et al. (2005).

Estimates of the impact of intercontinental transport on the surface air quality are mainly based on results from chemistry transport models and relatively few observations are available from surface stations to validate the model results. A trajectory-based analysis of observations at Mace Head (Ireland) revealed that pollutants from North America (single events) are hardly ever observed at ground (Derwent et al., 1998). Most likely the plume is merged with the background before it reaches the site. In contrast, influence from North American emissions was found at Porspoder (Britany), with a maximum in spring (Fenneteaux et al., 1999). Long-term ozone sonde data from Hohenpeißenberg (Germany) and Payerne (Switzerland) have been used to study the impact of intercontinental pollution transport (Naja et al., 2003). The results indicate a significant influence on ozone mixing ratios in the lower troposphere and the boundary layer over central Europe.

3.4.2 The effect of ship emissions

It is now recognized that ship emissions have global, regional and local impacts (Lawrence and Crutzen, 1999). As shown by Derwent et al. (2005), the contributions to ozone air quality on the western fringes of Europe resulting from the nitrogen oxides emissions arising from international shipping are currently not negligible (Figure 10). Based on the STOCHEM model they estimated that the contribution from international shipping to surface ozone concentrations during the summer peaks at about 6 ppb over the western-most area of Europe (Ireland, Brittany and Portugal). Endresen et al. (2003) quantified the shipping contribution to global emissions of NO_x , SO_2 , CO, CO_2 and VOCs and calculated maximum perturbations in ozone of up to 10 ppb in the North Atlantic Ocean by a global scale CTM.

However, sales of marine diesel oil to international shipping, so-called bunkers, are not attributed to individual countries or regional groupings and so are not presently included in the emission inventories compiled by the UN ECE and EU and do not come under the aegis of the Kyoto Protocol to the United Nations Framework Convention on Climate Change. It is thus likely that ship emissions will play a progressively more important role in achieving regional and global environmental policy targets in the future as land-based emissions are further controlled.



Figure 10. The contribution to the mean surface ozone levels during the summer months (June, July, August) from international shipping calculated with the STOCHEM model by Derwent et al. (2005).

Jonson et al. (2000) calculated the effect from international shipping on a $50 \text{km} \times 50 \text{km}$ grid resolution over Europe using the EMEP model. With this finer scale model resolution they found a pattern with very different characteristics over Europe with reduced ozone in the English Channel, the North Sea and the Baltic Sea and increased ozone in the Mediterranean area as shown by Figure 11. These regional differences are explained by the differences in the environment between these areas. The Mediterranean is characterized by lower levels of NO_x and more

intense solar radiation favoring the formation of additional ozone, whereas the English Channel, North Sea and Baltic Sea have less sunshine and higher levels of NO_x . The maximum calculated contribution of more than 12 ppb mean ozone in summer over the Atlantic Ocean is in line with Lawrence and Crutzen (1999) who used a global model and with the STOCHEM calculations by Derwent et al. (2005).

However, as seen by Figure 11, the EMEP model calculations indicate that the effect of the shipping emissions is mostly confined to the sea areas. Over land the effect is calculated to be much smaller. This reflects the differences in vertical mixing over sea and continents in summer. Whereas the marine boundary layer is typically very shallow and frequently show a stable stratification in summer due to the cooling from below, the continents experience a maximum in vertical turbulent exchange in summer. Furthermore, while there is no deposition of ozone to the water surface, maximum dry deposition rates over land occur in summer. Thus, polluted air masses that stay over the ocean in clear-sky, anticyclonic conditions will have a large potential for ozone formation, whereas this potential will be effectively reduced if the air mass is transported onto the continental areas.



Figure 11. The contribution from international shipping to the mean surface ozone levels in July (left panel) and to the 3-months' AOT40 for crops (right panel) calculated with the EMEP model by Jonson et al. (2000). The contributions are calculated from a model run with all emissions included and a model run excluding the shipping emissions.

4. Future development of surface ozone in Europe

The future development of surface ozone concentrations in Europe depends on the European and global changes in anthropogenic emissions, the change in climate conditions and the changes in land use practices. The IPCC emission scenarios for the 21^{st} century span a wide range of projections, giving a corresponding wide range of likely future ozone scenarios. Modelling studies within IPCC estimated a near-surface ozone increase over much of the northern hemisphere of about 5 ppb by 2030 and a range from -4 ppb to more than +20 ppb by the end of the century. Intercontinental transport is seen to have a key role in the future trend in surface ozone in Europe.

Global warming will normally lead to increased surface ozone levels because of the reduced atmospheric mixing, reduced cloud cover, increased temperature and reduced dry deposition. Increased biogenic emissions may also be important. Modelling studies based on the IPCC climate scenarios predict ozone increases over southern and central Europe that are significant compared to the ozone reductions expected from the emission regulations currently in force. In the northernmost part of Europe a decrease in ozone is predicted related to increased cloudiness, precipitation and changes in atmospheric circulation for that region. By 2020 it is expected that the manmade climate change signal will remain of the same magnitude as the inherent variability in the climate system itself, while towards the end of the 21st century the anthropogenic signal is expected to be ubiquitous.

The 2003 heat wave in Europe had shape "of things to come" in terms of an altered air quality regime under climate change. Ozone observations reported within EEA and EMEP indicate exceptionally long-lasting and spatially extensive episodes of high ozone concentrations in 2003. Trend analyses for the past 12-14 years show the highest number of ozone exceedance and AOT40 values than in any of the previous years. Climate model studies indicate that in response to greenhouse-gas forcing the year-to-year variability in European summer climate may increase, and that the unusual European summer of 2003 may be an example of what is to come.

The development in world wide surface ozone concentrations until 2020 will be controlled both by the development of anthropogenic precursor emissions of oxides of nitrogen and volatile organic compounds in regions like China and the far East, south-east Asia, India, EurAsia and North America, the change with time in the global sum and composition of these emissions, changes in agricultural and forestry practises (in particular related to biomass burning in the tropics and subtropics), and it will be related to changes in climate and climate variability. By 2020 it is expected that the underlying climate change signal from growing releases of anthropogenic greenhouse forcing agents will remain of the same magnitude as the inherent variability in the climate system itself (IPCC, 2001), while towards the end of the 21st century the anthropogenic signal is expected to be ubiquitous.

Several mechanisms will contribute to the future development of the surface ozone concentrations. One mechanism is the effect of regional and global changes in anthropogenic precursor emissions. Another is the contribution due to changes in the cultivation and use of terrestrial ecosystems. A third mechanism is the contribution to ozone variability from climate variability and the underlying global warming. These mechanisms are not independent of each other and therefore their contribution to ozone concentration changes is not strictly additive. European anthropogenic emissions of ozone precursors are projected to decrease in the following decade due to EU's national emission ceilings directive and UNECE's Gothenburg protocol under the CLRTAP. In spite of this, the development of tropospheric ozone and exposure to surface ozone in Europe is uncertain. In the future the effects of climate change and increased global emissions may gradually outweigh the effect of the reduced European precursor emissions for the European ozone levels.

4.1 Future changes in ozone due to changes in anthropogenic emissions

In the Special Report on Emission Scenarios (SRES) the Intergovernmental Panel on Climate Change (IPCC) has worked out a set of emission projections for the 21st century based on various assumptions on the type of economic, cultural and national development to come (Nakicenovic et al., 2000). Six illustrative emission scenarios for NO_x, NMVOC, CO and methane are shown in Figure 12. Briefly explained, the A1 scenarios represent a world of very rapid economic growth, low population growth and a rapid introduction of new and more efficient technologies. Some of the A1 scenarios are fossil-fuel intensive projections, as shown by the A1F1 scenario in Figure 12. The A2 scenarios describe a very heterogeneous world with a regionally orientated economic development resulting in a high population growth. The B1 scenarios represent a very opposite development with emphasis on global solutions to ecological, social and environmental sustainability resulting in a low population growth. The B2 scenarios describe a world with emphasis on local solutions resulting in a moderate population growth. It describes a world with intermediate levels of economic development, less rapid technological change than in B1 and A1 and focus on environmental protection.

As seen from Figure 12 the resulting emission scenarios span a wide range from the low-emission B1 scenario with a modest increase until 2020 followed by a substantial decline afterwards, to the fossil-fuel intensive high-emission A1F1 scenario predicting an approximate three-fold increase in the main ozone precursors by the end of the 21st century. This implies a wide range in the development of the future global tropospheric ozone levels.

Many research groups participated in IPCC's OxComp global modelling study calculating the effect of emission projections for the development of tropospheric ozone (and OH) during the next century (Prather et al., 2001), using the high-end A2 scenario as a basis. The OxComp group computed a linearized relationship between the tropospheric ozone column and the emission changes:

$$\Delta(O_3) = 6.7 \Delta ln CH_4 + 0.17 \Delta E_{NOx} + 0.0014 \Delta E_{CO} + 0.0042 \Delta E_{NMVOC}$$

where $\Delta(O_3)$ is the change in the tropospheric ozone column (in Dobson units), $\Delta lnCH_4$ is ln to the change in the methane concentration and ΔE_{NOx} , ΔE_{CO} and ΔE_{NMVOC} are the changes in global emissions. The OxComp study did not, however, discuss the implications for surface ozone and air quality issues.



Figure 12: IPCC SRES global emission scenarios (Nakicenovic et al., 2000) of ozone precursors for the 21st century. Units: Mt yr^{-1} (*Mt* (*N*) yr^{-1} *for NO_x*).

The implication of the SRES scenarios for surface ozone concentrations has subsequently been discussed by Prather et al. (2003). Based on the same OxComp modelling results they estimated a near-surface ozone increase by 2030 over much of the northern hemisphere of about 5 ppb (+2 to +7 ppb over the range of scenarios analysed). By 2100 the two more extreme SRES scenarios projected baseline ozone increases of more than 20 ppb, while four other scenarios gave -4 to +10 ppb. These calculations did not account for the response of the climate system to the overall anthropogenic forcing including carbon dioxide. This response is expected to alter natural ecosystems and their emissions of ozone precursors significantly. The physical climate system response also includes changes in dynamics, temperature and humidity in the troposphere, with potentially important implications for the spatial and temporal distribution of ozone episodes in the atmospheric boundary layer.

NEGTAP (National Expert Group on Transboundary Air Pollution) used the SRES A2 scenario to calculate the future global ozone baseline level over central England (NEGTAP, 2001). The modelled changes in mean seasonal cycle is shown in Figure 13. NEGTAP estimated a relative increase in annual mean ozone over central England of 14% in 2030, 27% in 2060 and 51% in 2100 compared to the 1990 level.



Figure 13: Current and future seasonal cycles in ozone for central England, predicted using the STOCHEM global model with the IPCC SRES A2 emission scenario. (Adopted from Fowler et al., 2001)

Figure 14 displays modelled surface ozone concentrations from one example study that contributed to the IPCC Third Assessment report. The increasing future continental surface concentrations can be contrasted with near constant concentrations over the oceans. In the presence of NO_x , ozone production is stimulated in continental air masses and elevated ozone concentrations are often found in air mass flowing out of the continents and into oceanic regions during summertime.

Understanding the mechanisms underpinning the increase in future surface ozone concentrations shown in Figure 14 is intimately connected to our understanding of intercontinental transport. Transport of ozone is efficient in the upper troposphere and transit times around latitude circles at this altitude are of the order of several days. Convective processes in tropical and mid-latitude regions are important in lofting NO_x into the middle and upper troposphere. Once there, NO_x can act as an efficient ozone source as one molecule of NO_x can form many ozone molecules during its lifetime. Weather systems in mid-latitudes with their associated fronts can provide another mechanism by which surface-emitted pollutants can be transported to the middle and upper troposphere.

The missing element of the story has been the identification of those meteorological processes that bring upper tropospheric air laden with ozone down to the surface over the continents. These processes may in turn involve convection and the large-scale weather systems that fill mid-latitudes of the troposphere. Man's activities will create a global-scale pool of increased upper tropospheric ozone and this in turn will bring increased surface ozone concentrations to all the Northern Hemisphere continents through the cycle of constantly changing weather patterns.



Figure 14: Modelled surface ozone concentrations over Europe during July in ppb for a) the years 2000-2009; b) 2030-2039; c) 2060-2069 and d) 2090-2099. Adopted from Derwent et al., 2002.

These ozone increases are more directly relevant to the long-term exposure levels of crops and vegetation than to human exposures. However, urban- and regionalscale pollution episodes are built upon these global baseline values. Their future increase may therefore work against regional pollution control strategies, set to reduce exposure levels of both man and vegetation.

4.2 Future changes in ozone due to climate change

The ozone concentration generated in the atmospheric boundary layer in warmer and dryer conditions are higher than normal due to several reasons. In anticyclonic conditions the residence time in the atmospheric boundary layer of a given air parcel is longer than average since the vertical exchange is suppressed allowing prolonged chemical build-up. The absence of clouds favours the transfer of visible and UV-radiation enhancing the chemical reactivity of precursors. In drought or near drought conditions the stomata opening of plants is reduced and consequently the dry removal mechanisms for ozone become less effective (Pleijel et al., 2004). In addition the emissions of biogenic organic species (isoprene, terpenes etc.) increase due to temperature and drought stress on vegetation, also contributing to increased chemical formation of ozone.

4.2.1 Model predictions of the climate change and its implications for ozone

Scenarios for the development of the climate on a global scale are available from a wide range of global general circulation models, GCMs (IPCC, 2001). Due to limitations in computer resources GCM studies have so far usually been performed with rather coarse horizontal resolution e.g. 2.5° by 2.5°. For studies of the impact of climate change on regional air pollution higher resolution is required and a natural source of meteorological information to drive CTMs are regional climate models (RCM). RCMs typically cover a limited area with a high horizontal resolution using results from a global climate model as boundary conditions (e.g. Rummukainen et al., 2001).

The UK Met Office unified model for climate and weather prediction, coupled to a detailed chemistry scheme was used by Zeng and Pyle (2003) to calculate changes in tropospheric ozone between 2000 and 2100. They did three 28 months' calculations, one for 2000 emissions and present-day climate forcing (A), one for 2100 emissions based on the A2 SRES scenario and present-day climate forcing (B), and one for 2100 emissions with double CO_2 climate forcing based on the A2 SRES emissions (C). They calculated the tropospheric ozone burden to be 317 Tg for run A, 489 Tg for B and 505 Tg for C. For surface ozone averaged over latitude bands, in C there was a reduction compared to B over low latitudes (from 50°S to 50°N), and a small increase at higher latitude bands in both hemispheres. It is likely that the model response to climate forcings and emission for years in between 2000 and 2100 would have resulted in ozone budget figures and surface ozone changes somewhere between the results for 2000 and 2100, and for 2020 much closer to the results for 2000 than those for 2100.

The potential impact of regional climate change on the distribution and deposition of air pollutants in Europe has been studied using a regional chemistry/transport/ deposition model, SMHI-MATCH (Langner et al., 2001, 2004). MATCH was set up using meteorological output from climate scenarios made with the Rossby Centre regional climate model version 1, RCA1 (Rummukainen et al., 2001; Räisänen, 2001). Two different regional climate scenarios, H88 and E88, were used. These were derived through downscaling with RCA1 of two different GCMs: HadCM2 (Johns et al., 1997) and ECHAM4/OPCY3 (Roeckner et al., 1996; Roeckner et al., 1999). The H88 scenario corresponds to the downscaling of HadCM2 and E88 corresponds to downscaling of ECHAM4/OPCY3 using RCA1 to a 88 x 88 km horizontal resolution over Europe. RCA1 was run for 10-year control and scenario time slices from both the HadCM2 and ECHAM4 simulations. The control time slices correspond to present climate, while the scenario time slices correspond to periods 50-70 years ahead depending on the GCM. The scenario time slices correspond to changes in greenhouse gases of 150% (HadCM2) and 100% (ECHAM4) compared to the control period. The corresponding changes in global mean temperature are about 2.6 K in both GCMs.

Results from Langner et al. (2004) are shown in Figure 15 for 10-year simulations with SMHI-MATCH using 10-year control (present climate) and scenario (future climate) in both the H88 and E88 scenarios (40 years of simulation in total). Figure 15 shows the calculated 10-year average April-September AOT40 for the control simulations (present climate) for H88 and E88 respectively, as well as the relative difference between the scenario and the control simulation. Relative

differences for the April-September mean of daily maximum (MDM) ozone are shown in Figure 16. Both scenarios show an increase in AOT40 and mean of daily maximum (MDM) over southern and central Europe and a decrease in northern Europe. The increase is larger in the E88 simulation where it exceeds a factor of two for AOT40 over western Europe and 15% in MDM. These changes in surface ozone can be understood in terms of changes in the factors regulating the production of surface ozone.

Increased air concentrations of oxidised nitrogen compounds are simulated over central and southern Europe due to decreased wet scavenging. Decreased precipitation also implies decreased cloudiness and increased solar radiation, which drives the photochemical production of ozone. Increased temperature also leads to increase in biogenic emissions of isoprene, a biogenic precursor of ozone. Domain-total emissions of isoprene for the April-September period has increased from 622 Gg to 917 Gg or by 47% on average in the H88 and from 907 Gg to 1438 Gg or by 59% on average in the E88 simulations (Langner et al., 2004). All these factors promote an increase in surface ozone concentrations over southern and central Europe.

This increase also extends partly over northern Europe but in the northernmost part a decrease in AOT40 and MDM is simulated. In the H88 simulation this is probably related to the strong increase in precipitation and cloudiness in this area during summer, which leads to removal of ozone precursors and less solar radiation. Increase in precipitation could also partly explain the decrease in the E88 simulation but changes in circulation are probably also involved. It should be noted here that the interaction between the dry deposition velocity of ozone and soil moisture via vegetation uptake was not included in the model. Inclusion of this effect would probably tend to enhance the changes in ozone concentrations.

The bottom panel of Figure 15 and Figure 16 shows the results of significance tests for the calculated changes in AOT40 and MDM, respectively, for April-September. Changes in AOT40 are significant in relation to internal variability over large areas, especially in the E88 simulation which shows the largest changes. Both simulations show significant changes over the northernmost parts of Europe. The changes in AOT40 and MDM simulated in the scenarios presented here are substantial and the increases over central Europe are significant compared to the reductions expected from the emission regulations currently in force. In the simulations presented above only the meteorological fields were changed while background atmospheric composition and emissions were unaffected.



Figure 15: Calculated 10-year average (April-September) AOT40 for the control and the scenario for the H88 (left) and E88 (right) simulations. Relative difference (top), AOT40 in the control simulations (middle) test variable for significance of the simulated changes in relation to internal variability (bottom). Values above 1.8 for the test variable indicate significance above the 95% confidence level. Units: ppm hours and percent.



Figure 16: Calculated 10-year average (April-September) relative difference in mean of daily maximum for the control and the scenario for the H88 (left) and E88 (right) simulations (top) and test variable for significance of the simulated changes in relation to internal variability (bottom). Values above 1.8 for the test variable indicate significance above the 95% confidence level. Units: percent.

Tuovinen et al. (2002) have presented a sensitivity analysis of the different factors affecting the future concentrations of ozone in Europe. They used the Lagrangian photochemical model of EMEP (Simpson 1992). Apart from the major importance of increases in background concentrations of ozone they found that increased biogenic emissions of VOC due to increased surface temperatures would significantly counteract the effects of reduced anthropogenic emissions of ozone precursors. The effect of temperature increase on chemistry would be less significant. Although we have not made a separate study of the impact of increased biogenic VOC emissions here it is likely that the substantial increases in biogenic VOC emissions simulated in both the E88 and H88 scenarios are responsible for a large fraction of the simulated increases in surface ozone. However, the increases in surface concentrations of oxidised nitrogen compounds as well as reduction in cloud cover over central and southern Europe are certainly also of importance.

4.2.2 The European summer of 2003 – an example of what to come?

The European summer of 2003 can serve as an example of what may be expected in time to come in terms of an altered air quality regime under climate change. The European summer of 2003 was exceptionally warm, in particular in Central Europe. Based on a temperature reconstruction of monthly (back to 1659) and seasonal (from 1500 to 1658) temperature fields for European land areas (25°W to 40°E, 35°N to 76°N) Luterbacher et al. (2004) concluded that the summer of 2003 was very likely warmer than any other summer back to 1500.

The reconstruction was based on a comprehensive data set that included a large number of homogenized and quality-checked instrumental data series, a number of reconstructed sea-ice and temperature indices, and a few seasonally resolved proxy temperature reconstructions from Greenland ice cores and tree rings from Scandinavia and Siberia. Compared to the 1901-1995 average surface temperature, the summer of 2003 exceeded that average by about 2°C (4 standard deviations). Luterbacher et al. (2004) calculated the return period of such an event. This calculation is sensitive to the underlying trend in the observational material as well as of the assumption of Gaussian distribution of the residuals. The return period of a European-scale summer event exceeding 2°C relative to the 1901-1995 average was calculated to be more than 5000 years for mid 18th century summer conditions. It increases to millions of years at the turn of the 20th century and decreases to less than 100 years for the most recent summers. However, Schär et al. (2004) calculate a return period of the summer 2003 of 46 000 years, with a considerable uncertainty range and a lower bound of the 90% confidence interval of 9 000 years.

The surface ozone distribution over Europe in the summer of 2003 as reflected in the ozone observations reported within EEA indicate that there were exceptionally long-lasting and spatially extensive episodes of high ozone concentrations, mainly in the first half of August, and covering the regions with the highest temperatures, as shown by Figure 17 and Figure 18 (Fiala et al., 2003). It was also found in a trend analysis over the past 12 years that the average number of hours of ozone exceedance above the information threshold of 180 μ g/m³ for any given monitoring site was higher in the summer of 2003 than in any of the previous years.

In case studies of the number of deaths related to the summer of 2003 heat wave in the United Kingdom and the Netherlands, Stedman (2003) estimated that for the first two weeks of August 2003 there were 2045 excess deaths over the 1998-2002 average, and between 423 and 769 of these were related to elevated ambient ozone and PM_{10} concentrations (21-38% of total excess deaths). The population-weighted mean concentration of ozone over the UK for these two weeks was 103 µg m⁻³ in 2003 compared to 58 µg m⁻³ in 2002. In a similar study for the Netherlands Fischer et al. (2003) found that of an excess 1000-1400 deaths in the Netherlands during the summer of 2003, 400-600 were air pollution related (ozone and PM_{10}). These studies indicate that at the current level of precursor emissions, there is a significant climate variability induced change from one year to another in seasonal surface ozone over Europe. It is of at least the same magnitude as the expected effect of a significant change (taken to mean more than the commitments for emission reductions in current legislation) in the precursor emissions.



Figure 17: Number of exceedances of the threshold value for the information to the public (one hour ozone concentration > 180 μ g m⁻³) observed at rural and background stations summer 2003 (April-August), interpolated using inverse distance weighting. Adopted from Fiala et al. (2003).





Rural monitoring data (preliminary) from the EMEP network shown in Figure 19 (Solberg et al., 2005b) indicate that in Central Europe an indicator like the 6-months AOT40 was higher in 2003 than in any other year since 1990, and that the AOT40 values in 2003 were almost a factor 2 higher than the average during the 1990s. The time series of ozone at two EMEP sites (Payerne, Switzerland and Yarner Wood, southern UK) in 2003 are compared with the long-term average and 10- and 90-percentiles from the 13-years period 1990-2002 in Figure 20 (Solberg et al., 2005b). This shows that the general ozone levels were elevated during most of the summer season in Central Europe and, for southern UK, particularly in July/August.

The 2003 heat wave in Europe had shape "of things to come". Schär et al. (2004) and Beniston (2004) carried out regional climate simulation calculations to investigate if summers like the one in 2003 could become more prevalent in a greenhouse-gas scenario calculation. Schär et al. found that in a greenhouse-gas scenario representing 2071-2100 conditions (SCEN) for a grid point in northern Switzerland for June-July-August (JJA) the JJA-average is shifted 4.6°C towards warmer temperatures, and there is a pronounced widening of its statistical distribution with the standard deviation increasing by 102%. This widening is highly significant.



Figure 19: Six-months (April-September) AOT40 for three EMEP sites in Central Europe in 2003. The data are preliminary. Adopted from Solberg et al. (2005b).



Figure 20: Surface ozone measured at Payerne (left) and Yarner Wood (right) in 2003 (red) compared with the average and the 10- and 90-percentiles during the period 1990-2002 (blue). All data are given as 15 days' running mean values (preliminary data from EMEP). Adopted from Solberg et al. (2005b).

The warm summers in SCEN show signs of drought, with semi-arid Mediterranean climate progressing towards central Europe. Drought conditions develop as anticyclonic conditions prevail, and with declining soil moisture solar radiation absorption leads to enhanced sensible heat flux since evapotranspiration is suppressed. Schär et al. conclude that in response to greenhouse-gas forcing the year-to-year variability in European summer climate may increase, and that the unusual European summer of 2003 may be an example of what is to come. Benisto (2004) draws a similar conclusion: "For many purposes the 2003 event

can be used as an analogue of future summers in coming decades in climate impacts and policy studies".

By implication, we can expect the summer surface ozone distribution for 2003 over Europe to become an analogue of future "ozone summers" in the coming decades. Future ozone summers over Europe will, however, also be influenced by changes in the European as well as global precursor emissions, since the ozone distribution in the European boundary layer contains contributions both from ozone formed outside of Europe and of ozone formed over Europe from emissions elsewhere.

In the summer of 2002 the number of high hourly ozone concentrations was much lower than in 2003, supporting the claim that climatological variability from one summer to the next can cause a change in the number of high ozone concentrations monitored across Europe by a factor of two or more.

4.3 Feedback effects between emission change and climate change on ozone

A future increase in biogenic VOC emissions due to increased temperature and drought stress caused by global warming is a likely scenario. One example of such an ecosystem response was published by Sanderson et al. (2003) who investigated the effect of climate change on vegetation, isoprene emissions and surface ozone levels using a GCM coupled to dynamic vegetation and chemistry models. The calculated isoprene emissions rose from 549 Tg/a (1990s) to 736 Tg/a (2090s) as a result of climate change but with fixed vegetation. When vegetation responses were accounted for the isoprene emissions rose to only 697 Tg/a in the 2090s. Surface ozone levels rose by 20-30 ppb in some locations in the fixed vegetation case and by 10-20 ppb if the vegetation changes were included. In particular in parts of China, Korea and eastern US the rise in ozone was calculated to be significant with serious implications for air quality and human health. This result indicated that the climate stress induced increase in biogenic emissions may give rise to an increase in surface ozone by 2020 which is comparable to the rise from increases in anthropogenic precursor emissions.

The EU FP5 project NOFRETETE has investigated the link between climate change and the exchange of Nitrogen trace gases between forest ecosystems and the atmosphere (Smith, 1997). Predicted changes in climate will exert strong feedback effects on these N-exchange processes, since the processes involved in production, consumption and emission of N-trace gases from soils are strongly temperature and moisture dependent. A first study on the effects of predicted changes in climate on microbial processes and N-trace gas emissions from forest soils has shown that N_2O as well as soil NO-emissions will increase by approx. 7% and 27%, respectively, within the next 50 years, mainly due to increases in rates of nitrification. This prediction has carried out for a small area only and did not consider future changes in atmospheric N-deposition.

5. Methane and CO control

Methane and CO are, together with NO_x , controlling the hemispheric background level of ozone. This baseline level is important not only for longterm cumulative exposure values as AOT40, but also for peak ozone episodes which sits on top of the background level. Recent studies indicate that methane emission control is an effective way of simultaneously meeting air quality standards and abating global warming. Methane and CO are closely coupled and should be seen together. However, the effect of CO emission control is likely to be less effective than methane control, both with respect to surface ozone and the climate issue. The effect of methane control will be global and independent on the location of the emission reduction. Thus, reduction of methane emissions in Europe is as good as anywhere else. A reduction of European methane emissions to pre-industrial levels would reduce the background ozone concentration by up to 2 ppb and would at the same time reduce the contribution to global warming.

Photochemical oxidation of methane and carbon monoxide in the presence of NO_x is a potent source of tropospheric ozone on the global scale as discussed already in the early work of Levy (1971) and of Crutzen (1974). However, neither methane nor carbon monoxide is considered ozone precursors on the short-term, regional scale because of their long atmospheric lifetimes. The lifetimes of methane and CO with respect to oxidation by OH in the northern troposphere is of the order of 8-9 years and 1-3 months, respectively. Understanding has changed in recent years and modelling studies indicate that global methane and carbon monoxide emission controls is beneficial both for the issue of global warming and for meeting ozone air quality standards.

The present global concentration of methane (CH₄) of 1700-1800 ppb is more than doubled since the pre-industrial concentrations of around 750 ppb (Prather et al., 2001). Much of the ozone in the global troposphere has been produced by the oxidation of methane and carbon monoxide in the presence of NO_x and this ozone constitutes a global ozone background.

The methane growth rate during the last decades has varied considerably from year to year. The reason for these variations is not clear but may reflect changes in emissions due to varying climate conditions as the methane emissions are strongly linked to the meteorological conditions. 60-70% of the global emissions are estimated to be of anthropogenic origin, whereas the rest is from natural ecosystems (mainly wetlands). Monitoring data indicate a reduced global methane trend during the last years and it has been speculated that this reflects a reduction in the global emission source strength (Lelieveld et al., 1998; Dentener, 2004)

More than 50% of atmospheric CO emissions today are caused by human activities, and the Northern Hemisphere contains about twice as much CO as the Southern Hemisphere. Through UN-ECE's Convention of Long-Range Transboundary Air Pollution (LRTAP) there has been a reduction in anthropogenic CO emissions of the order of 44% in the EMEP region (Solberg et al., 2004) since 1980.

Emission source	CH₄ Emission (Tg yr⁻¹)	CO Emission (Tg yr ⁻¹)
energy use	110	540
biomass burning	40	400
vegetation		100
CH₄ oxidation		845
natural NMHC		325
anthropogenic NMHC		120
wildfires		30
domestic ruminants	80	
wild ruminants	5	
rice paddies	80	
animal wastes	30	
landfills	40	
wastewater	25	
wetlands	145	
oceans	10	4
freshwaters	5	
termites	20	
CH ₄ hydrates	10	
total	600	2400

Table 1:Estimated global emissions of methane and CO for 1992 (from
Lelieveld et al., 1998).

The bulk of the methane oxidation takes place in the warmer regions of the troposphere, close to the surface and in low and mid-latitudes due to the strong temperature dependence of the reaction rate coefficient for the reaction of methane with tropospheric hydroxyl OH radicals. Net ozone production can only occur when methane is oxidised in the presence of a NO_x concentration of at least 25-50 ppt (Lelieveld et al., 1998) and this limits the efficiency of methane as a global source of ozone. Broadly speaking, about two-thirds of the methane is oxidised in the presence of enough NO_x to drive net ozone production.

In the Northern Hemisphere the hemispheric background concentration of ozone has experienced a 2-3 fold increase from the 10-20 ppb concentration level in the early stages of industrialization to the present levels of 30-40 ppb (Volz and Kley, 1988; Lelieveld and Dentener, 2000). It is very likely that this increase is caused by the increase in anthropogenic NO_x emissions together with the global rise in methane and CO concentration.

According to IPCC (IPCC, 2001), tropospheric ozone increased from 25 Dobson units (DU) to 34 DU from the pre-industrial era and up to 2000. Of this increase of 9 ppb 4 DU is attributed to the global growth in atmospheric methane. In the northern hemisphere there has been a uniform ozone increase of around 30 ppb since pre-industrial times. If 4/9 of this increase is due to methane, 13 ppb of the tropospheric ozone increase in the northern hemisphere since pre-industrial times are due to methane growth.

According to the EDGAR-HYDE 1.4 emission database (Van Aardenne et al. (2001) adjusted to Olivier and Berdowski (2001)) the European contribution to the global methane emissions has decreased from 15% to approx. 10% from preindustrial times to now. Assuming an upper value of 15% of European methane emission contribution, 2 ppb of the growth in tropospheric ozone in the northern hemisphere is caused by European methane emissions. This means that a reduction in European methane emissions alone would contribute to tropospheric ozone reductions. It would also contribute to a lowering of the radiative forcing from methane, and the effect would come quickly since methane has a fairly short decay time (8-10 years) compared to e.g. anthropogenic CO_2 (100 years).

As pointed out by Prather et al. (2003) the European ozone standards for cumulative exposures to crops and vegetation (AOT40) is particularly sensitive to changes in the baseline ozone levels. This is because the present background level is close to the threshold level of 40 ppb. The origins of this ozone background are beyond the geographical borders of Europe and involve emission sources beyond the control of policy-makers within the EU. Peak ozone concentrations are formed in regional-scale ozone episodes within Europe and abatable by European NO_x and VOC emission control. These peak episodes however sit on top of the ozone background so their magnitude is not entirely controlled by processes occurring within Europe. Without this global background from the oxidation of methane and carbon monoxide, episodic peak ozone concentrations would be substantially lower.

Recent studies of the importance of intercontinental transport of ozone (Stohl, 2001) and reports of increased background ozone in the northern hemisphere (Simmonds et al., 2004; Jaffe et al., 2003) have strengthened the interest in global ozone precursors as methane and CO. Using the GEOS global chemistry-transport model, Fiore et al. (2002) show that reducing man-made methane emissions globally enables the simultaneous pursuit of ozone air quality and climate change objectives. Reducing methane emissions lowers the global ozone background and improves surface air quality everywhere. Increasing emissions of methane in the IPCC SRES A1 scenario led to a greater incidence of regional ozone pollution episodes in the USA and a longer ozone pollution season by the year 2030, despite decreasing USA emissions of ozone precursors. The modelling of Fiore et al. (2002) showed that reducing the US anthropogenic emissions of NO_x by 50% nearly eliminated the occurrences of grid-square days in excess of 70 ppb in the US, and that a similar cut in the methane emissions reduced these days by 45%. Fiore et al. (2002) stress the need for a global perspective in the design of future regional pollution control strategies. These conclusions are likely to apply with equal force to Europe, where ozone air quality targets are more stringent compared with the USA and are set closer to the global ozone background.

These results agree with a recent global modelling study by Dentener et al. (2004). They conclude that control of methane emissions is an efficient option to reduce tropospheric ozone as well as radiative forcing. The study by Dentener et al. (2004) is based on more updated emission scenarios than the IPCC SRES scenarios and they conclude that with the current legislation scenario (CLE) surface ozone over the Northern Hemisphere is calculated to increase by 3-5 ppb to the 2020s, overcompensating for the effects of NO_x, VOC and CO emission

control in large parts of USA and Europe. Dentener et al. (2004) also argues that under a changed future climate the increase in natural methane emissions may be of the same order as the reduction in anthropogenic emissions.

In the OxComp workshop of the Third Assessment report of the Intergovernmental Panel on Climate Change (IPCC/TAR) (Prather and Ehhalt, 2001) the increase in methane concentration from 1750 ppb to 4300 ppb predicted for the 21^{st} century drives almost half of the calculated tropospheric ozone increase, and NO_x drives most of the other half.

In contrast to methane, the reaction rate coefficient for the reaction of carbon monoxide with hydroxyl radicals shows no significant temperature dependence and carbon monoxide can be oxidised readily throughout the troposphere. However, for its oxidation to be a net source of ozone, it too has to be oxidised in the presence of a sufficient level of NO_x just as with methane. Because of this restriction, the efficiency of carbon monoxide as a global ozone source is limited.

The impact of an across-the-board 50% reduction in global man-made carbon monoxide emissions on surface ozone within Europe has been investigated by Derwent et al. (2004) using the global Lagrangian chemistry transport model STOCHEM. This reduction lead to an average 0.8 ppb or 3% reduction in annual mean ozone across Europe and to a 600 ppb hours or 4% reduction in AOT40 exposure levels for crops. Thus, as for methane, global carbon monoxide emission controls may produce benefits within Europe in terms of reducing future ozone exposures and protecting crops and natural vegetation. Furthermore, carbon monoxide is a major degradation product of methane oxidation and hence these two trace gases are closely coupled together in the photochemistry of the troposphere and in their influence on the global ozone budget.

6. Monitoring strategies

One drawback in using observations to assess trends is the uncertainty of data quality and representativeness. A number of studies have revealed that data provided by the regular monitoring networks can only be used for trend studies after a thorough and extensive screening. There are also a number of local or even national monitoring networks reporting data to different databases, not so easily accessible to the scientific community. In addition, the quality of the data is not always established and documented. At many of the ozone monitoring sites in Europe, no other parameters are measured, not even basic meteorological quantities, such as temperature, wind speed and direction, humidity, and global radiation. This presents a serious obstacle in the evaluation of the data and their scientific understanding. In addition, the growing awareness of the importance of continental outflow calls for a change in ozone monitoring strategy.

The monitoring performed within Co-operative Programme for the Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (EMEP) is presently focused on air pollution in rural and background areas (Figure 21). The data can be downloaded from the EMEP web page (www.emep.int). As a complement, a large number of measurements are carried out in cities/agglomerations, industrial areas and rural areas/stations, by national or local authorities in 29 countries within EuroAirnet (www.eea.eu.int). The data can be collected from AirBase/APIS (Air Pollution Information System; air quality data). The EuroAirnet data quality objectives are set in order to ensure that mapping and assessment based on the data are of sufficient accuracy and reliability. All network operators with stations included in EuroAirnet should strive to accomplish these objectives. However, it is recognised that further work is needed to define data quality objectives and overall quality should be determined.

World-wide observations of ozone are performed within the World Meteorological Organisation - Global Atmosphere Watch – Global Ozone Observing System (WMO-GAW-GO3OS) program, including ground based stations, ozone soundings and total column ozone (see Figure 21). Ozone related data are stored at the World Ozone Data Centre (WODC) at the Atmospheric Environment Service in Canada. Data gathering, retrieval and reporting procedures are standardised by the WMO, with support from the International Ozone Commission of the International Association of Meteorology and Atmospheric Physics (IAMAP). Quality assurance is both addressed in this way along with periodic recalibrations against a given standard. In the future, the issue of quality assurance is to be increasingly addressed.

Through the WMO Global Atmosphere Watch GAW, GO3OS is tightly linked with the WMO Background Air Pollution Monitoring Network BAPMoN with which it shares a number of stations. Quality assurance is being addressed through the establishment of Quality Assurance/Science Activity Centres (QA/SACs), the Secretariat of which is located in Germany and is responsible for BAPMoN data covering the area of Europe and Africa. Similar QA/SACs are under consideration.

The MOZAIC program (Measurement of OZone and water vapour by AIrbus inservice airCraft) was initiated in 1993 by European scientists, aircraft manufacturers and airlines to collect experimental data. Its goal is to help understand the atmosphere and how it is changing under the influence of human activity, with particular interest in the effects of aircraft. MOZAIC consists of automatic and regular measurements of ozone and water vapour by five long range passenger airliners flying all over the world. MOZAIC data provide, in particular, detailed ozone and water vapour climatologies at 9-12 km where subsonic aircraft emit most of their exhaust and which is a very critical domain (e.g. radiatively and S/T exchanges) still imperfectly described in existing models. This will be valuable to improve knowledge about the processes occurring in the upper troposphere/ lower stratosphere (UT/LS), and the model treatment of near tropopause chemistry and transport. LA/CNRS carefully controls the ozone measurement quality: (1) periodical absolute calibration against a reference laboratory instrument before and after each flight period (about one year), (2) automatic in-flight calibrations at 3 levels, and (3) check of inlet and line cleanness to ensure that ozone is not destroyed before entering the analyser.

The EMEP Steering Body has adopted a new EMEP monitoring strategy (<u>http://www.nilu.no/projects/ccc/monitoring_strategy/</u>) and measurement programme 2004-2009, which to some extent also pays regard to suggestions presented by EUROTRAC-2 as a 'European Integrated Monitoring Network for Atmospheric Change' (Volz-Thomas et al, 2003).

The EMEP monitoring programme will be organised to allow for monitoring stations having measurements at different levels of scope and complexity. Three levels are proposed each targeting EMEP objectives in different ways. The main objective of monitoring at level-1 sites is to provide long-term basic measurements of the traditional EMEP parameters. Level-2 sites will provide additional parameters essential for process understanding and further chemical speciation of relevant components. Level-1 and level-2 sites will typically be operated by institutions nominated by the respective Parties for implementing their monitoring obligations. Level-3 activities are research-oriented, with the main objective to develop the scientific understanding of the relevant physicochemical processes in relation to transboundary pollution and its control. Level-3 sites are a voluntary component of the new monitoring network, and the activities will typically be undertaken by research groups and may also include campaign data.

The strategy has the potential to form a long-term baseline-monitoring scheme for Europe through which quality, availability and basic evaluation is ensured. The strategy opens also for changes in priorities, methods etc. due to new knowledge and shift in priorities. In relation to ozone monitoring, the addition of meteorological parameters at Level-1 sites is an important improvement. Other important parameters, not yet included, are radiation and CO, the later as a tracer of polluted air.

The subdivisions of the suggested 'European Integrated Monitoring Network for Atmospheric Change' are based on different geographical scales:

- 1. *Background sites.* It is proposed to integrate the existing GAW sites, with a possible expansion in Southern and Eastern Europe or Russia. The standard measurement programme should include O₃, NO_x, NO_y, CO, VOCs, PAN, temperature, wind, humidity, radiation, turbulence, aerosol composition, Brewer Dobson.
- 2. *Regional master sites.* It is proposed to upgrade initially at least 5-10 (EMEP) stations with extensive measurements to monitor and understand regional-scale ozone pollution, i.e., O₃, NO_x, NO_y, CO, PAN, peroxides, HNO₃, acid deposition, VOCs, carbonyls, meteorology, radiation, ABL height. These sites should fulfil high standards in terms of data quality, establish close cooperation with scientific institutions and provide a basis for intensive field campaigns.
- 3. *Local monitoring networks*. It is proposed to integrate the existing networks of regional authorities. The sites should be equipped with measurements of at least O₃, meteorology, NO_x, CO, aerosols and possibly VOCs (in situ or via a centralised sampling programme).
- 4. Aircraft measurements. The regional surface measurements should be augmented by routine observations (preferably weekly) from small research aircraft equipped with O_3 , CO, NO_x , NO_y , PAN, VOC, radiation, HCHO instruments. The small aircraft would survey the vertical structure in the vicinity of the station and also serve for satellite validation purposes (flight schedule should match satellite overpasses where appropriate). An alternative to a climatological survey would be to characterise the stations for typical meteorological situations, which are then investigated by aircraft flights.

Aside from the subdivision bases, the main difference in this program as compared with the new EMEP strategy is the focus on ozone alone and the selection of parameters (and platforms) in connection with that. It is also recommended that interactions between monitoring agencies and scientists should be strongly encouraged when it comes to quality control, data analysis and model evaluation.

Environmental measurements are crucial for scientific understanding and policy development, and it is important that CAFE recognises its importance in the development of long-term air pollution monitoring strategies in connection with its thematic strategy. The Commission has unique opportunities to influence the direction and quality of long-term measurements in support of European atmospheric science and air pollution policies. The Commission may use both legislative instruments and instruments that support scientific research and science-driven measurements. One such instrument that may fit the requirements from CAFE may be Research Infrastructures. NEPAP has taken an initiative in order to develop the possibility of using RI as a tool for to ensure long-term high quality data.



Figure 21: EMEP and WMO-GAW sites in Europe. Candidate joint supersites are marked with black bullets (status as of March 2004).

An obvious advice is to strengthen the links between the various monitoring networks (EMEP, EuroAirnet, GAW, national networks) and the various environmental issues (climate change, surface ozone etc). It is important to consider the possibility for a monitoring network to meet several objectives:

- 1) Compliance monitoring (i.e. are the long-term trends in measured concentrations in line with the reported emissions?)
- 2) Monitoring of air quality (to be able to evaluate the effects of air quality for human health and vegetation, and to inform the public when necessary).
- 3) Process related monitoring to understand the physical and chemical processes in the atmosphere and thereby to be able to evaluate and improve the models.
- 4) Monitoring in support of further policy development.

It is highly recommended that a proper reference to the EMEP monitoring strategy is made in the CAFE strategy and other documents, which, when implemented, will provide the information required also for these frameworks in background areas. In addition, these sites perform a more comprehensive monitoring programme, which through the emission inventories, chemical transport models and integrated assessment models provides a basis for abatement policies (NEC and CLRTAP). In the 4th Daughter Directive such a direct reference is provided (ref.) and we strongly suggest similar references in revisions to follow.

The EMEP monitoring strategy put a large emphasis on the need for supersites, which will provide some fundamental information required understanding the processes relevant for LRTAP. For ozone some additional parameters need to be

included in the program, e.g. CO, radiation, turbulence and aerosol composition. It is also recommended that interactions between monitoring agencies and scientists should be strongly encouraged when it comes to quality control, data analysis and model evaluation.

The strategy does not consider the need for data to characterise ambient concentrations in the regions where most people live, and it does not reflect processes in more polluted areas. Such data are fundamental both for process understanding, but also for evaluating the effects on human health. It is thus suggested that a limited number of urban and suburban supersites are established for this purpose. Obviously, such supersites must be operated by specialised organisations like research institutes to assure the required data quality. Also here, a close collaboration with EMEP would be beneficial to allow the assessment of regional vs. local air pollution, linking of scales etc.

7. Abbreviations and acronyms

AirBase	European Air Quality information system of the European Community			
APIS	Air Pollution Information System			
BAPMoN	Background Air Pollution Monitoring Network			
CAFE	Clean Air for Europe			
CLRTAP	Convention on Long-range Transboundary Air Pollution			
EMEP	Co-operative programme for monitoring and evaluation of long- range transmission of air pollutants in Europe (also European Monitoring and Evaluation Programme)			
EUROTRAC	Transport and Chemical Transformation of Environmentally Relevant Trace Constituents in the Troposphere over Europe			
GAW	Global Atmosphere Watch			
GO3OS	Global Ozone Observation System			
IAMAP	International Association of Meteorology and Atmospheric Physics			
LA/CNRS	Laboratoire d'Aérologie/Centre National de la Recherche Scientifique			
MINOS	Mediterranean Intensive Oxidant Study			
MOZAIC	Measurement of OZone and water vapour by AIrbus in-service airCraft			
NEPAP	Network for the support of European Policies on Air Pollution			
NEGTAP	National Expert Group on Transboundary Air Pollution			
ppb	parts per billion by volume (10^{-9} v/v)			
ppbh	ppb hours, i.e. the concentration in ppb multiplied with a time scale in hours			
TOR	Tropospheric Ozone Research (EUROTRAC)			
TROTREP	TRopospheric Ozone and Precursors – TREnds, Budgets and Policy			
UNECE/ICP	United Nations Economic Commission for Europe/International Co-operative Programme			
VOC	Volatile Organic Compounds			
WCB	Warm conveyor belt			
WHO/ECEH	World Health Organisation/European Centre for Environment and Health			
WMO	World Meteorological Organisation			

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