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Assessment of ground-level ozone in EEA member countries, with a focus on long-term trends

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European Environment Agency

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Abbreviations and definitions

AOT40 Accumulated ozone exposure over the threshold of 40 parts per billion (ppb), i.e. 80 micrograms per cubic metre (µg/m³). If no subscript is given (see below) the default period for calculating accumulated exposure is three months ($AOT40_c$).

AOT40 (expressed in (µg/m³)·hours) is the sum of the amounts by which hourly mean ozone concentrations (in µg/m³) exceed 80 µg/m³ from 08.00 to 20.00 Central European Time each day, accumulated over a given period. The target value given in the ozone directive is 18 000 µg/m³·h and the long-term objective is 6 000 µg/m³·h (EC, 2002; EC, 2008).

Note that the modelled AOT40 is based on ozone at crop height (normally 1 m) or tree height (normally 20 m) and thus not directly comparable to AOT40 based on measured ozone (intake typically at 2 m).

$AOT40_c$ AOT40 measured over three months (May–July) is valid for crops like wheat and natural vegetation.

$AOT40_{df}$ AOT40 measured over six months (April–September) is valid for deciduous forest.

N_{180} The number of hours with ozone exceeding the information threshold of 180 µg/m³ (human health; EC, 2002; EC, 2008).

MTDM Mean of the ten highest daily maximum ozone concentrations (based on hourly mean data) during April–September, corresponding approximately to the mean of the data ≥ 95 percentile.

$N_{8hDM120}$ Number of days with a maximum eight hours running average ozone concentration exceeding 120 µg/m³.

This target value may be exceeded up to 25 days a year, as an average over the three preceding years (EC, 2002, EC 2008).

NMVOC Non-methane volatile organic compounds.

SOMO35 Accumulated ozone concentrations in excess of 70 µg/m³ (or 35 ppb).

SOMO35 is the sum of the amounts by which maximum daily 8-hour concentrations (in µg/m³) exceed 70 µg/m³ on each day in a calendar year (WHO, 2001).

Ground-level ozone and **surface ozone** are used synonymously in this report.

Tropospheric ozone refers to ozone in the whole troposphere, and will normally be used for ozone above the mixed boundary layer (i.e. above 500–2 000 metres).

Hemispherical background level of ozone: European ozone concentrations are the sum of background ozone (derived from emissions on other continents and ozone transported from the stratosphere) and Europe's anthropogenic ozone production. As such, Europe's contribution only increases surface ozone above the general background level by a factor of 2–3.

Executive summary

Objective of this report

Ground-level and tropospheric ozone is one of the most harmful air pollutants in Europe today. Elevated levels cause health problems, premature deaths, reduced agricultural crop yields, changes in ecosystem species composition and damage to physical infrastructure and cultural heritage.

Ozone (O_3) is not directly emitted to the atmosphere but formed in complex photochemical reactions from ozone precursor gases. O_3 formation depends strongly on meteorological conditions (e.g. solar intensity and temperature). The nitrogen oxide (NO_x) regime is the main factor determining whether O_3 is produced or removed in the troposphere. The major precursors emitted due to human activities — mainly transport — are NO_x, non-methane volatile organic compounds (NMVOC) and carbon monoxide (CO).

European countries have significantly reduced anthropogenic emissions of ozone precursor gases since 1990. In general, however, ambient air measurements in urban and rural areas of Europe do not show any downward trends in ground-level ozone. The main purpose of this report is to shed light on possible reasons.

Approach

In this study, long-term measurements of rural background ozone levels are analysed and compared with modelling results and sensitivity analyses. Rural background stations are used in order to rule out the influence of local conditions close to sources of ozone precursors (mainly traffic). Reference is also made to several national and regional studies of ozone levels in Europe, which have generally reached the same conclusions as the present Europe-wide analysis.

On the basis of the data available in the European air quality database, AirBase, trend analyses were run for several ozone metrics used in the EU ozone directive: the public information threshold, target value, and long-term objective. Another metric, MTDM, was also analysed. This is not referred to in the Directive but is a proxy for photochemical ozone formation, well suited to trend analysis ([1]).

For this study, 198 rural stations in 18 countries with at least eight years of data were considered. In addition to the length of the time series, data had to meet certain quality criteria to be included in the study, specifically cover at least 75 % of hourly values each year and visual inspection of the time series for 'suspicious' events. Nearly all stations are located in central Europe (Austria, the Czech Republic, Germany and Switzerland), the United Kingdom and the Nordic countries. The biased geographical coverage of the monitoring network limits this assessment and its conclusions to certain regions of Europe.

40 stations in eight countries with at least 10 years of data and at least two stations per country, and 46 stations in four countries with 14–16 years of data were chosen for (statistical) trend analyses. Limited information on data quality issues in AirBase adds a level of uncertainty to the data.

These trends from measured data were compared with the output from scenarios generated using the regional EMEP Unified Chemical Transport Model, centred over Europe ([2]).

The reference scenario covers the years 1995 to 2005 and is based on the yearly emissions officially reported by European countries for this period. The model outputs are, for example, 50 x 50 km^2 maps for the ozone metrics AOT40 ([3]) or SOMO35 ([4]). Both

([1]) MTDM is the mean of the ten highest daily maximum ozone concentrations (based on one hourly mean data) during April–September, corresponding approximately to the mean of the data ≥ 95 percentile.

([2]) The AirBase stations used in this analysis are not in general the same as the EMEP stations used to calibrate the model, and the two sets of data can therefore be regarded as independent sources of information.

([3]) AOT40 is accumulated exposure over a threshold of 40 parts per billion (ppb) ozone during daylight hours over the vegetation period (addressing effects on vegetation). 40 ppb corresponds to 80 micrograms per cubic metre (µg/m^3).

([4]) SOMO35 is accumulated exposure over a threshold of 35 ppb for each day in a calendar year (addressing effects on human health). 35 ppb corresponds to 70 µg/m^3.

metrics are measures of accumulated exposure of vegetation or humans to high ozone levels over a certain period.

To assess the variability of the AOT40 and SOMO35 metrics due to inter-annual variations in meteorology alone, the scenario analysis was run holding the anthropogenic emissions constant at the 1995 level. A statistical approach was applied to the results of every 50 x 50 km^2 grid cell to express the mean and variability from the inter-annual variations in meteorology alone, independent of anthropogenic emission changes.

To assess the possible influence of biogenic isoprene emissions ([5]) in Europe, the EMEP reference scenario was run without isoprene emissions. Isoprene, a highly chemically reactive hydrocarbon emitted by plants, is considered to be the single most important natural NMVOC. Biogenic emissions are in general strongly dependent on environmental factors such as temperature, soil moisture, light, humidity and vegetation cover (plant species).

Finally, to assess the possible influence of pollutants from outside Europe the EMEP reference scenario was run with O_3 boundary concentrations fixed to the mean values for the years 1990–2000 over the modelling period 1995–2005.

Key findings

The constant emissions analysis shows that inter-annual variations in weather conditions have a significant impact on yearly ozone levels. This implies that long time series of measurement data are needed from stable networks of monitoring stations in order to discern the effect of reduced ozone precursor emissions. Such long time series are in general not available, particularly not in southern Europe where ozone pollution is a major problem. The results of this study must therefore be treated with caution. It is estimated that another 5–10 years of stable observations from the same stations (assuming they are maintained) are necessary to support a more robust analysis.

While keeping that in mind, the results of the modelled reference scenario, which predict reduced ozone levels across Europe in the period 1995–2005, match the measured data across the 18 countries as a whole better than the constant emissions scenario. There is therefore some evidence that emissions of ozone precursors and ozone levels are in fact falling across Europe. The modelled reduction in ozone metrics is approximately 20–30 % over the period.

However, this overall picture hides important regional variations. The best correlation between the reference scenario and measured data in the period 1995–2005 is found in north-western Europe (Belgium, Germany, the Netherlands and the United Kingdom), while for countries such as Austria and Switzerland there is a better match between the measured data and the constant emissions scenario.

Data from the longest measurement time series of 14–16 years present the same overall picture. Trend analyses using these data indicate significant reductions in ozone concentrations for British and Netherlands stations (falling during the 1990s and levelling off thereafter). For the Austrian and Swiss stations, no significant trends were identified.

The constant emissions analysis shows furthermore that a significant emission-driven trend over the period 1995–2005 should only in parts of central and southern Europe be discernible from natural fluctuations due to weather conditions.

The clearest measured downward trends were found in areas where, according to the model, meteorological variability is largest and likely to mask emission-induced trends. No downward trends were found in regions where the model predicts that meteorology will have least influence. At the country level, the available measured data thus seem to contradict the model predictions. The reasons for this are unclear and need further analysis.

The relatively large discrepancies between measured and modelled trends in ground-level ozone in Austria and Switzerland might be explained by poor model performance, e.g. because the spatial resolution of the model (50 x 50 km^2) does not address the complex topography of the Alps.

The biogenic emissions sensitivity analysis demonstrates that major uncertainties in modelled ozone levels are also related to the size and distribution of isoprene emissions from plants. For some metrics, the uncertainty can be as high as a factor of two or more.

([5]) Isoprene is a volatile organic compound (VOC) emitted by terrestrial vegetation. It plays an important direct and indirect role in the production of ground-level ozone and the formation of organic aerosol (contributing *inter alia* to $PM_{2.5}$ air pollution).

Finally, sensitivity analysis suggests that ozone 'imported' into Europe via intercontinental transport accounts for some 10–30 % of surface ozone levels in western Europe and the Scandinavian countries and less than 10 % in central Europe. Uncertainties in the magnitude and distribution of this inflow can therefore be an important factor in modelling ozone levels in regions exposed to Atlantic air masses.

Other considerations — the influence of climate change

The possible influence of climate change was not considered in the analysis. The strong dependency of ozone levels on meteorology suggests however that predicted changes in climate could also lead to increased ground-level ozone concentrations in many regions of Europe. A clear peak in measured ozone concentrations can be seen in 2003, which had a very hot summer.

Higher temperatures, (more frequent) heat waves, changes of rainfall distribution and reduced cloudiness may:

- accelerate ground-level O_3 production in the atmosphere (depending on the NO_x regime);
- increase biogenic isoprene emissions, which lead to higher ozone concentrations in regions with high NO_x levels;
- increase nitrogen monoxide (NO) emissions from soils and methane (CH_4) emissions from wetlands;
- deplete soil water and thus reduce deposition of O_3 to plant surfaces (e.g. closure of plant stomata under water stress);
- lead to increased incidence of forest fires (emissions of NO_x, CO and non-methane VOC).

Changing climate conditions may influence long-range (intercontinental) transport, leading to increased movement of air masses across areas with large O_3 precursor emissions. In addition, changes in circulation and bigger temperature differences between troposphere and stratosphere may lead to increased influx of O_3 from the stratosphere into the troposphere.

Ground-level ozone has become a hemispheric or even global air pollution problem. According to measurements at remote sites (e.g. in Ireland), the O_3 background concentration has increased by about 2 ppb (approximately 6 %) per decade since 1980 and is expected to rise further. The background concentration in the Northern Hemisphere is now 35–40 ppb. At the same time, ozone is an important greenhouse gas, ranked third behind carbon dioxide and methane. There are therefore good arguments for integrating ozone abatement into local and regional, but also global strategies and measures addressing air pollutants and greenhouse gas emissions simultaneously ([6]).

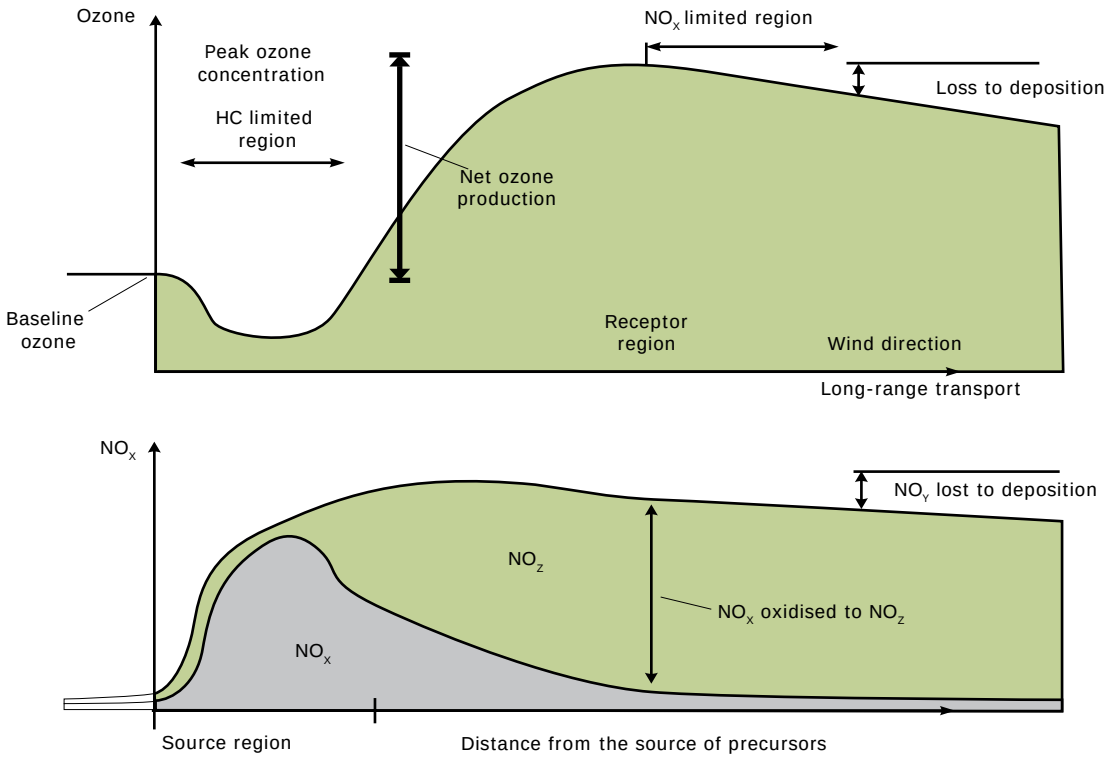
([6]) A comprehensive analysis addressing ozone as a global air pollution and greenhouse gas problem was recently published by the United Kingdom's Royal Society (2008).

1 Introduction

Ground-level ozone is among the most serious air pollutants in Europe today. Elevated levels of ozone cause health problems, premature deaths, reduced agricultural crop yields, changes in biodiversity and damage to materials. Ozone is formed in the atmosphere through chemical reactions between nitrogen oxides (NO_x) and volatile organic compounds (VOC) in the presence of sunlight during a timescale from hours to days. Ozone is lost from the atmosphere through dry deposition to surfaces (including the human respiratory tracts), uptake by vegetation and chemical reactions in the atmosphere with a timescale of hours to weeks.

Meteorological conditions have a decisive influence on the concentration of ground-level ozone. Net formation of ozone requires solar ultra violet (UV) radiation, high temperatures increase the efficiency of the photochemical formation and surface drought reduces deposition to the ground. Furthermore, as indicated in Figure 1.1, ozone formation is related to precursor emissions in a non-linear fashion. Close to emission sources, NO_x reduces ozone by titration ($NO + O_3 \rightarrow NO_2 + O_2$), while a net formation in ozone is found some distance downwind of the sources, depending on temperature and atmospheric dispersion. Thus, the influence on ozone by the

Figure 1.1 Schematic representation of the non-linearity of ozone formation in the atmosphere



Note: $NO_x = NO + NO_2$; NO_y = the sum of all oxidised nitrogen species; $NO_z = NO_y - NO_x$.

Source: Solberg *et al.*, 2004.

prevailing meteorological conditions in Europe from one year to another will easily mask the changes in ozone caused by precursor emission changes.

An additional complicating factor is the influence of the hemispherical background level of ozone. Above the mixed boundary layer (typically 500–2 000 m depending on surface roughness, wind speed and vertical temperature gradient) ozone has a lifetime of weeks. Background ozone levels are therefore a mix of ozone produced from emissions on other continents and ozone transported from the stratosphere as shown in Figure 1.2. Total European ozone concentration levels thus comprise the sum of this background and Europe's own emissions.

Whereas the concentration of primary emissions such as NO_X spans many orders of magnitude from polluted regions to remote areas, ozone emissions only elevate concentrations above the general background level by a factor of 2–3.

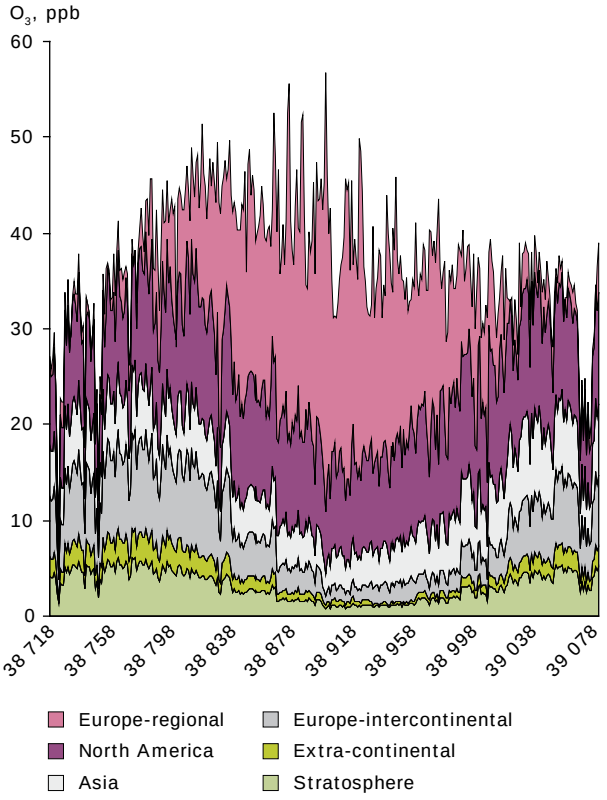
The seasonal cycle in Figure 1.2 is based on model calculations for one site in southern England only, and just for one year (2006). The main message is, however, that only a part of the European ozone levels are controlled by the European precursor emissions. Sources in North America, Asia and stratospheric ozone also contribute significantly. Nonetheless, in summer when the strongest ozone episodes are observed, the influence from European emissions dominates.

The 'Third Daughter Directive' (EC, 2002) defines as part of the Air Quality Framework Directive (EC, 1996) long-term objectives, target values, alert and information thresholds for ground-level ozone within the European Union. The control of precursor emissions is regulated by various directives and standards, such as the Solvent Directive and the EURO standards for road traffic, and by international protocols under the United Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (CLRTAP).

However, while considerable emission reductions have been reported, a corresponding reduction in ozone has apparently not been seen in Europe. According to the EEA's core set indicator 'Emissions of ozone precursors' ([7]) anthrogenic emissions of ozone precursor gases were reduced by 37 % across the EEA-32 countries between 1990 and 2006 while population exposure to ozone levels above the target value set in the EU legislation for protection of human health did not decrease ([8]). Peak ozone concentrations reported to AirBase dropped in the first part of the 1990s while between 1997 and 2006 there was a year-on-year increase in daily 8-hour maximum concentration at most stations

The recently published draft of the fifth report from the Air Quality Expert Group (AQEG) in the

Figure 1.2 Attributed source of ozone at a rural location in southern England in 2006



Note: 'Europe-regional' refers to the ozone advected directly; 'North America' refers to that formed over that continent and over the western North Atlantic and eastern Pacific; 'Asia' refers to that formed over that continent and over the western Pacific; 'Europe — intercontinental' refers to that advected around latitude circles and back into Europe.

Source: Derwent, 2008.

([7]) Available at: http://ims.eionet.europa.eu/IMS/ISpecs/ISpecification20081014123013/IAssessment1226322854001/view_content [Accessed 14 May 2009].

([8]) See Figure 7 at: http://ims.eionet.europa.eu/IMS/ISpecs/ISpecification20080701123452/IAssessment1219309276318/view_content [Accessed 14 May 2009].

United Kingdom (DEFRA, 2008) draws another conclusion looking only at the United Kingdom sites: 'Reductions in precursor emissions in the European region have led to reductions in peak ozone concentrations at rural sites, although there are significant variations from year to year due to the weather'. According to the AQEG draft report and Jenkin (2008), the observations at rural ozone measurement sites in Europe can be viewed as the net effect of three major influences:

- first, a gradual increase in the hemispherical background ozone concentration ([9]), most visible at sites on the Atlantic coast;
- second, a reduction in ozone metrics due to European emission abatement, most visible at measurement sites in countries influenced by regional transport of ozone (precursors) such as the Netherlands;
- third, an increase in ozone at sites exposed to more local traffic emissions due to reduced depletion of ozone by NO_X.

However, the trend in the hemispheric background ozone concentration is somewhat unsettled. Derwent *et al.* (2007) analysed long-term measurements from Mace Head on the west coast of Ireland and found an increase in background ozone until 1999 and a stabilisation or decline thereafter. Using statistical tests ([10]) they note the influence of boreal fires in 1998–1999 and 2002–2003 in elevating background ozone levels, with stronger trends in winter and spring and less in summer for the period 1987–2007.

Scientific literature has reported various ozone trend studies for other regions in Europe. Based on ozone measurements in Finland for the years 1989–2001, Laurila *et al.* (2004) found stable or increasing mean ozone concentrations for the months May–July. For the ninety-ninth percentile of hourly ozone data from the summer months they found consistently negative slopes for the Finnish stations, i.e. downward trends, but at statistically non-significant values, indicating what they call 'a marginal decline of the highest concentrations'. They found no decrease in AOT40 values, but argued that trend assessment was difficult due to the short time series and large year-on-year variations.

Ordóñez *et al.* (2005) analysed Swiss ozone measurement data from 1992 to 2002 and studied the links between local meteorological data and ozone using an ANCOVA (analysis of covariance) method. They found no significant downward trends in seasonal medians or the ninetieth percentiles of daily O_3 or O_X ($O_X = O_3 + NO_2$) maxima. However, for six sites in the industrial regions around Zurich they found a significant downward trend in the summer ninetieth percentile. They argue that the lack of downward trends at the rural sites could be due to an increase in the background ozone level.

Solberg *et al.* (2005) compared model results and measurements for the Nordic countries and found a better agreement between the model and the measurements using real (decreasing) emissions compared to using emissions fixed to the 1990 level for Norway and Sweden, indicating that ozone concentrations have declined in these countries.

A modelling study by Jonson *et al.* (2006) predicted reductions of some 5–10 ppb (10–20 μg/m^3) in mean daily maximum summer ozone concentrations (June to August) in large parts of Europe and up to 12 ppb (24 μg/m^3) in Germany for the period 1990–2004, clearly larger than seen in the measurement data. They argued that most of the monitoring sites were located in areas where NO_X emission reductions were expected to have less effect and in the areas most sensitive to changes in background concentrations (north-western Europe).

The lack of reductions in ozone concentrations have also been debated outside Europe. According to a survey by the United States Environmental Protection Agency in 2004, there was no change in the United States ozone concentrations despite a 12 % decline in NO_X and 25 % cut in VOC emissions in the preceding decade. Increased background ozone concentrations and increased emissions of biogenic VOC have been proposed as reasons for the more or less unchanged ozone levels (Holloway *et al.*, 2003; USEPA, 2006).

A main goal of the present report is to explore the links (or the lack of links) between trends in precursor emissions and measured ground-level ozone. Do the contradicting trends in precursor

([9]) The hemispheric background ozone concentration is a mixture of precursor pollutants emitted in Europe, on other continents of the northern hemisphere (e.g. in Asia) and ozone transported from the stratosphere (see also 'Abbreviations and definitions' on page 5).

([10]) A non-parametric Mann Kendall/Sen slope estimate indicated a positive trend in the mean baseline ozone of 0.31 ± 0.12 ppb per year (corresponding to 0.62 ± 0.24 μg/m^3 per year) with stronger trends in winter and spring and less in summer for the period 1987–2007.

emissions and ozone indicate major shortcomings in our understanding of ozone (formation) chemistry? Or can this be explained by other processes, such as meteorological influence, biogenic emissions? Or is it simply a result of not having monitoring time series of sufficient length?

Another aim is to study measurements and model predictions separately and together. Do the model calculations agree with the measured data or are there systematic discrepancies? The report is focussed on processes and uncertainties, not on effects. Thus, human exposure to ozone, health effects, reduction of crop yields etc. are not discussed.

2 Ozone precursor emissions

Key messages

- Emissions data for the EU-27 countries as a whole show a steady decline in anthropogenic emissions of NO_x and NMVOC from 1990 to 2005.
- Annual emission reductions were largest during the first part of the 1990s and have become smaller after 2000.
- Significant differences are seen between the individual countries. While some countries report strong reductions of both NO_x and NMVOC, other countries report increases in NO_x.
- The emission cuts are larger for NMVOC than for NO_x, implying a change in the NO_x to NMVOC ratio, which could have important consequences for the efficiency of ozone formation.

2.1 Trends in emissions by EU Member States

Emission estimates for the EU-27 were compiled by the EMEP Meteorological Synthesizing Centre-West (MSC-W), a centre under the Convention on Long-range Transboundary Air Pollution (CLRTAP), on the basis of official data reported by countries to EMEP/CLRTAP for the period 1990–2005 (see Table 2.1) ([11]). The respective emission trend lines for NO_x and NMVOC (excluding biogenic emissions) are shown in Figure 2.1. These data shows a steady decline in anthropogenic emissions during the whole period. However, the reductions in emissions were largest during the first years both in absolute and relative terms. From 1990 to 1995 the total emissions of NO_x and NMVOC dropped 17 % and 19 %, respectively, whereas during 2000–2005 the cuts were 11 % and 12 % (for details see also EEA, 2007b).

In general, however, there are large differences between the individual countries. Some countries, such as Greece, Portugal and Austria, realised virtually no change in emissions or increases in emissions, whereas others secured substantial reductions. The trends in emissions of NO_x and NMVOC for seven selected countries are given in Figure 2.2 and Figure 2.3. The seven countries were selected because long-term ozone measurement data (AirBase) were available.

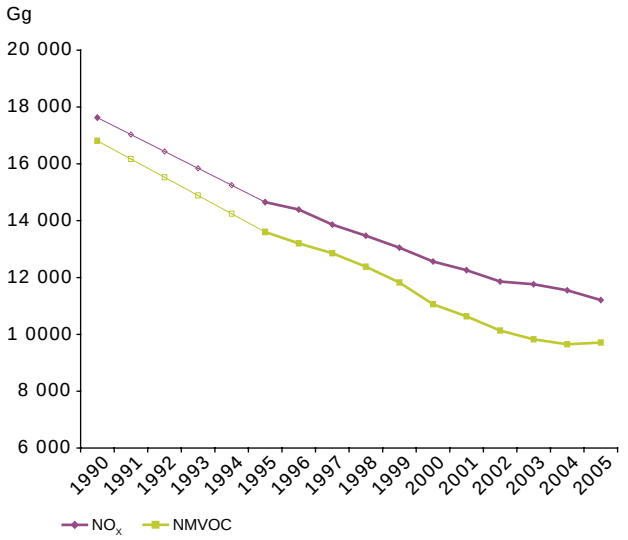
Marked reductions are apparent for France, Germany, Italy, Switzerland and the United Kingdom, whereas NO_x emissions increased in Austria and Spain.

Table 2.1 Emission from EU-27 in Gg (based on 2007 official data submission)

	1990	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005
SO_x	27 403	17 143	15 796	14 496	13 153	11 488	10 371	10 238	9 775	9 256	8 397	8 148
NO_x	17 628	14 653	14 394	13 862	13 473	13 049	12 564	12 260	11 858	11 762	11 549	11 208
NH_3	5 048	4 313	4 301	4 303	4 332	4 296	4 252	4 248	4 208	4 164	4 164	4 130
NMVOC	16 811	13 604	13 200	12 856	12 382	11 823	11 059	10 635	10 135	9 827	9 649	9 712
CO	61 549	48 610	47 886	45 932	43 861	41 901	38 477	37 350	35 042	33 816	33 456	32 091
$PM_{2.5}$							1 735	1 712	1 655	1 644	1 635	1 625
PM_{10}							2 726	2 707	2 621	2 604	2 582	2 495

([11]) The data used are so-called 'expert estimates': officially reported data are reviewed and used if regarded to be of sufficient quality (expert review). They are complemented with data from non-official estimates, i.e. Regional Air Pollution Information and Simulation/Greenhouse Gas and Air Pollution Interactions and Synergies (RAINS/GAINS) model data, for countries, sectors and/or years. For details see e.g. Vestreng *et al.*, 2007b.

Figure 2.1 Trends in EU-27 emissions of photo-oxidant pollution precursors



Note: NO_x is expressed here as NO_2.
The values for 1991–1994 are linearly interpolations of the country data for 1990 and 1995.

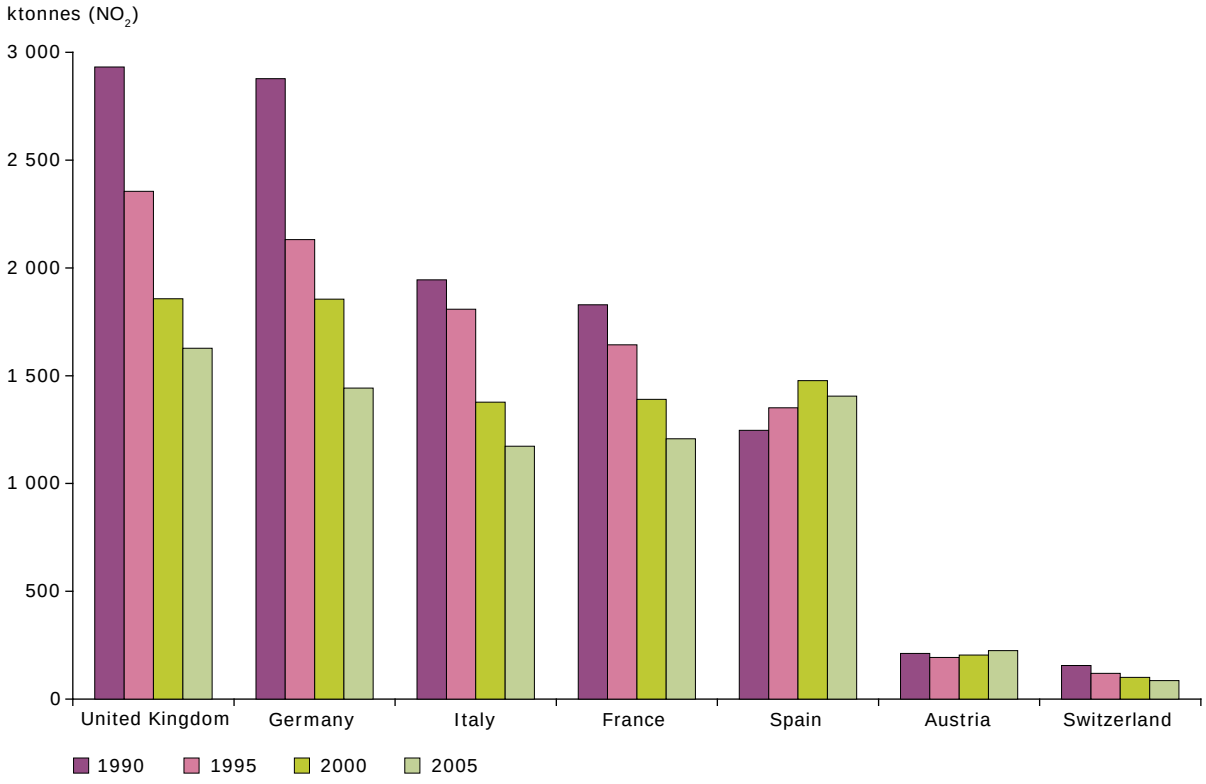
Source: EMEP, 2008.

The emission estimates for NMVOC are at least as uncertain as for NO_x. The largest sources of NMVOC are traffic and solvents. Emissions have been reduced in all relevant sectors but the largest reductions have been in the transport sector. There are large differences between the individual countries, however, with virtually no change in emissions in Greece and a reduction of more than 80 % in traffic emissions in Germany and the United Kingdom.

2.2 Changes in NO_x to NMVOC emission ratios

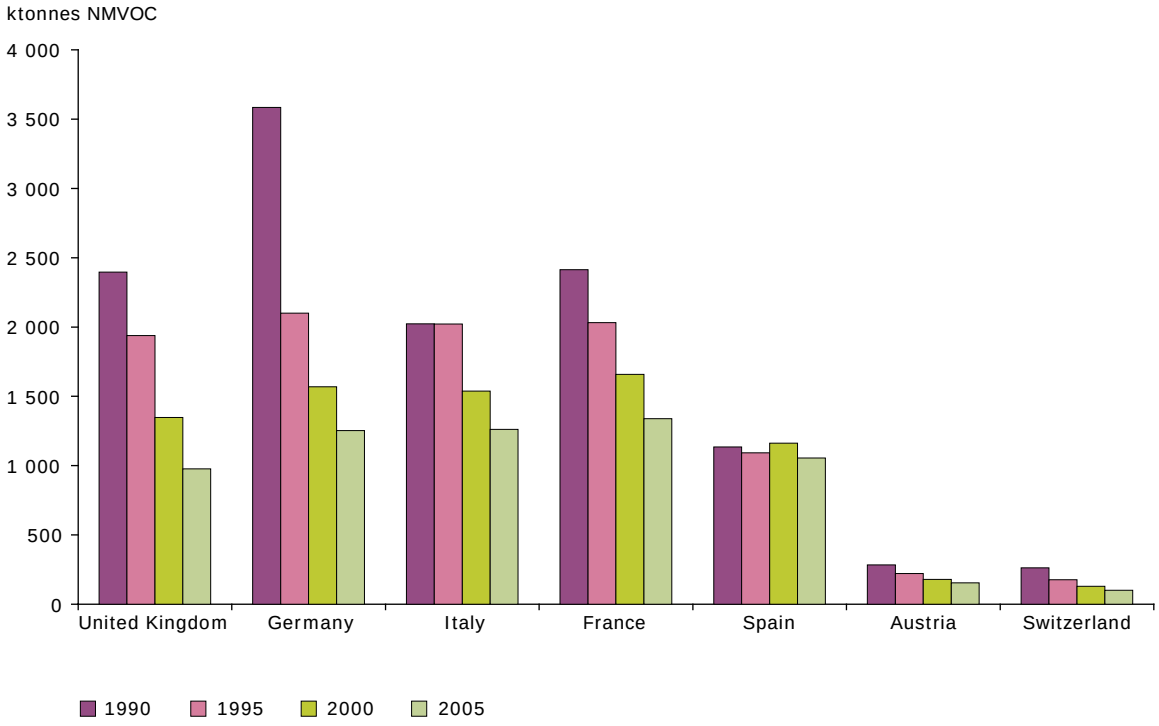
In addition to the trend in total emissions, the data reveal an apparent increase in the NO_x:NMVOC emission ratio in almost all countries as illustrated in Figure 2.4. This is caused by a larger reduction in NMVOC emissions since 1990 than in NO_x emissions. Due to the non-linearity of ozone formation this could be of significant importance although the actual effect on surface ozone in Europe is difficult to assess. Box model calculations

Figure 2.2 Total national emissions of NO_x (as tonnes NO_2) for 1990, 1995, 2000 and 2005 for seven selected countries



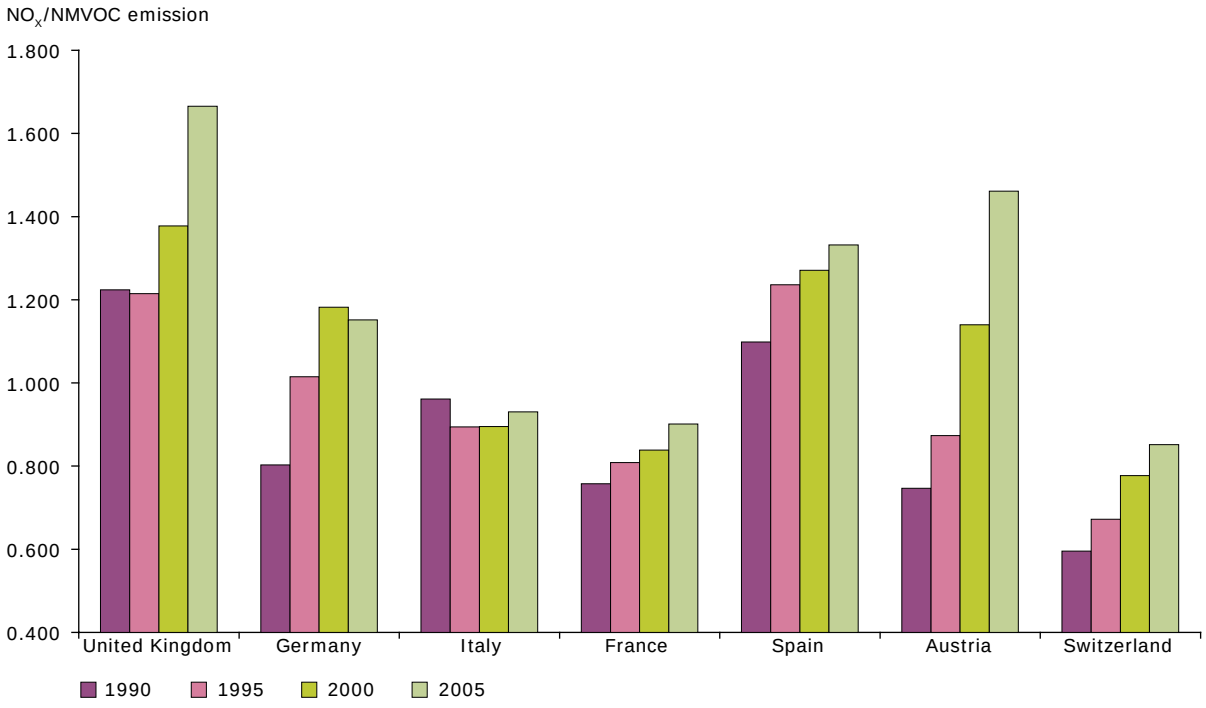
Source: EMEP, 2008.

Figure 2.3 Total national emissions of NMVOC for 1990, 1995, 2000 and 2005 for seven selected countries



Source: EMEP, 2008.

Figure 2.4 Net ratio of NO_x and NMVOC emissions for seven selected countries in 1990, 1995, 2000 and 2005



Source: EMEP, 2008.

simulating a slow moving polluted air mass under European conditions (Derwent *et al.*, 2003) indicate that reduced NMVOC emissions alongside constant NO_x emissions leads to efficient ozone reductions after some hours, whereas NO_x reductions with minimal changes in NMVOC emissions lead to an increase in ozone followed by a net ozone reduction after a few hours.

A larger reduction in NMVOC emissions coupled with a smaller reduction in NO_x could increase ozone close to the emission regions (through reduced titration, e.g. $NO + O_3$) and reduced ozone levels further away (less net ozone formation). On the other hand, the increased NO_x:NMVOC ratio could lead to a 'slower chemistry', i.e. the ozone formation could occur over a wider area (due to reduced OH) and thereby counteracting some of the benefits of the ozone reduction. Whether these phenomena can help explain the trends in ozone observed in Europe remains a hypothesis.

2.3 Uncertainties in ozone precursor emissions

It is beyond the scope of this report to address uncertainties in emissions inventories in detail. The uncertainty in the national total NO_x emissions are expected to be in the 10–20 % range for western European countries and around 25 % for Eastern European countries (Vestreng *et al.*, 2008 and references therein).

Uncertainties for the individual sectors are likely to be considerably larger. Emissions from different sectors have varying seasonal cycles. Traffic emissions generally peak in summer, whereas emissions from power production (at least north of the Alps) peak in winter. As NO_x emissions generally lead to ozone production in summer and ozone titration in winter, the seasonal emission cycle is of great importance. About 40 % of the NO_x emissions in the EMEP model domain are believed to be from road transport. Emissions for the years 1990–2005 are discussed in Vestreng *et al.* (2008) with a focus on the contribution from road traffic.

The NO_2:NO_x ratio is a further source of uncertainty. Diesel powered vehicles have a larger NO_2:NO_x emission ratio than petrol fuelled cars (Carslaw and Beevers, 2005). This is due to particulate matter filters that actually increase NO_2 concentrations during the process of removing soot from the exhaust emissions, thereby increasing direct NO_2 emissions. The penetration of diesel light duty vehicles with such filters is rising in Europe's car fleets. Thus an increase in the share of diesel vehicles in many countries in Europe would be expected to cause an increase in the overall NO_2:NO_x ratio in the emission areas which could potentially influence the ozone formation.

2.4 Long-term NO_2 and NMVOC measurements

Jonson *et al.* (2006) showed that both modelled and measured NO_2 concentrations had a consistent downward trend at EMEP monitoring sites with continuous measurements from 1990 to 2002. Fagerli and Aas (2008) showed that reductions in measured nitrate in precipitation can be explained by reductions in NO_x emissions between 1990 and 2003. Konovalov *et al.* (2008) compared summer GOME and SCHIAMACY NO_2 column satellite measurements and model calculated columns for the decade 1995–2005. They found that for most European countries measured and calculated trends in NO_x emissions over the 10 year period were in close agreement. Differences in trends were seen, however, for some areas including the Balkan countries, Italy, Russia and Turkey. In Italy the measured NO_2 columns remained virtually unchanged from 1995 to 2005 whereas emissions reported to EMEP went significantly down over this period.

In contrast to NO_2, long-term NMVOC measurement data are very sparse and scattered in time. Thus it is more difficult to make a reliable evaluation of the NMVOC emission trend based on the observed concentrations. NMVOC has been measured at a number of EMEP background stations since the early 1990s and for some compounds, such as toluene, the data show a marked reduction over the last 10–15 years.

The data for other compounds indicate a decline during the 1990s with more variable concentrations thereafter (Solberg *et al.*, 2008). A recent review of the Automated Hydrocarbon Monitoring Network in the United Kingdom, based mostly on measurements at urban stations relatively close to emission sources, showed substantial reductions in all NMVOC compounds related to road traffic emissions during the period 1993–2004 (Dollard *et al.*, 2007). This was attributed to the dramatic reduction in hydrocarbon concentrations observed across the United Kingdom due to implementing exhaust catalysts and other control technologies on petrol fuelled cars.

2.5 Emissions data used as EMEP model input

The (gridded) emissions for 1990–2004 used in the EMEP model (see Chapter 4) were derived from the 2006 official data submissions by the Parties to the UNECE CLRTAP, covering most EEA member countries. Parties to the Convention must report emissions every year and gridded emissions at five years intervals. The gridded distributions of the 1990–2004 emissions have been derived by scaling with respect to gridded data distributions in year 2000. The emissions for 2005 have been derived from the 2007 official data submissions pursuant to CLRTAP. The gridded distributions of these emissions were derived by scaling with respect to gridded data distributions in year 2005. Emissions from international shipping are assumed to increase by 2.5 % per year. For 2005, gridded emissions from international shipping is removed and replaced by 2004 emissions, increased by 2.5 %. Emissions are distributed temporarily and vertically depending on source category. National total emissions split into defined sectors from individual countries and gridded emissions data are available from the website of the EMEP Centre on Emission Inventories and Projections (EMEP-CEIP, 2009). The emissions inventories are described in detail by Vestreng *et al.* (2007a).

Biogenic emissions of isoprene are calculated in the model as a function of temperature and solar radiation modified by the total cloud cover. The calculations depend on land class (forest types) using the land use datasets described in the comprehensive description of the EMEP model (EMEP, 2009). Calculations are performed at every model chemical time step. The method is based upon Guenther *et al.* (1993, 1994).

3 Analysis of AirBase measurements

Key messages

- Screening of the AirBase ozone data with respect to length of time series, comprehensiveness and quality was considered crucial prior to the assessment.
- AirBase currently includes more than 2 500 surface ozone measurement stations throughout Europe but the majority have insufficient monitoring history for long-term trend analysis.
- The network of the stations with the required length of observations has an uneven geographical coverage, making the study biased to certain parts of Europe.
- The lack of information on data quality (such as calibration of ozone measurement instruments) adds an unknown level of uncertainty to the data.
- The longest time series (1990–2005) show reductions in various ozone metrics for the Netherlands and the United Kingdom, and to a less extent Austria for 1990–1998, with unclear trends after 1998. There is no indication of trends in the Swiss data.
- The large annual variations in ozone metrics make trend estimates based only on measurements uncertain.

3.1 Exchange of Information Decision (EoID)

The reciprocal exchange of information among EU Member States and the European Commission is based on a series of Council Decisions. The European Council adopted the Exchange of Information Decision (EoID) (EC, 1997) in 1997. The EoID requires a large set of meta-information and air quality data to be delivered to the Commission. Part of this information is mandatory and other items are to be delivered to the Commission 'to the extent possible' and 'as much information as feasible should be supplied'.

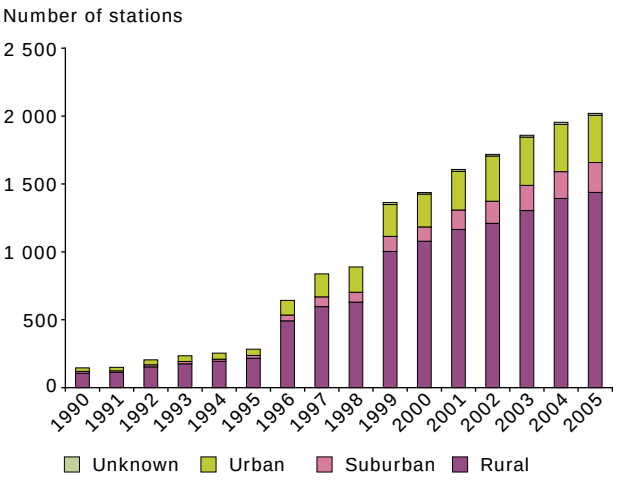
According to the EoID, the Commission will each year prepare a technical report on the meta-information and air quality data exchanged, and make the information available to EU Member States (e.g. Mol *et al.*, 2008). The Decision states that the Commission will call on the EEA with regard to the operation and practical implementation of the information systems. The ETC/ACC, under contract to EEA, manages the database system AirBase (AirBase, 2008).

3.2 Data overview and quality

For the present study, all AirBase NO_2 and ozone data were extracted from the official AirBase site for raw data download (AirBase, 2008) at the beginning of 2008. At that time the data were complete until 2005 and there were no data for 2006.

Regular reporting of ozone monitoring data to AirBase started in the mid-1990s and the number of stations until 1995 was a few hundred, as shown in Figure 3.1. From 1996 to 2005 the number of sites reporting ozone increased from less than 700 to more than 2 000. For trend studies the number of stations with sufficiently long and continuous time series is more important than the number of sites

Figure 3.1 Number of AirBase stations reporting ozone measurements each year during 1990–2005



Source: AirBase, 2008.

itself. Figure 3.2 shows the number of ozone stations as a function of the length of their time series using 1995 as the initial year. In a similar fashion, Figure 3.3 shows the number of countries that have ozone stations with the indicated length of time series. Approximately 250 sites and 10 countries have continuous ozone monitoring data for all 11 years.

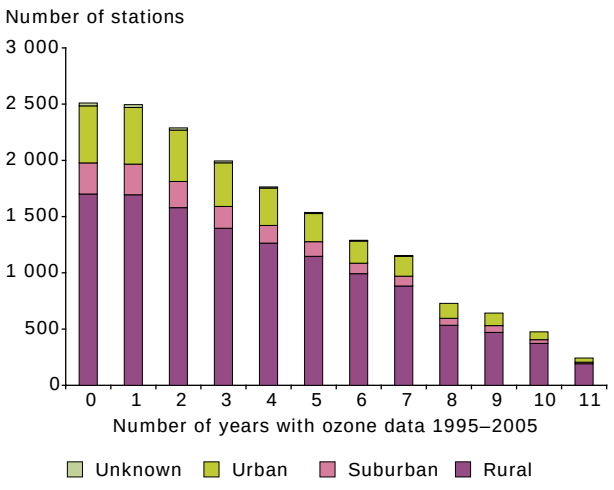
It should be added that longer time series do exist outside AirBase. For example, AirBase currently lacks ozone data before 1998 for Norway and Sweden but data back to 1990 and earlier are available from the EMEP web page (EMEP, 2009). The same is presumably true for other countries and stations. Thus, future trend studies would benefit from efforts to complete the historical ozone data in AirBase.

In the assessment study presented here a subset of the AirBase ozone stations was prepared based on the requirements that:

- they should have at least eight years of data in the period 1995–2005;
- they should cover at least 75 % of hourly values each year;
- the data pass a quality check based on visual inspection.

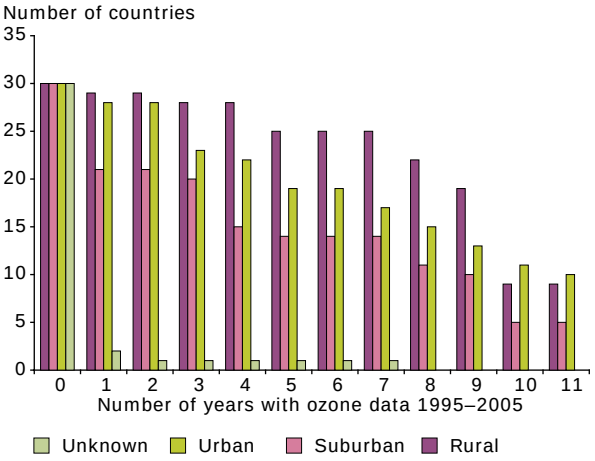
At the time of preparing this study, AirBase contained ozone data from 2 495 stations for the period 1995–2005. Discounting those without at least eight years of data for that period reduced the number to 729 stations. Discounting those without 75 % data capture reduced it to 624.

Figure 3.2 Number of AirBase ozone stations as a function of the number of years with reported data during 1995–2005



Source: AirBase, 2008.

Figure 3.3 Number of countries that have AirBase ozone stations with at least the indicated number of years of data during 1995–2005



Source: AirBase, 2008.

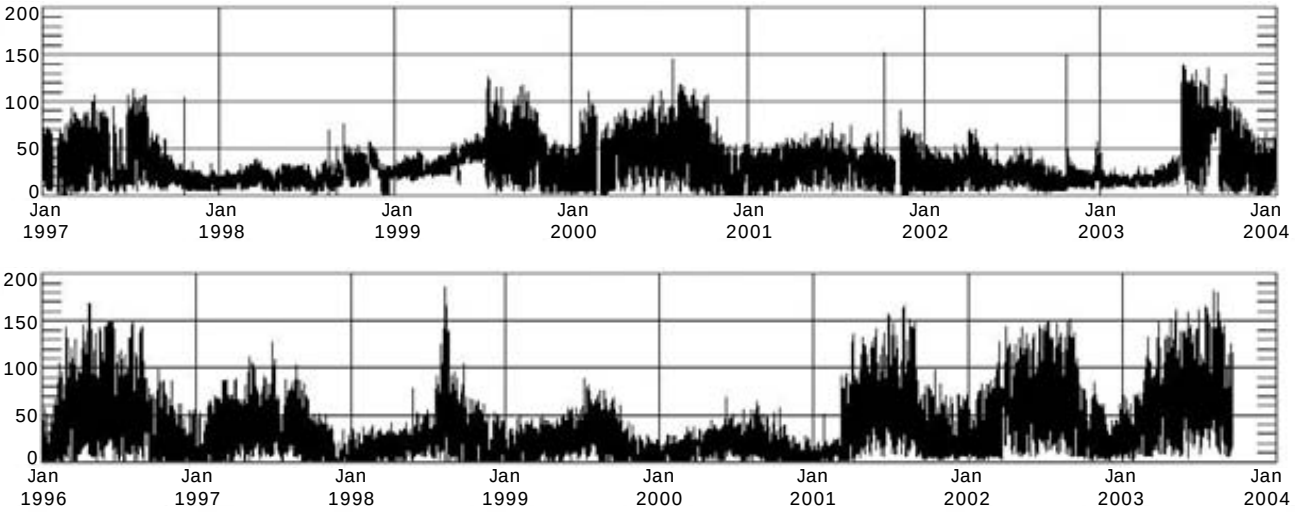
The data reported to AirBase are quality controlled and checked prior to submission by the countries providing the data. The data are then subject to certain automated checks by those responsible for AirBase (ETC/ACC). These automated checks have been applied to data back to 2002 while older data have not been checked in this way.

In the present study no automated data quality checks were carried out. Instead a visual inspection of each of the 624 ozone time series was possible. Such a check may actually be superior to 'blind', automated scripts for detecting suspicious data because peculiarities in the time series can take many forms, some of them difficult to capture using an automated script.

Based on this inspection, 38 stations were rejected, corresponding to 6 % of the 624 time series. The main peculiarities discovered were sudden shifts in the concentration level, extreme peaks of short duration (1–2 hours), baseline offsets or changes with a factor of 2–3 in the general concentration level from one year to another. Two examples of such data are shown in Figure 3.4.

Following screening and selection there was a limited number of data sets available but the time series all surpassed minimum length and quality requirements. A more detailed investigation of

Figure 3.4 Examples of AirBase ozone time series rejected after visual inspection. The values shown are hourly ozone concentrations in µg/m³



Source: AirBase, 2008.

the technical data quality based on documented calibration of the ozone monitors, quality assurance, quality control procedures etc., should ideally be done but was not feasible within the present project.

Running ozone monitors that have been carefully calibrated against traceable standards and following specified procedures for technical maintenance may be less critical for day-to-day air quality monitoring. This quality assurance is crucial, however, for long-term trend studies. The lack of such information represents a limitation of the findings and conclusions presented here, adding an unknown level of uncertainty to the monitoring data and calculated trends.

Ozone trends were studied in the research project TROTREP, funded within the EU's Fifth Framework Programme (FP5). It has been concluded that 'data quality aspects are extremely important to trends, and especially to small trends, such as those of ozone' (Roemer, 2001).

3.3 Regional coverage of ozone stations with long-term data series

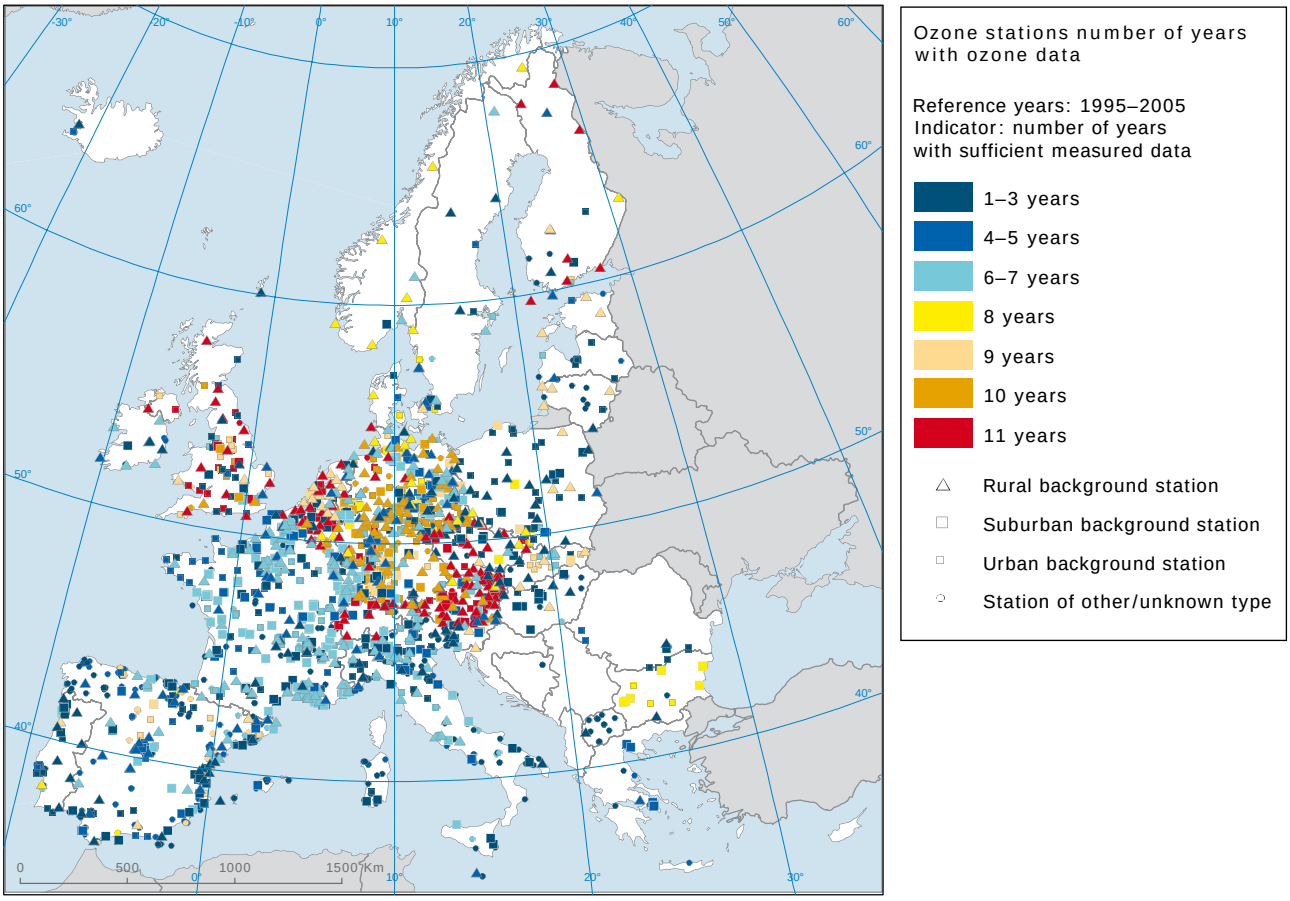
Data screening has another consequence. Map 3.1 shows an ozone station map, differentiating between sites with the required length of data and those with shorter time series. Map 3.1 shows that in large areas of Europe the ozone monitoring history reported to AirBase is shorter than what is required here for a meaningful trend study. This includes countries like France and Italy, as well as many regions in Spain, the Baltic States and eastern Europe.

As shown in Table 3.1, around one-third of the stations meeting the three criteria for selection were classified as background rural (198 sites) and their location is shown in Map 3.2. Such stations are the ones least sensitive to local conditions (roads, buildings, etc.) and also less influenced by the $NO + O_3$ reaction depleting ozone close to the emissions. They are therefore considered best suited for evaluating long-term trends on a regional scale. As Map 3.2 shows, these stations constitute a highly uneven coverage of Europe. Nearly

Table 3.1 Number of AirBase ozone stations after screening and selection of stations

	Rural	Suburban	Urban	Sum
Background	198	93	153	444
Industrial	12	26	14	52
Traffic	9	9	72	90
Sum	219	128	239	586

Map 3.1 Number of years with ozone data in the period 1995–2005



Note: Shades of blue indicate that the number of years is less than required (i.e. less than eight years) for the present trend study. Shades of red/yellow indicate a sufficient number of years.

Source: AirBase, 2008.

all are located in central Europe (Austria, the Czech Republic¸ Germany and Switzerland), the United Kingdom and the Nordic countries. The biased geographical coverage of the monitoring network limits this assessment and its conclusions to certain regions of Europe.

3.4 Trend calculations using AirBase data

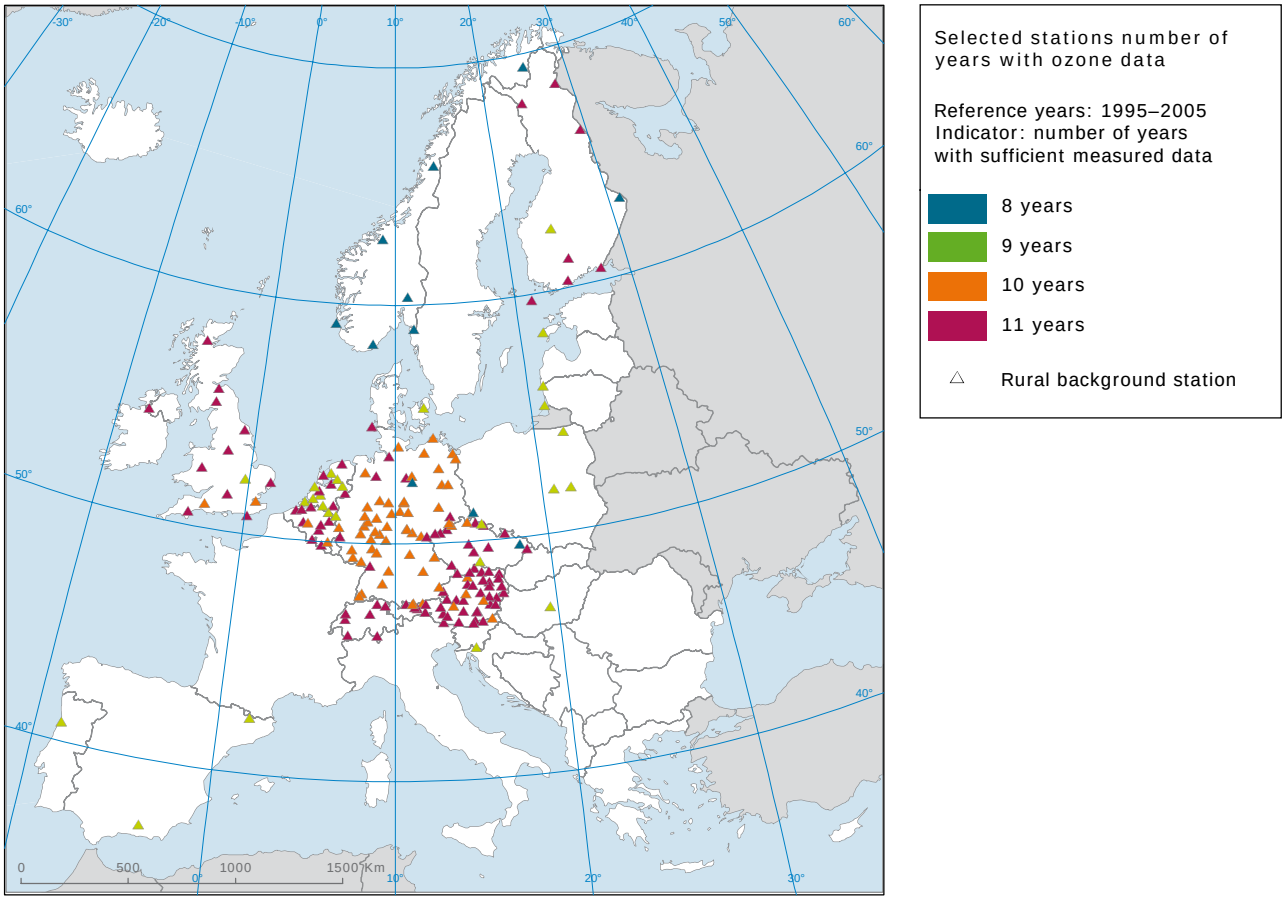
The present chapter sets out long-term trend analyses (standard linear regressions) based on the AirBase measurement data alone. Model-measurement studies are presented later in Chapter 5. The longest ozone time series in AirBase start in 1990, which is also the reference year for the Ozone Daughter Directive (EC, 2002). To evaluate the longest time series and compliance with the ozone directive rural background sites were identified with at least 75 % data capture for at least 14 of the 16 years from 1990 to 2005. Only a few sites fulfilled these criteria and national averages for various ozone metrics were thus calculated for four countries based on 22 sites in Austria, eight sites in the Netherlands, six sites in Switzerland and 10 sites in the United Kingdom.

Figures 3.5–3.8 show the national averages for:

- N_{180} (number of hours exceeding the information threshold of 180 µg/m^3);
- $N_{8hDM120}$ (number of days with an 8-hour maximum concentration exceeding 120 µg/m^3);
- AOT40 (accumulated exposure, May–July, over the threshold of 80 µg/m^3);
- MTDM (mean of the ten highest daily maximum concentrations during April–September).

The first three of these metrics are linked to parameters in the ozone directive (information

Map 3.2 Background rural ozone stations meeting the criteria for inclusion in this study



Source: AirBase, 2008.

threshold, target values, long-term objective). The last metric, MTDM, is included as an indicator of photochemical processes, i.e. it is used as a proxy for photochemical ozone formation. The results given in Figures 3.5–3.8 show differences among the countries and among the metrics.

For all metrics and countries, elevated values are seen in 2003 linked to the extreme summer conditions in Europe that year (Fiala *et al.*, 2003; Solberg *et al.*, 2008; Tressol *et al.*, 2008). Furthermore, the results in general indicate a decline in the ozone metrics during the first six or seven years and a stabilisation from 1998 onwards. The inter-annual variability is so large that a trend calculation only based on the years 1998–2005 seems meaningless (see also Section 4.3).

Using 1990 as a reference year (or 1992 for the Swiss sites) these results indicate:

N_{180} a marked reduction for British and Netherlands sites during 1990–1998 with a stabilisation thereafter and no apparent trend for the Swiss and Austrian sites;

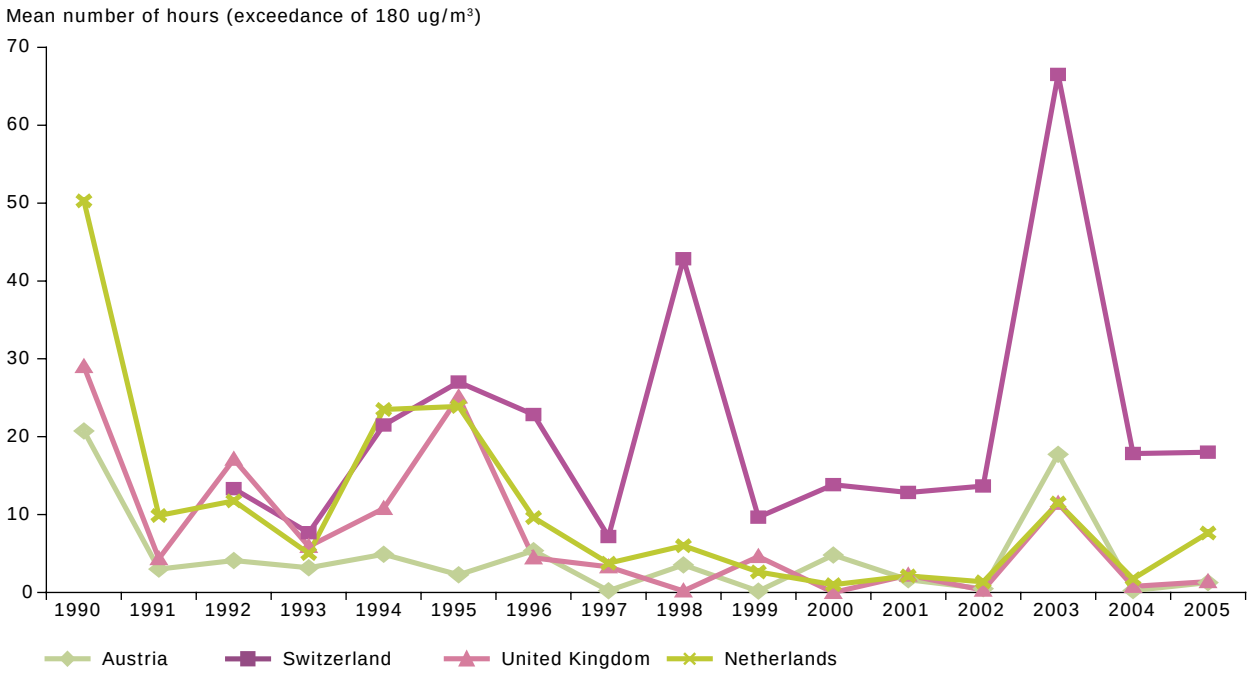
$N_{8hDM120}$ reductions for British, Netherlands and Austrian sites during 1990–1998 with uncertain trends thereafter and no apparent trend for the Swiss sites;

AOT40 marked reductions for the British and Netherlands sites during 1990–1998 followed by uncertain trends; no clear trends for the Swiss and Austrian sites but indications of increasing values in Switzerland during the last period;

MTDM a strong decline at British and Netherlands sites during 1990–1998 and uncertain trends after that; signs of reductions also at Austrians sites during 1990–1998 but no apparent trend for the Swiss sites.

Some words of caution should be given, however. The mean of the N_{180} parameter could be rather

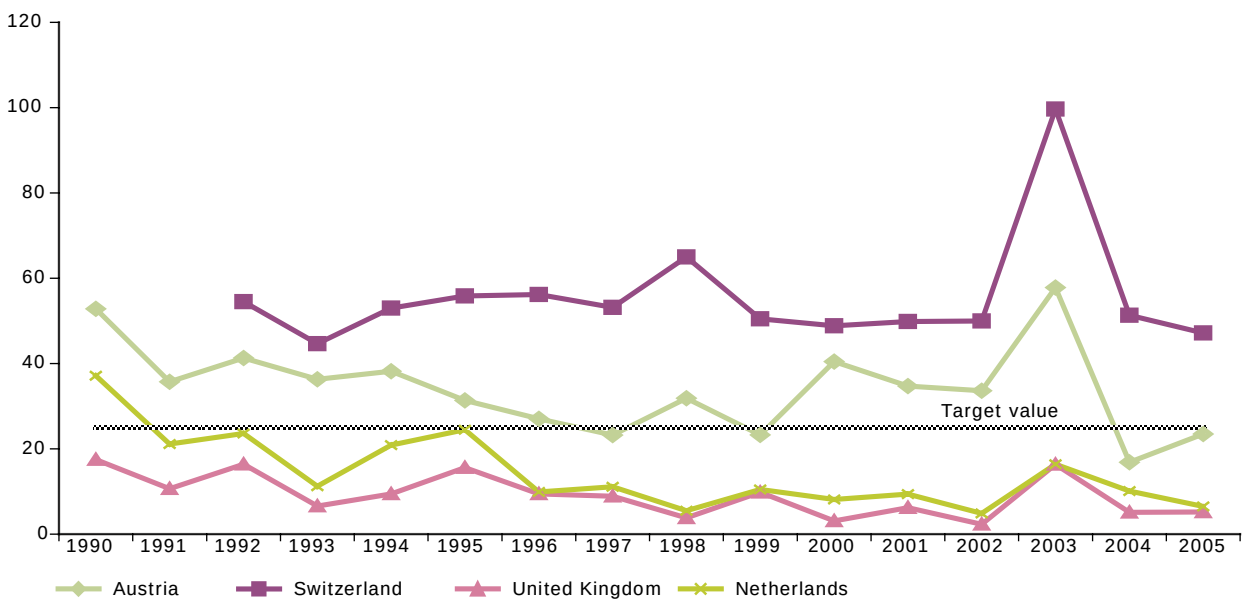
Figure 3.5 National mean hours exceeding the information threshold of 180 µg/m³ at rural background stations with nearly complete data coverage



Note: The numbers of sites with nearly complete data were as follows: Austria, 22 sites; the Netherlands, 8 sites; Switzerland, 6 sites; United Kingdom, 10 sites.

Source: AirBase, 2008.

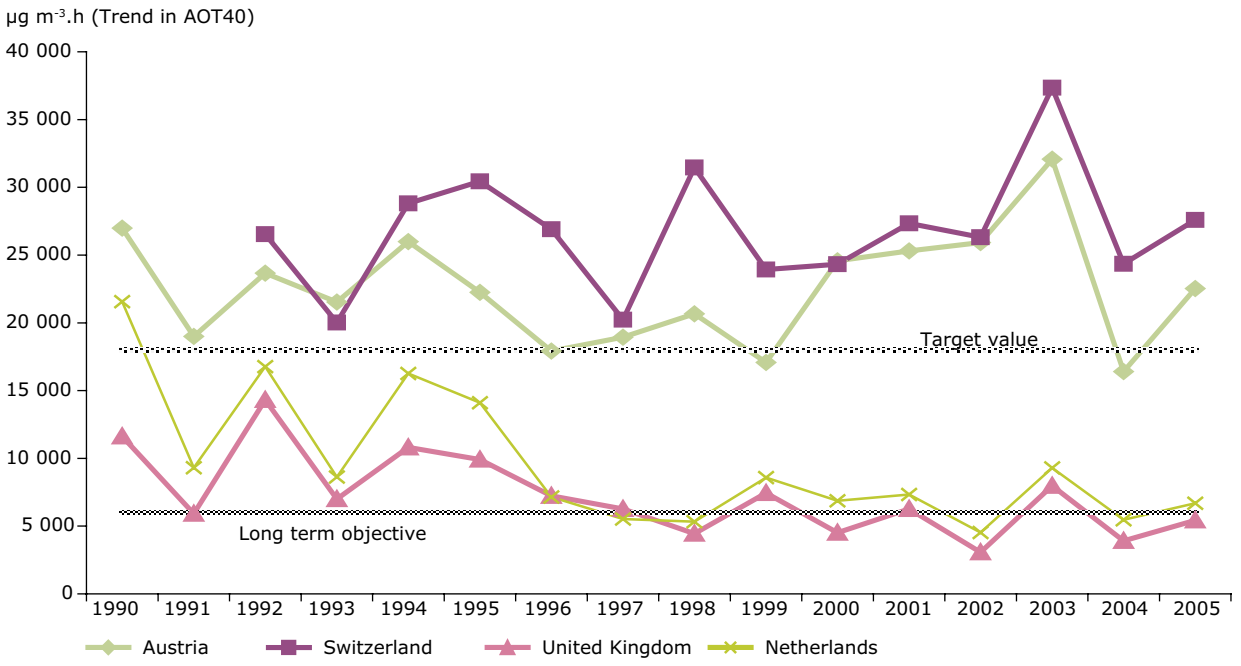
Figure 3.6 National mean days with a maximum 8-hour mean concentration exceeding 120 µg/m³ at rural background stations with nearly complete data coverage



Note: The numbers of sites with nearly complete data were as follows: Austria, 22 sites; the Netherlands, 8 sites; Switzerland, 6 sites; United Kingdom, 10 sites. The target value of 120 µg/m³ may be exceeded up to 25 days a year.

Source: AirBase, 2008.

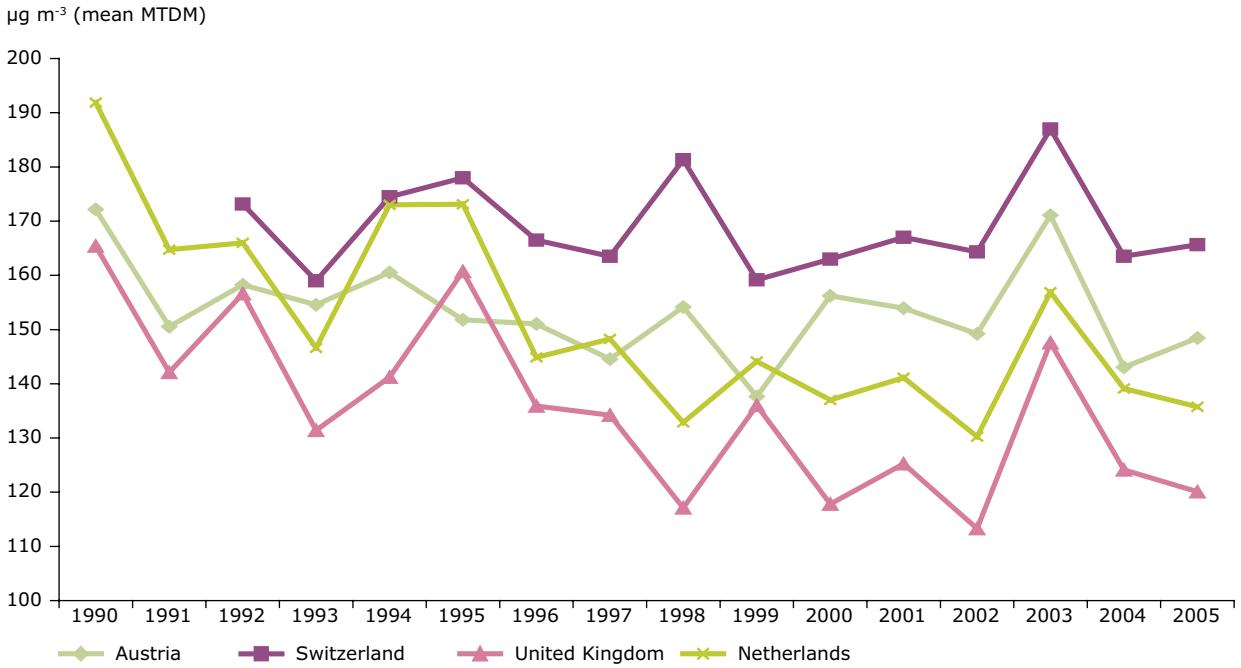
Figure 3.7 National mean AOT40 at rural background stations with nearly complete data coverage



Note: The numbers of sites with nearly complete data were as follows: Austria, 22 sites; the Netherlands, 8 sites; Switzerland, 6 sites; United Kingdom, 10 sites. The target value given in the ozone directive is 18 000 µg/m³.h and the long-term objective is 6 000 µg/m3.h.

Source: AirBase, 2008.

Figure 3.8 National mean MTDM at rural background stations with nearly complete data coverage



Note: The numbers of sites with nearly complete data were as follows: Austria, 22 sites; the Netherlands, 8 sites; Switzerland, 6 sites; United Kingdom, 10 sites.

Source: AirBase, 2008.

meaningless in a country with large variations in ozone concentrations at different stations, for example the United Kingdom. If many stations do not record any ozone concentrations above the threshold during a summer then averaging over all stations would indicate a smaller 'mean trend' than the trend at the stations with $N_{180} > 0$. In 2003 two of the ten British sites in this analysis exceeded the 180 µg/m^3 threshold more than 40 times whereas four of the sites never exceeded the threshold.

The differences among the stations were particularly large in Switzerland. As an example at CH0033A (Magadino) the information threshold was exceeded 127 times in 1996 and 173 times in 2003, whereas at CH0024A (Saxon) the threshold was never exceeded in either year. Both sites are classified as background rural and are located a little over 100 km apart. This shows that there could be substantial differences among stations on a fairly fine spatial scale even for background rural sites.

A statistically significant linear downward trend was found for the period 1990–2005 for the means of the British sites and of the Netherlands sites for all four ozone indicators discussed above. The means of Austrian sites and of Swiss sites showed no significant trend for any of the four indicators. Non-significant negative slopes (downward trends) were calculated, though, for $N_{8hDM120}$ and MTDM for both Austria and Switzerland. Non-significant positive slopes were found for AOT40 and mixed results for N_{180}. There is however, no reason to expect linearity with time in these parameters as the emissions themselves have not dropped linearly (see discussion above) and as there are large interannual fluctuations due to meteorology.

4 Regional EMEP model scenarios

4.1 Model scenario description

The EMEP Eulerian photochemistry model has a polar stereographic projection with a horizontal resolution of 50 km^2 true at 60°N and 20 vertical layers below 100 hectopascals (hPa). The model domain is centred over Europe and also includes most of the North Atlantic and the polar region. The EMEP model uses three-hourly resolution meteorological input data from a dedicated version of HIRLAM (High Resolution Limited Area Model). The model was run using EMEP/CLRTAP emissions inventory data, where available. In some areas where this was not the case expert estimates were used, i.e. for emissions from international shipping (for more details see Chapter 2).

The model is publicly available and can be downloaded from the EMEP website. A comprehensive description of the model is also available there. A model description and its application to an ozone trend study is included in Jonson *et al.* (2006).

Initial and lateral boundary conditions, especially of ozone, represent key inputs to the EMEP model, in particular when used for ozone trend studies. For ozone, these boundary conditions are derived from the three-dimensional ozone climatology of Logan (1999), modified in order to accommodate inter-annual variability in air masses arriving from the upwind Atlantic region. The modifications are based on the measurements at Mace Head, Ireland. Those measurements are filtered using trajectory analysis to obtain clean sector O_3 values. The adjustment (in ppb ozone) in lateral boundary concentrations is the same for the whole model domain and at all vertical levels. As ozone mixing ratios generally increase with altitude, the relative adjustment is largest near the surface and becomes very small in the upper free troposphere. This procedure was chosen since data from Mace Head cannot in principal be used to correct mid- to upper-tropospheric ozone. The Mace Head adjustment was also used in Jonson *et al.* (2006).

The assumption that all lateral boundaries are affected equally by the Mace Head adjustment is rather crude but works well because the main bulk of the model domain is subject primarily to dominating westerly winds. Lateral boundary concentrations are also specified for other (air pollutant) species (see the model description at the EMEP website for details).

The EMEP model was used for this report to perform multi-annual calculations and sensitivity runs for ozone and other air pollutants for Europe.

The purpose of applying the EMEP model was:

- to estimate ozone statistics (air concentrations and aggregated quantities such as AOT40, SOMO35) for many years;
- to evaluate the variability in these statistics due to certain main processes such as meteorological variability and anthropogenic emission changes;
- to calculate long-term trends in these statistics and uncertainty in such trends;
- to determine the agreement between modelled and observed data.

A set of model scenarios was defined, which are explained in more detail in Table 4.1.

As Table 4.1 indicates, the scenario REF was the reference scenario based on actual emissions and boundary conditions varying from year to year.

The E90 scenario was included to estimate air pollution changes due to anthropogenic emission changes since 1990, the base year for the EU ozone directive (EC, 2002).

The E95 scenario was included mainly to allow a detailed comparison between model scenarios and observations because most measurements at the sites included in AirBase started after 1995.

The NOBIO scenario was run to estimate the importance of biogenic isoprene emissions, and the FIXBND to estimate the importance of inflowing pollutants from outside the model domain (i.e. Europe). The last two scenarios refer to

Table 4.1 EMEP model scenarios used in this study

Scenario	Description	Years
REF	Standard model runs	1990, 1995–2005
E90	Same as REF but with anthropogenic emissions as in 1990	1990, 1995, 2000, 2005
E95	Same as REF but with anthropogenic emissions as in 1995	1995–2005
NOBIO	Same as REF but without biogenic isoprene emissions	1995–2005
FIXBND	Same as REF but with ozone boundary concentrations fixed to the mean values for the years 1990–2000.	1995–2005

'external processes', not influenced by the European countries emission abatement policies.

The EMEP model runs produced daily and annual values, such as mean and max atmospheric concentrations of O_3, NO_2, NO_x and a number of other trace compounds, as well as aggregated quantities such as AOT40 and SOMO35. Note that the modelled AOT40 presented in this report should not be compared directly with AOT40 calculated from the measurements as the modelled values are based on ozone concentrations interpolated to crop height (normally 1 m) and tree height (normally 20 m). On the other hand the measurement of ozone is normally done at 2 m height. This difference in height could be important for ozone due to effective dry deposition.

Only output for ground level was taken out, i.e. the lowest layer in the model. For components subject to surface dry deposition the concentrations were reduced to 2 m height through a standard module in the EMEP model, taking into account vertical transport and turbulent diffusion between the ground and the lowermost model layer. The reason for not extracting hourly and vertically distributed data was extensive data storage and handling. Furthermore, the diurnal cycle at a monitoring site is largely controlled by local, sub-grid, scale effects not well captured in a regional scale photochemical CTM. Hourly model data would therefore provide limited benefits.

4.2 Modelled trends in ground-level ozone concentrations

Key messages

- The model predicts maximum mean values of $AOT40_c$, $AOT40_{df}$ and SOMO35 in southern Europe, particularly in Italy and the Mediterranean area.
- Reductions in these ozone statistics of the order of 20–30 % are predicted due to the anthropogenic emission reductions during 1995–2005. The strongest decline is modelled for $AOT40_c$, amounting to some 30–50 % in parts of France, Germany and elsewhere in central Europe.

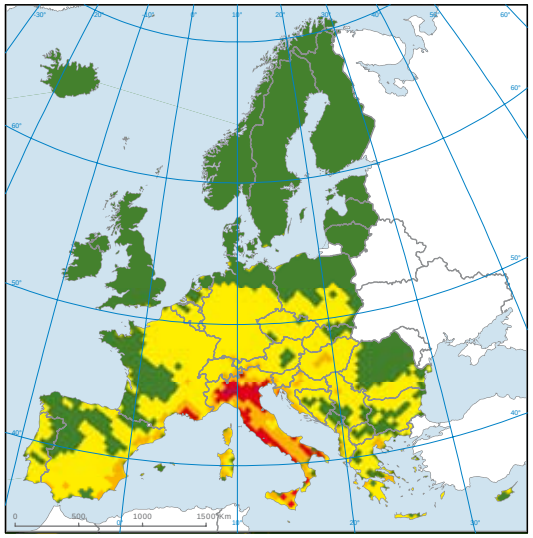
The modelled mean $AOT40_c$, $AOT40_{df}$ and SOMO35 for the period 1995–2005 is given in Map 4.1 together with the modelled reduction in these metrics in 2005 due to the European anthropogenic emission reductions during 1995–2005. The modelled reductions are given by the plain difference E95(2005) — REF(2005). It should be noted that the difference in ozone parameters will vary annually due to the emission trend and also the varying influence of photochemical ozone formation from one year to another. The change in total emissions of ozone precursors from 2004 to 2005 was fairly small (Figure 2.1) however 2005 was a more 'ozone potent' year with emission changes producing a larger effect than in 2004.

The mean modelled $AOT40_c$ shows a maximum of around 30 000 µg/m³.h in northern Italy and less than 6 000 µg/m³.h in Scandinavia and the United Kingdom ([12]). The modelled AOT40df shows maximum values of 60 000–100 000 µg/m³.h in southern and central Europe, while the modelled fields of SOMO35 show a maximum of 12 000–15 000 µg/m³.h, mainly in Italy.

The model predicts significant reductions in all three ozone metrics for 2005 based on the reported emission changes during 1995–2005. The strongest relative reductions are estimated for $AOT40_c$. The model indicates a reduction in $AOT40_c$ of some 10 000 µg/m³.h in northern Italy and 4 000–7 000 µg/m³.h in

([12]) The target value given in the ozone directive is 18 000 µg/m³.h and the long-term objective is 6 000 µg/m³.h.

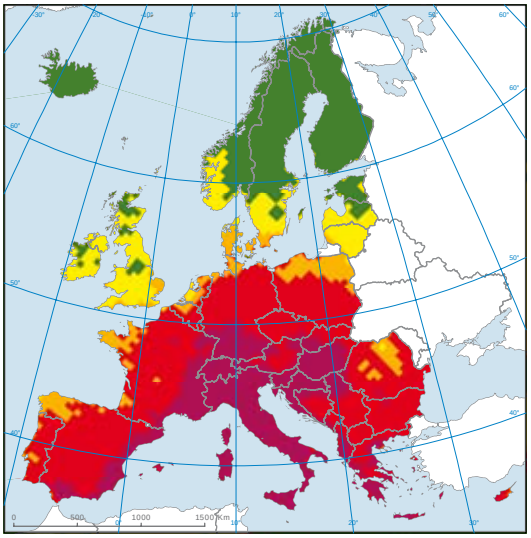
Map 4.1a Modelled mean $AOT40_c$, $AOT40_{df}$ and SOMO35 for 1995–2005



Ozone — AOT40
protection of crops

Reference year: 1995–2005 average
EMEP model output

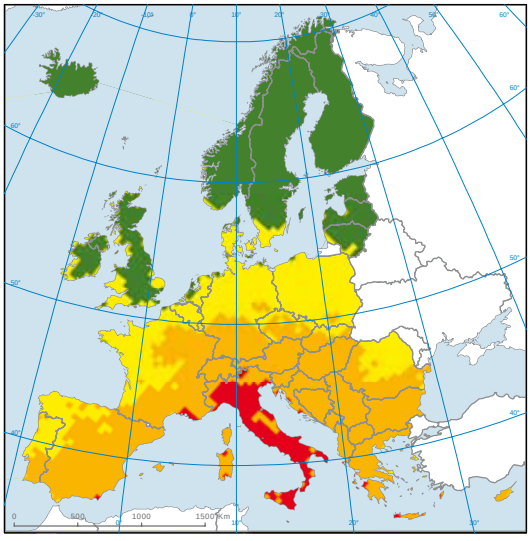
- < 6 000 µg.m^{-3}.h
- 6 000–12 000 µg.m^{-3}.h
- 9 000–18 000 µg.m^{-3}.h
- 18 000–27 000 µg.m^{-3}.h
- > 27 000 µg.m^{-3}.h
- Countries excluded from study



Ozone — AOT40
protection of forests

Reference years: 1995–2005 average
EMEP model output

- < 10 000 µg.m^{-3}.h
- 10 000–20 000 µg.m^{-3}.h
- 20 000–30 000 µg.m^{-3}.h
- 30 000–50 000 µg.m^{-3}.h
- > 50 000 µg.m^{-3}.h
- Countries excluded from study



Ozone — SOMO35

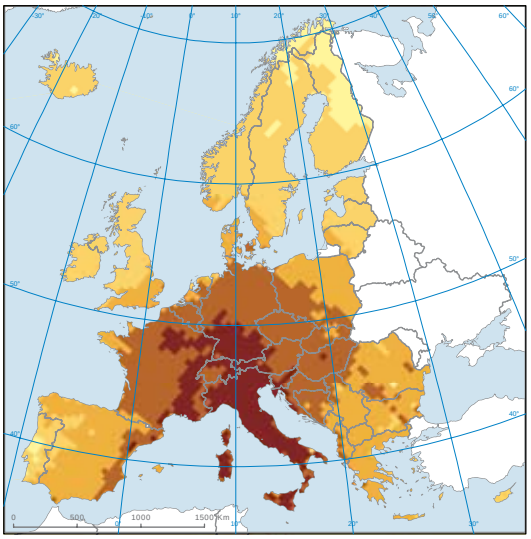
Reference years: 1995–2005 average
EMEP model output

- < 3 000 µg.m^{-3}.d
- 3 000–6 000 µg.m^{-3}.d
- 6 000–10 000 µg.m^{-3}.d
- 10 000–15 000 µg.m^{-3}.d
- > 15 000 µg.m^{-3}.d
- Countries excluded from study

Note: All values are in µg/m^3 hours. The colour scales differ.

Source: EMEP, 2008.

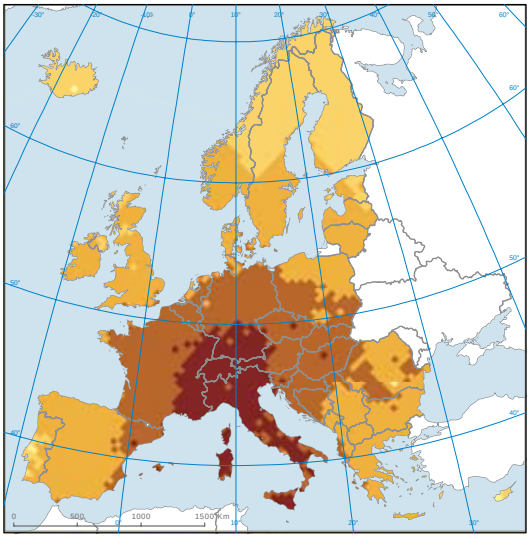
Map 4.1b Calculated change between 1995 and 2005 due to anthropogenic emission reductions in Europe during that period



Ozone — AOT40
protection of crops

Reference years: 1995–2005
Indicator: difference E95-REF
EMEP model output

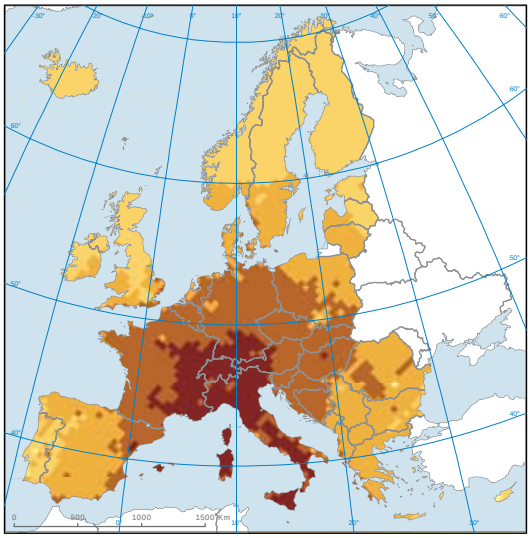
- < 0 µg.m^{-3}
- 0–1 000 µg.m^{-3}
- 1 000–3 000 µg.m^{-3}
- 3 000–6 000 µg.m^{-3}
- > 6 000 µg.m^{-3}
- Countries excluded from study



Ozone — AOT40
protection of forests

Reference years: 1995–2005
Indicator: difference E95-REF
EMEP model output

- < 0 µg.m^{-3}
- 0–2 000 µg.m^{-3}
- 2 000–10 000 µg.m^{-3}
- 10 000–20 000 µg.m^{-3}
- > 20 000 µg.m^{-3}
- Countries excluded from study



Ozone — SOMO35

Reference year: 2005
Indicator: difference E95-REF
EMEP model output

- < 0 µg.m^{-3}
- 0–500 µg.m^{-3}
- 2 000–1 000 µg.m^{-3}
- 10 000–2 000 µg.m^{-3}
- > 2 000 µg.m^{-3}
- Countries excluded from study

Note: All values are in µg/m^3 hours. The colour scales differ.

Source: EMEP, 2008.

parts of central Europe, corresponding to a relative reduction of 30–50 % compared to the 11 year average.

$AOT40_{df}$ reductions of some 25 000 μg/m³.h are calculated for northern Italy and Switzerland and around 12 000–15 000 μg/m³.h in central Europe (Czech Republic, France and Germany), corresponding to around 20–30 % of the mean value for 1995–2005.

The modelled reduction in SOMO35 is of the order of 2 000–3 000 μg/m³.h in northern Italy and southern France and 1 000–2 000 μg/m³.h elsewhere in central Europe, corresponding to 20–30 % of the mean for 1995–2005. For all three ozone metrics the model indicates little reduction in Spain, presumably reflecting the smaller emission changes there (Figure 2.2 and Figure 2.3).

4.3 Variation in ozone statistics due to meteorological variability

Key messages

- The meteorologically induced variability in AOT40 and SOMO35 was estimated and revealed a gradient decreasing from north-west to south-east Europe.
- Variability in $AOT40_c$ of approximately 10 % was estimated for parts of southern Europe, increasing to 20–30 % in central Europe and to 50 % or more in the United Kingdom, the Nordic countries and Russia. Similar patterns with lower values were found for $AOT40_{df}$ and SOMO35.
- The magnitude of emission trend's impact on ozone levels relative to the effect of meteorological variability indicates that the trend would only be discernible in certain regions of central Europe, most notably northern Italy.
- The findings above refer to the modelled data. The chances of separating emission trends from meteorological variability will be less for measured data.

The influence of meteorological variability on surface ozone levels is well known. Primary emissions, such as NO_x and CO, show a more direct link between changes in emissions and atmospheric concentrations, although there is also a link to meteorology through the efficiency of mixing and advection. Secondary pollutants, like ozone and particulate matter (PM) are controlled by the interplay of emissions and meteorology.

There are several processes responsible for this link. Increased temperatures speed up most of the chemical reactions in the atmosphere producing ozone; high-pressure situations may create soil drought and thereby reduce surface deposition of ozone; elevated temperatures may enhance biogenic emissions leading to higher ozone production; and subsidence may trap the photochemical pollution in the planetary boundary layer for an extended time (Solberg *et al.*, 2008). Reduced absolute humidity (during high-pressure situations) may lower ozone formation through less OH radicals giving less oxidation of VOC while increasing the lifespan of NO_x and thereby leading to more ozone formation.

The links between ozone and meteorological variability are complex and in reality require a chemical transport model (CTM) to provide meaningful relationships. The substantial year-on-year variation in prevailing meteorological conditions in Europe is a major challenge for evaluating improvements in ozone exposure due to abatement policies for NO_x and NMVOC. A simple year-on-year reduction in European ozone concentration levels may not take place despite a year-on-year reduction in precursor emissions.

This is without doubt the main problem in any ozone assessment study. It has often been addressed either by focussing on the results of model predictions (Jonson *et al.*, 2006) or measurements alone (Oltmans *et al.*, 2006). Some studies (e.g. Ordóñez *et al.*, 2005) have analysed ozone with respect to local meteorological conditions. Such methods may be fruitful for certain regions but will often encounter problems when applied to Europe as a whole.

In the following meteorologically induced variability in ozone is quantified and then compared to the predicted long-term trend in ozone caused by emission changes. The basic idea is to estimate how likely it is to be able to distinguish the emission induced trends from natural meteorological variability. Intuitively, this is linked to the length of the observational time series. With a short time series, the emission induced trend is masked by the meteorological variation, while with a long time series it is more likely that emissions will stand out from natural variability.

The variability of certain annual ozone metrics due to meteorology was estimated by calculating the gridded mean and standard deviation of these metrics for the years 1995–2005 using the E95 scenario. As described above, the E95 scenario

is run with the same annual set of emission data (valid for 1995). Thus the mean (*xm*), standard deviation (*std*) and relative standard deviation (*rstd*=*std*/*xm*) express the mean and variability from the interannual variations in meteorology alone, independent of emission changes. As noted in Section 2.5, the gridding of the emissions for the year 2005 differs from the other years and this will affect calculated ozone for this year. However the effect should be small compared to the effects of meteorological variability.

The resulting relative variations, *rstd* (expressed in per cent), for $AOT40_c$, and SOMO35 based on the years 1995–2005 are shown in Map 4.2. Areas with a high relative variation in this parameter indicate areas where the changes in meteorology from year to year have a decisive influence on the statistic.

In general these results show a north to south gradient. This is explained by the fact that in general ozone concentrations in the north are closer to the threshold values of 80 and 70 µg/m^3 (AOT40 and SOMO35) than in central Europe. For $AOT40_c$ the meteorological variability for 1995–2005 measured by *rstd* is approximately 50 % or more of the mean in the Nordic countries, Russia and the United Kingdom. In western France, Portugal and Spain the variability is also high at 40–50 %. In central Europe the variability is 20–30 % for the period 1995–2005, while in coastal areas of parts of southern Europe the variability is the lowest at only around 10 %.

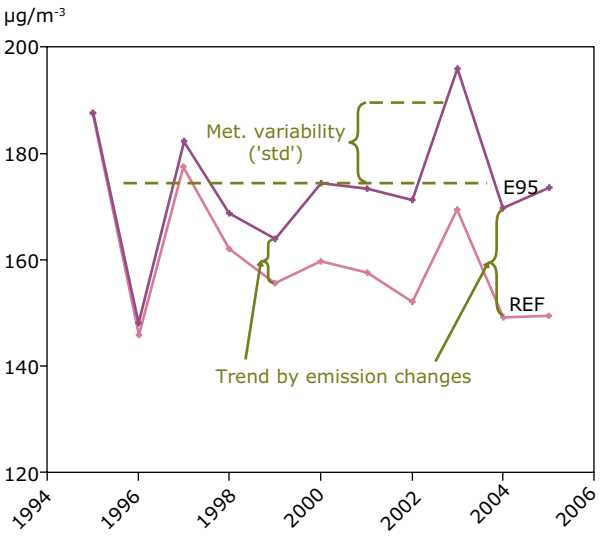
The $AOT40_{df}$ values (not shown) present a somewhat different regional distribution and generally lower values. In the Nordic countries, United Kingdom and western France the estimated variability in $AOT40_{df}$ due to meteorology is 20–30 %. In parts of southern Europe the variability is less than 10 %.

The meteorological variability of SOMO35 (Map 4.2) shows a pattern similar to $AOT40_{df}$ for forests but with even lower values. In large parts of the continent the relative variability is 10 % or less and lies between 10 % and 20 % in Denmark, western France and western Germany. In the Nordic countries and the United Kingdom the relative variability is 20–30 % except for the far north where it is 50 % or higher.

These model results give an indication of the expected range of 'natural variability', i.e. variability in these metrics caused by inter-annual variations in meteorology in different parts of Europe. One should keep in mind that this is based on the period 1995–2005 only, which is a very short period for assessing meteorological variability in a climate perspective. Furthermore, it contains the year 2003, which was an outlier in terms of weather and air pollution in Europe with a summer that was very likely the warmest in the last 500 years (Luterbacher *et al.*, 2004). Within weather forecasting 30-year periods are used as the basis for calculating trends.

The modelled trend in the ozone metrics caused by anthropogenic emission changes during 1995–2005 was compared to the natural variability estimated above. The linear trend in the ozone metrics due to the emission changes was estimated in every grid point by performing a least squares linear regression on the differences in the annual ozone metrics for the scenario difference REF – E95 as exemplified in Figure 4.1. As the ozone response due to emission changes will not necessarily be linear with time, grid points with a linear correlation coefficient (r2) less than 0.9 were marked as non-significant in the analyses. Map 4.3 shows the modelled linear trend estimate for 1995–2005 divided by the estimated meteorological variability in these statistics, expressed by the standard deviation (2-σ).

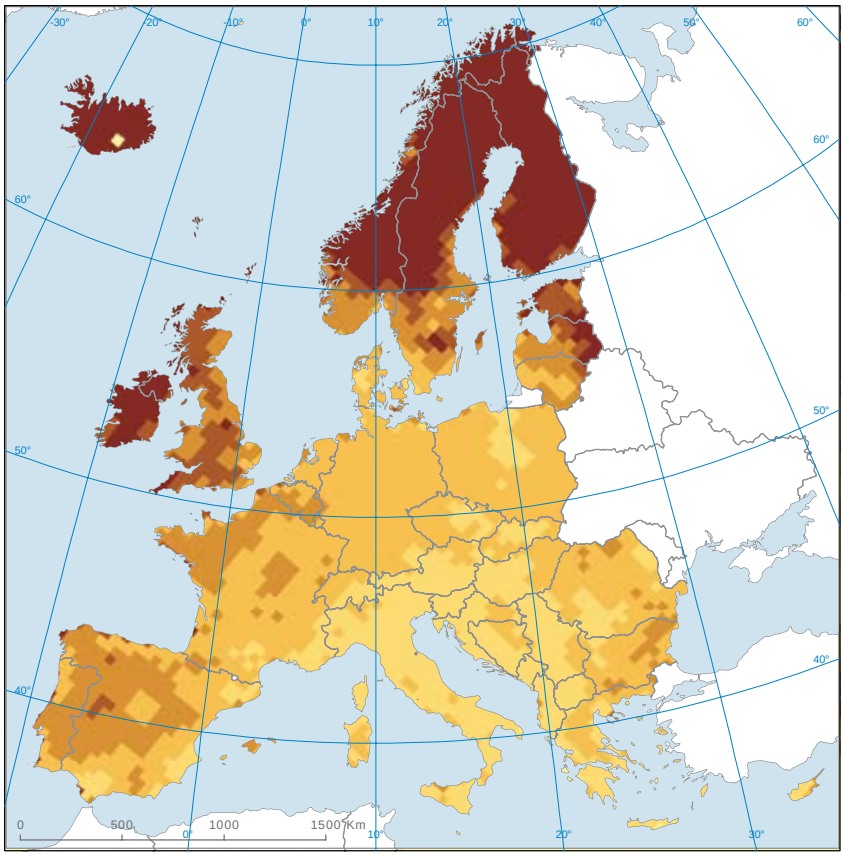
Figure 4.1 Example of the separation between meteorological variability as inferred from the standard deviation of the E95 scenario (red curve), and the trend due to emission changes as inferred from the difference between the REF (blue curve) and E95 scenarios



Note: The example plot shows the calculated values for MTDM for a site in Belgium.

Soure: EMEP, 2008.

Map 4.2 Relative annual variability (standard deviation / mean) of ATO40 and SOMO35 induced by meteorological variability, 1995–2005

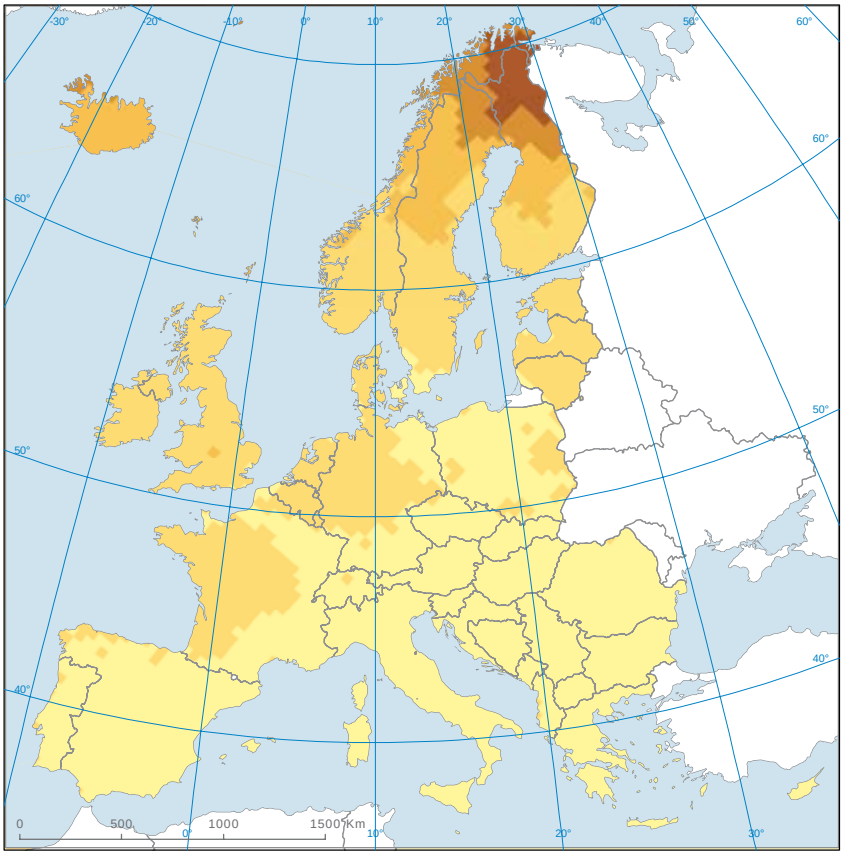


Ozone — AOT40
protection of crops

Reference years: 1995–2005
Indicator: relative standard deviation
(constant 1995 emissions)
percentage non fixed
boundary conditions
EMEP model output

< 10 %
10–20 %
20–30 %
30–40 %
40–50 %
> 50 %

Countries excluded from study



Ozone — SOMO35

Reference years: 1995–2005
Indicator: relative standard deviation
(constant 1995 emissions)
EMEP model output

< 10 %
10–20 %
20–30 %
30–40 %
40–50 %
> 50 %

Countries excluded from study

Source: EMEP, 2008.

$$f = b/2s$$

where

b = slope calculated from a linear least squares regression of X_y(E95) – X_y(REF) y = [1995, …, 2005]

X_y = annual ozone metric (AOT40, SOMO35, …) year y

s = 1σ st. dev. of Xy(E95), y = [1995, …, 2005]

These results express the magnitude of the modelled emission induced trend relative to the natural variability and thus indicate the chance of being able to detect an emission induced trend. It should be stressed that the criteria used ($r2 > 0.9$; using the 2-σ for the meteorological variability; etc.) were subjectively chosen and not part of a rigorous statistical analysis.

The results suggest that the expected trend in the ozone statistics due to emission reductions will only be distinguishable from the natural variability in certain areas of central Europe. The values in Map 4.3 indicate that the signal from the emission reductions should be most pronounced in northern Italy (the Po Valley) and discernible from the natural variability in Italy, Switzerland, Austria, eastern parts of France and southern parts of Germany and the Czech Republic. In parts of Belgium and Hungary the modelled trend is of the same order as the meteorological variability. The values are generally higher for SOMO35 than for AOT40, indicating that any emission induced trend should be easier to detect and separate from the natural variability for SOMO35 than for AOT40. This reflects the large meteorological variability of AOT40 compared to SOMO35 (Map 4.2).

It is important to note that the analyses presented here only concern the modelled data, not the measurements. The plots shown in Map 4.3 indicate the chance of being able to see the signal from the emission-induced trend on certain ozone metrics given the meteorological variability, based on the modelling data alone. It is well known, however, that the agreement between modelled and measured ozone metrics such as AOT40 is often poor, and highly sensitive to small offsets in absolute ozone levels. Thus, the chance of being able to distinguish the emission trend from the meteorological variability by consulting measurement results is likely to be less than indicated by Map 4.3. This has not been quantified here.

4.4 Factors outside the influence of European emission reduction measures

Key messages

- Model calculations indicate that the biogenic isoprene emissions represent a major uncertainty in AOT40 with a factor of at least two, while less so for SOMO35.
- The model indicates that changing boundary concentrations ([13]) of ozone based on the Mace Head correction have a relatively small influence on AOT40 and SOMO35 in most parts of Europe (less than 10 %). In Scandinavia, the United Kingdom and western parts of the continent the influence could be larger.

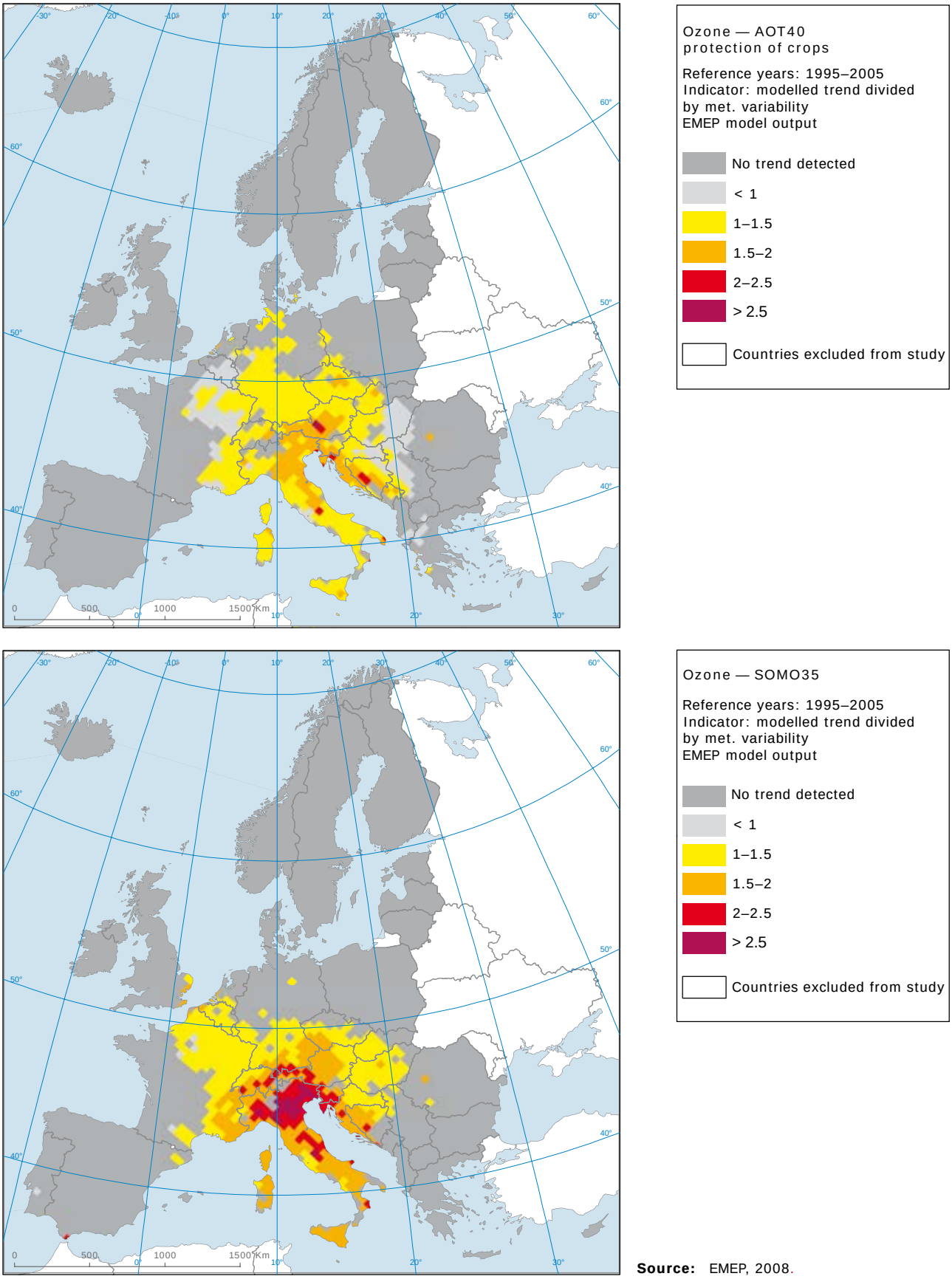
Anthropogenic emissions are one source of ozone observed at ground sites in Europe. However, biogenic emissions of precursors as well as the baseline ozone concentration transported into Europe are also important, as discussed by e.g. Jonson *et al.* (2006). The magnitude of these processes is of interest as they are outside the control of EU emission abatement legislation. The scenarios NOBIO and FIXBND, described above, were used to estimate these influences.

Additionally, there are further processes outside the emission abatement directives and protocols that will have an effect on European ozone levels which have not been studied here. Forest fires and biomass burning, either within Europe or on other continents, is known to have an important influence on ozone during episodes (e.g. Stohl *et al.*, 2007).

The possible role of forest fires and biomass burning events for European ozone concentrations has not been considered in the present study. Both intercontinental transport of fire plumes and European fires could contribute significantly to ozone formation during episodes. The importance of such incidences in different regions of Europe for the ozone indicators specified by the Ozone Daughter Directive is not addressed by EU air quality legislation, however.

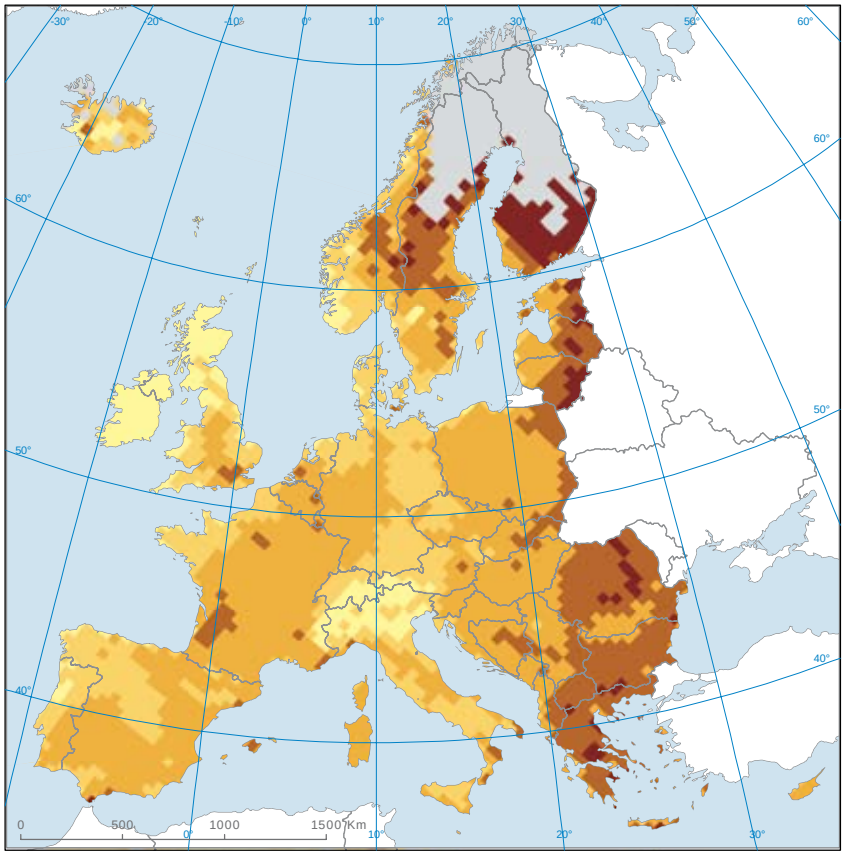
([13]) Here referring to the inflow of pollutants from outside of the model domain (i.e. from outside of Europe). The definition of these conditions in the EMEP model is *inter alia* based on ozone measurements at one monitoring site (Mace Head in Ireland).

Map 4.3 Ratio of the emission-induced trend relative to the variability induced by meteorology (expressed by the standard deviation) of two ozone statistics during 1995–2005



Source: EMEP, 2008.

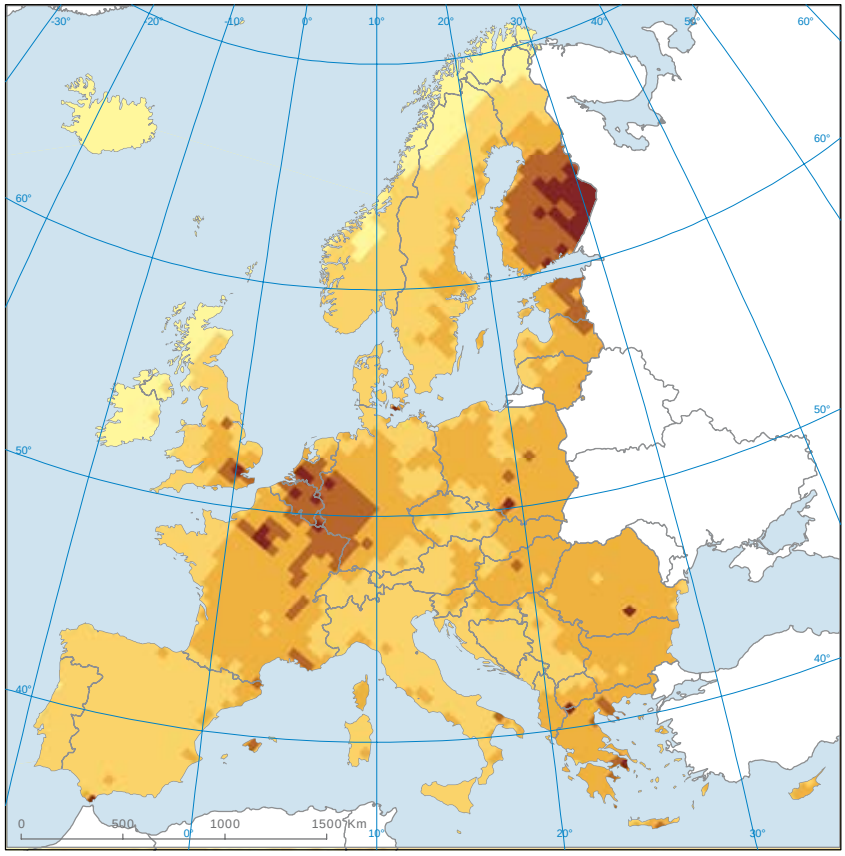
Map 4.4 **Modelled mean relative difference, (REF-NOBIO)/REF, in AOT40c (top) and SOMO35 (bottom) caused by neglecting all biogenic isoprene emissions, 1995–2005**



Ozone — AOT40
protection of crops

Reference years: 1995–2005
Indicator: mean percentage caused by biogenic emissions
EMEP model output

- 0–30 %
- 30–40 %
- 40–50 %
- 50–60 %
- 60–100 %
- No AOT40 modelled
- Countries excluded from study



Ozone — SOMO35

Reference years: 1995–2005
Indicator: mean percentage caused by biogenic emissions
EMEP model output

- 0–10 %
- 10–20 %
- 20–25 %
- 25–30 %
- 30–100 %
- Countries excluded from study

Note: The scales differ for the two maps.

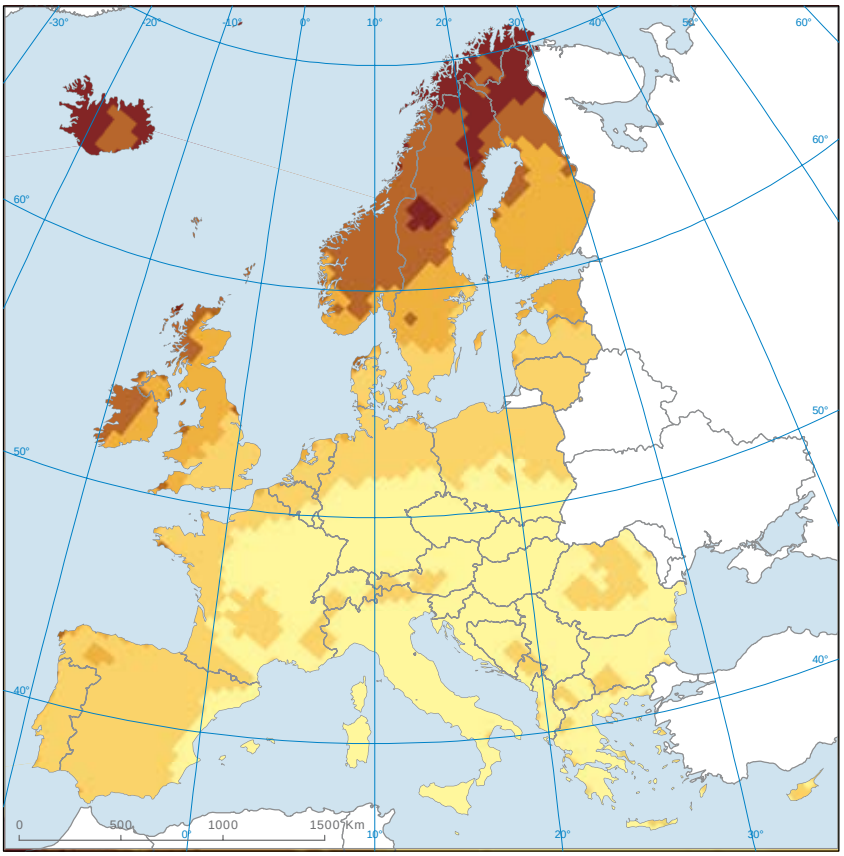
Source: EMEP, 2008.

Map 4.5 Modelled mean relative difference, (REF-FIXBND) / REF, in $AOT40_c$ (top) and SOMO35 (bottom) for 2003 caused by keeping the lateral boundary concentrations fixed

Ozone — AOT40
protection of crops

Reference years: 1995–2005
Indicator: mean percentage
non fixed boundary conditions
EMEP model output

- < 0 %
- 0–2 %
- 2–5 %
- 5–10 %
- 10–30 %
- No AOT40 modelled
- Countries excluded from study



Ozone — SOMO35

Reference year: 2003
Indicator: percentage non fixed
boundary conditions
EMEP model output

- < 0–5 %
- 5–10 %
- 10–15 %
- 15–20 %
- 20–40 %
- Countries excluded from study

Note: The scales differ for the two maps.

Source: EMEP, 2008.

Biogenic isoprene emissions

The modelled mean influence of biogenic isoprene emissions for AOT40 and SOMO35 for 1995–2005 is shown in Map 4.4. These results suggest that isoprene has a substantial influence on $AOT40_c$ and contributes approximately 30–50 % in large parts of Europe. Only in Ireland and north-western United Kingdom is a significantly lower influence (10–20 %) found.

The modelled influence on SOMO35 is, on the other hand, much lower at 10–30 % in most areas. The main reason for this difference is that the SOMO35 metrics are based on the whole year, whereas $AOT40_c$ is only based on daytime hours during three summer months when the isoprene emissions are peaking.

With uncertainty in the European biogenic isoprene emissions of a factor of 3–5 (Simpson *et al.*, 1999) these results indicate that the biogenic emissions constitute also a major uncertainty in the modelled $AOT40_c$ with a factor of at least two in the calculated AOT values. One should be careful, though, with statements about the contribution of biogenic emissions to ozone formation in Europe. The ozone formation process is non-linear and the sum of contributions from individual precursors calculated in this way would sum up to much more than 100 % in a sensitivity exercise. Viewed in this way, it could be misleading to state that 50 % of the AOT40 is due to biogenic emissions.

Intercontinental transport of ozone

The modelled difference in $AOT40_c$ and SOMO35 caused by keeping the lateral boundary concentrations of ozone fixed is indicated in Map 4.5. This shows the calculated change in the ozone parameters for one single year, 2003, which had generally higher ozone background concentrations as measured at Mace Head than the average. As documented also by Jonson *et al.* (2006), boundary conditions have a fairly minor influence on $AOT40_c$ and SOMO35 except in the Nordic countries and north-west United Kingdom. The calculated difference in these statistics using the true 2003 background compared to using a 10 year mean background (Table 4.1) indicates less than 10 % change in large parts of central Europe.

For AOT40 the calculated difference is 10–30 %, however, for Scandinavia, the United Kindom and the Nordic countries. For SOMO35 the calculated differences are generally smaller than for AOT40. This indicates the uncertainty range of AOT40 and SOMO35 when applying the so-called Mace Head correction in the model. It is important, however, to be aware of the limitations of the Mace Head correction. Intercontinental transport events, e.g. biomass fire plumes, may occur above the Mace Head station. This is not discussed further here.

The calculated difference in $AOT40_c$ and SOMO35 is only a few per cent in most parts of the continent, surely less than other uncertainties in these metrics. For the major inflow regions (Iceland, Ireland, Scandinavia and Scotland) $AOT40_c$ and SOMO35 is estimated to be approximately 10 % lower than the reference with the FIXBND scenario.

5 Modelled and measured trends

Key messages

- The overall model results agree better with the measured values when assuming (changes in) actual annual emission data than when assuming constant (1995) emissions for the whole period 1995 to 2005.
- The measured trend in MTDM (mean of the ten highest daily max concentrations during summer) show a mixed pattern in Europe with reductions at some sites and increases or no trend at others.
- On a country average basis the modelled trend in MTDM during 1995–2005 agrees well with the observations (approximately –20 μg/m^3 per decade) for the the Czech Republic, the Netherlands and the United Kingdom. For other countries the mean measured trend is smaller than modelled but large differences are seen within single countries.

5.1 EMEP model performance

The agreement between the measurements reported to AirBase and the two EMEP model scenarios REF and E95 (Table 4.1) has been checked. Based on the daily maximum ozone concentrations measured and calculated (REF and E95 scenarios; background rural stations) various statistical indicators for checking agreement were computed. Only the summer half year (April–September) was used in the calculations. The results are presented in Table 5.1.

Averaged over all 198 stations the REF (standard) scenario gives a slightly better agreement with the measurements than the E95 scenario (emissions constant at 1995 level). This is shown by all the statistics, indicating less bias and better correlation for the REF scenario than for E95. The difference is not very large, though. A bootstrap technique could be used to further evaluate the statistical significance of these differences as shown in Solberg *et al.* (2005) for Nordic ozone stations but is beyond the scope of this report.

Table 5.1 shows that there are differences for individual countries. For some countries (Austria, Slovenia and Switzerland) the observations actually show slightly poorer correlation (r^2) with REF than with E95. Furthermore, the RMS (root mean square) is higher for the REF compared to the E95 scenario for sites in Estonia, Spain and Switzerland. There are only one Estonian and two Spanish sites included in the analysis, implying that the results cannot be considered country averages. The best correlation is found for sites in Belgium, Germany, the Netherlands and the United Kingdom.

This statistical summary shows that on average the model results agree better with the measured values when assuming actual annual emissions data than when assuming constant 1995 emissions for the whole period 1995–2005. These results can be seen as an indication that the emissions data (see Chapter 2) that show clear reductions in ozone precursor emissions (NO_x and NMVOC) are for most countries closer to reality than assuming fixed emissions during 1995–2005. The difference is not very large, however, and there seem to be systematic differences, i.e. southern-central Europe (Austria and Switzerland) differs from north-western Europe.

5.2 Observed and modelled trend in high ozone concentrations

AOT40 and SOMO35 are ozone indicators used to evaluate the effects of ozone pollution on vegetation and human health. However, they are not necessarily the best suited parameters for evaluating long-term trends. It is well known that these statistics are sensitive to the general ozone level and that small offsets in this level can cause large deviations in AOT40. In the following, MTDM ([14]) is used as a metric for evaluating the trends. This is in line with the study by Jonson *et al.* (2006). One could argue that averaging over many sites should reduce the uncertainty and the fluctuations caused by single stations. On the other hand, such averaging could

([14]) MTDM is used as a proxy for photochemical ozone formation.

Table 5.1 Statistical summary of the EMEP model performance for the scenarios REF (1) and E95 (2), based on daily maximum ozone concentrations during April–September in 1995–2005 for background rural sites

Country	N	Mo	M1	M2	RMS1	RMS2	NMD1	NMD2	MAE1	MAE2	r12	r22
Austria	44	109 006	108 250	113 077	23 284	23 976	– 0.004	0.041	0.188	0.198	0.239	0.244
Belgium	9	94 945	97 634	102 792	24 920	27 499	0.028	0.082	0.217	0.242	0.430	0.425
Switzerland	7	114 966	109 788	116 099	28 685	28 471	– 0.042	0.013	0.241	0.249	0.225	0.239
Czech Republic	12	106 961	104 032	108 752	21 884	22 876	– 0.027	0.017	0.176	0.187	0.348	0.324
Germany	63	102 418	103 289	109 079	22 843	25 019	0.012	0.068	0.190	0.211	0.448	0.435
Denmark	1	89 634	93 093	96 829	18 069	20 858	0.039	0.080	0.161	0.184	0.328	0.309
Estonia	1	96 813	84 844	88 418	21 200	20 188	– 0.124	-0.087	0.170	0.163	0.265	0.265
Spain	2	116 732	102 342	104 092	26 586	26 029	– 0.123	-0.108	0.174	0.171	0.164	0.163
Finland	9	103 934	100 409	104 892	23 434	24 364	– 0.030	0.013	0.190	0.201	0.306	0.301
United Kingdom	14	84 859	83 702	85 911	18 974	20 359	– 0.011	0.014	0.170	0.183	0.446	0.432
Hungary	1	115 263	112 449	116 582	34 373	35 698	– 0.024	0.011	-3.026	-3.198	0.140	0.104
Lithuania	1	90 689	98 284	101 840	19 121	22 057	0.084	0.123	0.175	0.201	0.277	0.270
Latvia	1	80 206	87 548	90 644	20 025	22 402	0.092	0.130	0.238	0.266	0.234	0.209
Netherlands	18	86 595	92 930	97 795	24 626	28 322	0.073	0.129	0.232	0.264	0.410	0.402
Norway	8	80 984	74 898	76 987	17 986	18 465	– 0.078	-0.053	0.181	0.187	0.287	0.270
Portugal	1	90 631	101 803	102 611	29 194	29 703	0.123	0.132	0.172	0.178	0.354	0.352
Poland	5	101 439	98 146	102 838	20 221	20 894	– 0.032	0.014	0.168	0.177	0.364	0.350
Slovenia	1	112 881	110 563	116 386	20 606	21 027	– 0.021	0.031	0.157	0.165	0.347	0.359
All	**198**	**99 750**	**98 976**	**103 603**	**22 807**	**24 297**	**– 0.006**	**0.040**	**0.178**	**0.192**	**0.357**	**0.349**

Note: The statistics are: N = number of stations; M = mean; RMS = root mean square; NMD = normalized mean difference; MAE = mean absolute difference; r^2 = linear correlation coefficient squared. M_o = the observed mean. The values refer to µg/m³.

Source: EMEP, 2008.

mask errors or non-representative station behaviour. The model-to-measurement comparison is therefore conducted on an individual site basis.

5.3 Comparison of measurement and EMEP modelling results

Comparison plots for a selection of sites for various countries are depicted below (Figures 5.1–5.8). Only stations classified as rural background sites with complete or almost complete data coverage during 1995–2005 are shown. Data coverage of at least 75 % during April–September was used as the criterion for choosing the years included in the analysis. This selection excludes many countries.

In addition to the measured values (black) the REF (blue) and E95 (red) model scenarios are given in the plots. Thus the difference between the blue and the red curves indicates the modelled change due to the trend in anthropogenic emissions during 1995–2005. An estimate of the trend in the measured data based on a robust least absolute deviation method is also given (the function LADFIT based on Press *et al.*, 2007 in the software IDL®). The statistical significance of this trend is not considered here, i.e. a trend estimate is given for all sites although the time series in many cases show that the inter-annual variation is so large that the given value is certainly not significant.

First of all the plots show that even with a complete 11 year data series the inter-annual variations in the measured data is so large that the trend due to emission changes is easily masked. The difference in modelled MTDM, i.e. REF – E95, indicative of the modelled trend, was larger than the standard deviation of the measured values of this statistic for nearly all sites. In other words, the magnitude of the modelled trend due to emission changes in the period 1995–2005 was of the order of standard deviations of the observations for this period. If the additional uncertainty due to lack of model

performance is included then this magnitude of trend signal is at the edge (or below) what could be expected to be discernible from total variability. Thus, the short length of the measurement time series of ozone data is a major obstacle for the trend analyses. Based on the magnitude of the modelled trend and the standard deviation of the observations, a time series 5–10 years longer would facilitate a substantially more robust study.

Keeping these observations in mind, the trends do show interesting features, although no clear overall picture for Europe. Austria has a large number of stations including rural background sites with complete or almost complete time series between 1995 and 2005. A selection of eight sites is shown below (Figure 5.1).

A negative slope, i.e. a reduction in MDTM, is calculated for most sites but the inter-annual variation in the ozone metric makes these estimates unreliable. Furthermore, for several sites the measurement data is apparently closer to the E95 than the REF model results (e.g. AT0004A, AT0058A and AT134A). The data also indicate that, as in many other countries, the model generally underestimated ozone concentrations in the extreme summer 2003. The mean modelled trend in MDTM for all of the Austrian rural background sites was –18 $\mu g/m^3$ over this time period while the mean (1σ) standard deviation was 12 $\mu g/m^3$.

A negative slope is found for two of the four Belgian sites shown (Figure 5.2), but with a large inter-annual variation in the data, making the trend unreliable. The modelled trend due to emission changes is 21–25 $\mu g/m^3$ while the standard deviation based on the measurements is somewhat less, 18–23 $\mu g/m^3$.

For the Swiss sites (Figure 5.3), the magnitude of the modelled trend (approximately 20 $\mu g/m^3$) is clearly larger than the standard deviation of the measurements (8–13 $\mu g/m^3$). Based on this result Switzerland should be an area where downward trends in the MTDM can be expected. That this is not the case may indicate poor model performance due to small scale topography and sub-grid scale vertical transport processes or it could indicate a more fundamental lack of understanding of emissions and photochemical processes.

The Swiss ozone measurement concentrations are generally higher than the modelled values. This could partly be explained by mountain sites (e.g. CH0005) not well captured in the regional model with a spatial resolution of 50 x 50 km^2.

Several of the Czech sites show a fairly large decline in the measured MTDM (e.g. CZ0001R and CZ0049A) while some, such as CZ0062, show a closer similarity with the E95 scenario without any negative trend. The estimated trend (E95 – REF) is generally larger than the standard deviation of the measurements (15–22 $\mu g/m^3$ as compared to 10–20 $\mu g/m^3$) and the estimated slope based on the observations is negative for most of the sites. Thus, some of the Czech sites indicate a real reduction in peak ozone values due to emissions reductions but there is no clear picture for the whole country.

The German sites also show a mixed picture. Some sites, such as DE0679A in the south-east and DE0960A in the north-east show a marked decline in MTDM and a good agreement with the model results. However, for other sites (e.g. DE0422A) the agreement is poor and the observations show an upwards trend. The estimated trend in MTDM is 20–30 $\mu g/m^3$, i.e. somewhat higher than the standard deviation of the measurements (10–20 $\mu g/m^3$).

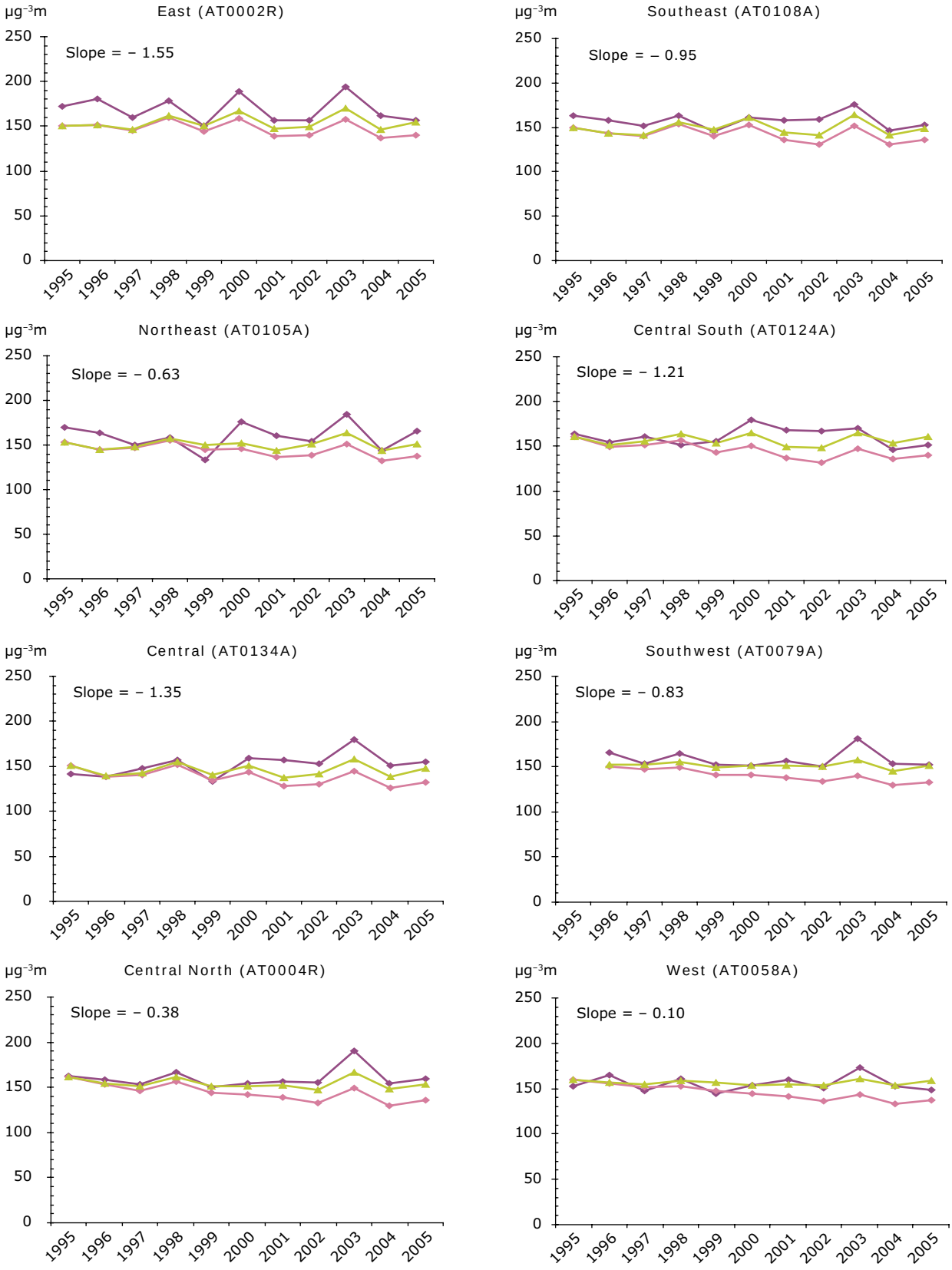
For the Finnish sites the estimated trend in MTDM is at most sites clearly smaller than the standard deviation of the measured values, thus one would not expect to see an emission-induced trend signal from the measurements alone.

For the British sites differences are large. While the modelled trend is larger than the standard deviation from the measurements in some areas, they are comparable in other areas. Several sites do however show a marked decline in measured MTDM, like GB0014R and GB0002R. As seen from Figure 5.7 the inter-annual variation is large.

A reduction in MTDM is calculated for all the Netherlands sites (Figure 5.8). Again, the year-to-year variation is large, indicating that the estimated slope is not significant. NL0227A is an exception to this and shows a very big reduction in measured MTDM.

In summary, the results presented above give a very mixed picture for different regions in Europe. Reductions in MTDM are found at some ozone measurement sites, while other sites show no changes or even increases in concentrations. Year-on-year variations in observed MTDM are larger than the modelled emission induced trend during the analysed time period (1995–2005) at many sites. This indicates that in most regions 10–11 year time series are too short to permit conclusions on the long-term trend caused by anthropogenic emission reductions.

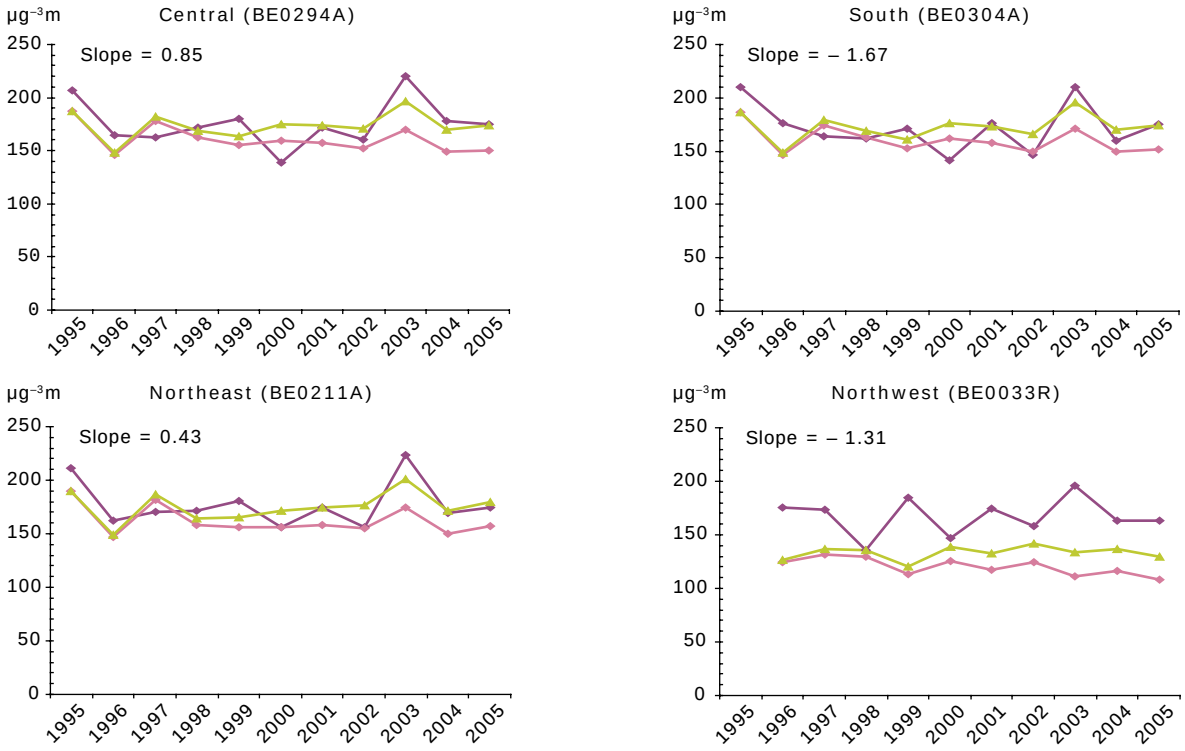
Figure 5.1 Austria: observed and modelled MTDM



Note: The purple line represents observed MTDM values, the pink line represents the REF scenario results and the green line represents the E95 scenario results.

Source: AirBase, 2008; EMEP, 2008.

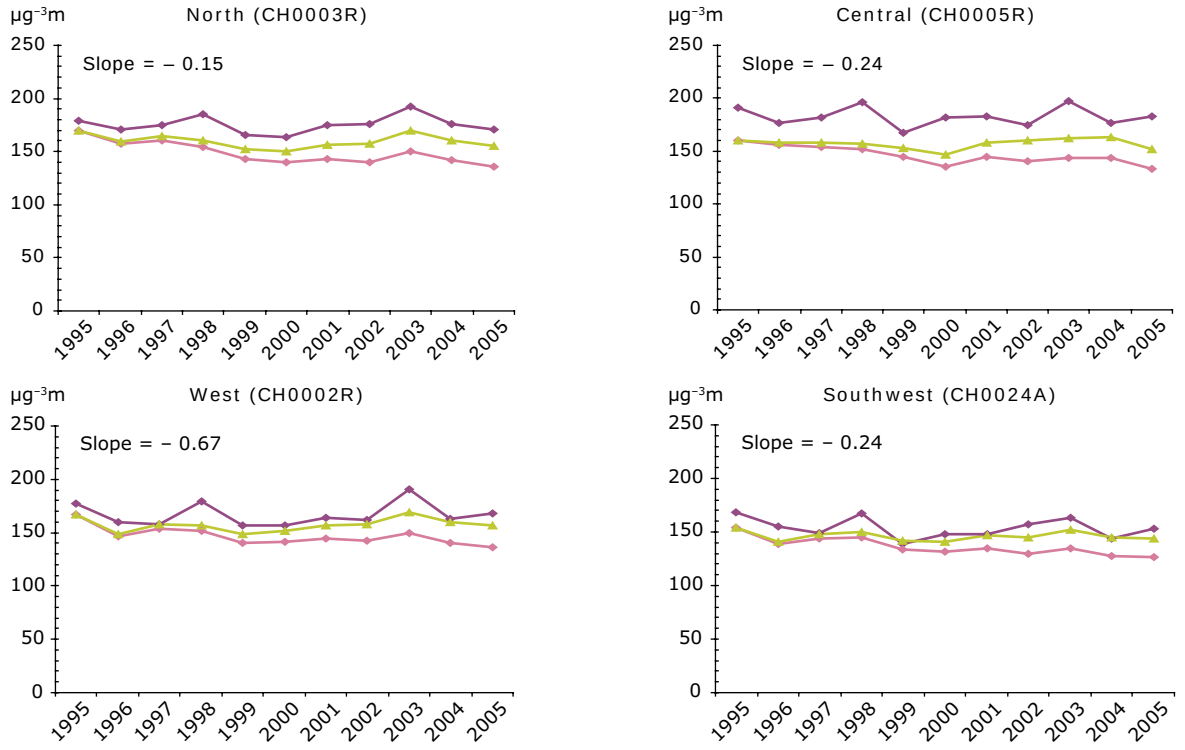
Figure 5.2 Belgium: observed and modelled MTDM



Note: The purple line represents observed MTDM values, the pink line represents the REF scenario results and the green line represents the E95 scenario results.

Source: AirBase, 2008; EMEP, 2008.

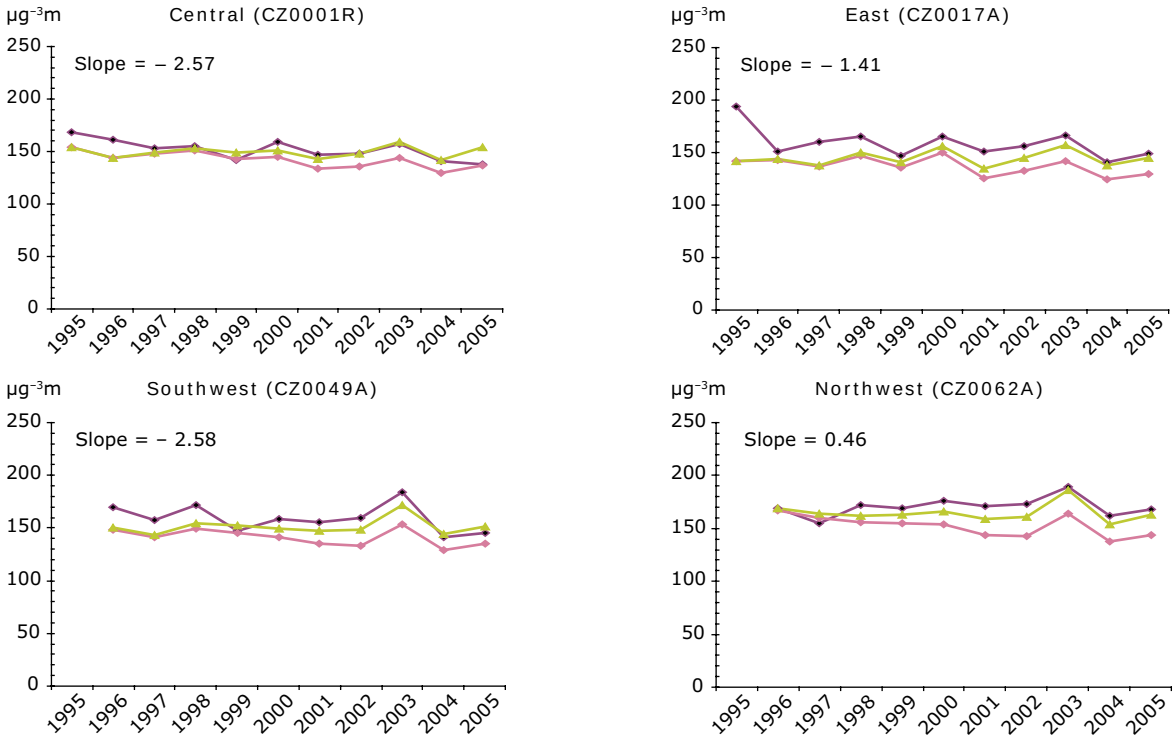
Figure 5.3 Switzerland: observed and modelled MTDM



Note: The purple line represents observed MTDM values, the pink line represents the REF scenario results and the green line represents the E95 scenario results.

Source: AirBase, 2008; EMEP, 2008.

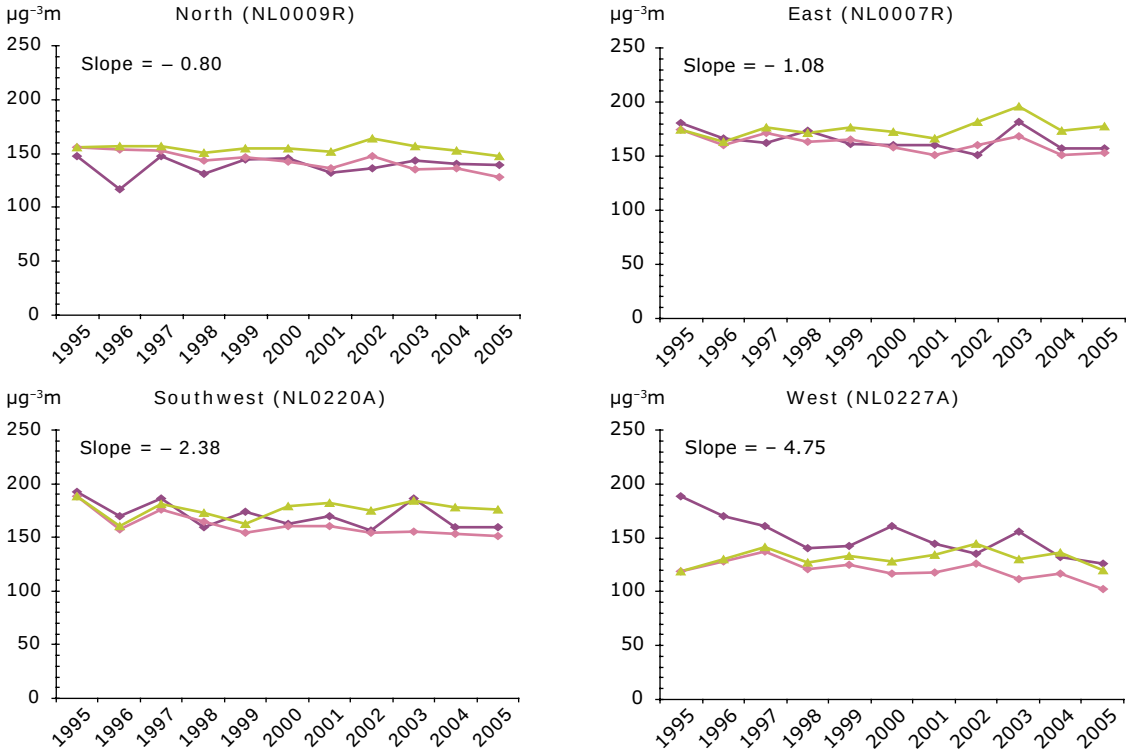
Figure 5.4 Czech Republic: observed and modelled MTDM



Note: The purple line represents observed MTDM values, the pink line represents the REF scenario results and the green line represents the E95 scenario results.

Source: AirBase, 2008; EMEP, 2008.

Figure 5.8 Netherlands: observed and modelled MTDM



NNote: The purple line represents observed MTDM values, the pink line represents the REF scenario results and the green line represents the E95 scenario results.

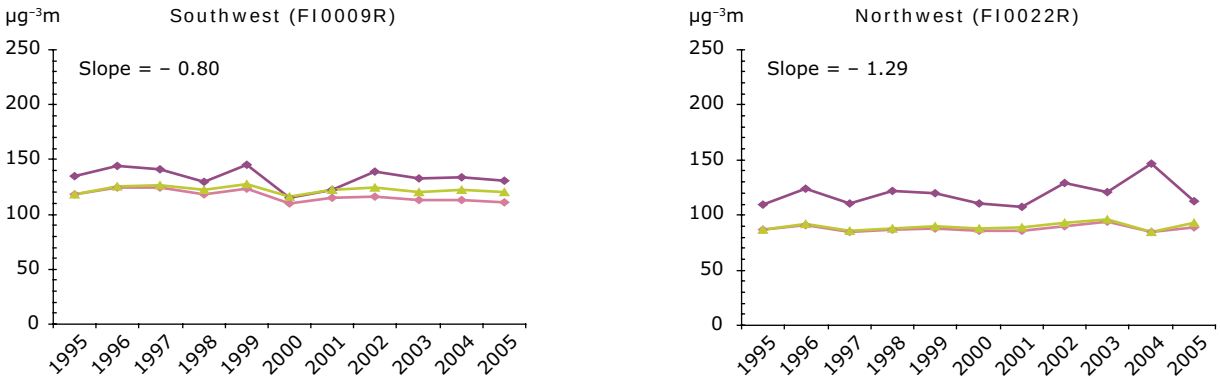
Source: AirBase, 2008; EMEP, 2008.

Figure 5.5 Germany: observed and modelled MTDM

Note: The purple line represents observed MTDM values, the pink line represents the REF scenario results and the green line represents the E95 scenario results.

Source: AirBase, 2008; EMEP, 2008.

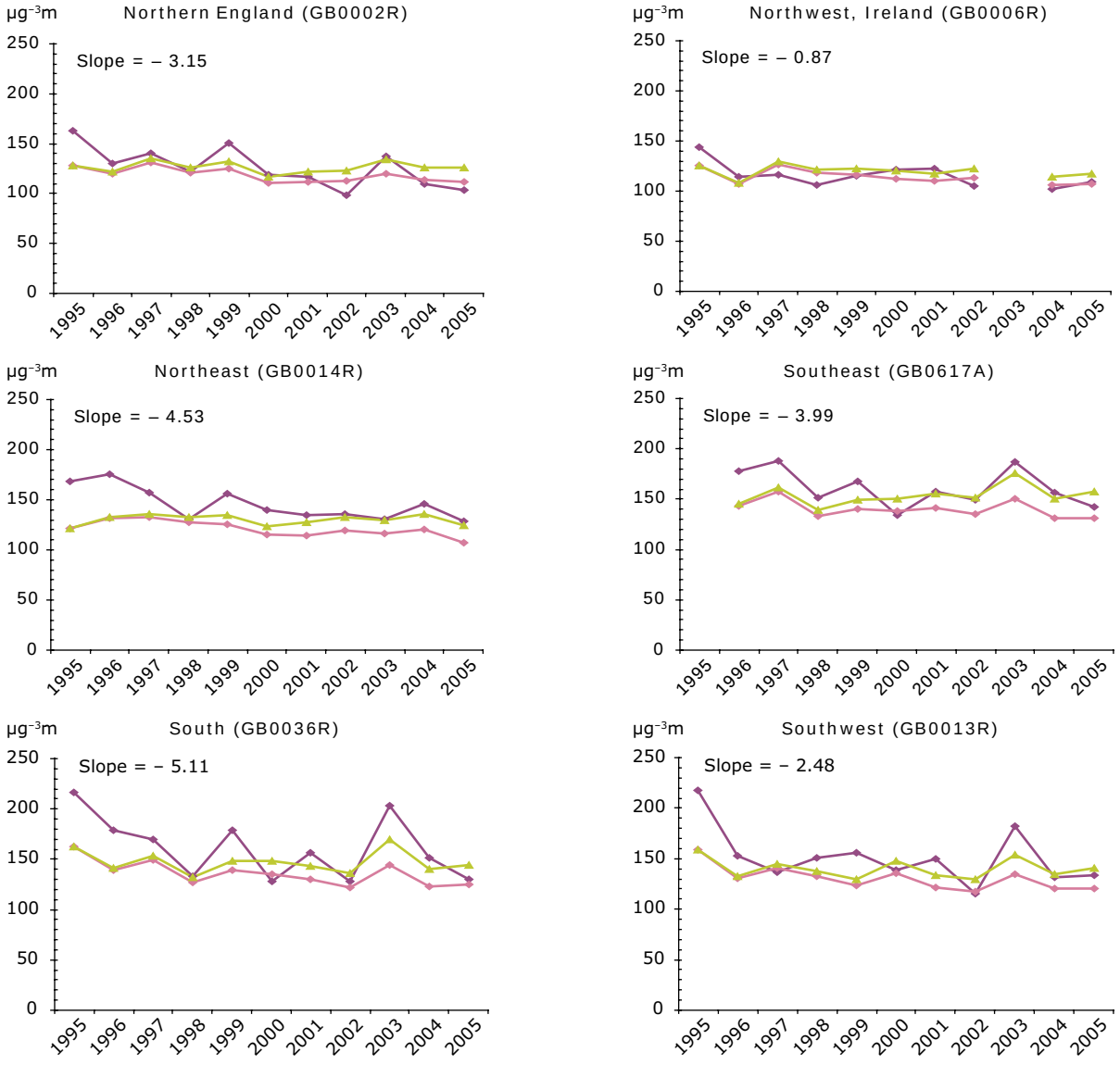
Figure 5.6 Finland: observed and modelled MTDM



Note: The purple line represents observed MTDM values, the pink line represents the REF scenario results and the green line represents the E95 scenario results.

Source: AirBase, 2008; EMEP, 2008.

Figure 5.7 United Kingdom: observed and modelled MTDM



Note: The purple line represents observed MTDM values, the pink line represents the REF scenario results and the green line represents the E95 scenario results.

Source: AirBase, 2008; EMEP, 2008.

5.4 Europe-wide modelled and measured trends

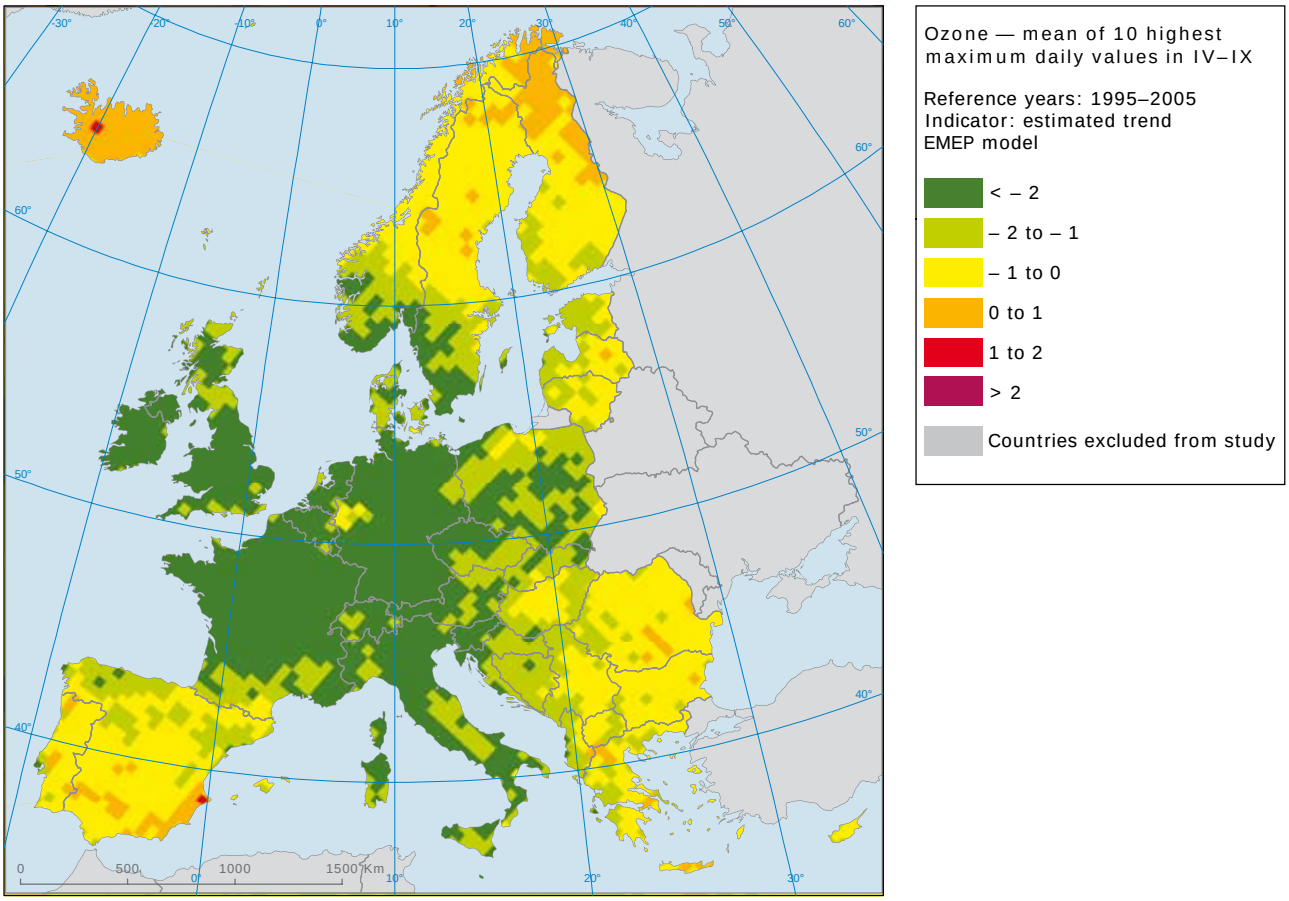
The Europe-wide trends predicted by the model and given by the rural background stations with sufficient data are shown in Maps 5.1 and 5.2. The model (Map 5.1) predicts a reduction in this metric of approximately 2 µg/m^3 or more per year in central and north-western Europe (1995–2005). Smaller reductions are predicted outside this region (Spain, Balkan countries, the Baltic and the Nordic countries).

As noted above, the agreement with the measurements is very mixed and could only be studied for certain areas of Europe. Map 5.2 shows the calculated trend in the measurements and the model predicted values for the selected sites with sufficiently long monitoring histories. Note that these model values can differ from the ones shown in Maps 5.1 as they are only based on the years with monitoring data in that grid cell. Thus, differences in the model values between Maps 5.1 and 5.2 indicate the sensitivity of the trend estimates to individual years.

Map 5.2 shows that the model in general predicts larger reductions than found in those measured. This is particularly true for Switzerland, western Austria and parts of Germany. In Belgium, the Netherlands, the Nordic countries and the United Kingdom the agreement is better but with some exceptions.

The results of the analyses of MTDM are summarized in Table 5.2. Here we show the country average values using only sites with at least 10 years of measurement data. Only eight countries fulfilled this criterion. A negative slope is found for all countries except for Finland. However, as mentioned above, the estimated slope at the Finnish sites is clearly non-significant when compared with (1σ) standard deviation of the measurements (10 µg/m^3).

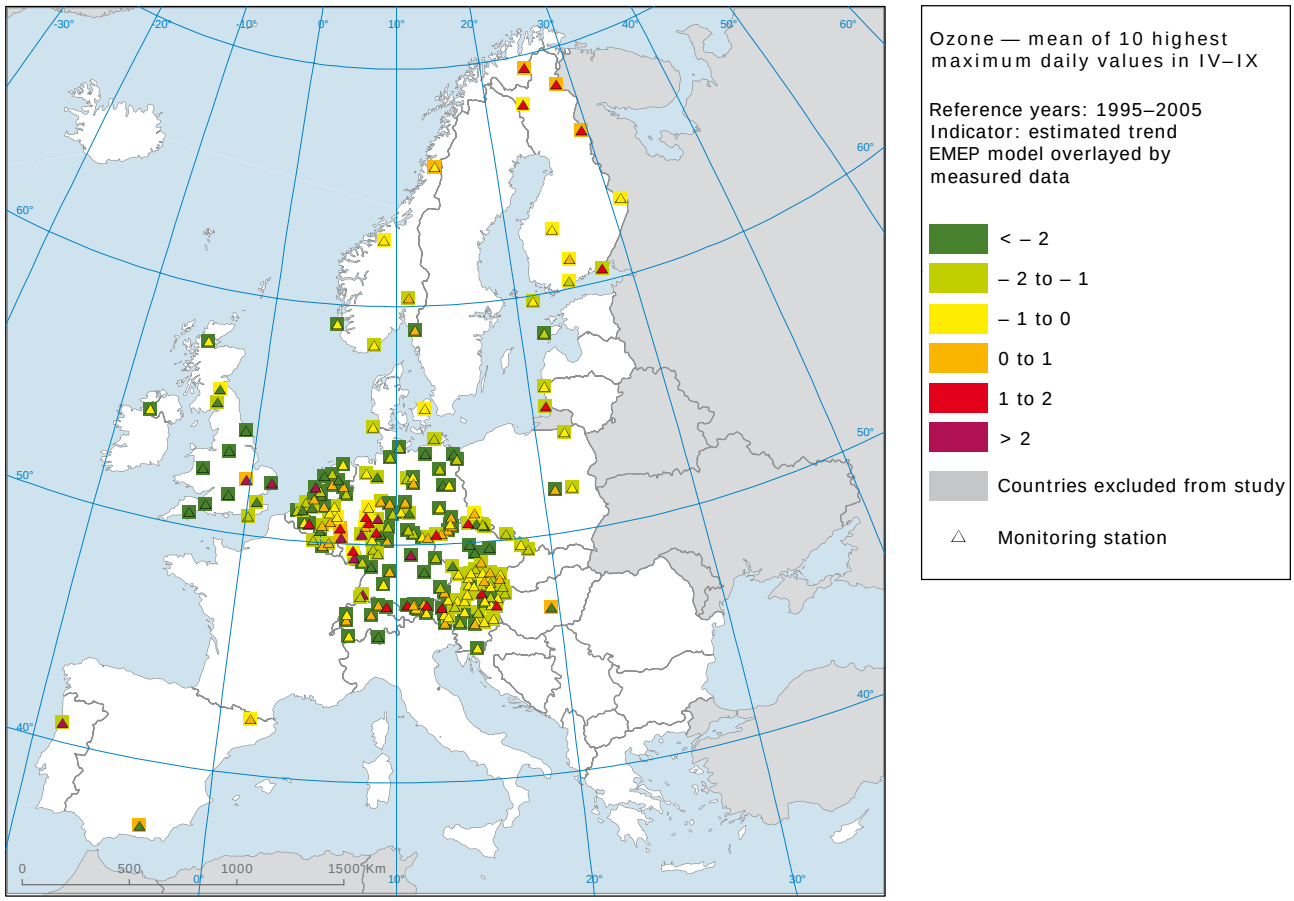
Map 5.1 Estimated trend in MDTM calculated by the EMEP model



Note: Unit: µg/m^3 per year. IV–IX = April to September.

Source: EMEP, 2008.

Map 5.2 Estimated trend in MDTM based on the rural background AirBase monitoring sites (triangles) shown on top of the values from the EMEP modelling using only the years with measured data



Note: Unit: µg/m³ per year. IV–IX = April to September.

Source: EMEP, 2008; AirBase, 2008.

The largest national reductions in MTDM are found for the Czech Republic, the Netherlands and the United Kingdom, with a decline of approximately 20 µg/m³ per decade. That is of the same order as modelled (REF scenario) and slightly greater than standard deviation of the measurements. A reduction of 11 µg/m³ per decade is found for the Belgian sites, which is less than modelled with the REF scenario and less than standard deviation.

The largest discrepancy compared to the model results is found for Switzerland in particular and also for Austria. The REF model scenario predicts a mean reduction in MDTM of some 25 µg/m³ per decade for the Swiss sites and standard deviation of the measurements is fairly small (11 µg/m³). The observations, however, do not show any reductions. Similar results are found for the Austrian sites, though with a small calculated reduction of 2.3 µg/m³ per decade. This difference in agreement between modelled and measured trends is consistent with the findings of Vautard *et al.* (2006), who found a similar systematic difference between north-west Europe and central Europe.

The mean of the German sites is similar to the results for the Austrian sites, with a slightly stronger decline (3.0 µg/m³ per decade). There are, however, large differences between the individual German sites, with both reductions and increases in MTDM.

The reason for these national differences is unclear, although the situation in the Alpine countries is obviously more difficult to model properly with a regional scale model with a resolution of 50 x 50 km².

In general, the regions where the model is expected to predict trends most reliably (i.e., those where meteorological variability is smallest relative to emission-induced trends) are actually the areas with the largest discrepancy between the measurements

and the modelled values. On the other side, the areas where the meteorological variability is expected to be larger relative to the emission based trend (e.g. the United Kingdom) the agreement between the model and the measurements is the best. The reasons for this apparently contradiction is not clear.

Table 5.2 Actual and modelled trends in MTDM — national averages based on sites with 10–11 complete years of data

	Measured trend (µg/m³ per decade)	Modelled trend (REF) (µg/m³ per decade)	Modelled trend (E95) (µg/m³ per decade)	Standard deviation of measured MTDM data (µg/m³)
Austria	– 2.3	– 19	– 0.8	12
Belgium	– 11	– 20	+ 1.5	21
Switzerland	– 0.2	– 25	– 1.3	11
Czech Republic	– 20	– 20	– 1.4	14
Germany	– 3.0	– 21	+ 4.0	15
Finland	+ 5.7	– 5.1	– 0.1	10
United Kingdom	– 22	– 20	– 2.4	19
Netherlands	– 18	– 21	+ 4.4	17

6 Open questions and further recommendations

This report has presented various analyses based on ozone measurement data reported to AirBase and EMEP modelling results and the key findings are summarized in the beginning of the report. Although the results indicate certain relationships and answers, several questions remain open:

- Why do we see the clearest downward ozone trends in the regions where the meteorology has the strongest influence (the United Kingdom and north-west continental Europe) and no trends in the regions with the least influence of meteorological variability (Switzerland and western Austria)?
- Do the discrepancies between the model results and the measurements in southern and central Europe indicate flaws in the emission data or in the model?
- The observational data indicate a clear reduction in ozone concentrations during 1990–1998 followed by a stabilisation from 1999 onwards. Is this simply an artefact due to the less emission reductions in this period or is it a real effect due to changes in other controlling parameters?
- Is the role of the hemispherical background ozone much more crucial than suggested by the estimates based on Mace Head surface measurement data? Could part of the discrepancies between model predictions and observations be due to intercontinental transport, e.g. plumes of forest fires above the boundary layer (and above Mace Head), mixed down to the surface over the continental sites?
- What are the effects on ozone of an increase in the NO_2/NO emission ratio caused by an increase in the share of diesel vehicles in Europe? Do direct NO_2 emissions only affect the $NO/NO_2/NO_x$ reactions along busy roads? Are urban and rural background stations also affected?
- How certain are the (gridded) emissions inventory data used for modelling? Uncertainties in emission inventories have not been explicitly addressed in this report. However, both spatial and temporal inconsistencies arising from different methods for calculating national emissions or their spatial disaggregation in different European countries and in different years can contribute masking 'real' trends in emissions and in modelled ozone concentrations ([15]).

Additionally, the study leads to certain recommendations:

- The lack of long-term observational time series of ozone data in AirBase is a major problem for assessing the trends. To some extent this can only be solved by 'waiting' until existing monitoring stations offer the required length of time series. However, historical data do exist for many of the AirBase ozone stations and such data should be included in the European database.

- In-depth inspection of individual time series has revealed a number of dubious ozone data in AirBase. Although it is probably a small fraction of the whole dataset, and one probably has to accept that all databases contain errors, it points to the value of having data with a certain degree of documented quality.

- The role of meteorology and intercontinental transport of ozone should be studied more than was possible in the present assessment. The importance of 'scale issues' on modelled ozone, i.e. the influence of model resolution on predicted ozone, should also be given greater attention. The 50 x 50 km^2 resolution of the present version of the EMEP model may be inadequate in certain regions of Europe.

([15]) See for example EMEP, 2008.

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