BOREAL ENVIRONMENT RESEARCH 9: 167–184 ISSN 1239-6095
Helsinki 7 May 2004 © 2004

Trends and scenarios of ground-level ozone concentrations in Finland

Tuomas Laurila[1)], Juha-Pekka Tuovinen[1)], Virpi Tarvainen[1)] and David Simpson[2)]

[1)] *Finnish Meteorological Institute, Air Quality Research, Sahaajankatu 20 E, FIN-00880 Helsinki, Finland*
[2)] *EMEP/MSC-W, Norwegian Meteorological Institute, P.O. Box 43 Blindern, N-0313 Oslo, Norway*

Laurila, T., Tuovinen, J.-P., Tarvainen, V. & Simpson, D. 2004: Trends and scenarios of ground-level ozone concentrations in Finland. *Boreal Env. Res.* 9: 167–184.

This paper presents an overview of the changes in ground-level ozone and vegetation exposure occurring in Finland, both as observed in the recent decade, and as estimated for the period 1900 to 2100. A trend analysis of ozone and total nitrate concentrations is carried out for the 1989–2001 period. Future and past concentrations are modelled based on chemistry-transport model simulations, the SRES (Special Report on Emissions Scenarios) scenarios of the Intergovernmental Panel on Climate Change and emission inventories. Measured summertime ozone shows no decreasing trend despite reported precursor emission reductions. In central Finland, AOT40 (accumulated exposure over a threshold of 40 ppb) over April–September is presently about 6300 ppb h and is estimated to decrease by 570 ppb h as a result of the agreed European emissions reductions by 2010. A similar but opposite change results from the enhanced biogenic emissions of volatile organic compounds due to increased temperatures by 2050. According to the SRES scenarios, the tropospheric background concentrations will increase considerably until about 2050. After this, exposures begin to decline in those scenarios which emphasise new technologies and environmental aspects (8100 ppb h by 2050 in the B1 scenario), but increase monotonically in the A scenarios driven by economic growth.

Introduction

Economic growth since the industrial revolution has been largely based on the increasing use of fossil fuels. This has resulted in various adverse effects on the environment and human health due to gaseous and particulate air pollutants. Recently, increasing atmospheric concentrations of greenhouse gases have been recognised as a major problem. Radiative forcing of these gases may result in climate warming that will have widespread consequences for the environment and for society. To address these long-term changes in Finland, the Finnish Global Change Research Programme (FIGARE) established the FINSKEN project for making future projections of various environmentally important factors (Carter *et al*. 2004). The starting point of these projections is the global emission scenarios developed under the Intergovernmental Panel on Climate Change (IPCC) (Nakićenović *et al*. 2000) and the modelling of the climatic effect

of these emissions (IPCC 2001). In this paper, we will investigate changes in the ground-level concentrations of ozone, which is a key air pollutant and also acts as a greenhouse gas. Within FINSKEN, scenarios for Finland were also produced for socio-economic factors (Kaivo-oja *et al*. 2004), climate change (Jylhä *et al*. 2004), sea level (Johansson *et al*. 2004) and acidifying deposition (Syri *et al*. 2004).

In the troposphere, approximately the lowest 10-km layer of the atmosphere, ozone is photochemically produced from precursor compounds including methane, other reactive organic species and carbon monoxide in the presence of nitrogen oxides (NO_x). Surface ozone concentrations have increased substantially since the pre-industrial times due to increased precursor emissions associated with industrialisation (Volz and Kley 1988, Guicherit and Roemer 2000). For example, global anthropogenic NO_x emissions are estimated to be presently six times as high as they were a hundred years ago (van Aardenne *et al*. 2001). Guicherit and Roemer (2000) evaluated a large set of observations and concluded that at the turn of the 20th century the surface ozone concentrations were about 10–15 ppb in continental Europe, while the present annual average in similar areas is now about 35–50 ppb (Hjellbrekke and Solberg 2001), and in Finland about 30–40 ppb (Laurila 1999). Atmospheric chemistry-transport modelling supports the view that the global ozone concentrations in the troposphere have increased significantly. According to a review of various modelling studies (Prather *et al*. 2001), the tropospheric ozone burden has globally increased 36% since the pre-industrial times, the increase being higher in the northern hemisphere. During the past twenty years, the rate of increase has declined (Scheel *et al*. 1999), and in the western parts of Europe high concentrations during photochemical air pollution episodes in summer have even decreased as a result of emission abatement policies (Roemer 2002). Despite this improvement, the critical levels considered harmful for human health and vegetation are presently exceeded in practically all parts of Europe (Hjellbrekke and Solberg 2001, de Leeuw and de Paus 2001).

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 *et al*. 2000, *see* Carter *et al*. 2004), based on which atmospheric composition scenarios have been developed (IPCC 2001). Within these scenarios, information from different disciplines related to global change forms a consistent chain of actions and responses, beginning from the social and economic drivers and ending at responses in the environment.

All SRES scenarios for the 21st century show increasing methane concentrations that lead to a significant increase in the average tropospheric ozone concentrations. Emissions of NO_x, which is critical for the rate of photochemical ozone production, are increasing rapidly in Asia, Latin America and Africa. This leads to high average and episodic ozone concentrations over these continental regions and facilitates the long-range transport of polluted air to the 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). Superimposed on this increasing background level, the European emissions of precursor compounds will cause high episodic concentrations (Simpson 1992, 1995, Jonson *et al*. 2001). The increasing tropospheric background concentration counteracts the European efforts to combat photochemical air pollution by emissions reductions.

Climate change will affect chemical processes and concentrations of many trace species in the atmosphere. For example, increasing temperatures increase water vapour concentrations in the troposphere, which will reduce the rate of ozone formation in photochemical processes (Brasseur *et al*. 1998, Stevenson *et al*. 2000). On the other hand, the emissions of biogenic Volatile Organic Compounds (VOCs) 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 significantly, thus contributing to the amount of ozone precursors in the atmosphere.

The aim of this study is to present future scenarios of ozone concentrations and vegetation exposure in Finland that are consistent with the up-to-date understanding that the global SRES scenarios and the related climate scenarios represent. We also present an estimate of the long-term changes in the past. In addition to these modelling-based projections, we analyse observed trends in the surface ozone concentrations and investigate how these relate to reported emission changes.

Material and methods

Observations of ozone and total nitrate

The Finnish Meteorological Institute (FMI) has monitored ozone concentrations using commercial instruments based on UV absorption since 1985. The measurement practices have been kept relatively unchanged since the late 1980s. We adopted data from the 13-year period of 1989–2001 for our trend analysis. The monitors are calibrated four times a year using a field calibrator. Previously, the field calibrator was checked annually against the standard instrument of the Tropospheric Ozone Research network at the Institute for Applied Environmental Research at the University of Stockholm. Presently, the laboratory calibrator of the accredited FMI laboratory is used to calibrate the field calibrator. During a transition period in 1998–1999, the laboratory calibrator was also calibrated in Stockholm, but is nowadays traced to a primary calibrator at EMPA (Swiss Federal Laboratories for Materials Testing and Research).

Total nitrate (the sum of gaseous HNO_3 and particulate NO_3^-) was collected on a daily basis using filter-pack samples. The samples were analysed by ion chromatography in accordance with the EMEP manuals (Uggerud *et al*. 2002) at the FMI laboratory.

In this study, we use data from nine monitoring sites (Table 1). The southernmost site, Utö, is located on a small island in the Baltic Sea, where prevailing strong winds efficiently mix the atmospheric boundary layer. Virolahti, Jokioinen and Ähtäri are in a rural, agricultural environment, while Lammi, Ilomantsi, Oulanka and Inari are forested sites. Virolahti is located about 120 km from St. Petersburg. The easternmost site at Ilomantsi is on a hill about 60 m above the surrounding terrain. The station of Oulanka is located close to the Arctic Circle on a hillside within a landscape typical of the northern boreal region. Pallas is on a treeless subarctic mountain about 250 m above the surroundings. The northernmost site, Inari, is on a forested hill. Most of the stations belong to the international air pollution monitoring networks of the European Monitoring and Evaluation Programme (EMEP), the Global Atmospheric Watch (GAW) programme of the World Meteorological Organization (WMO) and the Integrated Monitoring (IM) programme of the United Nations Economic Commission for Europe (UN/ECE).

Linear trends in ozone and nitrate concentrations were estimated both from linear regression and by using the nonparametric Mann-Kendall

Table 1. Characteristics of the monitoring stations.

Station	Coordinates	Elevation (m a.s.l.)	Network	Site characterisation
Utö	59°47´N, 21°23´E	7	EMEP	Marine
Virolahti	60°32´N, 27°41´E	4	EMEP	Rural
Jokioinen	60°49´N, 23°30´E	106		Rural
Lammi	61°13´N, 25°08´E	133	IM	Forested
Ähtäri	62°32´N, 24°13´E	160	EMEP	Rural
Ilomantsi	63°08´N, 31°03´E	230	IM	Forested, hill top
Oulanka	66°19´N, 29°24´E	310	EMEP	Forested
Pallas	67°58´N, 24°07´E	566	GAW	Subarctic, mountain top
Inari	68°29´N, 28°18´E	262		Forested

a.s.l. = above sea level

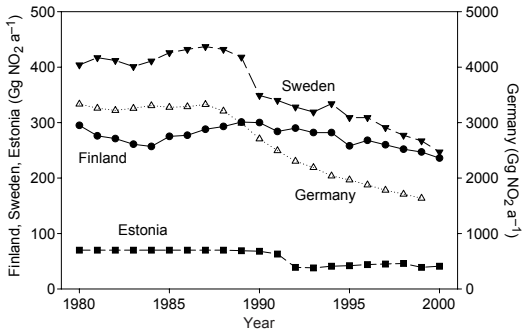


Fig. 1. Annual NO_x emissions in some countries around the Baltic Sea (Vestreng and Klein 2002). Estimated annual emissions in 1990 from the ship traffic in the Baltic Sea were 352 Gg NO_2.

significance test for the existence of a trend together with the nonparametric Sen's method for the magnitude of the trend (Salmi *et al*. 2002).

Ozone precursor emissions in Europe

VOCs and NO_x are the primary precursors of photochemical smog. In Nordic countries, elevated ozone concentrations are mostly due to the long-range transport of ozone and precursors generated further south, in densely populated parts of Europe. Although VOC emissions play a role in this ozone generation, concentrations in Finland are estimated to be more sensitive to NO_x emissions in both Finland and neighbouring countries than to VOC (Simpson 1995, Simpson *et al*. 1997). According to calculations by Simpson *et al*. (1997), the anthropogenic changes in ozone concentrations in Finland should largely follow NO_x emissions in this area.

The emissions of NO_x mostly affecting Finland have changed since 1980 mainly for two reasons. Economic and technological changes during the end of the 1980s and the early 1990s in countries undergoing economical transition in eastern Europe resulted in the closing of many old industrial and power generation plants. After that, modern industrial activities and increasing traffic have altered the emission patterns. As an example, the Estonian emissions, located south of our measuring sites, show large reductions in 1992, when power generating capacity declined, and an increase thereafter, along with renewed economic growth (Fig. 1).

In the western part of Europe, the emission abatement measures introduced during the 1990s have included, for example, low-NO_x burners in combustion power plants and catalytic converters in automobiles, even though increasing traffic density has partly cancelled the benefits from the cleaner technology. As an example of this region, the German NO_x emissions were at a relatively constant level in the 1980s, while Swedish emissions show a broad maximum around 1987 (Fig. 1). In both these countries, the emission reductions in the 1990s were substantial. Finnish emissions have changed less, increasing towards the end of the 1980s and gradually declining thereafter.

Estimates of anthropogenic VOC emissions and their trends have considerable uncertainties. One may expect decreases in these emissions to have taken place because of the increasing number of cars with catalytic converters, introduction of reformulated gasoline and regulations of emissions from stationary sources. From the photochemical point of view, biogenic VOCs complicate the analysis of the importance of anthropogenic VOC emission changes for ozone formation because natural emissions surpass the anthropogenic ones during weather conditions favourable for ozone formation (Lindfors *et al*. 2000). The reported annual anthropogenic Finnish VOC emissions decreased by 30% between 1990 and 2000 (Vestreng and Klein 2002).

Emission scenarios

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 *et al*. 2000, *see* Carter *et al*. 2004). 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.

Within these four main scenario sets, there are different technologies for production of energy. 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.

The SRES emissions are reported for four aggregated areas of the world. Finland belongs to the "OECD90" region together with other parts of Western Europe, and for example North America, Japan and Australia. 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. The driving factors of the emission modelling within the SRES studies are general socio-economic and technological developments. In contrast to the AIR-CLIM study (Mayerhofer *et al*. 2002), actual information on the present and near future environmental legislation is not considered. This may result in conflicting emissions trends on a regional, for example European, level for the next two to three decades. Many SRES scenarios do not follow the already agreed European emission reduction protocols and the environmental legislation of the European Union (Syri *et al*. 2004). However, it is difficult to consider SRES emissions to be unrealistic because of their spatially aggregated nature. The AIR-CLIM emissions scenarios cover only sulphur and nitrogen dioxide, which makes them too limited for ozone formation studies.

Photochemical modelling

The ground-level ozone concentrations used in this study were modelled by Tuovinen *et al*. (2002) using the regional-scale Lagrangian photochemical model of EMEP developed at the Norwegian Meteorological Institute (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 (Nakićenović *et al*. 2000), as shown in Table 2. The methane concentrations and the boundary

Table 2. Global fossil CO_2 emissions[a], atmospheric methane concentrations[b] (in parts per billion), global NO_x emissions[a], tropospheric average ozone burden[b] (in Dobson Units), and NO_x and VOC emissions in the western (WEU) and eastern (REF)[c] parts of Europe in 1999[d], 2010[d] and 2050[a]. NO_x and VOC emissions are relative to those in 2010. Scenario emissions and concentrations are according to the MESSAGE model[a].

	Global CO_2 emissions (Pg C)	CH_4 conc. (ppb)	Global NO_x emissions (Tg N)	O_3 burden DU	WEU NO_x emissions rel. to 2010	REF NO_x emissions rel. to 2010	WEU VOC emissions rel. to 2010	REF VOC emissions rel. to 2010
1999	6.9	1760	31	34	1.20	0.75	1.53	0.75
2010	8.6	1860	32	34	1	1	1	1
2050								
A1C	20.6	2637	94	46.4	1.14	3	0.93	2.4
A1G	21.5	2537	80	43.3	1	2	0.81	2.3
A1T	12.3	2373	61	40.6	0.64	1.66	0.70	2.1
A2	15.1	2799	66	41.6	1.21	2	0.82	1.4
B1	9.2	2256	40	36.8	0.5	1.33	0.62	1.8
B2	11.2	2383	54	39.0	1.15	1.66	1.18	2.3

[a]Nakićenović *et al.* (2000); [b]IPCC (2001); [c]Central and eastern European countries undergoing economical reform and newly independent states of the former Soviet Union; [d]Vestreng (2001).

conditions for ozone concentrations were also derived from the IPCC results (Prather *et al.* 2001) (Table 2). Simulations of the present-day atmospheric composition use emissions reported by the European countries for 1999 (Table 2) and gridded meteorological input from a numerical weather prediction model for 1994. The model calculations for 2010 use these same meteorological data, while the emissions summarised in Table 2 are taken to follow the reductions agreed in the Gothenburg protocol of the UN/ECE (UN/ECE 1999, Vestreng 2001).

In addition to the input data of the photochemical model, the IPCC results were used for deriving background ozone concentrations for many other scenarios and periods than those actually simulated by the photochemical model. From the global ozone precursor emissions and atmospheric methane concentrations we calculated the change from the current tropospheric ozone burden of 34 Dobson Units (DU) using the relationship derived by Prather *et al.* (2001) (Table 4.11 in IPCC 2001). Based on the zonally averaged ozone concentrations presented by Prather *et al.* (2003), we assume that the relative changes in ozone concentrations in the free troposphere over northern Europe follow the mean global trends. We used mostly the emissions obtained from a single model (MESSAGE, Nakićenović *et al.* 2000) to make sure that we compare scenarios rather than the models used to produce them.

The same methodology can be used for estimating tropospheric ozone trends in the past as well. For the past input data, we employed the historical methane concentrations presented by Prather *et al.* (2001) and the global ozone precursor emissions since 1890 from the emission inventory of van Aardenne *et al.* (2001). The free tropospheric concentration in Europe was calculated by assuming a value of 20 ppb in 1900 based on the modelling studies of Simpson *et al.* (1997), Wang and Jacob (1998) and Lelieveld and Dentener (2000), and the observations summarised by Guicherit and Roemer (2000).

In order to identify the most important drivers of the concentration changes, a series of sensitivity tests were carried out with the EMEP model for the A1C scenario for 2050. This was accomplished by starting from the base situation and adding changes one by one until the full A1C scenario is reached. After the simulation using European emissions in 2050 (Table 2), free tropospheric ozone concentrations were multiplied by a factor of 1.46, corresponding to the A1C scenario (IPCC 2001), and the EMEP model was re-run. Then surface temperature was increased to find out how increased biogenic VOC emissions affect ozone concentrations, and finally temperature in the chemistry module of the EMEP model was increased.

From these sensitivity runs we calculated how changes of the tropospheric background concentrations (b) affect the boundary-layer concentrations (c) in each grid cell. This was expressed as relative transfer coefficients $(dc/c)/(db/b)$, which were used in the subsequent scenario calculations. When the background ozone concentrations were increased by a factor of 1.46, transfer coefficients of 0.831, 0.784, 0.749 and 0.739 were obtained for the measurement sites Oulanka, Ähtäri, Virolahti and Utö, respectively. An additional sensitivity test with a ±10% perturbation in the boundary conditions showed that the response of the ground-level is nearly linear.

The effect of global tropospheric warming on ozone burden was estimated from simulation results presented by Stevenson *et al.* (2000) who modelled global ozone concentrations for the A2 and B2 scenarios. They estimated the effect of climate warming on methane, the OH radical and ozone concentrations from additional simulations in which the global warming was specified. We used those results to obtain an independent estimate of ozone changes and of the effect of global warming on these.

Estimation of vegetation exposure

Ozone effects on vegetation most likely occur during the growing season, when stomatal gas exchange is active (Emberson *et al.* 2000). The accumulated exposure of vegetation to ozone over the growing season can be described by the AOT40 index, which represents the sum of the positive differences between the hourly mean concentration and a threshold of 40 ppb, multiplied by the one-hour averaging time, during daylight hours (Fuhrer *et al.* 1997, Tuovinen

2000). The critical levels of ozone effects on vegetation, which are widely used in the European air pollution abatement strategy and legislation work, are defined in terms of AOT40 (Fuhrer *et al.* 1997, Amann and Lutz 2000, EU 2002). For forests, a critical level of 10 000 ppb h is set for the AOT40 calculated over April–September (AOT40f), while agricultural crops have a critical level of 3000 ppb h for the AOT40 accumulated over May–July (AOT40c), both averaged over a five-year period (Kärenlampi and Skärby 1996). Within the recently-endorsed European directive on ozone (EU 2002), the critical level for agricultural crops is adopted as the long-term objective value for the protection of vegetation.

The AOT40 index has been shown to be highly sensitive to background ozone levels and local conditions (Tuovinen 2000, Laurila *et al.* 2001, Sofiev and Tuovinen 2001). This means that a systematic error in the concentration data will result in a considerable error in the value of AOT40. This is the case with our model calculations for the high northern latitudes, where the modelled ozone concentrations are too low (Fig. 2). The underestimation of the average concentrations is highest, around 4 ppb, in the northernmost parts of Scandinavia. We corrected for this by adding the average difference between modelled and observed concentrations at each measurement site.

For calculating the AOT40 index, we use the statistical method developed by Tuovinen (2002), which presents a relationship between parameters describing the frequency distribution (arithmetic mean and standard deviation) and AOT40. From the model calculations, we employ changes in the average concentrations between the runs, which are better simulated and are less affected by inaccuracies in the boundary conditions. The required standard deviations are calculated from the observed concentrations at each site and are assumed to remain unchanged in all basic simulations. The standard deviations of hourly daytime concentrations increase towards the south due to higher photochemical pollution there (Fig. 2). This suggests that for the scenario calculations, it seems reasonable to assume the standard deviation to be proportional to the mean concentration, the approach of which is taken in a later example.

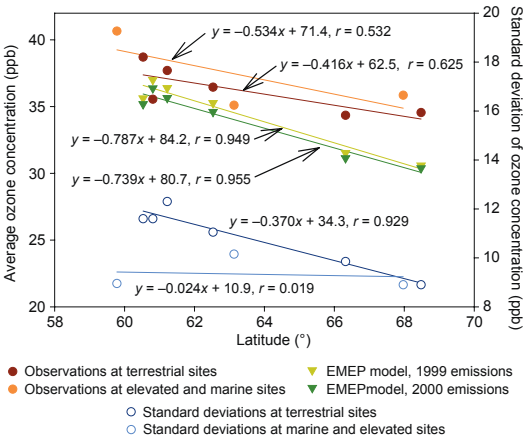


Fig. 2. Averages (upper points) and standard deviations (lower points) of the hourly ozone concentrations at the Finnish monitoring stations during daylight hours (8:00–20:00) in April–September in 1996–2000 as a function of latitude. The corresponding average ozone concentrations simulated by the EMEP model using the 1999 and 2010 emissions are also shown.

Climate change effects

Two types of climate change effects were considered in the model calculations. Increased temperatures affect biogenic VOC emissions and atmospheric chemistry in the boundary layer. On the global scale, changes in the tropospheric temperature and water vapour content influence concentrations of ozone and other trace species. Climate change scenarios, related to increasing surface temperature, were produced for 2050 by adding an average temperature change obtained from climate change simulations. The other meteorological fields of the EMEP model were kept unchanged. The reason for this relatively simple approach was that air temperature was the only parameter found to undergo a consistent change in summer in all climate model projections analysed (Jylhä *et al.* 2004). Precipitation amounts also tend to be increasing, but they are not so relevant for the ozone problem.

The temperature change (ΔT) was derived from the results of the ACACIA project (Hulme and Carter 2000). ACACIA employed climate simulations from eight general circulation models which were based on preliminary versions of the SRES scenarios. ΔT is expressed as a change in the mean summer (June–August)

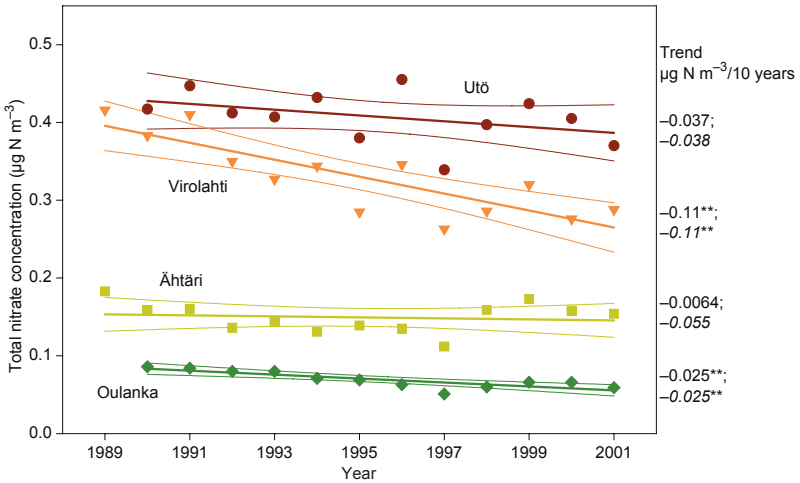


Fig. 3. Annual averages of the total nitrate concentrations in ambient air. Linear regressions together with the 95% confidence intervals are also shown. Trend estimates are calculated from the linear regressions and by the Sen's method (in italics). Two stars indicate significance at p = 0.01.

temperature and was derived by first estimating its mean dependence on latitude (ϕ) from the gridded medians of the different model simulations for the 'ACACIA A2-high' scenario for the 2050s as

$$\Delta T = \begin{cases} 4.5\ \text{K}, & \phi \le 42.5°\text{N} \\ 2.3(55° - \phi)/12.5 + 2.2\ \text{K}, & 42.5° < \phi < 55° \\ 2.2\ \text{K}, & \phi \ge 55° \end{cases} \quad (1)$$

This change is then linearly scaled according to the CO_2 concentrations projected for the SRES scenarios (IPCC 2001) and the estimated relationship between the mean global temperature and CO_2 concentration based on the four ACACIA scenarios.

The obtained temperature changes are comparable to those simulated by high-resolution models for northern Europe (Christensen *et al.* 2001, Räisänen *et al.* 2001). For example, the four regional climate models give, on average, a 1.7 K surface temperature increase in June–August between 1990 and 2050 in the Nordic region (Räisänen *et al.* 2001). According to the same study, the difference between the average temperature in April–October and the June–August temperature available from the ACACIA report is only 0.13 K. The climate change projections of FINSKEN (Jylhä *et al.* 2004) are very close to the summer temperature warming given by Eq. 1. The average increase in the June–August temperature in Finland between 1990 and 2050 is 2.1 K for the A2 scenario (range 1.6–2.5 K for different climate models).

Results

Trends of nitrate and ozone

Concomitantly with the photochemical processes catalytically producing ozone, NO_x is oxidised to HNO_3, which is partly taken up by aerosols and is then present as NO_3^-. Thus, the trend of these oxidised NO_x species may be expected to qualitatively follow that of NO_x. The ratios of NO_x emissions in 2000 to those in 1990 are 0.70 and 0.79 in Sweden and Finland, respectively, and about 0.6 in Germany and Estonia (Fig. 1). The Finnish records of total nitrate and ozone concentrations correspond to the decreasing phase of NO_x emissions (Fig. 3), but the time-series are relatively short for determining trends. The nitrate concentrations have decreased at all sites, but the trends are significant only at Virolahti and Oulanka. Interestingly, no significant trends can be detected at the two westernmost sites, even though countries to the west of Finland report substantial emission reductions (Vestreng and Klein 2002). The ratios of annual average total nitrate concentrations in 2000 to those in 1990 (0.92, 0.72, 0.97 and 0.70 at Utö, Virolahti, Ähtäri and Oulanka, respectively) are generally larger than the corresponding emission ratios.

With regard to ozone forming potential, a full year does not represent an appropriate averaging period, as photochemical ozone production takes place only in spring and summer, with highest intensity in May–July. The total nitrate concentrations averaged over these months are

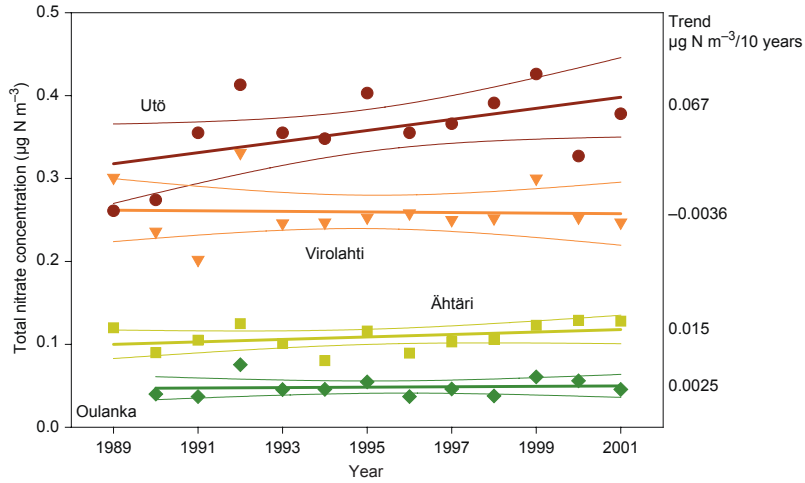


Fig. 4. Averages of the total nitrate concentrations in ambient air in May–July. Linear regressions together with the 95% confidence intervals are also shown. Trend estimates are based on the linear regression and are not significant at $p < 0.1$.

relatively stable or slightly increasing, but no statistically significant trends can be determined (Fig. 4). The reason for the discrepancy between annual and summer trends, and also that between the summer concentration and annual emissions, is not known. The lifetime and the corresponding source area of these nitrogen compounds are shorter in summer, suggesting that the emissions in Finland and the neighbouring countries have not decreased very much. The trends of ship emissions in the Baltic Sea are not accurately known, but the current emission levels are significant (Vestreng and Klein 2002, Entec 2002) and traffic densities increasing (Rytkönen *et al.* 2002). Unlike land-based sources, these emissions have not been subject to control measures to any great extent.

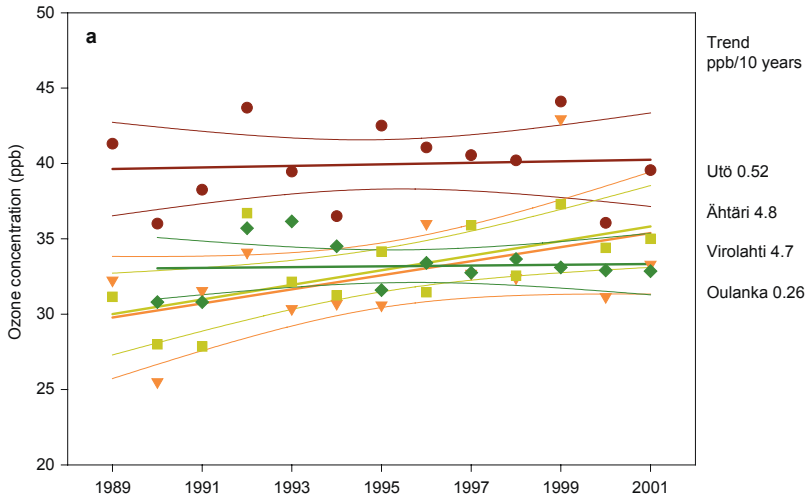
The average ozone concentrations during the May–July period are stable or increasing (Fig. 5). The trends of the April–September concentrations are similar (data not shown). At Oulanka in northern Finland, the average ozone concentrations are stable, and the shorter time-series from Pallas does not show any significant changes either. This is consistent with the weekly ozone sounding record from Sodankylä starting in 1988 (R. Kivi pers. comm.), which does not indicate any trend in the lower troposphere (the 850–700 hPa layer).

The trends of the highest ozone concentrations in summer, as represented by the 99th percentile of the hourly values, are statistically insignificant (Fig. 5b). However, the consistently negative sign suggests a marginal decline of the highest concentrations. According to model calculations using the Swedish MATCH model presented by Solberg *et al.* (2002), the reduction in the corresponding quantity for April–September should be about 6 ppb per 10 years, which is clearly in contrast to the observed trend.

The atmospheric concentrations of total nitrate were used to distinguish unpolluted air masses (Laurila 1999). Only ozone data for days with the total nitrate concentration below the 10th percentile were included in this analysis. On average, the ozone concentrations in unpolluted clean air masses show an increasing trend (approximately 5 ppb per decade) in the southern and central parts of Finland, but no trend at the northernmost site (Fig. 6).

The values of the AOT40 index, calculated according to the instructions given in the ozone directive of the European Union (EU 2002), are shown in Figs. 7 and 8 for forests (AOT40f) and crops (AOT40c), respectively. The AOT40 index effectively accumulates elevated concentrations, resulting in great variations between years and locations. In 1992 and 1999, high ozone concentrations were frequent, but in 1990, 1991, 1994 and 2000 they were uncommon. Highest exposures are observed in the southern parts of the country. The distance between the northernmost and southernmost sites is so great that the corresponding values are poorly correlated. A year with a relatively high AOT40 at the northern site may have a low value at a southern site.

At the sites representing northern Finland, *viz.* Oulanka and Pallas, AOT40f has been rather



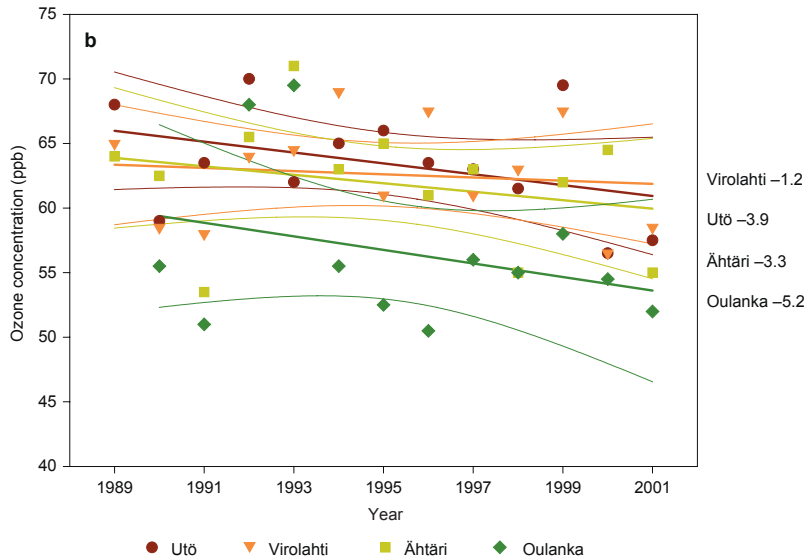


Fig. 5. Averages (**a**) and the 99th percentiles (**b**) of hourly daytime ozone concentrations in May–July. Linear regressions together with the 95% confidence intervals are also shown. Trend estimates are based on the linear regression and are not significant at $p < 0.1$ level.

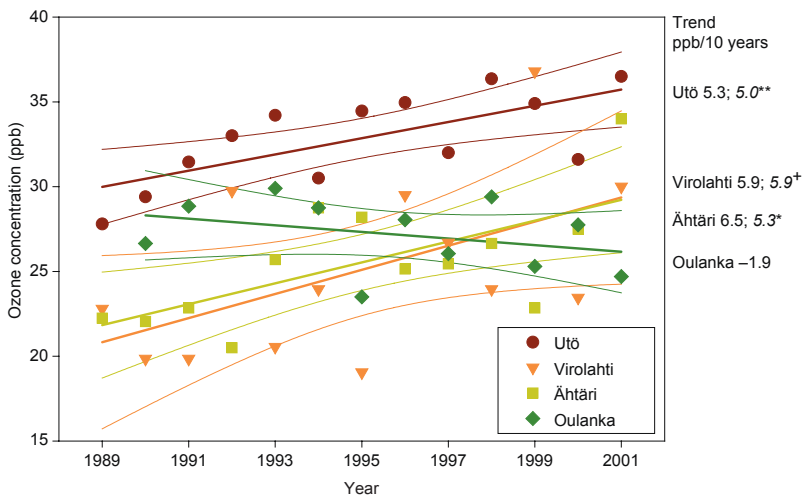


Fig. 6. Average ozone concentrations in unpolluted air masses in May–July. Trend estimates are calculated from the linear regressions and by the Sen's method (in italics). The significance levels are 0.1 (+), 0.05 (*) and 0.01 (**).

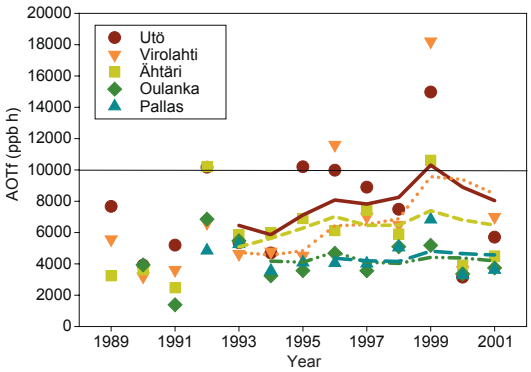


Fig. 7. Values of the AOT40 exposure index for forests (April–September) at the Finnish EMEP and GAW sites. The 5-year averages and the corresponding critical level of 10 000 ppb h are also indicated.

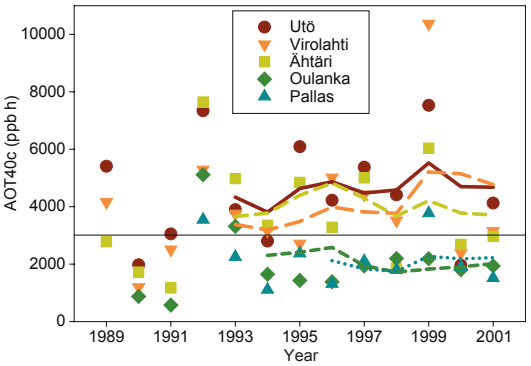


Fig. 8. Values of the AOT40 exposure index for crops (May–July) at the Finnish EMEP and GAW sites. The 5-year averages and the corresponding critical level of 3000 ppb h are also indicated.

steady at about 4000 ppb h during 1994–2000 but more variable during the early 1990s. At the more southern sites, AOT40f is about 7000 ppb h on average. The critical level (10 000 ppb h) is not exceeded as a 5-year average, even though in some individual years this value may be exceeded. For crops, the observed AOT40c exceeds the critical level (3000 ppb h) in the southern and central parts of the country.

As the time-series are short as compared to the year-by-year variation, it is difficult to estimate trends in the ozone exposure expressed in terms of the AOT40 index. Within the period 1989–1999 there would appear to be a significant trend at some southern sites, but adding the last two years having low exposures makes the trends statistically insignificant. We can conclude, however, that there has been no decrease in the AOT40 values that should result from reported precursor emission reductions.

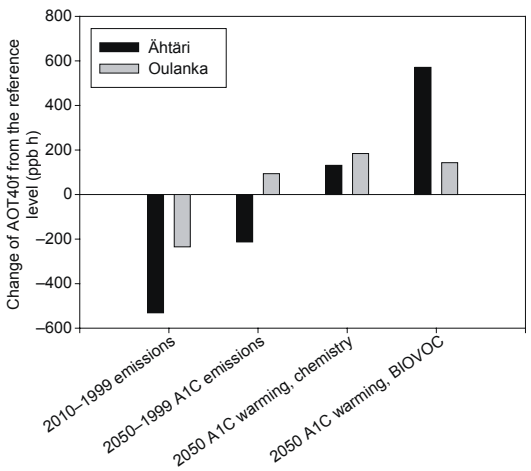


Fig. 9. Effects of European anthropogenic precursor emission changes between 1999 and 2010, and between 1999 and 2050 (the A1C scenario) at Ähtäri and Oulanka. The effects of climate warming via the boundary layer chemistry and the rate of biogenic VOC emissions are also illustrated.

Ozone exposure scenarios for the 21st century

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 AOT40 values were recalculated using the method of Tuovinen (2002). Figure 9 shows how much European emission changes estimated for 2010 and 2050 (A1C scenario) affect ozone exposures in Finland. We also illustrate the effect of increasing boundary layer temperature on ozone exposures separately for chemical reactions and biogenic emissions rates.

The simulations show that the European emission reductions expected by 2010, as agreed in the Gothenburg protocol (UN/ECE 1999), result in less than 10% decreases in the AOT40f index in Finland (Fig. 9). In the western part of

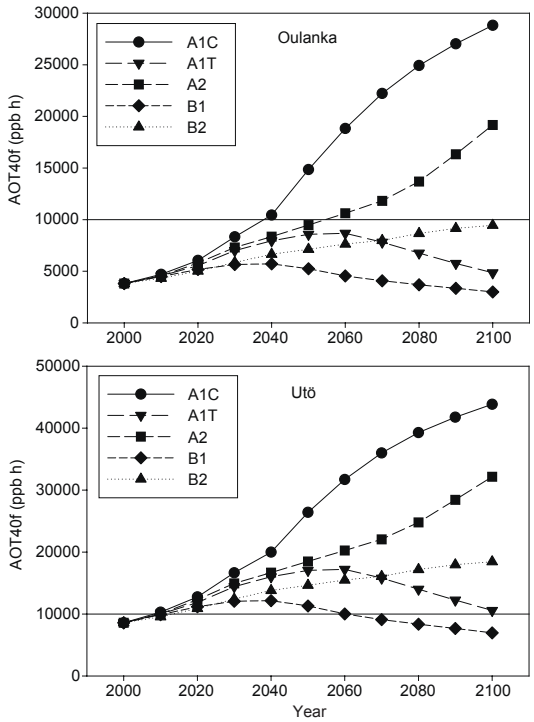


Fig. 10. Modelled development of AOT40f at Oulanka and Utö for different SRES scenarios. The critical level of 10 000 ppb h is also indicated.

Europe, the anthropogenic NO_x and VOC emissions in 2050 according to the A1C scenario are similar to those in 2010, while in eastern Europe they are larger by a factor of 2–3 (Table 2). This results in small decreases in ozone exposure in western Finland and increases in the eastern parts of the country. On a European scale, the highest increases are experienced in eastern Europe, where the growth rates of precursor emissions are highest (Tuovinen *et al*. 2002). In Finland, ozone concentrations seem to follow more closely precursor emission trends in western Europe including the domestic emissions.

The effect of increasing near-surface temperatures were considered for biogenic VOC emissions and for the direct effect on chemical reactions. The enhanced biogenic VOC emissions are estimated to increase AOT40f by about 600 ppb h in southern Finland, the effect being smaller in the northern parts of the country. Small positive changes are induced in the chemistry module by the higher temperatures. Interestingly, all these modifying factors are of a similar importance as the decrease of concentrations due to the emission reductions between 1999 and 2010 according to the Gothenburg protocol. The combined climate warming effects overcompensate the reductions attributable to the present environmental policies.

Widely differing ozone concentrations are obtained after the mid-century for different scenarios (Fig. 10). At the station of Utö, the exposures in all scenarios reach the critical level by 2010. The maximum is reached in 2040 and 2050 in the more environmentally friendly B1 and A1T scenarios, respectively, after which AOT40 values begin to decrease. However, only in the B1 scenario will AOT40 be below the critical level by 2100. In the A1C and A2 scenarios, the modelled exposures grow steadily to very high levels as a result of increasing background concentrations. In northern Finland (Oulanka), the exposures have a similar trend but at lower levels. The critical level is exceeded only in scenarios A1C and A2.

These model calculations show that the growth of the free tropospheric ozone concentration will be the most important factor influencing ground-level ozone concentrations. However, it must be noted that a negative feedback between warming of the troposphere and ozone production was not taken into account in our calculations. Increased water vapour concentrations increase concentrations of the OH radical and decrease ozone and methane concentrations (Prather *et al*. 2001). Based on simulations using the STOCHEM model, Stevenson *et al*. (2000) showed that tropospheric warming would decrease the average tropospheric 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.

We calculated exposures for Ähtäri using the tropospheric ozone concentration changes reported by Stevenson *et al*. (2000) for the A2 and B2 scenarios with and without the tropospheric warming (Fig. 11). The tropospheric ozone trends based on the STOCHEM model without climate change effects are close to those estimated here. In the end of the century, however, climate change will significantly influence the ozone concentrations. For example, the critical level of 10 000 ppb h would be exceeded in

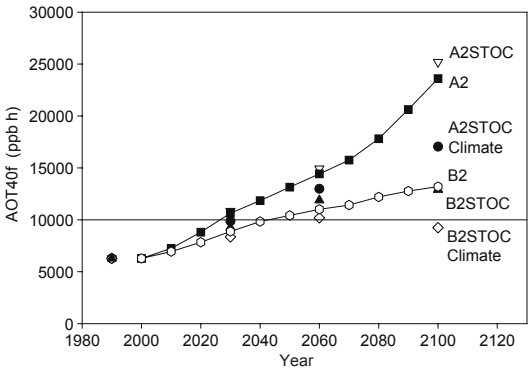


Fig. 11. Modelled development of AOT40f at Ähtäri for scenarios A2 and B2 assuming the tropospheric ozone change according to IPCC (2001) (A2 and B2) and Stevenson *et al.* (2000) without climate change (A2STOC, B2STOC) and with climate change (A2STOC Climate and B2STOC Climate).

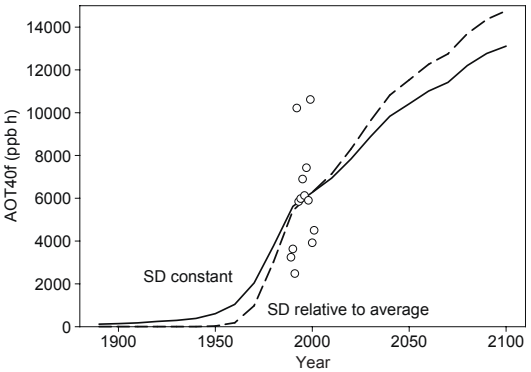


Fig. 12. Development of AOT40f at Ähtäri calculated on the basis of data on atmospheric composition and precursor emissions. The future projections are based on the SRES B2 scenario. Calculations have been carried out for both constant and variable standard deviation of ozone concentrations. The circles denote observed values in 1989–2001.

the B2 scenario, if tropospheric warming is not taken into account in the model calculations, but AOT40f remains below this value, if the negative feedback is included.

Ozone exposures in the past

Similar to the future scenarios, the ozone exposures in the past were calculated based on changes in the atmospheric composition and precursor emissions. The relationship between these driving forces and the change from the current ozone concentration were calculated using data on past methane concentrations and ozone precursor emissions, as described above. Figure 12 shows the historical development of AOT40f at Ähtäri since 1890 combined with the present-day observations and the future development according to the B2 scenario. The past trend reflects changes in both the global atmospheric composition and European precursor emissions. Simpson *et al.* (1997) estimate that roughly half of the change in the ozone concentrations in summer between 1900 and the present has been driven by enhanced ozone production in the atmospheric boundary layer, thus directly attributable to European precursor emissions. The rest is due to changes in free tropospheric ozone concentrations, which are also affected by these emissions.

The mean ground level concentration corresponding to that presented in Fig. 12 is 16 ppb in 1900, increasing to 25 ppb in 1960 and further to 33 ppb in 1980. Before the 1950s, ozone concentrations were practically always below 40 ppb, and hence the ozone exposure expressed in terms of AOT40 was very small then. After that, precursor emissions increased strongly as a result of rapid economic growth in the 1960s in particular, and ozone concentrations above 40 ppb became common. The rapid increase in the value of the AOT40 index illustrates the nonlinear response of this metric to concentration changes, which is also reflected in the observed data (Fig. 12). Within the future projection according to the B2 scenario, the growth rate of AOT40 (about 100 ppb h a^{-1}) is lower than that in the past. In this scenario, the critical level of 10 000 ppb h is reached around 2040.

A further nonlinearity related to the AOT40 index is due to a possible change in the frequency distribution of concentrations. When applying the statistical method of Tuovinen (2002) in this study, we assumed a constant value for the standard deviation as calculated from the observations at each site. However, it is likely that this quantity depends on air pollution conditions and has a higher value in a more polluted environment. As a first order estimation of this effect, we scaled the observed standard deviations linearly accord-

ing to the calculated mean concentrations. In the past record, the value of the AOT40f index is clearly reduced, if a variable standard deviation is used. For that period this may be a more proper way to estimate AOT40, as the ozone precursor emissions in Europe were much less than presently. At the end of the 21st century, the scaling by the average concentrations results in higher AOT40 values than in our base case. However, we feel that for these future scenarios, the approach based on a constant value is more appropriate, as the changes are mostly due to the increase of tropospheric average concentrations and much less affected by the changes in European precursor emissions.

Discussion and conclusions

In this study we used observations and model calculations to investigate the long-term development of the ground-level ozone concentrations and vegetation exposures in Finland, both in the past and in the future. The trends in the mean summertime ozone concentration during the past 13 years are statistically insignificant. However, we may conclude from the observations that, in qualitative terms, the tendency is towards higher levels.

There were no statistically significant trends in the episodic high ozone concentrations either. This is in contrast to the situation in north-western Europe including southern Sweden, where the highest concentrations in summer exhibit decreasing trends attributable to emission reductions (NEGTAP 2001, Roemer 2002, Solberg *et al.* 2002, Volz-Thomas *et al.* 2003). According to the MATCH model simulations presented by Solberg *et al.* (2002), we should observe a clearly decreasing trend in the highest concentrations in southern Finland, but this is not the case. However, the small, although statistically insignificant, negative trends in the peak concentrations observed at all sites may indicate a marginal benefit across the country from European emission reductions.

Urban measurements in Finland show clear reductions in NO_x concentrations and corresponding increases in the urban ozone concentrations (Aarnio *et al.* 2002). During the photochemically inactive seasons, the declining NO emissions may explain the increasing average ozone concentrations also in rural areas (Roemer 2002). However, ozone concentrations in southern and central Finland have increased in unpolluted air masses during the photochemically active season. A similar increase has also been observed in the U.K. (NEGTAP 2001). These changes may reflect changes in the atmospheric composition on a large scale. It seems that the two opposite effects, increased concentrations in unpolluted air and slightly decreased concentrations in the most polluted air masses, are of the same order of magnitude. A similar balance can be seen in the modelling results of Jonson *et al.* (2001), which suggest that by 2010 the increasing background level may have counteracted the benefit obtained in Finland from the emission abatement according to the Gothenburg protocol of the UN/ECE.

The annual averages of the total nitrate concentrations are decreasing at some of the Finnish monitoring sites, reflecting the emission reductions in northern Europe since 1989. A peculiarity in our trend analysis is that the two easternmost sites exhibit significant downward trends, while the trends at the two western sites are statistically insignificant, in spite of the fact that countries west of Finland have reported high reductions in their annual NO_x emissions. It should be noted that the annually averaged concentrations and emission rates are not optimal for the estimation of ozone formation potential. During the photochemically active season (May–July), the total nitrate concentrations are not decreasing, which suggests that the emission patterns in the Baltic Sea region, including Finland and the emissions from ships, may have changed in such a way that an increasing part of the annual emission occurs in summer, or that increases in the transport distance of nitrogen compounds may compensate for the reduced emissions. This complicates the analysis of factors behind the ozone trends. According to the SRES emission scenarios, which we have used in our model calculations, the rates of ozone precursor emissions will rise in eastern Europe during the first half of this century. The Gothenburg protocol also allows increases in some countries in that area. Our trend analyses suggest that the emission

patterns in north-eastern Europe may already be changing differently as compared to the situation in western Europe.

One of the factors omitted in this study is the influence of changes in the ozone flux from the stratosphere to the troposphere. The decline of ozone concentrations in the lower stratosphere of polar regions has resulted in decreasing tropospheric concentrations for example over Canada (Logan *et al.* 1999). A decreasing stratospheric flux counteracts the increasing tropospheric ozone production and may confound our ability to resolve the signal from tropospheric composition change (Lelieveld and Dentener 2000). These interactions should be taken into account in chemistry-transport simulations and trend analyses in the future.

Even if our methods for calculating the future ozone exposures involve many uncertainties, our results are in accordance with Stevenson *et al.* (2000) (Fig. 11) and other studies (Jonson *et al.* 2001, Ashmore *et al.* 2002). It is important to note that all the global transport-chemistry models that contributed to the IPCC assessment provide similar results, indicating an increase in the tropospheric background concentrations in the 21st century (Prather *et al.* 2003). This upward shift in the baseline level has important implications for pollution abatement strategies and related research needs, as has recently been pointed out (Derwent *et al.* 2002, Coyle *et al.* 2003, Prather *et al.* 2003, Volz-Thomas *et al.* 2003). The increase is largely dependent on the global fossil fuel consumption, because emissions of ozone precursors are connected to various combustion processes. According to the SRES scenarios, it will take many decades to introduce new environmentally friendly technology for energy production on a global scale. Thus, all scenarios indicate increasing emissions during the first half of the century, and the decreasing trends in some scenarios emerge only thereafter. However, emission reductions by 2010 are estimated to temporarily decrease exposure.

Climate change affects ozone concentrations in many ways. Increasing tropospheric temperature and absolute humidity may lead to a reduction in the net ozone production partly via radical chemistry and partly via lower tropospheric methane concentrations (Prather *et al.* 2001). In 2060, this effect is estimated to reduce the increase of tropospheric ozone concentrations by about 20% from those presented in this paper (Stevenson *et al.* 2000). Close to the earth's surface, increasing temperatures will enhance biogenic VOC emissions and thus fuel ozone production in the boundary layer. Our sensitivity tests suggest, however, that this has a much weaker effect than the increasing background concentrations.

One of the uncertainties of our future estimates of ozone concentrations stems from the fact that the emission inventories provided by Nakićenović *et al.* (2000) have a low spatial resolution and do not take into account European conditions in great detail. General trends for western and eastern parts of Europe are only available, and the effects of environmental policy leading to reductions in ozone precursor emissions, such as the Gothenburg protocol, are not taken into account. The AIR-CLIM study (Alcamo *et al.* 2002) provides more detailed scenarios for European NO_x emission, but not for VOCs and CO. The scenarios produced within AIR-CLIM, which assume the present emission reductions technologies and abatement strategies, show relatively small reductions in the European NO_x emissions from 2010 up to 2050. Advanced environmental policies are needed to reach substantial reductions. More refined scenarios for all precursor compounds would improve future projections, but would not change the main result of the global atmospheric chemistry simulations that economic and technical development in the fast growing areas of the world may have profound impacts globally.

The development of ozone concentrations and vegetation exposures over decades has been and will be highly dependent on global emissions of ozone precursors, methane concentrations and climate change. The increase of the ground-level concentration since the pre-industrial era has dramatically increased the human and vegetation exposure to ozone. This development will continue, if global economics follow materialistic pathways. Ozone precursor emission control strategies should acknowledge the global aspect, a view which is also supported by the fact that ozone is a greenhouse gas.

Acknowledgements: This study was supported by the Academy of Finland, the Ministry of Transport and Communication and the Nordic Council of Ministers.

References

Aarnio P., Haaparanta S. & Koskentalo T. 2002. Air quality in Helsinki Metropolitan area year 2001. *Pääkaupunkiseudun julkaisusarja PJS C* 2002:17. Helsinki Metropolitan Area Council, Helsinki.

Alcamo J., Mayerhofer P., Guardans R., van Harmelen T., van Minnen J., Onigkeit J., Posch M. & de Vries H.J.M. 2002. An integrated assessment of regional air pollution and climate change in Europe: Findings of the AIR-CLIM project. *Environ. Sci. Policy* 5: 257–272.

Amann M. & Lutz M. 2000. The revision of the air quality legislation in the European Union related to ground-level ozone. *J. Hazard. Mater.* 78: 41–62.

Ashmore M., Coyle M. & Fowler D. 2002. *Implications of increasing tropospheric background ozone concentrations for vegetation in the U.K.* A review for DEFRA under contract EPG 1/3/173, University of Bradford, U.K.

Brasseur G.P., Kiehl J.T., Müller J.-F., Schneider T., Granier C., Tie X.X. & Hauglustaine D. 1998. Past and future changes in global tropospheric ozone: Impact on radiative forcing. *Geophys. Res. Lett.* 25: 3807–3810.

Carter T.R., Fronzek S. & Bärlund I. 2004. FINSKEN: a framework for developing consistent global change scenarios for Finland in the 21st century. *Boreal Env. Res.* 9: 91–107.

Christensen J.H., Räisänen J., Iversen T., Bjørge D., Christensen O.B. & Rummukainen M. 2001. Synthesis of regional change simulations — a Scandinavian perspective. *Geophys. Res. Lett.* 28: 1003–1006.

Collins W.J., Derwent R.G., Johnson C.E. & Stevenson D.S. 2000. The impact of human activities on the photochemical production and destruction of tropospheric ozone. *Quart. J. Royal Meteor. Soc.* 126: 1925–1951.

Coyle M., Fowler D. & Ashmore M. 2003. New Directions: Implications of increasing tropospheric background concentrations for vegetation. *Atmos. Environ.* 37: 153–154.

de Leeuw F.A.A.M. & de Paus T.A. 2001. Exceedance of EC threshold values in Europe in 1997. *Water, Air, and Soil Pollut.* 128: 255–281.

Derwent R., Collins W., Johnson C. & Stevenson D. 2002. Global ozone concentrations and regional air quality. *Environ. Sci. Technol.* 36: 379A–382A.

Emberson L.D., Ashmore M.R., Cambridge H.M., Simpson D. & Tuovinen J.-P. 2000. Modelling stomatal ozone flux across Europe. *Environ. Pollut.* 109: 403–413.

Entec 2002. *Quantification of emissions from ships associated with ship movements between ports in the European Community.* Final Report for European Commission, Entec UK Limited, Northwich, UK.

EU 2002. Directive 2002/3/EC of the European Parliament and of the Council of 12 February 2002 relating to ozone in ambient air. *Official Journal of European Communities* L 67: 14–30.

Fuhrer J., Skärby L. & Ashmore M.R. 1997. Critical levels of ozone effects in Europe. *Environ. Pollut.* 97: 91–106.

Guenther A.B., Zimmerman P.R., Harley P.C., Monson R.K. & Fall R. 1993. Isoprene and monoterpene emission rate variability: model evaluations and sensitivity analyses. *J. Geophys. Res.* 98: 12609–12617.

Guicherit R. & Roemer M. 2000. Tropospheric ozone trends. *Chemosphere–Global Change Sci.* 2: 167–183.

Hjellbrekke A.-G. & Solberg S. 2001. Ozone measurements 1999. *EMEP/CCC Report* 1/2001, Norwegian Institute for Air Research, Kjeller.

Hulme M. & Carter T.R. 2000. The changing climate of Europe. In: Parry M. (ed.), *Assessment of potential effects and adaptations for climate change in Europe: The Europe ACACIA Project*, Jackson Environment Institute, University of East Anglia, Norwich, UK, pp. 47–84.

IPCC 2001. *Climate change 2001: The scientific basis.* Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge and New York.

Jacob D., Logan J.A. & Murti P. 1999. Effect of rising Asian emissions on surface ozone in the United States. *Geophys. Res. Lett.* 26: 2175–2178.

Johansson M.M., Kahma K.K., Boman H. & Launiainen J. 2004. Scenarios for sea level on the Finnish coast. *Boreal Env. Res.* 9: 153–166.

Jonson J.E., Sundet J.K. & Tarrasón L. 2001. Model calculations of present and future levels of ozone and ozone precursors with a global and a regional model. *Atmos. Environ.* 35: 525–537.

Jylhä K., Tuomenvirta H. & Ruosteenoja K. 2004. Climate change projections for Finland during the 21st century. *Boreal Env. Res.* 9: 127–152.

Kaivo-oja J., Luukkanen J. & Wilenius M. 2004. Defining alternative national-scale socio-economic and technological futures up to 2100: SRES scenarios for the case of Finland. *Boreal Env. Res.* 9: 109–125.

Kärenlampi L. & Skärby L. (eds.) 1996. *Critical levels for ozone in Europe: Testing and finalizing the concepts. UN-ECE Workshop Report.* Department of Ecology and Environmental Science, University of Kuopio, Finland.

Laurila T. 1999. Observational study of transport and photochemical formation of ozone over northern Europe. *J. Geophys. Res.* 104: 26235–26243.

Laurila T., Tuovinen J.-P. & Lindfors V. 2001. Frequency distributions of ozone concentration and the uncertainty in the AOT40 index. In: Midgley P.M., Reuther M. & Williams M. (eds.), *Transport and chemical transformation in the troposphere: Proceedings of the EUROTRAC-2 Symposium 2000*, Springer-Verlag, Berlin, Germany, CD-ROM, pp. 1300–1304.

Lelieveld J. & Dentener F.J. 2000. What controls tropospheric ozone? *J. Geophys. Res.* 105: 3531–3551.

Lindfors V., Laurila T., Hakola H., Steinbrecher R. & Rinne J. 2000. Modeling speciated terpenoid emissions from the European boreal forest. *Atmos. Environ.* 34: 4983–4996.

Logan J.A., Megretskaia I.A., Miller A.J., Tiao G.C., Choi

D., Zhang L., Stolarski R.S., Labow G.J., Hollandsworth S.M., Bodeker G.E., Claude H., De Muer D., Kerr J.B., Tarasick D.W., Oltmans S.J., Johnson B., Schmidlin F., Staehelin J., Viatte P. & Uchino O. 1999. Trends in the vertical distribution of ozone: A comparison of two analyses of ozone data. *J. Geophys. Res.* 104: 26373–26399.

Mayerhofer P., de Vries B., den Elzen M., van Vuuren D., Onigkeit J., Posch M. & Guardans R. 2002. Long-term, consistent scenarios of emissions, deposition, and climate change in Europe. *Environ. Sci. Policy* 5: 273–506.

Nakićenović N., Alcamo J., Davis G., de Vries B., Fenhann J., Gaffin S., Gregory K., Grübler A., Jung T.Y., Kram T., La Rovere E.L., Michaelis L., Mori S., Morita T., Pepper W., Pitcher H., Price L., Raihi K., Roehrl A., Rogner H.-H., Sankovski A., Schlesinger M., Shukla P., Smith S., Swart R., van Rooijen S., Victor N. & Dadi Z. 2000. *Emissions scenarios. A special report of Working Group III of the Intergovernmental Panel on Climate Change*. Cambridge University Press.

NEGTAP 2001. *Transboundary air pollution: acidification, eutrophication and ground-level ozone in the UK*. National Expert Group on Transboundary Air Pollution, CEH Edinburgh, U.K.

Prather M., Ehhalt D., Dentener F., Derwent R., Dlugokencky E., Holland E., Isaksen I., Katima J., Kirchhoff V., Matson P., Midgley P. & Wang M. 2001. Atmospheric Chemistry and Greenhouse Gases. In: Houghton J.L., Ding Y., Griggs D.J., Noguer M., van der Linden P.J., Dai X., Maskell K. & Johnson C.A. (eds.), *Climate change 2001: The scientific basis*. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge and New York, pp. 239–287.

Prather M., Gauss M., Berntsen T., Isaksen I., Sundet J., Bey I., Brasseur G., Dentener F., Derwent R., Stevenson D., Grenfell L., Hauglustaine D., Horowitz L., Jacob D., Mickley L., Lawrence M., von Kuhlmann R., Muller J.-F., Pitari G., Rogers H., Johnson M., Pyle J., Law K., van Weele M. & Wild O. 2003. Fresh air in the 21st century? *Geophys. Res. Lett.* 30, 1100, doi:10.1029/2002GL016285.

Räisänen J., Rummukainen M. & Ullerstig A. 2001. Downscaling of greenhouse gas induced climate change in two GCMs with the Rossby Centre regional climate model for northern Europe. *Tellus* 53A: 168–191.

Roemer M. 2002. Trends of ozone and precursors in Europe — an overview. In: Midgley P.M. & Reuther M. (eds.), *Proceedings from the EUROTRAC-2 Symposium 2002*, Margraf Verlag, Weikersheim, pp. 97–103.

Rytkönen J., Siitonen L., Riipi T., Sassi J. & Sukselainen J. 2002. Statistical analyses of the Baltic Maritime Traffic. *Research Report* VAL34-012344, VTT Technical Research Centre of Finland, Espoo.

Salmi T., Määttä A., Anttila P., Ruoho-Airola T. & Amnell T. 2002. Detecting trends of annual values of atmospheric pollutants by the Mann-Kendall test and Sen's slope estimates — the EXCEL template application MAKESENS. *Publications on Air Quality* 31, Finnish Meteorological Institute, Helsinki.

Scheel H.E., Sladkovic R. & Kanter H.-J. 1999. Ozone variations at the Zugspitze (2962 m a.s.l.) during 1996–1997. In: Borrell P.M. & Borrell P. (eds.), *Transport and chemical transformation in the troposphere: Proceedings of EUROTRAC Symposium '98*, WITPRESS, Southampton, UK, pp. 264–268.

Simpson D. 1992. Long-period modelling of photochemical oxidants in Europe. Model calculations for July 1995. *Atmos. Environ.* 26A: 1609–1634.

Simpson D. 1995. Biogenic emissions in Europe 2. Implications for ozone control strategies. *J. Geophys. Res.* 100: 22891–22906.

Simpson D., Guenther A., Hewitt C.N. & Steinbrecher R. 1995. Biogenic emissions in Europe 1. Estimates and uncertainties. *J. Geophys. Res.* 100: 22875–22890.

Simpson D., Olendrzyński K., Semb A., Støren E. & Unger S. 1997. Photochemical oxidant modelling in Europe: multi-annual modelling and source-receptor relationships. *EMEP/MSC-W Report* 3/97, Norwegian Meteorological Institute, Oslo.

Sofiev M. & Tuovinen J.-P. 2001. Factors determining the robustness of AOT40 and other ozone exposure indices. *Atmos. Environ.* 35: 3521–3528.

Solberg S., Bergström R., Langner J., Laurila T., Sjöberg K. & Lindskog A. 2002. Changes in ozone episodes due to emission reductions. A Nordic study. *EMEP/CCC-Report* 10/2002, Norwegian Institute for Air Research, Kjeller.

Stevenson D.S., Johnson C.E., Collins W.J., Derwent R.G. & Edwards J.M. 2000. Future estimates of tropospheric ozone radiative forcing and methane turnover — the impact of climate change. *Geophys. Res. Lett.* 27: 2073–2076.

Syri S., Fronzek S., Karvosenoja N. & Forsius M. 2004. Sulphur and nitrogen oxides emissions in Europe and deposition in Finland during the 21st century. *Boreal Env. Res.* 9: 185–198.

Tuovinen J.-P. 2000. Assessing vegetation exposure to ozone: properties of the AOT40 index and modifications by deposition modelling. *Environ. Pollut.* 109: 361–372.

Tuovinen J.-P. 2002. Assessing vegetation exposure to ozone: is it possible to estimate AOT40 by passive sampling? *Environ. Pollut.* 119: 203–214.

Tuovinen J.-P., Simpson D., Mayerhofer P., Lindfors V. & Laurila T. 2002. Surface ozone exposures in Northern Europe in changing environmental conditions. In: Hjorth J., Raes F. & Angeletti G. (eds.), *A changing atmosphere: Proceedings of the 8th European Symposium on the Physico-Chemical Behaviour of Atmospheric Pollutants*, European Commission, DG Research, Joint Research Centre, CD-ROM, Paper AP61.

Uggerud H., Hanssen J.E., Schaug J. & Skjelmoen J.E. 2002. The nineteenth intercomparison of analytical methods within EMEP. *EMEP/CCC Report* 1/2002, Norwegian Institute for Air Research, Kjeller.

UN/ECE 1999. Protocol to the 1979 Convention on Long-range Transboundary Air Pollution to Abate Acidification, Eutrophication and Ground-level Ozone. UN/ECE Document EB/AIR/1999/1, United Nations, New York, Geneva.

van Aardenne J.A., Dentener F.J., Olivier J.G.J., Klein Goldewijk C.G.M. & Lelieveld J. 2001. A 1° × 1° resolution data set of historical anthropogenic trace gas emissions for the period 1890–1990. *Global Biogeochem. Cycles* 15: 909–928.

Vestreng V. 2001. Emission data reported to UNECE/EMEP: Evaluation of spatial distribution of emissions. *EMEP/MSC-W Note* 1/01, Norwegian Meteorological Institute, Oslo.

Vestreng V. & Klein H. 2002. Emission data reported to UNECE/EMEP: Quality assurance and trend analysis and presentation of WebDab. *EMEP/MSC-W Note* 1/02, Norwegian Meteorological Institute, Oslo.

Volz A. & Kley D. 1988. Evaluation of the Montsouris series of ozone measurements made in the nineteenth century. *Nature* 332: 240–242.

Volz-Thomas A., Beekman M., Derwent D., Law K., Lindskog A., Prévôt A., Roemer M., Schultz M., Schurath U., Solberg S. & Stohl A. 2003. Tropospheric ozone and its control. In: Midgley P.M., Builtjes P.J.H., Fowler D., Harrison R.M., Hewitt C.N., Moussiopoulos N., Noone K., Tørseth K. & Volz-Thomas A. (eds.), *Towards cleaner air for Europe — science, tools and application, part 1. Results from the EUROTRAC-2 synthesis and integration project*. Margraf Verlag, Weikersheim, pp. 73–122.

Wang Y. & Jacob D.J. 1998. Anthropogenic forcing on tropospheric ozone and OH since preindustrial times. *J. Geophys. Res.* 103: 31123–31135.

Received 5 August 2003, accepted 8 December 2003