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Ozone exposure scenarios in the Nordic countries during the 21st century

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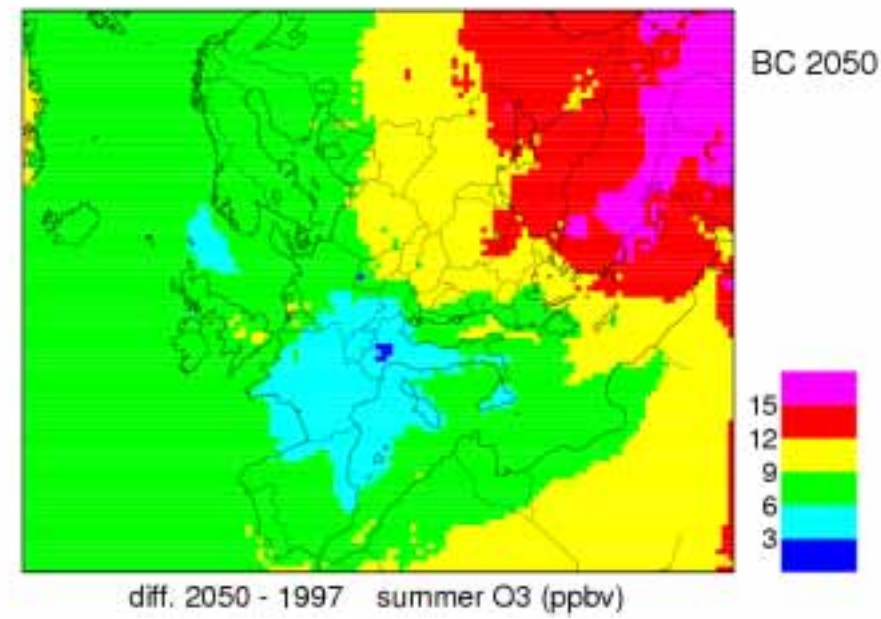

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Ozone exposure scenarios in the Nordic countries during the 21st century

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Preface and acknowledgements

Results from recent global atmospheric chemistry simulations on the future ozone concentrations indicate that there is risk for very high tropospheric concentrations towards the end of this century. Ozone concentrations in unpolluted air masses in the Nordic countries are already close to 40 ppb and seem to be increasing. Concern on the rising background ozone gave birth to a project "Ozone exposure scenarios in the Nordic countries during the 21st century" financed by the Nordic Council of Ministers. This project brings together four research groups who have studied future air pollution problems:

- University of Oslo
- Norwegian Meteorological Institute
- Swedish Meteorological and Hydrological Institute (SMHI)
- Finnish Meteorological Institute

This report is produced as a result of studies conducted within the ozone scenario project. Within the UN-ECE research policy, there is also a growing interest in atmospheric chemistry on extended spatial and temporal scales, as an increasing background level would counteract the European efforts to combat photochemical air pollution by emissions reductions.

We wish to acknowledge the Nordic Council of Ministers for financial support. The research group at the Finnish Meteorological Institute thanks Timothy Carter (Finnish Environment Institute) for encouraging studies on future environmental impacts and the Academy of Finland for financial support. The contribution of SMHI has been supported by the Foundation for Strategic Environmental Research through the Swedish Regional Climate Modelling Programme, SWECLIM.

Executive summary

This is the final report from the project "OZONE EXPOSURE SCENARIOS IN THE NORDIC COUNTRIES DURING THE 21 CENTURY" financed by the Nordic Council of Ministers.

In this study future emission scenarios for ozone precursors are discussed. Calculated effects on ozone of emission changes are shown. Furthermore calculated effects of climate change on ozone are described.

Based upon research within the IPCC different sets of emission scenarios (SRES scenarios) of greenhouse gasses and ozone precursors have been developed. These scenarios aggregate the world into only 4 areas. In this report we are focusing on the SRES A1T scenario for 2050, but some results are also presented for other scenarios. The year 2050 was selected for modelling because it was well beyond the horizon of the CAFÉ scenarios. The A1T scenario is characterized by rapid economic growth in all regions of the world, but with a rapid transition to non-fossil fuel technologies. Comparing the A1T scenario with emission scenarios up to 2030 estimated within the CAFÉ programme of the European commission, it seems that the A1Tscenario is quite realistic for Western Europe, but emissions estimated for Central and Eastern Europe seem to high.

Model calculations are performed with the Oslo CTM2 global model and the EMEP Eulerian photochemistry model with 1997 meteorology. The calculations are made with 1997 and 2050 emissions using the A1T scenario. The evolution of European ozone levels are to a large extent superimposed on the hemispheric pool of background ozone in the free troposphere. Emission reductions in Western Europe result in a decrease in surface ozone, but the reductions are in general more than compensated by the increase in background ozone. Thus mean ozone and ozone indicators as AOT40f are all expected to increase. In Central/Eastern Europe the combined effects of increased background concentrations and increased emissions of ozone precursors leads to a substantial increase in surface ozone.

The ozone exposure varies greatly, in particular after 2040, depending on scenario. With the fossil fuel intensive scenario A1C and A2 scenarios, AOT40 at Utö in Finland will increase to more than 30.000 ppb.h by the year 2100, mainly as a result of very high background concentrations. Only in the B1 scenario will AOT40f be below the critical level of 10.000 ppb.h by year 2100.

Calculations with the regional model MATCH indicates that ozone concentrations and ozone exposure expressed as AOT40f will increase substantially in Southern and Central Europe as a result of a warmer climate with less precipitation and cloudiness. In the Nordic countries calculated ozone decrease as a result of less intense heating, increased precipitation and changes in transport patterns not favouring advection of pollutants in this direction.

1. Introduction

Tuomas Laurila, Finnish Meteorological Institute

Measurements and modelling studies show that the present ozone concentrations over the Nordic countries are at a level which is potentially harmful for vegetation and human health (Hjellbrekke and Solberg, 2004; de Leeuw and de Paus, 2001; Simpson et al., 2003b). The abatement of ozone precursor emissions in Europe should result in lower levels of photochemical pollution in these areas (Simpson, 1995; Jonson et al., 2001). However, various global chemistry-transport model (CTM) simulations indicate that during the 21st century ozone concentrations may increase substantially (Prather et al., 2001). These simulations are usually based on the internally consistent set of global greenhouse gas (GHG) and ozone precursor emissions presented in the Special Report on Emissions Scenarios (SRES) of the Intergovernmental Panel on Climate Change (IPCC) (Nakićenović and Swart, 2000).

Most global socio-economic scenarios simulate high economic growth and increasing precursor emissions in the Far East, the developing world and the eastern parts of Europe. From these areas of enhanced photochemical production, elevated ozone concentrations can be long-range transported to other parts of the world (Jacob et al., 1999; Collins et al., 2000). As a consequence, tropospheric ozone concentrations also in unpolluted air masses in northern Europe may increase (Jonson et al., 2001; Derwent et al., 2002, 2004). Presumably, this will lead to a situation in which northern and Western Europe will experience less intensive photochemical episodes embedded on a gradually increasing background concentration.

Climate change will alter many factors which modify ozone production and transport (Prather et al., 2003). The flux from the stratosphere to the troposphere, accelerated oxidation of methane and other organic compounds by the intensified OH formation due to higher water vapour concentrations, increased emissions of biogenic Volatile Organic Compounds (VOCs), temperature dependent chemical reactions and changing weather patterns are among the phenomena which may affect ozone concentrations. We still lack a comprehensive analysis of all these processes, of which some act on a global and some on a local level. Climate model simulations predict increasing temperatures and changing weather patterns, including changes in cloudiness and precipitation in northern Europe.

Air pollution studies related to global change need special tools adapted to this complex problem. The global CTMs developed at the University of Oslo have been widely used for different applications including studies on atmospheric concentration changes and their effects on radiative forcing (Gauss et al., 2003), effects of Asian emissions on air quality in the Pacific area (Wild et al., 2003), and emissions of aviation (Isaksen et al., 2001) and marine traffic (Endresen et al., 2003) on atmospheric composition. All these show that human activities have global impacts on tropospheric ozone concentrations. The new Unified EMEP model developed at the Norwegian Meteorological Institute (Simpson et al., 2003a; Fagerli et al., 2004) has been shown to accurately simulate ozone concentrations in Europe (Simpson et al., 2003b). In this study, this regional model has been run using boundary conditions derived from the global Oslo CTM2 model of the University of Oslo. This will provide us with detailed ozone concentration fields over Europe for long-term scenarios extending the period in a similar previous study by Jonson et al. (2001). In another previous study, a wide range of

different future ozone scenarios were estimated for Finland from the SRES emission scenarios (Laurila et al., 2004). This was carried out within the FINSKEN project (Carter et al., 2004), which is an example of country-scale analysis of the socio-economic and technological alternatives behind the SRES scenarios and their related environmental impacts. In Sweden, regional photochemical pollution has been studied using the MATCH model, which has been developed at the SMHI (Robertson et al., 1999). For example, the decline of high ozone concentrations during the 1990s in southern Scandinavia has been simulated successfully (Solberg et al., 2002). In the present study, the MATCH model has been run using meteorological fields from the high resolution climate change simulations produced at the Rossby Centre of the SMHI (Rummukainen et al., 2001).

In this report, we first present an overview of the observed long-term changes in ozone concentrations (Section 2) and then discuss the global SRES emissions and how they are linked to the tropospheric ozone burden (Section 3). The SRES A1T scenario is explained in detail because the photochemical model simulations of this study are based on this scenario. To highlight problems related to using the aggregated SRES data on a regional scale, where local environmental strategies may play an important role, the SRES emissions are also compared to a projection of European emissions up to 2030 that was recently produced within the Clean Air For Europe (CAFE) programme (Amann et al., 2004). Next, we describe the photochemical model calculations that combine global (Oslo CTM2) and continental (Unified EMEP) scale CTMs and present simulated ozone concentrations and exposures in 2050 for the A1T scenario (Section 4). The effect of climate change on European ozone concentrations is assessed based on the model calculations with the MATCH model using meteorological output from a regional climate model (Section 5). Finally, we present the main conclusions of this study and some recommendations for future work (Section 6).

2. Past changes in ozone concentrations

Tuomas Laurila, Finnish Meteorological Institute

Ozone concentration in the background troposphere is formed as a net effect of the flux from the stratosphere, net production in the troposphere and deposition to the surface. It is highly sensitive to atmospheric concentrations of methane, carbon monoxide, volatile organic compounds and nitrogen oxides because the tropospheric photochemical production and loss terms are several times higher than transport from the stratosphere and deposition to the surface (Prather et al., 2001). During the pre-industrial times, the tropospheric concentrations have been estimated to be considerably lower than today (Guicherit and Roemer, 2000). In Europe, the ground level average concentrations have nearly doubled (Volz and Kley, 1988). We may expect that the highest episodic concentrations have experienced even more drastic changes because during past centuries there were hardly any photochemical smog episodes.

The increase in concentrations has followed from the industrial and traffic emission becoming more widespread. The highest increase rates of emissions occurred after the Second World War, especially during the 1950s, when economic growth was high (Simpson et al., 1997; van Aardenne et al., 2001). Unfortunately, quantitative ozone concentration measurements are extremely scarce from that era. The measurements made in the Alps (Staehelin et al., 1994) and on the coast of the Baltic Sea (Arkona) (Feister and Warmbt, 1987) are in accordance with the emission changes. The increasing trends have been stabilized in the U.S. after 1970s (Fiore et al., 1998), and in the western and central parts of Europe during the 1980s (Scheel et al., 1999; Guicherit and Roemer, 2000), evidently due to the precursor emission control. During the most recent years, the substantial precursor emission reductions have lowered the highest episodic concentrations in the western part of Europe (NEGTAP, 2001; Solberg et al., 2002).

In the remote areas of the world, in the absence of local emissions, ozone observations show the effects of photochemical production and loss in the free troposphere, intercontinental transport and stratospheric ozone depletion affecting the stratosphere-troposphere flux. Embedded on the increasing tropospheric ozone concentration, there seems to be variations on a decadal time scale. The Canadian ozone sounding data from four stations show mostly declining concentrations (–25%...+5%) in the lower troposphere during the period 1980–1990 (Tarasick et al., 2004). After that period, the ozone concentrations have increased at a high rate (+9%...+19% in 1991–2001) (Tarasick et al., 2004). Increasing tropospheric ozone concentrations in unpolluted air masses during the 1990s have been reported for many locations. A statistically significant increase of 1 $\mu g\ m^{-3}\ yr^{-1}$ in background air masses has been observed at Mace Head on the west coast of Ireland since 1987 (Simmonds et al., 2004). A similar trend, with an increase of 0.6 $\mu g\ m^{-3}\ yr^{-1}$ in the annual average, has been found at Esrange in northern Sweden (Lindskog, 2003).

Most interestingly, the summertime concentrations have also been increasing in the lower free troposphere at Sodankylä in northern Finland (Kivi et al., 2004). The reported increase by 0.6 % yr^{-1} corresponds to 0.5 $\mu g\ m^{-3}\ yr^{-1}$, which is about half of the increase of 1.0–1.2 $\mu g\ m^{-3}\ yr^{-1}$ observed during summer in unpolluted air masses in the southern and central parts of Finland (Laurila et al., 2004). We may conclude that after 1990 there has been an increasing trend in the background tropospheric ozone concentrations in the high and mid-latitudes of the northern hemisphere. This has occurred in spite of the mostly declining stratospheric ozone concentrations (Tarasick et al., 2004).

3. SRES scenarios

Tuomas Laurila, Juha-Pekka Tuovinen and Virpi Tarvainen, Finnish Meteorological Institute

3.1 Scenario families

Scientific research within the IPCC on the global development of population, social structures, economy and technology has produced a consistent set of future scenarios of emissions and atmospheric concentrations with a good documentation of the driving factors. The most recent set of future greenhouse gas and ozone precursor emissions has been reported in the Special Report on Emissions Scenarios (SRES) (Nakićenović and Swart, 2000), based on which atmospheric composition scenarios have been developed (Prather et al., 2001).

The IPCC SRES scenarios define four storylines of different future development of social and cultural interactions and of economic and technological progress: A1, A2, B1 and B2 (Nakićenović and Swart, 2000). The main features of the related scenario families are illustrated in Figure 3.1. In the scenario family A, economic growth is the major driving force, while environmental values are more important in the B scenarios. Scenarios 1 describe globally integrating futures, in which social and technological interactions are enhanced, while in scenarios 2 different regions of the world have different solutions to social and technical challenges. The global population growth is higher in the more regional scenarios 2, in which the low birth rates of industrialised countries are only slowly diffusing to developing countries. By the end of this century, the global population has increased to around 7 billion in the A1 and B1 scenarios and to 15 and 10 billion in the scenarios A2 and B2, respectively. World gross domestic product in 2100 is in A2 and B2 scenarios half and in B1 two thirds of that produced in A1 scenario.

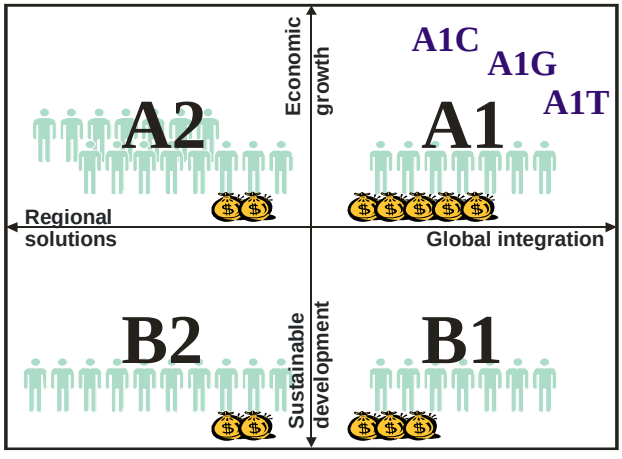

Figure 3.1. The four main scenario families. A1 is often referred to as ‘World markets’, A2 as ‘Provincial enterprise’, B1 as ‘Global sustainability’ and B2 as ‘Local stewardship’. Different scenarios produce different development of global population and gross domestic product.

Within the four main scenario sets, there are different technologies for production of energy. In addition, three alternative developments of energy technologies are defined within the A1 set. The fossil fuel intensive scenarios, for example A1C, produce increasing emissions, while the scenarios which place more weight to the new technologies (A1T) and environmental

factors (B1) will result in considerable reductions in greenhouse gas and air pollutant emissions. As the development and diffusion of new technologies will take time, in all scenarios the global emissions increase during the first decades of the century, and greater differences emerge mainly after 2040.

In this study, we simulated ozone concentrations based on the A1T scenario. Nakićenović and Swart (2000) describe that the A1 scenario is a case of rapid and successful economic development, in which regional average income per capita converge and current distinctions between "poor" and "rich" countries eventually dissolve. The transition to economic convergence results from advances in transport and communication technology, shifts in national policies on immigration and education, and international cooperation in the development of national and international institutions that enhance productivity growth and technology diffusion. According to the A1T scenario, transition to non-fossil technologies is fast. By the year 2050, there is no more traditional burning of fossil fuels. The most important energy sources are hydrogen produced using biomass, nuclear and solar power, hydrogen fuel cell and natural gas-fired combined cycle power plants.

The emphasis in SRES scenario development is to calculate future GHG emissions and to estimate long-term changes in energy production, agriculture and land use. Emissions of ozone precursors, NOx and VOC, are mainly produced by combustion processes. The main driving factor of emission changes is transition to new technologies. Traditional air pollution legislation and emission control technologies are considered in an incomplete way. This results in overestimation of ozone precursor emissions for the next few decades, at least in Europe, where trajectories of GHG and ozone precursor emissions diverge due to strict air pollution legislation.

3.2 Global emissions of ozone precursors

Of the ozone precursors, emissions of NO_x and methane are globally the most important ones (Prather et al., 2001). In all emission scenarios, there is an increasing development up to 2030 and 2050 for NO_x emissions and atmospheric methane concentrations, respectively (Figure 3.2). After that, the environmentally friendly scenarios show gradually declining trends. Prather et al. (2001) presents a simple relation between the global ozone precursor emissions and the tropospheric ozone burden, showing that the development of the ozone burden closely resembles that of the precursors. Great increases are calculated for scenarios in which energy is produced primarily by fossil fuels. High methane concentrations enhance photochemical ozone production all over the troposphere, and high emissions of NO_x and VOC produce photochemical smog in areas of fast economic growth such as Asia and Latin America.

The SRES emissions have been reported for four aggregated areas of the world. Western Europe, North America, Japan and Australia belong to the 'OECD90' region. The 'REF' region covers Central and Eastern European countries undergoing economical reform and the newly independent states of the former Soviet Union. The highest growth rates of economy and emissions take place in the 'ASIA' region covering China and the other Far and Middle East countries. Latin America and Africa form the 'ALM' region. The geographical distribution of emissions for different years is produced by scaling the emission map of 1990 homogeneously within each region. The emission data of the SRES A1T scenario used in this study is presented in Table 3.1.

Table 3.1. Emissions in 2000 and 2050 according to the SRES A1T (MESSAGE) scenario (Nakićenović and Swart, 2000). The regions are explained in the text.

Region	CH_4		CO		VOC		NO_x	
	2000 ($Tg\ yr^{-1}$)	2050 (Δ%)	2000 ($Tg\ yr^{-1}$)	2050 (Δ%)	2000 ($Tg\ yr^{-1}$)	2050 (Δ%)	2000 ($Tg\ N\ yr^{-1}$)	2050 (Δ%)
OECD90	74	–24	161	+25	36	–25	12	–25
REF	39	+38	41	+173	13	+162	3	+67
ASIA	125	+82	270	+190	37	+122	9	+211
ALM	85	+91	404	+66	55	+78	8	+150
Total	323	+55	877	+102	141	+71	32	+91

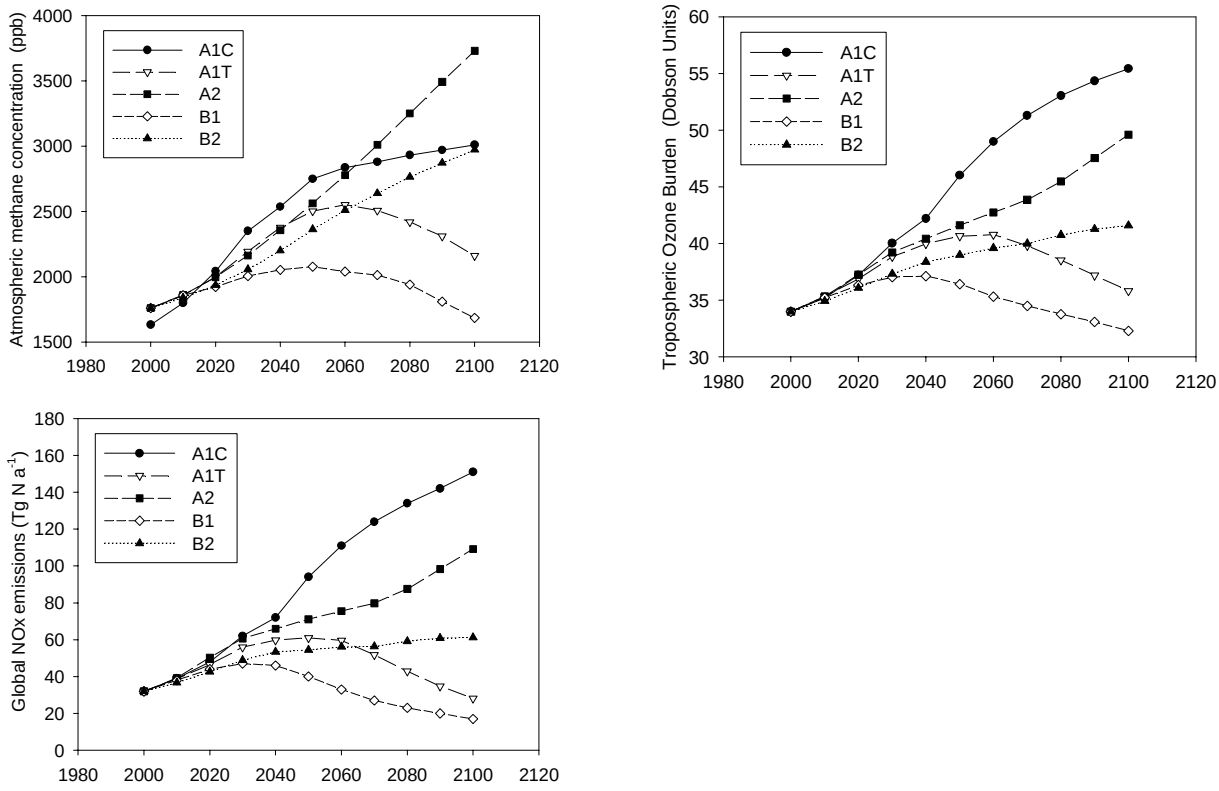

Figure 3.2. Atmospheric methane concentrations, global NOx emissions and tropospheric ozone burden according to some SRES scenarios (Prather et al., 2001).

3.3 European emissions of ozone precursors

In Figures 3.3 and 3.4, we show the trends of NOx emissions in the 'OECD90' and 'REF' regions and VOC emissions in the 'OECD90' region in various scenarios. These scenarios have been produced using the MESSAGE model (Nakićenović and Swart, 2000). Generally, emissions are lower in the environmentally oriented B and the globalization oriented 1 scenarios. The differences after 2050 are very large. For the A1 scenarios, very diverse trends are simulated depending on the energy production options. Between 2010 and 2040, NOx emissions increase in most scenarios in the 'OECD90' region, and in all scenarios in the 'REF' region. VOC emissions mostly decline after 2010, but in some scenarios, such as B2

and A1C, emissions are rather stable. Between 2000 and 2050, in the A1T scenario both NOx and VOC emissions decline by 25% in the 'OECD90' region. In the 'REF' region, NOx emissions increase by 70% and VOC emissions by 160% in the same period. However, it can be assumed that the socio-economic development and air pollution control legislation will be quite different in the 10 new EU countries, as compared to the other 'REF' countries.

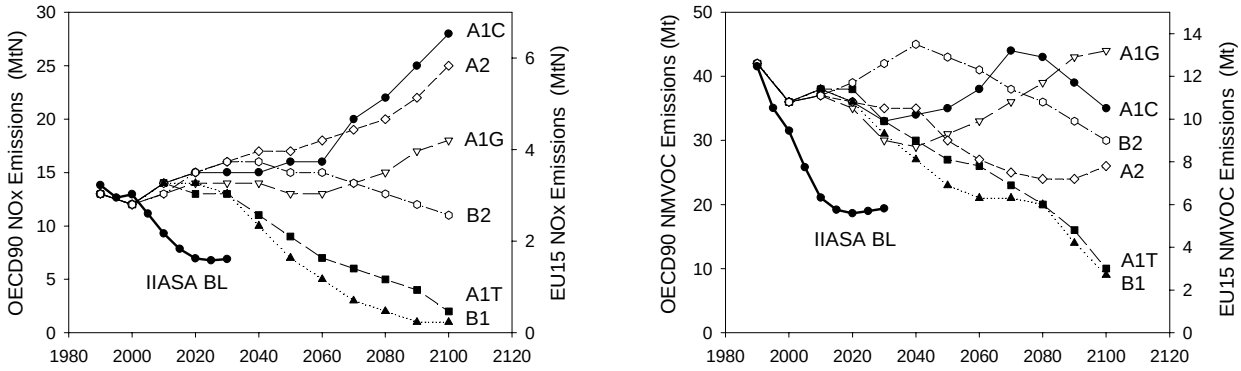

Figure 3.3. NOx and VOC emissions in the 'OECD90' countries according to different SRES scenarios (Nakićenović and Swart, 2000) and the projections of IIASA (Amann et al., 2004) for the 15 old EU countries assuming the CAFE baseline (BL) scenario up to 2030.

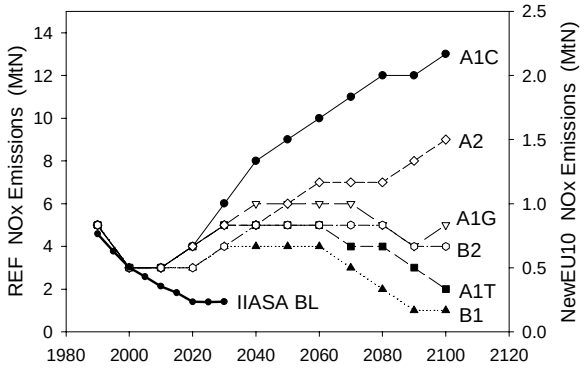

Figure 3.4. NOx emissions in the 'REF' countries according to different SRES scenarios (Nakićenović and Swart, 2000) and the projections of IIASA (Amann et al., 2004) for the 10 new EU countries in eastern central Europe assuming the CAFE baseline (BL) scenario up to 2030.

The role of the European air pollution legislation in the emission projections has been considered in two studies. The AIR-CLIM project (Alcamo et al., 2002) simulated changes in NO_x and SO_2 emissions in the A1 and B1 scenarios up to 2100 dividing Europe into three areas: Western Europe, the countries in Central and Eastern Europe undergoing economical reform and the newly independent states of the former Soviet Union. Under the present legislation, without any additional constraints to stabilize the atmospheric CO_2 concentration, NO_x emissions stay at the level of 2010 up to 2050 in all European areas except for the B1 scenario in Western Europe (Mayerhofer et al., 2002). In the 'advanced technologies' scenarios, they are roughly halved by 2050 compared to the 2010 level everywhere in Europe. In the central and western parts of Europe, emissions are considerably lower in the B1 scenario than in A1.

Within the CAFE programme of the European Commision, air pollution emissions of all EU and Western European countries have been recently estimated up to 2030 taking into account

environmental legislation (Amann et al., 2004). According to this study, NOx emission in the 15 old and 10 new EU countries will reduce by 26% and 34%, respectively, between 2010 and 2030. The corresponding reductions in the VOC emissions are 8% and 3%. Thus, it seems that the emissions produced by the SRES A1T scenario in 2050 may be quite realistic for the western part of Europe. Considering that the new EU10 countries, at least, follow a trajectory already diverging from the A1T scenario for the 'REF' region, the emissions estimated for Central Eastern Europe seem too high. In general, the division of the world into four regions for reporting the SRES emissions appears rather crude for mapping European emissions for regional air pollution studies.

3.4 Ozone exposures

3.4.1 Comparison of scenarios

Global ozone precursor emissions are the primary reason for the changes in tropospheric ozone burden. Prather et al. (2001) present an equation for calculating the tropospheric burden from global emissions. Comparison with the 3-D CTM simulations by Stevenson et al. (2000) shows that the relationship is valid in spite of the fact that ozone production efficiency varies regionally and seasonally. If we assume that regional ozone trends in the lower free troposphere follow the global tropospheric trends and that other factors may be taken into account as independent linear processes estimated from simulations from a regional CTM, we may develop a scheme for calculating local ozone exposure from global emission changes (Tuovinen et al., 2002; Laurila et al., 2004). We use this method to illustrate ozone exposures for different SRES scenarios and their temporal development.

The ground-level ozone concentrations were modelled by Tuovinen et al. (2002) using the regional-scale Lagrangian photochemical model of the EMEP/MSC-W (Simpson, 1992, 1995). For these calculations, the European precursor emissions reported for 2010 were scaled for different scenarios according to the data presented in the IPCC SRES report making division between the 'OECD90' and 'REF' countries (Nakićenović and Swart, 2000). The methane concentrations and the boundary conditions for ozone concentrations were also derived from the IPCC results (Prather et al., 2001). Surface temperature increase in climate change simulations is estimated based on Hulme and Carter (2000). Simulations of the present-day atmospheric composition use meteorological data for 1994 and emissions reported by the European countries for 1999, while the emissions for 2010 are according to the Gothenburg protocol of the UN/ECE (Vestreng, 2001).

To make it possible to obtain results for various scenarios, we extracted from the EMEP model simulations the effect of environmental and emission changes in Europe only. The effect of tropospheric background ozone concentrations, derived from the IPCC results, was then added to these changes for each scenario, and finally the values of the AOT40 index were recalculated using the method of Tuovinen (2002). AOT40 is a measure of the accumulated ozone exposure over a threshold concentration of 40 ppbv and is used to assess potential ozone effects on vegetation (Fuhrer et al., 1997). The accumulation period was 1 April – 30 September corresponding to the AOT40 used for forests (here denoted AOT40f).

Widely differing ozone concentrations are obtained after the mid-century for different SRES scenarios. We present results for the EMEP monitoring station of Utö, located in the Baltic Sea. The exposures in all scenarios reach the critical level by 2010 (Figure 3.5). The

maximum is reached in 2040 and 2050 in the more environmentally friendly B1 and A1T scenarios, respectively, after which AOT40f values begin to decrease. However, by 2100 only in the B1 scenario will AOT40f be below the critical level of 10,000 ppb h set for forests (Fuhrer et al., 1997). In the A1C and A2 scenarios, the modelled exposures grow to very high levels as a result of increasing background concentrations. We calculate that the exposure reduction resulting from the emission reductions according to the Goethenburg protocol using present-day boundary conditions is 740 ppb h from the present exposure of 8600 ppb h. The effect of even substantial European emission reductions is minor compared to the great increases resulting from the increases in the free tropospheric ozone concentrations.

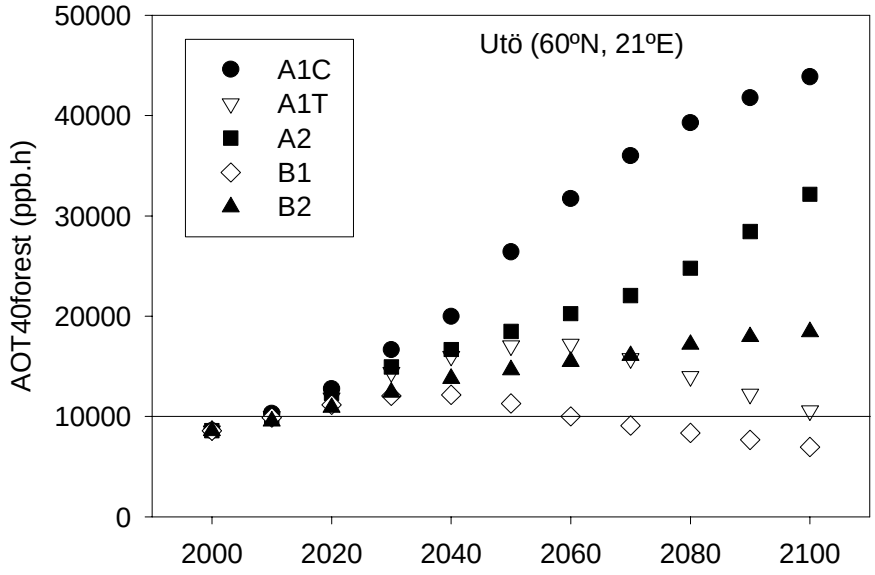

Figure 3.5. Scenarios of ozone exposure index AOT40f for the Utö monitoring station in the Baltic Sea. The critical level of 10,000 ppb h is also indicated.

3.4.2 Climate change effects

Climatological factors affect ozone concentrations in many ways including ozone precursor emissions, chemical reactions and transport processes. Warmer temperatures in the boundary layer accelerate biogenic VOC emissions and chemical reactions, increasing ozone concentrations in typical European conditions. On a global scale, climate warming increases water vapour concentration in the troposphere and the stratosphere-troposphere exchange, modifying ozone concentrations in the troposphere. Anthropogenic emissions of halogenated compounds have depleted stratospheric ozone concentrations increasing UV radiation in the troposphere that accelerates photochemical reactions. We may very well expect that UV radiation will have further variations during the 21st century.

Biogenic VOC emissions are strongly controlled by temperature (Guenther et al., 1993; Simpson et al., 1995), and an increase in surface temperatures in Europe may enhance the emission rates contributing to the amount of ozone precursors in the atmosphere. Tuovinen et al. (2002) showed that a simple climate warming scenario, in which temperature was increased by a constant value corresponding to the SRES A1C scenario for 2050, resulted in an enhancement of biogenic VOC emissions which increased ozone exposures significantly. The effect was greater in southern Europe than in the Nordic countries because climate

warming and photochemical activity are higher there. The simulations show that the enhanced biogenic VOC emissions are estimated to increase AOT40f by about 600 ppb h (for A1C in 2050) from the present day level of 6000–9000 ppb h in southern Finland. The effect is smaller in northern Scandinavia. For comparison, we estimated that the European emission reductions expected by 2010 according to the Gothenburg protocol will result in less than 10% decreases in the AOT40f index calculated using the 1999 emissions. In addition to biogenic VOCs, evaporative emissions of anthropogenic VOCs will be higher in warmer climate. However, this process and its effect on ozone formation have not been quantified.

Most rates of chemical reactions involved in the ozone formation are temperature dependent. Higher temperature in the boundary layer tends to enhance photochemical ozone formation in the temperature range 10–35 ºC (Tarvainen et al., 2004; Laurila et al., 2004). The positive effect is highest in conditions corresponding to typical Nordic sunny summer days when temperatures at noon are between 20 and 30 ºC. Considering the troposphere globally, under warmer conditions net ozone formation is lower because water vapour concentration will be higher. Increased water vapour concentrations increase concentrations of the OH radical and decrease methane and ozone concentrations (Brasseur et al., 1998; Stevenson et al., 2000). Based on simulations with the STOCHEM model, Stevenson et al. (2000) showed that tropospheric warming would decrease the average tropospheric ozone concentration increase by about 20% by the middle of the century, and by 35% and 50% by the end of the century in the A2 and B2 scenarios, respectively (Figure 3.6). This effect enhances towards the end of the 21st century because there is a lag between GHG emissions and warming. Figure 3.6 also indicates that the simple relationship by Prather et al. (2001) used here gives estimates that are very close to those based on the 3-D CTM simultations by Stevenson et al. (2000) for years 1990, 2030, 2060 and 2100.

There are also other climate factors that may act to the opposite direction. Climate warming accelerates stratosphere-troposphere exchange increasing the tropospheric ozone of stratospheric origin (Collins et al., 2003). Zeng and Pyle (2003) and Sudo et al. (2003) found that in climate change simulations up to 2100 for the SRES A2 emissions, the Brewer-Dobson and the Hadley circulations intensify causing enhanced ascent in the tropics and descent in the subtropical lower stratosphere. Future climate change induces increases in downward cross-tropopause ozone flux overcoming enhancements in upward tropospheric ozone transport due to increased ozone in the troposphere with emission increases. Sudo et al. (2003) estimate a 80% increase in the ozone transport from the stratosphere to the troposphere during 1990–2100. Ozone exposures calculated for central Finland (Figure 3.6) using the simulation results by Zeng and Pyle (2003) are higher than those based on Prather et al. (2001) and Stevenson et al. (2000). In contrast to Stevenson et al. (2000), ozone exposure in a warmer climate is higher because increased stratospheric flux surpasses enhanced tropospheric ozone loss by reaction of water vapour and photolytically excited ozone $O(^1D)$.

Stratospheric ozone depletion is an important climate change related issue which impacts ozone concentrations in the troposphere as well. Decreased ozone concentrations in the lower stratosphere diminish the flux to the troposphere. Total ozone reduction increase UV radiation fluxes enhancing photochemical activity in the troposphere (Fuglestvedt et al., 1994). Simulations by Isaksen et al. (2004) show that in the unpolluted low-NOx region of the atmosphere, net ozone destruction is enhanced, and in polluted regions in summer ozone formation is lower. In mid and high latitudes in winter, when UV radiation levels are low and rate limiting, ozone formation speeds up when total ozone is low. Sudo et al. (2004) show that the stratospheric ozone depletion since about 1980 has had a significant negative effect on the

tropospheric ozone trend. In response to the stratospheric ozone depletion during 1970–2000, stratospheric ozone input to the troposphere decreased significantly, and the global mean chemical lifetime of tropospheric ozone also decreased by 5–20% owing to increased UV radiation.

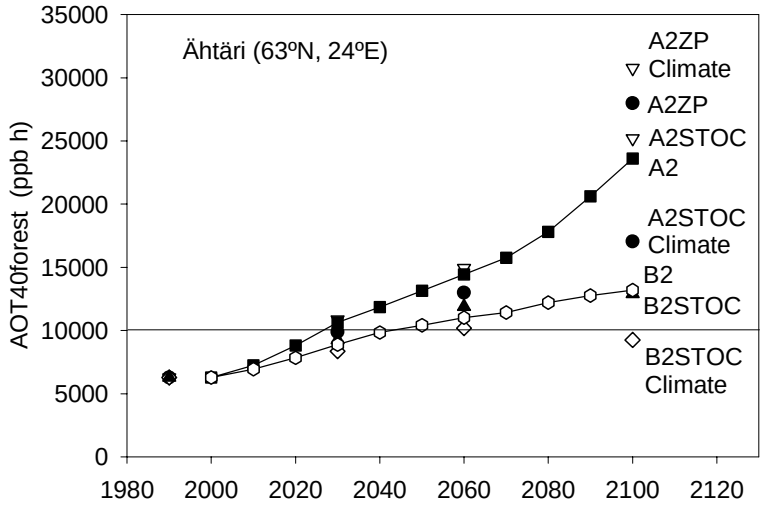

Figure 3.6 Modelled development of AOT40f at Ähtäri for the A2 and B2 scenarios assuming the tropospheric ozone change according to Prather et al. (2001) (A2 and B2) and Stevenson et al. (2000) without climate change (A2STOC, B2STOC) and with climate change (A2STOC Climate and B2STOC Climate). Shown are also the AOT40f for the A2 scenario with (A2ZP Climate) and without (A2ZP) climate change effects for 2100 based on the ozone burden simulations by Zeng and Pyle (2003).

Climate change will alter distributions and routes of low and high pressure areas that have a direct impact on occurrence of weather conditions favouring ozone formation. Estimation of advection and wet and dry deposition of ozone and its precursors during the transport from a source to a receptor calls for a 3-dimensional CTM study. The impacts of regional climate change are more difficult to estimate because natural variability inherent in meteorological fields may mask the climate change signal. Multi-year simulations using well documented meteorological fields have to be conducted to reach confident results (see Section 5).

4. Impact of emission changes on European ozone levels

4.1 Global model calculations

Jostein Sundet and Ivar S.A. Isaksen, University of Oslo

4.1.1 Description of the Oslo CTM2 model

The global chemical transport model Oslo CTM2 is used to study the global oxidant impact of possible future emission changes. Previously this model has been employed in a range of atmospheric studies including oxidant chemistry and natural and anthropogenic aerosols (Sundet, 1997; Jonson et al., 2001; Kraabøl et al., 2002; Gauss et al., 2003; Wild et al., 2003; Hsu et al., 2004).

The horizontal resolution of the Oslo CTM2 model is 2.8° × 2.8°, and the vertical resolution is 600 m in the troposphere decreasing to 2 km at 10 hPa, the grid comprising 40 hybrid-pressure levels. The transport data are generated with a time resolution of 3 h using the ECMWF model (http://www.ecmwf.int). The chemistry scheme is based on lumping and the QSSA integrator (Hesstvedt et al., 1978; Berntsen and Isaksen, 1997). Photodissociation is calculated on-line with the "Fast-J" scheme (Wild et al., 2000). Convection is based on the convective statistics from the ECMWF model, and is calculated according to surplus or deficiency of mass in an air column. The anthropogenic surface emissions are taken from the EDGAR database (http://arch.rivm.nl/env/int/coredata/edgar) and natural emissions mostly from Müller and Brasseur (1995). The non-surface emissions are obtained from a variety of sources. Aircraft emissions are based on the NASA 1992 inventory and scaled up to 1997 levels, while lightning emissions of NOx are based on Price et al. (1997). The surface-based emissions of NOx are divided into low-level (below 100 m) and high-level sources, the latter comprising a significant fraction (75%) of the total emissions. The stratospheric boundary condition of ozone is initialized every day with data from the Oslo CTM2 model run with a full stratospheric chemistry included, but at a lower horizontal resolution (5.6° × 5.6°). Thus, there is a large degree of consistency between the two models when the same meteorological data are used, which is the case in this study. NOy is calculated from the ozone distribution in the lower stratosphere, and scaled according to a measured ratio.

4.1.2 Modelling set-up

For this study, the baseline experiment is run for 1997 with meteorological and emission input for the same year. A spin-up of 12 months is used, based on a previous 1997 run. This spin-up time is more than sufficient, but is applied to facilitate comparison with a run for an emission scenario for 2050. The scenario run is made for 2050 based on the emission data of the SRES A1T scenario discussed in Section 3 and shown in Table 3.1.

4.1.3 Results

4.1.3.1 Present (1997) ozone levels

Many of the chamical species treated in the model have been analyzed in several EU projects (POET, RETRO, TRADEOFF) and have been shown to agree well with measurements. In polluted regions in summer, the surface ozone concentrations are high since the available solar radiation is high and the chemical system is active (Figure 4.1). The highest concentrations are found in the industrial areas over Europe, North America and South East Asia. In Europe, the highest ozone values occur over the Mediterranean with the mean values above 70 ppbv in July. In winter, the highest concentrations are found over ocean areas in the Northern Hemisphere since ozone loss is smaller there than over land. Values above 60 ppbv are reached in oceanic regions around India and South East Asia. Over Europe, the Mediterranean stands out with values above 40 ppbv. The modelled distribution fits well with measurements around the world (e.g., Wild et al., 2003).

The seasonality of ozone in the upper troposphere is mostly determined by the stratospheric supply of ozone into the troposphere. Thus the free tropospheric ozone is mostly determined by the stratospheric boundary condition of ozone, which is found to be good for most of the year, but possibly with too high values in the northern spring at mid-latitudes.

For CO concentrations, the seasonal variability is smaller than observed. This is probably related to the lack of seasonal variability in the emissions from biomass burning, resulting in CO concentrations that are too high in summer and too low in winter, in particular at high northern latitudes. Too small OH concentrations could also be a candidate for explaining this, but if one looks at the lifetime of methyl chloroform in the Northern Hemisphere, this does not seem likely.

4.1.3.2 Future (2050) ozone levels

Compared to the baseline run, we see a significant increase in the free tropospheric ozone concentrations from our scenario run with the SRES A1T emissions for 2050. Most of this increase can be attributed to emission increases in India and South East Asia, where the increase in NOx is more than 200%. Increase in the surface ozone concentrations shows more regional features, since the lifetime of ozone is shorter in the boundary layer than in the free troposphere (Figure 4.2). This also shows that the sensitivity to free tropospheric ozone in the boundary layer is rather limited (Wild et al., 2004). Regional limitation of emissions for pollution control issues is thus rather efficient, even if other regions increase emissions resulting in increased ozone and aerosol concentrations.

The A1T emissions for 2050 result in a significant increase in the surface ozone concentrations. Over large parts of Asia, the increase is from about 25 ppbv up to 55 ppbv. with the largest values over India and the Arabian Peninsula. The largest relative increase of about 40% is simulated over South East Asia. We also observe a small reduction over the Mediterranean and over parts of Eastern U.S., which results from the reduction in local emissions. From the zonally averaged ozone distribution, we observe that the largest increase occurs in the upper troposphere (Figure 4.3). One reason for this is the larger ozone production efficiency in this region since the primary components are readily transported into the upper troposphere by convection in the summer season. The largest relative increase of

ozone is found lower down in the free troposphere with a maximum increase of 20% at about 5–6 km.

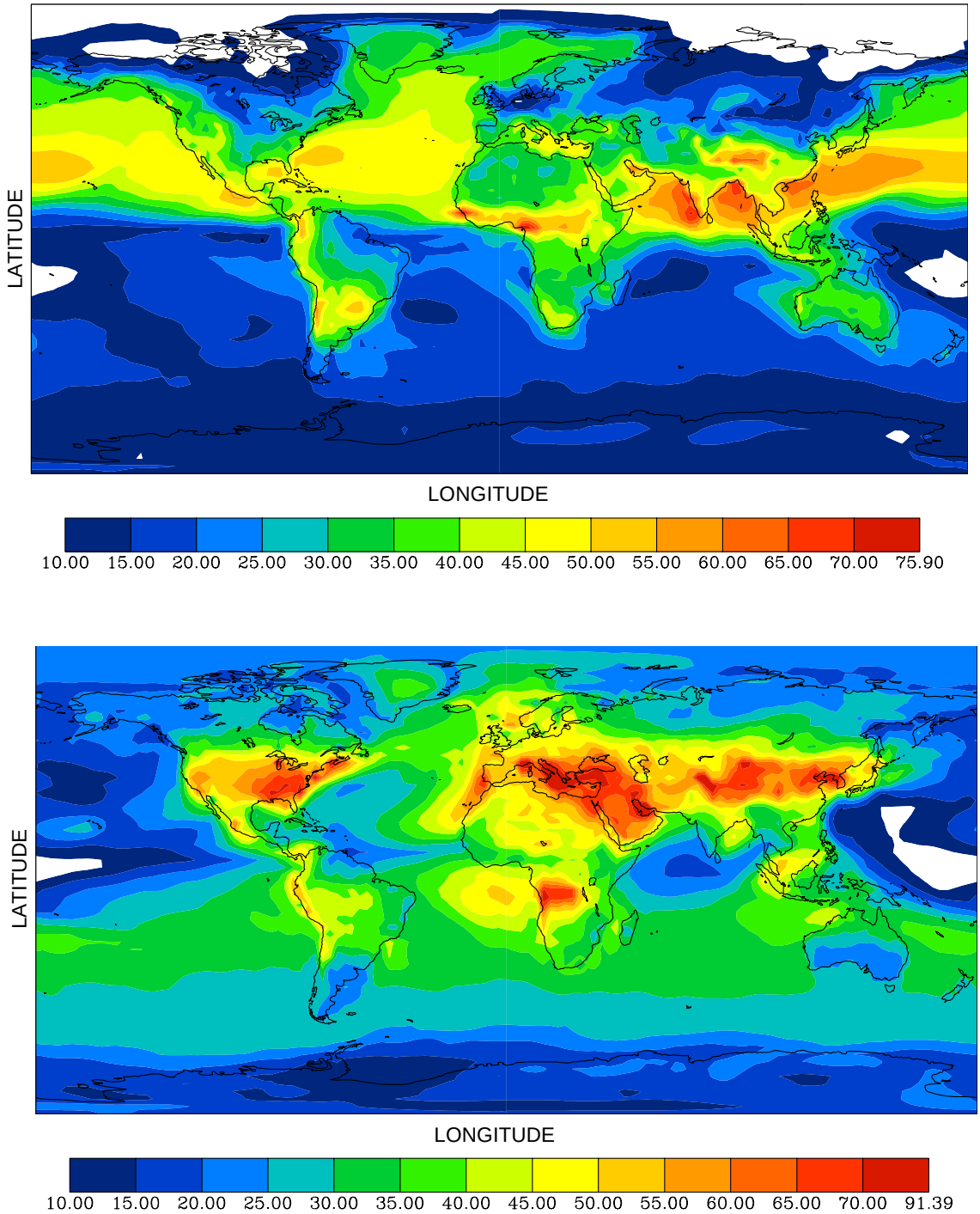

Figure 4.1. Surface ozone concentration (ppbv) in January (top) and July (bottom) 1997 modelled using the Oslo CTM2 model. White areas correspond to values below 10 ppbv.

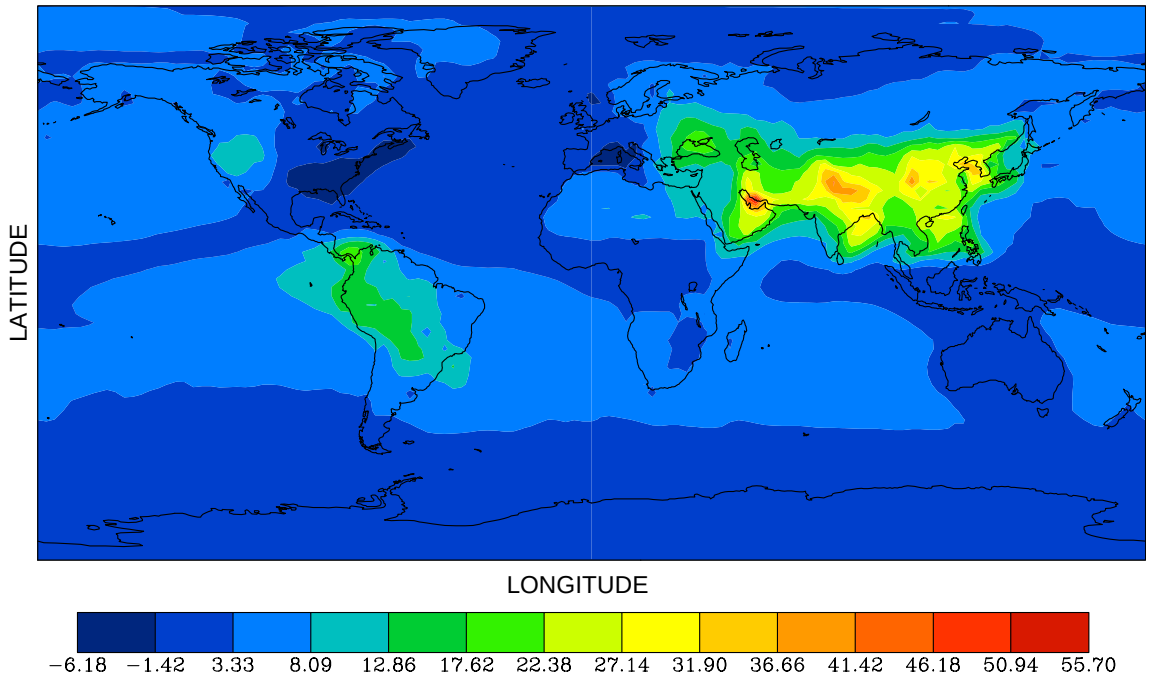

Figure 4.2. Absolute difference in the mean surface ozone concentration (ppbv) between 2050 and 1997 resulting from different emission data, modelled using the Oslo CTM2 model.

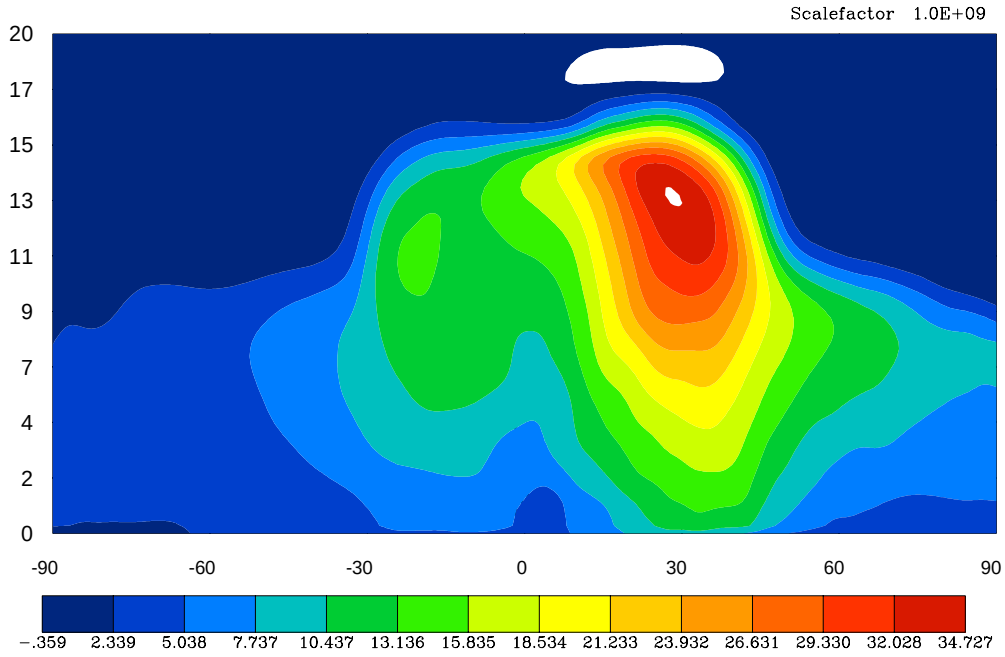

Figure 4.3. Absolute difference in the zonally averaged ozone concentration (ppbv) between 2050 and 1997 resulting from different emission data, modelled using the Oslo CTM2 model.

4.2 Regional model calculations

Jan Eiof Jonson, EMEP/MSC-W

4.2.1 Description of the Unified EMEP model

The Unified EMEP photochemistry model has been developed as a tool for calculating long-term source-receptor relationships of air pollutants at the European regional scale as part of the EMEP program. It describes the transport, chemical transformation and deposition of approximately 75 species, among them ozone, CO, NO_x, SO_2, NH_3, numerous VOCs, HNO_3, primary (unspeciated) fine and coarse particulate matter. The model domain is centred over Europe and also includes most of the North Atlantic and the polar region with a horizontal resolution of approximately 50 km × 50 km and 20 vertical layers below 100 hPa. Dry deposition is modelled using a resistance approach. Wet scavenging is set to be proportional to the precipitation intensity using species-specific scavenging coefficients. Photolysis rates are tabulated for clear sky and cloudy conditions. A detailed model description can be found in Simpson et al. (2003a) and Fagerli et al. (2004). In the present model version monthly averaged lateral boundary concentration of several species including ozone are extracted from the global model calculations described in Section 4.1.

The model has been extensively tested (Fagerli et al., 2003; Simpson et al., 2003b; Jonson et al., 2004) and reviewed (ECE, 2004a; Velders et al., 2003) and earlier versions of the model have participated in several model intercomparison studies (van Loon et al., 2004; City-Delta 2003–2004; Euro-Delta, 2004).

4.2.2 Modelling set-up

For this study, the EMEP photochemistry model has been run with present (1997) and a future (2050) emission scenario for SOx, NOx, non-methane VOCs, CO and NH_3. All model runs have been made with the 1997 meteorology, the same meteorological year as was used for the Oslo CTM2 model. Two sets of monthly averaged lateral boundary concentrations from the Oslo CTM2 model have been used. The first set has been calculated with present global emissions, whereas the second set has been calculated with the emissions of the SRES A1T scenario for 2050 (Table 3.1). Three cases have been modelled:

1. Present (1997) emissions and 1997 lateral boundary concentrations
2. Future (2050) emissions and 2050 lateral boundary concentrations
3. Future (2050) emissions and 1997 lateral boundary concentrations

The difference between the first two model runs will give an indication of the effects of the projected changes in global emissions on Europe. The difference between model runs 2 and 3 will illustrate to what extent the calculated changes in air pollution can be attributed to European emission changes alone.

4.2.3 Results

4.2.3.1 Present (1997) ozone levels

In order to assess how well the year 1997 represents present ozone levels, we compare the ozone concentrations modelled for the summer months of this year with the corresponding 6-year average for 1995–2000 (Figure 4.4). For most of Europe, the calculated ozone levels in 1997 were close to the 6-year average. Over large parts of Norway, parts of Sweden and over the southern parts of Finland, the concentrations were slightly higher in 1997 than in the average calculated for the 6 years period 1995–2000.

The average modelled daily maximum concentrations in April–September 1997 are shown in Figure 4.5 (top left). Over the Nordic countries, these concentrations are typically in the 35–45 ppb range. As was the case in the global model calculations, the highest levels (in Europe) are calculated for the central parts of the Mediterranean. The average over the 7 days with the highest ozone concentrations (Figure 4.5, top right) shows a similar distribution, but high levels are also calculated over north-western parts of Europe. In the Baltic, ozone levels above 70 ppbv are calculated.

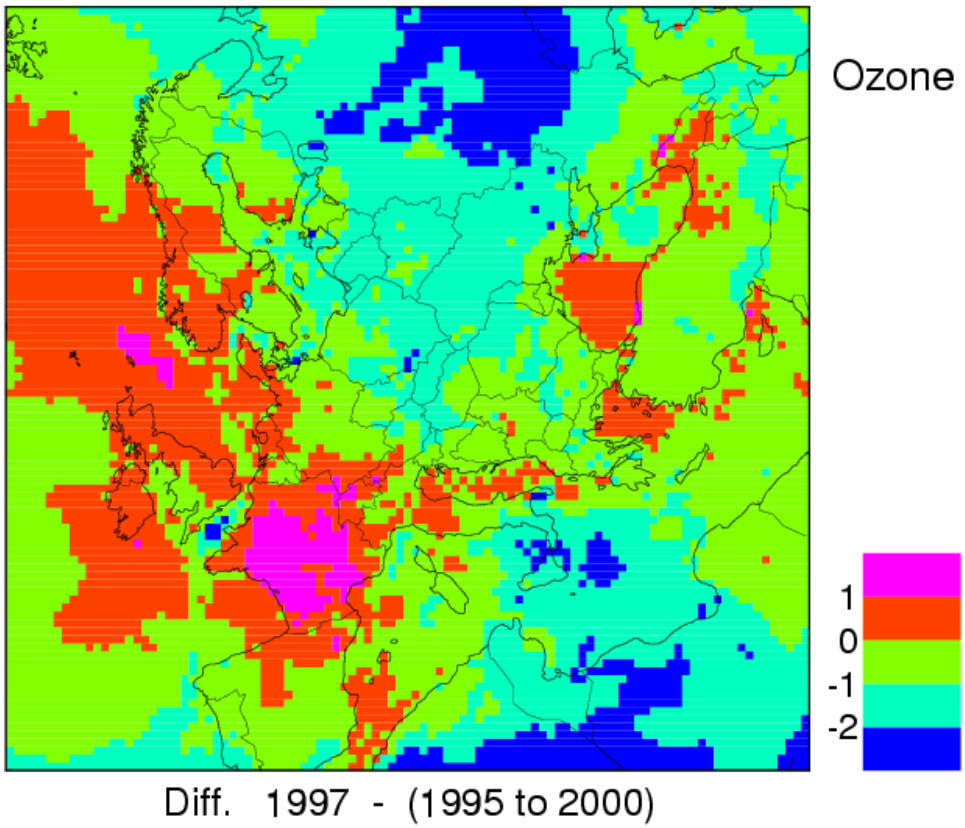

Figure 4.4. Difference in the average daily maximum ozone concentrations in April–September between 1997 and the 6-year average 1995–2000.

Figure 4.6 depicts the values of the ozone exposure in terms of the AOT40f index (top left) and the SOMO35 index (top right). AOT40f is explained in Section 3.4.1. SOMO35 denotes the accumulated daily maximum of the 8-hour running average above 35 ppbv integrated over the whole year and is used as a measure of human exposure to ozone (ECE, 2004b). As for the average daily maximum concentrations, high AOT40 and SOMO35 levels are in

particular seen in the Mediterranean and over Italy. Over the Nordic countries, AOT40f and SOMO35 levels are rather low and, AOT40f being well below the critical level of 10,000 ppbh.

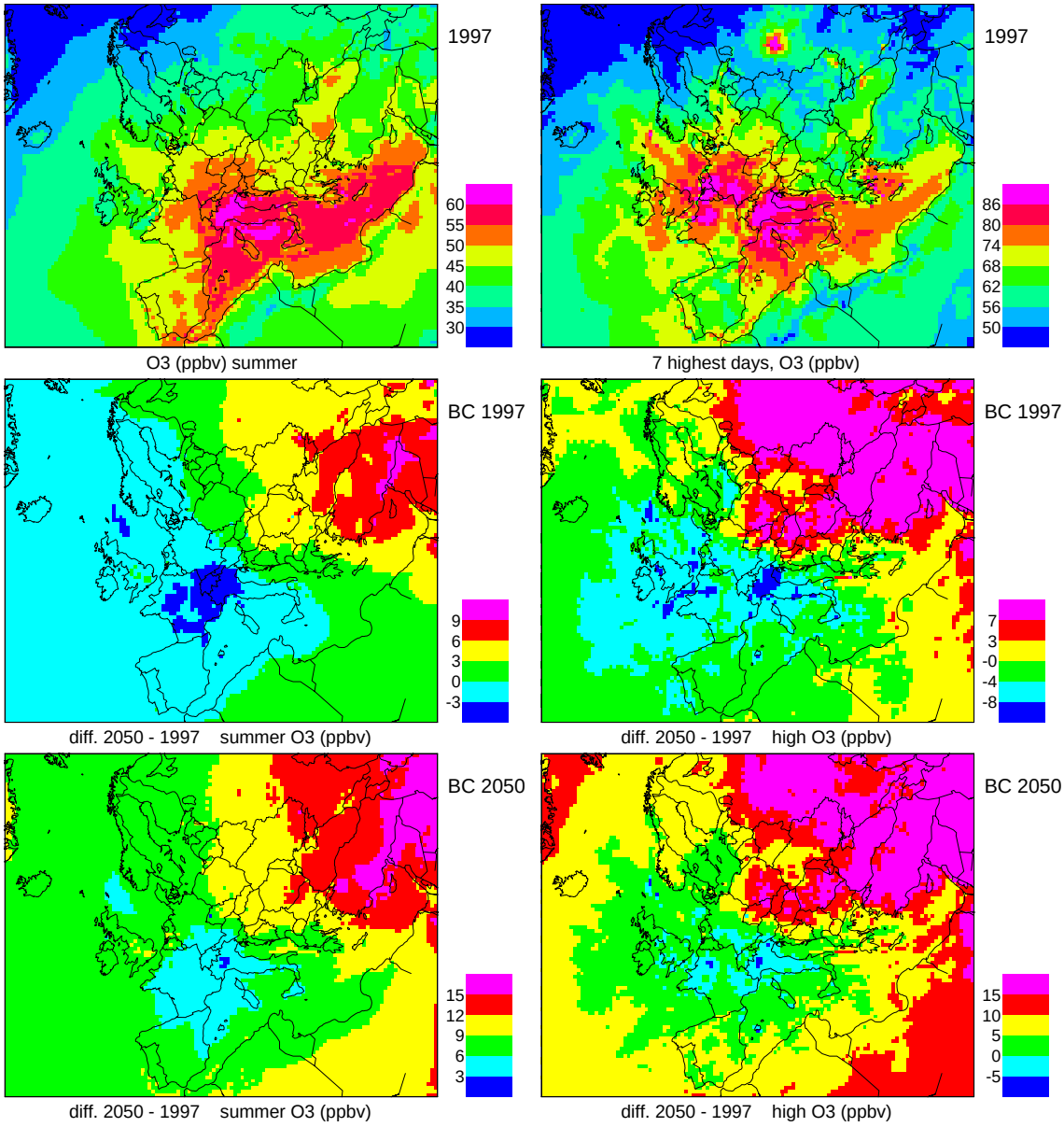

Figure 4.5. Left column, the average daily maximum ozone concentration in April–September: 1997 (top), difference between 2050 and 1997 with lateral boundary concentrations calculated with the global model using the 1997 emissions (middle) and the 2050 emissions (bottom). The right column depicts the same differences averaged over the 7 days with the highest ozone concentrations.

4.2.3.2 Future (2050) ozone levels

In the emission scenario used in this study (see Table 3.1) there is a distinct difference in the emission trends for the western and the eastern European countries. In Western Europe, emissions are expected to decrease by the year 2050, as compared to 2000, whereas in Eastern Europe they are expected to increase. This difference is clearly seen in the ozone

concentrations calculated with the 2050 emissions in Europe and the 1997 lateral boundary concentrations from the global model. In 2050, all quantities (mean daily maximum, maxima, AOT40f and SOMO35) are markedly lower over Western Europe and higher over Eastern Europe. Over the Nordic countries, the mean daily maximum in summer is lower. For high ozone events the tendency is mixed, reflecting the fact that these events may be generated over either Western or Eastern Europe. Even though the mean summer concentrations have increased, the high ozone events are reduced over parts of Western Europe, and in particular in Northern Italy.

Repeating the calculations with the lateral boundary concentrations calculated with the 2050 emissions in the global model has a large effect on ozone levels. The mean daily maximum, AOT40f and SOMO35 increase all over Europe as a result of an increase in lateral boundary concentrations. Even though the mean daily maximum in summer has increased substantially high ozone events are virtually unchanged, and for some regions lower than 1997 levels in Western Europe. On the other hand they are higher than when calculated with 1997 lateral boundaries and 2050 emissions. This suggests that the high ozone events are controlled by European emissions, superimposed on the background ozone levels.

The effects of emission chances on AOT40 and SOMO35 reflect the chances in ozone concentrations. However, the changes are larger for AOT40 than for SOMO35 demonstrating the high sensitivity of this parameter even to small or moderate changes in ozone levels.

4.3 Comparing global and regional calculations over Europe

The concentration fields in the two models are not directly comparable. In the global model July concentrations are presented, whereas in the regional model an average of the summer months April through September is depicted. This may to some extent explain the higher ozone levels in the global model. In both models the highest ozone levels are seen in and around the Mediterranean ocean. In both models concentrations decline as we go north. In the regional model very high ozone levels are seen in Northern Italy. This is not the case in the global model. Such local differences may be caused by a higher horizontal resolution in the emission inventories and in the model calculations in the regional model.

The change in surface ozone from 1997 to 2050 over the Atlantic is smaller in the global model compared to the regional model calculations. Even so differences in European ozone levels are similar in both model calculations, with small changes over Western parts of the continent and a substantial increase in Eastern Europe. The main difference in the calculations for Europe is over the Mediterranean where the global model calculates a decrease in ozone levels, whereas the regional model calculates an increase.

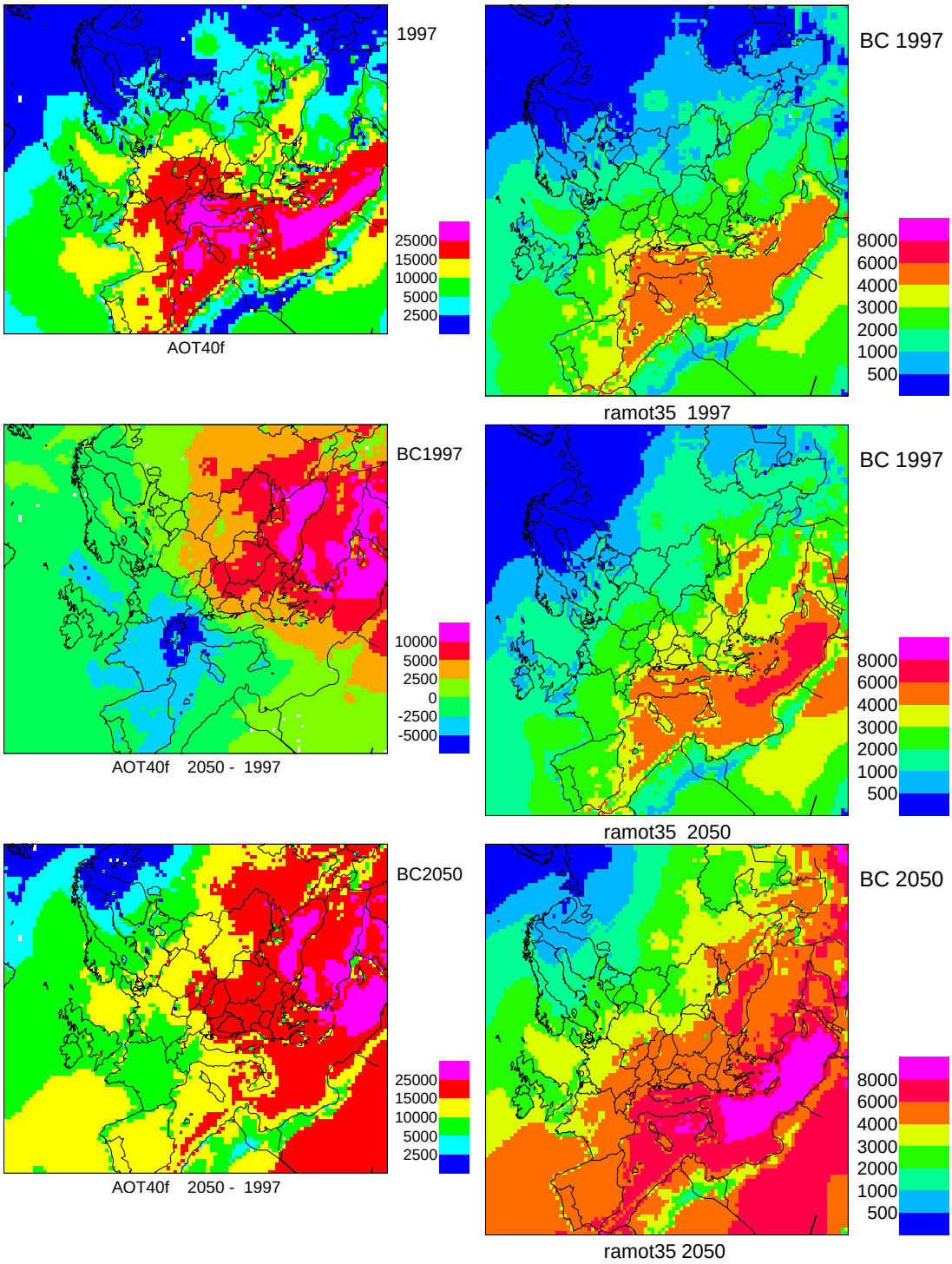

Figure 4.6. Left column, AOT40f exposure index: 1997 (top), difference between 2050 and 1997 with lateral boundary concentrations calculated with the global model using the 1997 emissions (middle) and the 2050 emissions (bottom). The right column, depicts the same for the SOMO35 (ramot35) exposure index.

5. Impact of climate change on European ozone levels

Joakim Langner, Robert Bergström and Valentin Foltescu, SMHI

5.1 Description of the MATCH model

The MATCH (Multi-scale Atmospheric Transport and Chemistry) model (Robertson et al., 1999) has been developed as a flexible, offline, chemistry/transport/deposition model for atmospheric pollutants. MATCH has been extensively evaluated on the European scale and compared well with observations and was also well in line with a range of European CTMs in two recent European model inter-comparisons (Roemer et al., 2003; Hass et al., 2003). A specific model evaluation for ozone in the Nordic area has also shown good agreement with observations for two different years (Solberg et al., 2002).

Advection in MATCH is modelled using a Bott-type advection scheme (Bott, 1989, 1992). Fifth order integral functions were used in the horizontal and an upstream scheme in the vertical direction. Dry deposition is modelled using a resistance approach. Wet scavenging is set to be proportional to the precipitation intensity using species-specific scavenging coefficients. The gas-phase chemical mechanism used is based on the EMEP/MSC-W model chemistry (Simpson et al., 1993) with some changes in the isoprene oxidation (Langner et al., 1998). The photochemical model includes about 130 thermal and photochemical reactions between 59 chemical components. Production of ammonium sulphate, ammonium nitrate and nitrate on coarse aerosols is modelled following Hov et al. (1994). Aqueous-phase oxidation of SO_2 by O_3 and H_2O_2 in cloud water and gas-aqueous-phase equilibria for these components is modelled following Berge (1992). Further details on the basic transport model can be found in Robertson et al. (1999). More details about the photochemistry version of the model as well as comparisons with observations can be found in Langner et al. (1998).

Anthropogenic emissions for the simulations were derived from the 50 km × 50 km emission data provided by the EMEP/MSC-W The emissions for 1996 for NO_x, SO_x, non-methane hydrocarbons, CO and NH_3 were used in the model calculations. Simple variations of the emissions with the time of day and with the day of the week were used. Also monthly variations were specified. Boundary conditions in terms of concentrations for all simulated species were set to representative values for present conditions based on observations where available. Biogenic emissions of isoprene were calculated in MATCH, based on 2-meter temperature and radiation from the regional climate model, using the E-94 isoprene emission methodology presented by Simpson et al. (1995).

5.2 Regional climate change scenarios for Europe

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

taken from climate change experiments performed with the regional climate model RCA version 1 (RCA1) (Rummukainen et al., 2001; Räisänen et al., 2001). These regional climate scenarios used boundary conditions from two different GCMs: HadCM2 (Johns et al., 1997) and ECHAM4/OPCY3 (Roeckner et al., 1996, 1999).

RCA1 was run for 10-year control and scenario time slices from both the HadCM2 and ECHAM4 simulations, hereafter referred to as H88 and E88, respectively. The control time slices correspond to present climate, while the scenario time slices correspond to periods 50–70 years ahead depending on the GCM. The scenario time slices correspond to changes in greenhouse gases of 150% (HadCM2) and 100% (ECHAM4) compared to the control period. The corresponding changes in global mean temperature are about 2.6 K in both GCMs. MATCH used 6-hourly output from RCA1 for winds, temperature, humidity, cloud cover and cloud water on model levels, and a range of surface parameters. Data from all the 19 levels of the RCA1 model were used for vertical wind calculations, but the transport calculations in MATCH extended only over the eight lowest levels of the original RCA1 data or up to a height of about 5.5 km. MATCH version 3.9.1 was used.

5.3 Results

Comparisons with observations were made between the 10-year control simulations and observed deposition of sulphur and nitrogen and ozone concentrations. Observed data for this comparison were drawn from the EMEP database. There are significant trends in emissions and consequently in air concentrations and deposition over Europe over the last decades. We have therefore compared the 10-year MATCH simulations with average statistics for the observed data over a five-year period centred on 1996, corresponding to the year for the anthropogenic emission data used in the MATCH simulations. Statistics for the comparison are given in Table 5.1. A complete presentation of the comparisons with observations can be found in Langner et al. (2004).

Table 5.1. Statistics for the comparison of five-year average observed and simulated annual non-seasalt sulphate wet deposition, nitrate wet deposition, April–September mean of daily maximum ozone and April–September AOT40 (AOT40f) for the H88 and E88 control simulations.

	Sulphate wet deposition		Nitrate wet deposition		Mean of daily maximum ozone		AOT40f	
	H88	E88	H88	E88	H88	E88	H88	E88
Spatial correlation	0.37	0.38	0.50	0.44	0.48	0.58	0.78	0.83
Fractional bias (%)	10	2	2	3	-2	-1	-29	-21

For wet deposition, the model simulations are close to observed values on average for model simulations based on both H88 and E88. The spatial correlation for annual deposition is however quite low and there is considerable scatter. This is not surprising given the strong inter-annual variability in precipitation, both in the RCA1 simulation and in reality. Spatial correlation coefficients for single years using analysed meteorological data to drive MATCH result in substantially higher values in the range 0.7 to 0.8 for a similar set of stations (Langner et al., 1998; Zlatev et al., 2001; Hass et al., 2003). For surface ozone, the model simulations predict average values in good agreement with the observations with some

tendency for underestimation. The spatial correlation for AOT40f is high, around 0.8, while the correlation for the mean of daily maximum is lower due to the more narrow range of values.

Figure 5.1 shows the calculated 10-year average total deposition of oxidised nitrogen for the control simulations with both H88 and E88, as well as the relative difference between the scenario and the control simulation. There are both differences and similarities between the two scenarios. Both scenarios show decrease in deposition over central Europe, however, with a more pronounced decrease for the E88 scenario than for H88. These reductions in deposition are caused by decrease in wet deposition and are consistent with the reductions in annual precipitation in the E88 and H88 scenarios in the same areas. In the northern and eastern part of Europe the two scenarios give rather different results. The H88 scenario shows an increase in deposition over large parts of northern and eastern Europe, while the E88 scenario shows an increase only over north-eastern Europe and north-western Europe including western Norway. The decrease in wet deposition over central Europe leads to higher air concentrations of both primary and secondary oxidised nitrogen components over large parts of the modelling domain. This results in increased dry deposition, which actually counteracts the decrease in wet deposition in southern and central Europe. The changes in wet deposition dominate, however.

A problem with the interpretation of the simulated changes in deposition is the strong inter-annual variability in the results mainly driven by inter-annual variability in precipitation. The changes in total deposition in the climate scenarios are only statistically significant in relation to internal variability for limited areas of the model domain, as shown in the bottom panel in Figure 5.1. Given the low significance of the simulated changes, longer time periods should also be used when assessing changes. Mayerhofer et al. (2002) have presented a similar study on deposition using results from a regional climate model coupled to the one-layer Lagrangian CTM developed at the EMEP/MSC-W for source-receptor calculations in Europe (Barrett and Berge, 1996). The results presented by Mayerhofer et al. (2002) are generally similar to the ones presented here for the E88 scenario, although they find mainly reduced deposition (within a 5% reduction), while we find both decrease and increase in different areas (mainly within a 5% change).

Figure 5.2 shows the calculated 10-year average AOT40f for the control simulations with both H88 and E88, as well as the relative difference between the scenario and the control simulation. Relative differences for the April–September mean of daily maximum (MDM) ozone concentration are shown in Figure 5.3. In this case, there is greater similarity between the results using H88 and E88. Both scenarios show an increase in both AOT40f and MDM over southern and central Europe and a decrease in northern Europe. The increase is however larger in the E88 simulation in which it exceeds a factor of two for AOT40f over western Europe and 15% for MDM. These changes in surface ozone can be understood in terms of changes in the factors regulating the production of surface ozone.

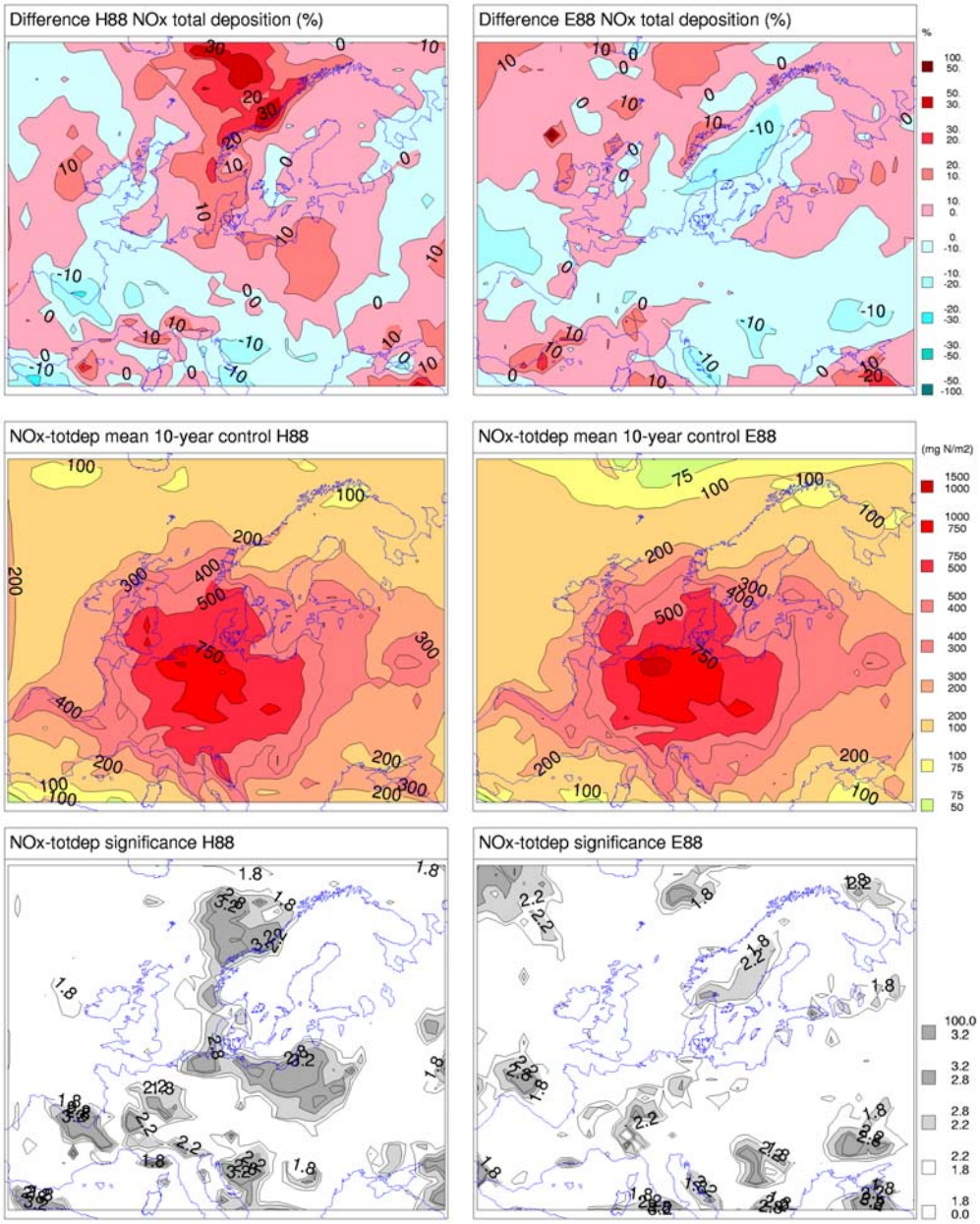

Figure 5.1. Calculated 10-year average deposition of oxidised nitrogen for the control and the scenario for the H88 (left) and E88 (right) simulations. Shown are the relative difference (top), the deposition in the control simulations (middle) and a test variable for significance of the simulated changes in relation to internal variability (bottom). Test variable values above 1.8 indicate statistical significance above the 95% confidence level.

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

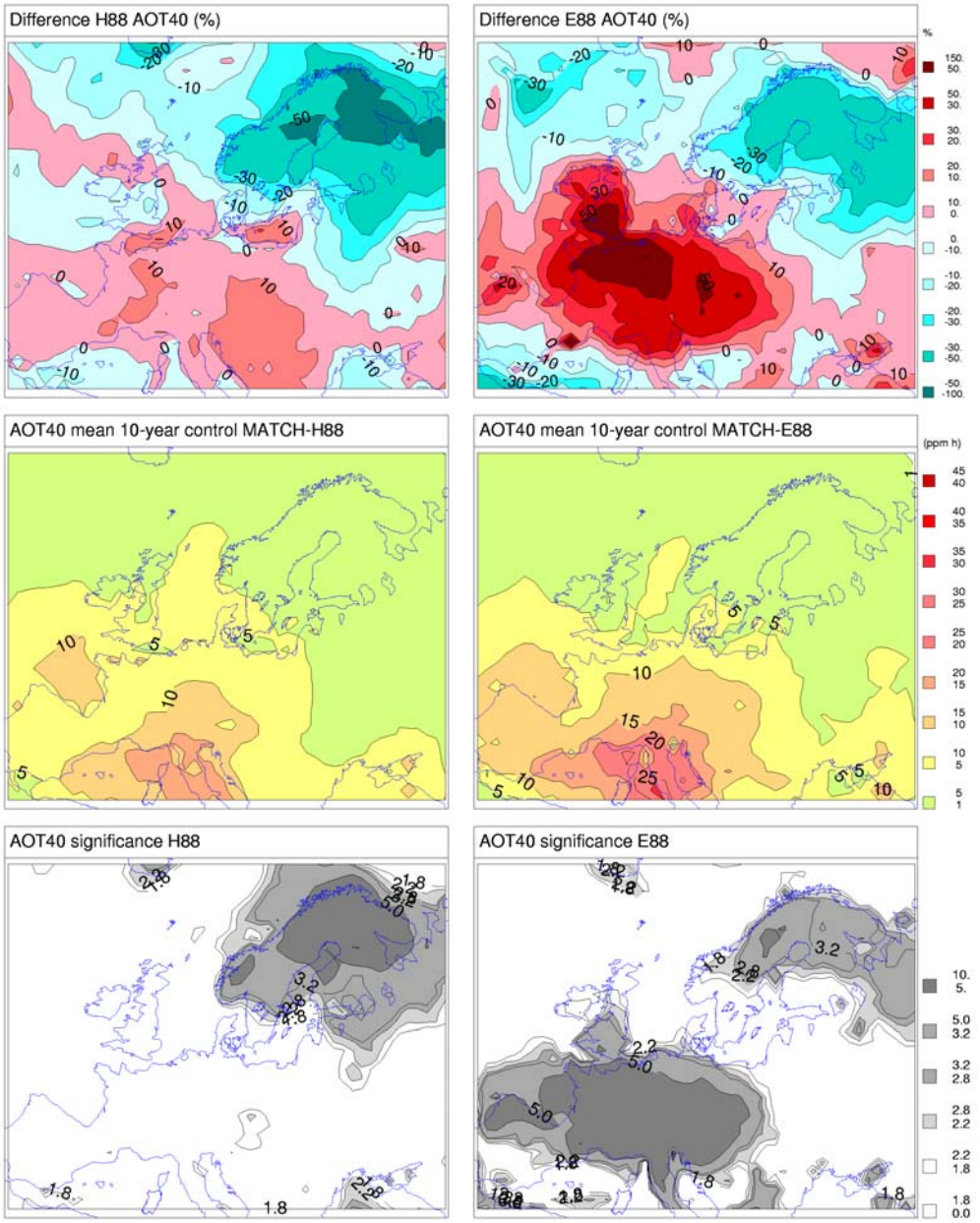

Figure 5.2. Calculated 10-year average AOT40f (April–September) for the control and the scenario for the H88 (left) and E88 (right) simulations. Shown are the relative difference (top), AOT40f in the control simulations (middle) and a test variable for significance of the simulated changes in relation to internal variability (bottom). Test variable values above 1.8 indicate statistical significance above the 95% confidence level.

Simulated relative changes in the mean ozone concentration for April–September look very similar to the changes in the mean of daily maximum. Changes in the annual mean ozone (not shown here) have a similar spatial pattern, but with a smaller extent of the area of increase. The changes in annual mean ozone are generally less than 5%.

This increase also extends partly over northern Europe, but in the northernmost part a decrease in AOT40f and MDM is simulated. In the H88 simulation, this is probably related to the strong increase in precipitation and cloudiness in this area during summer, which leads to removal of ozone precursors and less solar radiation. Increase in precipitation could also partly explain the decrease in the E88 simulation, but changes in circulation are probably also

involved so that less ozone and ozone precursors produced and emitted over southern and central Europe is transported north.

The bottom panel of Figure 5.2 shows the results of significance tests for the simulated changes in AOT40f. In contrast to total deposition of oxidised nitrogen, the changes in AOT40f are significant in relation to internal variability over quite large areas, especially in the E88 simulation which shows the largest changes. Both simulations show significant changes over the northernmost parts of Europe. The changes in AOT40f and MDM simulated in the scenarios presented here are substantial and the increase simulated over central Europe would be quite significant compared to the expected reductions resulting from emission reduction protocols currently in force.

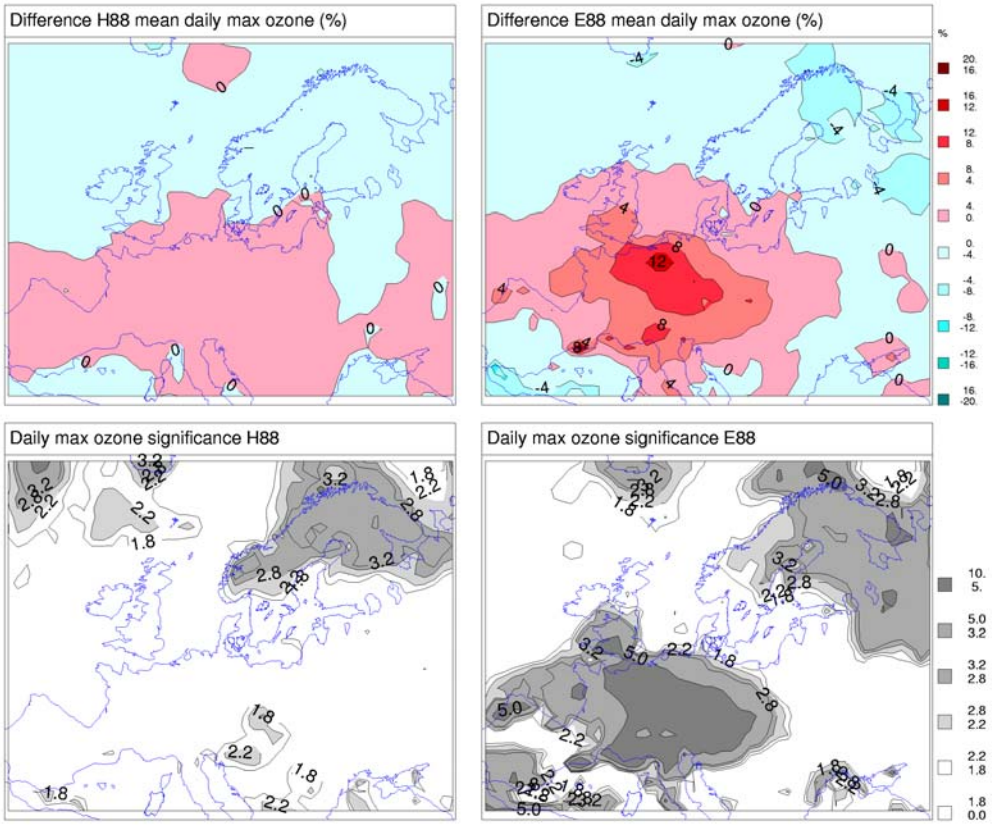

Figure 5.3. Calculated 10-year average relative difference in the mean of daily maximum (April–September) for the control and the scenario for the H88 (left) and E88 (right) simulations (top) and a test variable for significance of the simulated changes in relation to internal variability (bottom). Test variable values above 1.8 indicate statistical significance above the 95% confidence level.

Tuovinen et al. (2002) have presented a sensitivity analysis of the different factors affecting the future concentrations of ozone in Europe using the Lagrangian photochemical model of EMEP (Simpson, 1992). Apart from the major importance of increases in background concentrations of ozone, they found that increased biogenic emissions of VOC due to increased surface temperatures would significantly counteract the effects of reduced anthropogenic emissions of ozone precursors. The effect of temperature increase on chemistry would be less significant. Although we have not made a separate study of the impact of

increased biogenic VOC emissions here, it is likely that the substantial increases in biogenic VOC emissions simulated in both the E88 and H88 scenarios are responsible for a large fraction of the simulated increases in surface ozone. However, the increases in surface concentrations of oxidised nitrogen compounds as well as reduction in cloud cover over central and southern Europe are certainly also of importance.

6. Discussion and conclusions

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The prospects for long-term estimation of future ozone concentrations beyond the horizon defined by the current environmental legislation have advanced considerably during the recent years. This kind of estimation largely relies on the well-documented set of alternative future emission scenarios presented in the SRES report (Nakićenović and Swart, 2000) and on global CTM simulations. Consistent results may be drawn from studies on future atmospheric pollution, but significant uncertainties still remain. The main scope of this study has been to address how northern Europe is impacted by ozone pollution under long-term atmospheric global change.

A great number of CTM simulations (e.g., Prather et al., 2001; Collins et al., 2000) show that atmospheric methane concentration and global nitrogen oxide emissions are the most important determinants of tropospheric ozone concentrations. The SRES scenarios foresee great growth of energy demand in the world resulting in high ozone precursor emissions all over the world, in particular in those scenarios which rely on the use of fossil fuels. In all scenarios, including the environmentally friendly ones, precursor emissions are increasing particularly in the Far East and tropical areas where ozone production efficiency is high. High ozone concentrations are advected in the middle and upper troposphere from these areas to other parts of the world. Most of the scenarios produce so high growth rates of tropospheric ozone concentrations that environmental and health problems for example in Europe would be severe (Derwent et al., 2002).

The SRES scenarios were developed for estimation of emissions of GHGs and their precursors, ozone being one of the GHGs. The primary goal was to simulate the transition of energy production technology from traditional ways of fossil fuel combustion to more modern technologies including hydrogen, solar and wind energy, biofuels, and natural gas-fired combined cycle and nuclear power plants. Emissions of GHGs and ozone precursors follow mostly these technological developments. Environmental legislation for protecting human health and environment is not, as such, included in the modelling system. This is the main weakness of the SRES scenarios when they are used for simulation of ozone concentrations in the troposphere. The models used for the SRES studies include socio-economic factors such as standard of living and environmental awareness, which potentially could be used for modelling the development of environmental legislation.

Nitrogen oxide emissions originate mainly from combustion sources and their emissions are nowadays regulated in the industrialized countries. Amann et al. (2004) estimates that according to the current legislation NOx emissions in the western part of Europe, belonging to the ‘OECD90’ group in the SRES modelling, will be reduced by 47% between 2000 and 2020. The corresponding reduction in the 10 new EU countries is 54%. These countries are mostly located in eastern central Europe and belong to the ‘REF’ group in the SRES modelling. In all SRES scenarios, NOx emissions of the ‘REF’ group are increasing or stable. The same is true for nearly all scenarios for the ‘OECD90’ group. In Europe, we are clearly on a different track compared to the SRES emissions for the next few decades because strict regulations and the use of the end-of-pipe technology reduce emissions of air pollutants.

The atmospheric methane concentration is one of the key factors behind the tropospheric ozone burden. The emission scenarios for methane are highly uncertain, and even the present emissions are not accurately quantified. Wetlands constitute the greatest source category, followed by ruminants, energy production, rice agriculture, landfills and biomass burning. Climate and land use change will affect the most important source categories but, for example, there is not yet consensus about which direction the emissions from natural wetlands will change. All SRES emission scenarios simulate increasing methane concentrations in the beginning of the 21st century, while observations show that the increase has levelled off (Dlugokencky et al., 2003), for reasons which have not yet been resolved.

Using a simple modelling approach and the SRES emissions, we estimated that the ozone exposures in 2100 are high or extremely high in the northern parts of Europe. The variation between the scenarios is relatively low during the first part of the century, but after 2040, when widespread transition to new energy production technologies occurs, the variation increases, the highest scenarios showing unbearably high exposures. For example, the AOT40f ozone exposure index calculated for a monitoring station in the Baltic Sea varies between 11,000 and 26,000 ppb h in 2050, while the current value is below 9000 ppb h. In 2100, the estimated AOT40f varies between 7000 and 40,000 ppb h showing the potential for very high photochemical pollution, if emission trends follow their highest scenarios. However, these estimates were based on simplifying assumptions on the complex atmospheric processes, which called for global and regional CTM simulations for the most interesting cases.

For the modelling carried out within this study, we chose the SRES scenario A1T because the global air pollution emission trends of A1T are close to the median of different scenarios in 2050 and by this they decline in the 'OECD90' region. The emission estimates developed within CAFE (Amann et al., 2004) for the EU for 2030 are close to the A1T emissions for 2050 following the trend of the 'OECD90' region. The year 2050 was selected for modelling because it is beyond the horizon of the CAFE scenarios.

The global chemistry-transport model Oslo CTM2 shows high increases in ozone concentrations in the middle and upper troposphere in 0–50 ºN, and in the surface ozone concentrations in Asia. In Scandinavia, the annual average surface ozone concentration increases about 5 ppbv, or 15%. In April–September, the increase is higher, about 20%. This is in accordance with the increase of 20% for the A1T scenario, obtained using the simple relationship between the global emissions and the tropospheric ozone burden (Section 3.4).

The regional simulations with the Unified EMEP model show that the average daily maximum ozone concentrations in April–September exceed 50 ppbv in large areas in the central and southern parts of Europe. The average of 7 highest days exceeds 60 ppbv nearly everywhere in Europe, excluding northern Scandinavia and northern and eastern Russia. The simulated changes for 2050, assuming the A1T emissions in Europe but unchanged background ozone concentration, reflect the decreasing emissions in Western Europe and the increases in the eastern parts of the continent. There average concentrations are generally over 3 ppbv higher than presently, and in areas surrounding the Black Sea over 6 ppbv higher. In Western Europe, the average ozone concentrations typically decrease by about 3 ppbv, and the highest concentrations a little more. In the Nordic countries, the changes are small.

The boundary conditions of 2050 from the global model influence the European ozone concentrations greatly. In Scandinavia, the average concentrations increase by 6–9 ppbv and the decline in Western Europe turns into a 3–9 ppbv increase. The highest increases are simulated in Russia and in the Black Sea region. The emission reductions in Western Europe significantly reduce the increase of average ozone concentrations, but they are even more effective in the abatement of the highest ozone concentrations. In spite of the increasing background ozone and emissions in Eastern Europe, the averages of the 7 highest days remain invariable. In the Nordic countries ozone levels are largely determined by advection, and an increase of 5–10 ppbv is simulated, in spite of their own emission reductions. The results of these model calculations emphasise the importance of further emission reductions in Europe to protect the environment and human health. In Eastern European countries, in particular, photochemical pollution will get worse, if the increase of air pollutants due to high economic growth is not sufficiently controlled.

Furthermore the calculations also show that surface ozone in Europe is strongly affected by sources on other continents. In order to meet the targets for ozone reductions, European emission reductions should be supplemented by emission control measures also on other continents.

Climate change effects on atmospheric composition are an intrinsic and complex part of the future air pollution problems. The inadequate understanding of the basic processes affecting the ozone budget of the troposphere severely hampers quantification of the trend of global tropospheric ozone burden. Even though tropospheric ozone concentrations are simulated well by different models, the different components of the budget, including the stratosphere-troposphere exchange and photochemical production and loss, which are affected by climate change, vary very much between the models (Prather et al., 2001). This results in deviating trends in climate warming simulations (Zeng and Pyle, 2003). Moreover, the response may be highly variable depending on the relative rates of climate warming and emissions of ozone precursors.

On a continental scale, climate change affects emissions, advection, photochemical production and loss, and dry and wet scavenging of precursors and ozone, requiring a sufficient spatial resolution for proper simulations. The simulations with MATCH that are based on 10-year control and scenario integrations from RCA1 with forcing from two different global climate models indicate substantial impact of regional climate change on both deposition of oxidised nitrogen and concentrations of surface ozone. In these simulations, the meteorological fields were changed, while atmospheric composition and emissions were unaffected. Ozone concentrations and exposures expressed as AOT40 increase very much in the southern and central parts of Europe because there the surface warming in summer is high and the precipitation amounts decrease. Biogenic VOC emissions increase together with ozone production, as there is less cloudiness and wet scavenging of pollutants. In Scandinavia, ozone concentrations decrease because in summer warming is less intense, the precipitation amounts increase and the transport patterns do not favour advection of pollutants to Scandinavia. Statistical tests for the simulated changes in AOT40f show that the changes are significant in relation to internal variability over extended areas. In spite of the high natural variability of meteorological fields, we might expect that, on a broad scale, these results represent the correct trend, as the more recent high resolution simulations of the Rossby Centre for 2070–2100 indicate similar climate change patterns for temperature, cloudiness and precipitation in Europe (Räisänen et al., 2003). Changes in deposition of

oxidised nitrogen are less coherent due to the strong inter-annual variability in precipitation in the RCA1 simulations and differences in the regional climate change simulated with RCA1 in the two regional scenarios. Longer simulation periods are necessary to establish changes in deposition.

The tropospheric ozone concentration has increased significantly since the preindustrial times, and according to the most recent data this trend is continuing. All studies based on the SRES emissions simulate increasing ozone concentrations for the next few decades. If we aim at acceptable air quality in Europe, strict emission reduction programmes are needed to counteract the effect of the increasing precursor emissions elsewhere. The countries in eastern and south-eastern parts of Europe should be especially aware of the future scenarios because climate change and local emission scenarios imply signinificantly enhanced photochemical pollution. Ozone production in the Nordic countries is relatively inefficient and thus these areas are more dependent on the development in other areas. Adding the ozone precursors to the Kyoto GHG list would help reaching emission reductions on a global scale. The highest uncertainties originate from the insufficient understanding of global ozone precursor emissions, taking into account the possibilities of environmental policy and end-of-pipe emission reduction technology, on future methane emissions and on interactions between climate change and atmospheric composition.

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