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Magnuz Engardt, Robert Bergström and Camilla Andersson

Climate and Emission Changes Contributing to Changes in Near-surface Ozone in Europe over the Coming Decades: Results from Model Studies

We used an off-line, regional, model of atmospheric transport and chemistry to investigate current and future levels of near-surface ozone and accumulated ozone exposure over a threshold of 40 ppb(v) (AOT40) in Europe. To describe the current situation and enable an evaluation of the model's performance we simulated a number of years around 2000. To assess changes in ozone concentrations due to possible emission changes in Europe, the model was run with the meteorology of the early 2000s and precursor emissions from a set of Clean Air for Europe (CAFE) emissions scenarios. By extrapolation of the observed increase in near-surface O_3 at coastal locations in northwest Europe we constructed model boundaries that were used to simulate the impact of increasing hemispheric background in 2020. To assess changes in ozone concentrations due to climate change, the model was run with recent (2000) emissions but using meteorology from a regional climate model simulating a control (1961–1990) and a future (2021–2050) climate. The results indicate that climate change will have a small impact on ozone concentrations and AOT40 in the Nordic countries. Changes in hemispheric background concentrations and changes in precursor emissions in Europe will have a larger effect on ozone in Northern Europe. The situation is quite different in southern Europe, where climate change is expected to result in a very large increase in near-surface ozone concentrations.

INTRODUCTION

Ozone (O_3) constitutes a threat to human health (1) and is causing damage to vegetation (2) in regions with elevated concentrations. The present, near-surface, O_3 concentration in Europe is estimated to be at least double that in preindustrial times (3). In order to mitigate the consequences of elevated O_3 across Europe, international agreements (e.g., the Gothenburg Protocol) based on scientific evaluations of air pollutants' atmospheric cycle and their damage to humans and ecosystems are now in place aiming at reducing the emissions of O_3-generating species.

O_3 is formed through photochemical reactions in environments rich in ozone precursors, i.e., nitrogen oxides (NO_X), carbon monoxide (CO), and volatile organic compounds (VOCs). High temperature, intense solar radiation, stable atmosphere, and low wind speed—frequently occurring in southern Europe during summer—promote the production and accumulation of O_3 in the planetary boundary layer. Near-surface O_3 concentrations in northern Europe (the Nordic and Baltic countries) are generally much lower than in central and southern Europe as a result of the lower precursor concentrations here, and as a result of weather conditions less favorable for O_3 production at these latitudes. Episodes of enhanced near-surface O_3 in northern Europe typically occur during spring and summer following the advection of O_3-laden air from continental Europe after periods of O_3 buildup there.

The lifetime of boundary-layer O_3 is on the order of hours to days; changed precursor concentrations or weather conditions translate into O_3 changes within a few hours. This is evident when examining the diurnal variation of near-surface O_3, which typically features maximum concentrations in the afternoon and low concentrations during night and early morning. O_3 is taken up at the surface and subsequently destroyed when it reacts with organic or other materials. At night, the photochemical production is turned off, while the vertical mixing—responsible for replenishing the O_3 continuously destroyed at the surface—is operating less efficiently than during daytime.

In this work we used the Multiple-scale Atmospheric Transport and Chemistry Model (MATCH)—a regional off-line, chemistry transport model (CTM)—to investigate current and future concentrations of O_3 in Europe. We investigated changes in near-surface O_3 brought about by altered precursor emissions in Europe, as well as increasing hemispheric O_3 and changes in climate following anthropogenic emissions of greenhouse gases and aerosol particles. We will restrict our discussion to accumulated ozone exposure over a threshold of 40 ppb (AOT40), which is one of the measures that is used in Europe to quantify damage to crops and natural vegetation brought about by near-surface O_3. AOT40 is defined as the sum of the differences between the hourly mean O_3 concentration (in ppb[v] and the threshold 40 ppb(v), when the concentration exceeds 40 ppb(v) during daytime. AOT40 crops (AOT40c) has traditionally been used as a measure of crop impacts in Europe and should be deduced at canopy height and accumulated over a 90-d growing season, which varies between different climatic regions. In this work, we set the growing season for crops to May–July and define daytime as between 06.00 and 18.00 coordinated universal time, across all of Europe. We aim at reproducing background measurements, which are taken at 2–10 m above ground and not specifically at canopy height. AOT40 forests (AOT40f) has been used as a measure of O_3 impacts on natural forests and is accumulated over the 6-mo period April through September. AOT40 will in this presentation be expressed in kppb(v)·h, i.e., $=10^3$ ppb(v)·h, sometimes denoted as ppm(v)·h.

The aim of this work is to study how O_3, and AOT40 in particular, will develop in the Nordic and Baltic countries compared to the rest of Europe and to investigate the relative importance of the different factors controlling future O_3 in Europe. AOT40 is selected to illustrate the current and future impact of O_3 on vegetation. AOT40 is a concentration-based measure that we deduce from the modeled hourly O_3 concentrations (Fig. 1). The work can be regarded as a continuation and refinement of Laurila et al. (4), which used coarser-resolution models and older emission inventories to estimate changes in O_3 concentrations in the Nordic countries following global emission changes and regional climate change. The work also complements Simpson et al. (5), who use the

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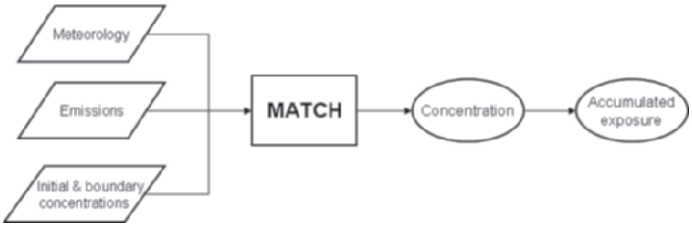


Figure 1. Layout of model system.

European Monitoring and Evaluation Program (EMEP) Meteorological Synthesizing Center—West (MSC-W) model to investigate the O_3-induced impacts on vegetation in Europe in 2000 and in 2020 following one CAFE emission scenario as well as changed background concentrations of O_3 and methane.

METHODS

For the mapping of current and future O_3 in Europe we use MATCH (6, 7)—an off-line CTM developed at the Swedish Meteorological and Hydrological Institute (SMHI). The model is flexible in that it accepts input from different sources and, depending on application, operates on different scales and generates different types of output, see Figure 1. Running the model over Europe is a standard task for MATCH, and the system's ability to calculate near-surface O_3 in the region is described and evaluated in a number of previous publications (8–10). The interested reader is directed to these papers for general descriptions of model setup and performance.

Year-to-year variation in meteorological conditions has a profound impact on near-surface O_3 in Europe (11, 12). Annual mean, near-surface, O_3 concentration for an arbitrary year typically deviates 5–10% from the long-term average, mainly as a consequence of the variability in the driving meteorology. In order to discriminate trends in O_3 brought about by changes in precursor emissions, background concentrations, or climate it is therefore essential to construct robust estimates of present and future concentrations by appropriate averaging of O_3 concentrations and selected indices (e.g., AOT40) over a number of years.

The present work is a compilation and extension of two previous (partly unpublished) studies where we examined the influence of decreasing emissions as well as increasing hemispheric background concentrations and the impact of climate change on European near-surface O_3 concentrations. The two studies used similar configurations of MATCH but operated with different input to produce hourly, three-dimensional fields of tropospheric O_3 across Europe. The first study dealt with the change in O_3 concentrations from the early 2000s to 2020 following changes in European precursor emissions and hemispheric background concentrations of O_3; the model resolution was $0.4° \times 0.4°$, and the model atmosphere was divided into 22 layers up to 5 km. The second study (13) investigated the impact of one possible climate change realization from 1961–1990 to 2020–2050 by driving MATCH with meteorological data representing recent and future climate; here the model resolution was $0.44° \times 0.44°$ and the atmosphere was divided into 15 layers up to 6 km where the top boundary of the CTM was set. The geometries of the two model setups differ slightly and can be deduced from the maps below. Both studies utilized recent compilations of European emissions. The anthropogenic emissions were released with different temporal and spatial variation depending on sector. Biogenic isoprene emissions are calculated on-line in MATCH (7, 14). The experiments are summarized in Table 1 and further explained below.

Model Evaluation

To evaluate the model's ability to simulate O_3 and AOT40 we ran MATCH with meteorology representing 4 y around the turn of the century (1999, 2000, 2001, 2003). The weather data were generated with the operational weather forecast model in use at SMHI (the high-resolution limited area model [HIRLAM]). Emissions of anthropogenic NO_x, SO_x, NH_x, NMVOC, and CO were taken from EMEP's expert emissions (15), which reproduce the anthropogenic emissions occurring in Europe during these years. Monthly varying model boundary concentrations of all species of concern in the CTM were estimated from a combination of ground-based measurements and vertical soundings and should describe the conditions during each respective year. The results from this experiment constitute our best estimate of the situation during the early 2000s, which we will compare with measurements collected during the same years.

Emission Reductions in Europe

To isolate the impact of changing European anthropogenic precursor emissions, we first ran MATCH using meteorology and boundary concentrations representing four different years (1999, 2000, 2001, 2003). In these calculations the model was fed with EMEP's expert emissions valid for one close-to-present year, 2004 (*BASE_2004*). These simulations were contrasted with simulations where we ran MATCH with the same meteorological input and boundary values, but utilizing emission data from two different scenarios constructed by the International Institute for Applied Systems Analysis (IIASA) in the CAFE project.

The emissions in *D23A* are described in Amann et al. (16), where the scenario is denoted Case A, joint optimization. These gridded emissions constitute an optimized emission scenario where the aim was to reach a number of EU environmental targets (covering years of life lost due to particulate material smaller than 2.5 μm (PM2.5), acidification, eutrophication, and ozone) at the lowest possible cost. D23A is similar to the thematic strategy scenario in Amann et al. (17). MFR-DEEP stands for maximum technically feasible reduction plus a deep-cutting EU climate-taxation of 90 € per tonne CO_2 (18). This is currently believed to be close to what is possible to reach in

Table 1. Model experiments.

Experiment	Meteorology	Emissions	O_3 boundaries
Early 2000s	1999–2001, 2003 (HIRLAM)	1999–2001, 2003 (EMEP)	1999–2001, 2003
BASE_2004	1999–2001, 2003 (HIRLAM)	2004 (EMEP)	1999–2001, 2003
D23A	1999–2001, 2003 (HIRLAM)	D23A (IIASA/CAFE)	1999–2001, 2003
MFR-DEEP	1999–2001, 2003 (HIRLAM)	MFR-DEEP (IIASA/CAFE)	1999–2001, 2003
2020 Background	1999–2001, 2003 (HIRLAM)	D23A (IIASA/CAFE)	1999–2001, 2003 +4 ppb(v)
Control Climate	1961–1990 (RCA3/ECHAM4 A2)	2000 (EMEP)	2000
Future Climate	2021–2050 (RCA3/ECHAM4 A2)	2000 (EMEP)	2000

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Table 2. MATCH's ability to reproduce the early 2000s and control climate. Observations refers to data from EMEP monitoring sites (25). Model results are extracted from the grid cells encompassing the respective monitoring site.

	Observations (kppb[v]·h) (period)	Model results (kppb[v]·h) (period)	Correlation	Bias (%)	RMSE (kppb[v]·h)	No. samples
Early 2000s*						
AOT40c	7.0 (1999–2001, 2003)	7.8 (1999–2001, 2003)	0.78	+10.1	2.2	363
AOT40f	12.4 (1999–2001, 2003)	13.4 (1999–2001, 2003)	0.83	+8.0	3.5	355
Control climate†						
AOT40c	7.7 (1997–2003)	8.2 (1961–1990)	0.86	+6.3	2.8	60
AOT40f	13.8 (1997–2003)	13.3 (1961–1990)	0.90	−2.8	4.1	61

* Model data reduced to 3 m aboveground. † Model data from lowest model layer (ca. 30 m).

Europe in 2020 and can therefore be regarded as the cleanest possible option.

Increasing Hemispheric Background of O_3

A number of authors (19, 20) have reported that background concentrations of O_3, represented by data collected at remote locations around Europe, seem to have been increasing during the last few decades. The increase occurs in spite of recent emission reductions in North America and Europe and is thought to be caused by increasing emissions of O_3 precursors in the emerging economies of Asia, Africa, and Latin America, or increasing emissions from international shipping. Other factors, such as increased frequency of boreal forest fires, increased stratospheric-tropospheric exchange, or climate change, are also suggested. The increase is not uniform across all background stations, and the trend is not always clear. Measurements collected on western Ireland (21) indicate an average annual increase of ~0.25 ppb(v) y^{-1} for baseline air for the period 1990–2001. Derwent et al. (22) report a similar value (0.31 ±0.12) ppb(v) y^{-1} for the 20-y period 1987–2007. At background stations in southern and southwestern Sweden, annual mean ozone levels show upward trends of about ~0.24 ppb(v) y^{-1} for the period 1990–2006 (23).

To investigate the consequences of increasing hemispheric concentrations of O_3 on AOT40 in Europe during 2020 we executed the model with increased O_3 concentrations on the boundaries of the model domain (*2020 background*)—everything else (including the boundary concentration of other species) as the *D23A* simulation. We consider D23A to be a reasonably realistic emission scenario for Europe in 2020 and therefore use this experiment as the base for exploring the consequence of increasing hemispheric O_3 concentrations. The increased O_3 boundaries were deduced from an extrapolation of the 0.25 ppb(v) y^{-1} trend of near-surface O_3, which results in a 4 ppb(v) increase of O_3 from 2004 to 2020. The +4 ppb(v) was applied uniformly to all five boundaries (which all lie in the troposphere), during all 12 months.

Climate Change

In the final analysis, we used long time series of meteorological data generated by a regional climate model, to drive MATCH. Control climate corresponds to the climatological average and variability during a recent, close to present, period. Future climate is constructed by running MATCH with an identical setup as the control climate simulation but using meteorology from a regional climate model simulating one possible climate change realization.

The driving meteorological data for the climate change experiments were generated by the Rossby Centre regional atmospheric climate model (RCA3) driven by boundary data from the global model ECHAM4/OPYC3 forced by the SRES A2 emission scenario (24). The control climate and future climate both form 30-y averages representing roughly 1961–1990 and 2021–2050 in terms of climate but ca. 2000 in terms of emissions and boundary values. In this work we are interested in the change in AOT40 caused by climate change and will therefore consider the difference between these two 30-y averages.

The ability of the regional climate model to realistically simulate recent climate is discussed in Kjellström et al. (24), while the performance of MATCH forced by RCA3 data is evaluated in Andersson and Engardt (13). A separate evaluation of MATCH's ability to reproduce AOT40 measured at EMEP stations across Europe is discussed below along with the evaluation of the model's ability to reproduce the situation during the early 2000s.

RESULTS

Model Performance

Table 2 presents a summary of MATCH's capability of reproducing measured AOT40 during the early 2000s and the control climate. Since the data used for generating the statistical measures are not from the same locations and time periods, the two sets of results are not expected to be identical. The scores are also generated somewhat differently. For the early 2000s, the correlation coefficient (r), the bias (average relative model deviation), and the root mean-square error (RMSE) are calculated from the annual values at each station for each individual year. In the control climate these measures are formed by comparing the long-term averages (7 and 30 y, respectively) of observations and model results at each station. The latter method thus only takes the spatial variation into account, while the former also considers temporal variation between the years 1999, 2000, 2001, and 2003. For the statistics we only include those sites where the model and station elevation are within 250 m, and we use only stations that have at least 90% data coverage during the 3 and 6 months needed for the AOT40c and AOT40f calculations. The first criterion excludes a number of high-elevation sites, which MATCH cannot locate vertically because the model's limited horizontal resolution averages out the complex topography of the real world.

The observational data are typically collected at ca. 3 m above ground. In the climate change simulations, the statistical scores were marginally better if we used the average concentration in the lowest model layer instead of model values downscaled to 3 m, which is the normal quantity we use for "near-surface" O_3 in MATCH. The reason, we believe, is a consequence of the coarser vertical resolution in the meteorological data from the climate model (RCA3) compared to the weather forecast model (HIRLAM). Consequently, O_3 from the lowest model layer will be used to represent near-surface O_3 and

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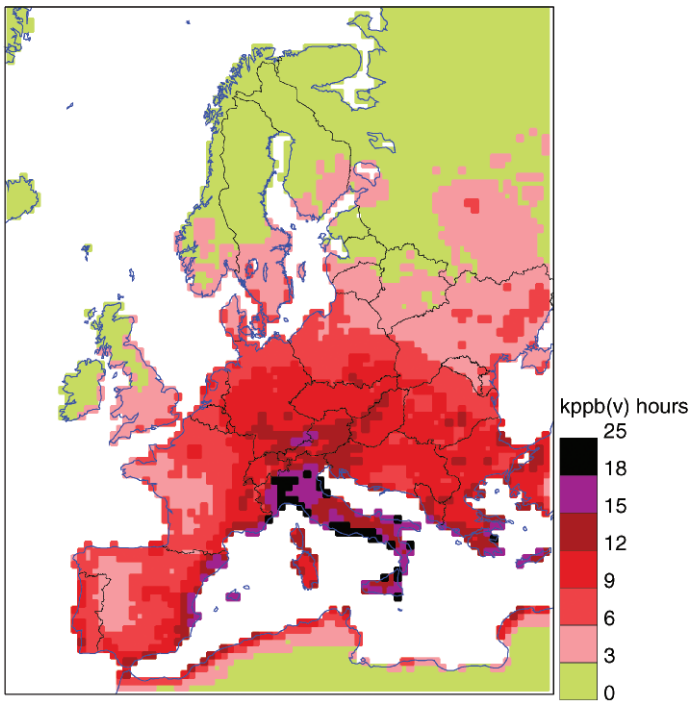


Figure 2. Average modeled AOT40c over Europe, using 2004 emissions during the meteorological years 1999–2001, 2003. Units: kppb(v)·hours.

for the calculations of AOT40c in the climate change simulations.

The correlation coefficient between observations and model result is close to or above 0.8, which means that MATCH can reproduce the spatial variability of AOT40 across Europe. The bias is within ±10%, indicating that, on average, the model realistically describes AOT40 values over Europe. The absolute error may, of course, be much larger at individual stations, which is also indicated by the relatively high RMSE. For the early 2000s the model slightly overestimates the values observed across Europe. For the climate simulations the 3-mo value is overestimated while the 6-mo value is underestimated. At first glance, it appears that the climate simulations perform better than the model's realization of the early 2000s. It is, however, more difficult to reproduce individual years than averages over a number of years, which the climate simulation scores are calculated for.

Current Situation

Figure 2 shows average AOT40c over Europe using meteorology for 1999–2001 and 2003 and emissions valid for 2004 (*BASE_2004*). According to the analysis above, the model, on average, overestimated measurements during the early 2000s by ~10%. The spatial distribution, with high AOT40c in southern Europe and low values in the Nordic countries, the Baltic countries, and the British Isles was, however, well reproduced.

A recent compilation of dose-response relationships (26) established a linear relationship between growing season AOT40 and relative yield loss of several crops. For wheat, AOT40c of 3 kppb(v)·h corresponds to 5% yield loss, 6 kppb(v)·h cause 10% yield loss, etc. This means that substantial parts of Europe are currently at risk for severe ozone-related crop damages. New research has, however, pointed out that the AOT40 index is not optimal to assess crop losses and that stomatal uptake calculations are a far more realistic measure to assess the ozone-induced damages to vegetation (27). For example, during periods with reduced water availability (which is typical in southern Europe during summer), the plants will close their stomata to save water and, as a consequence, they will also be less affected by high O_3 concentrations. Simpson et

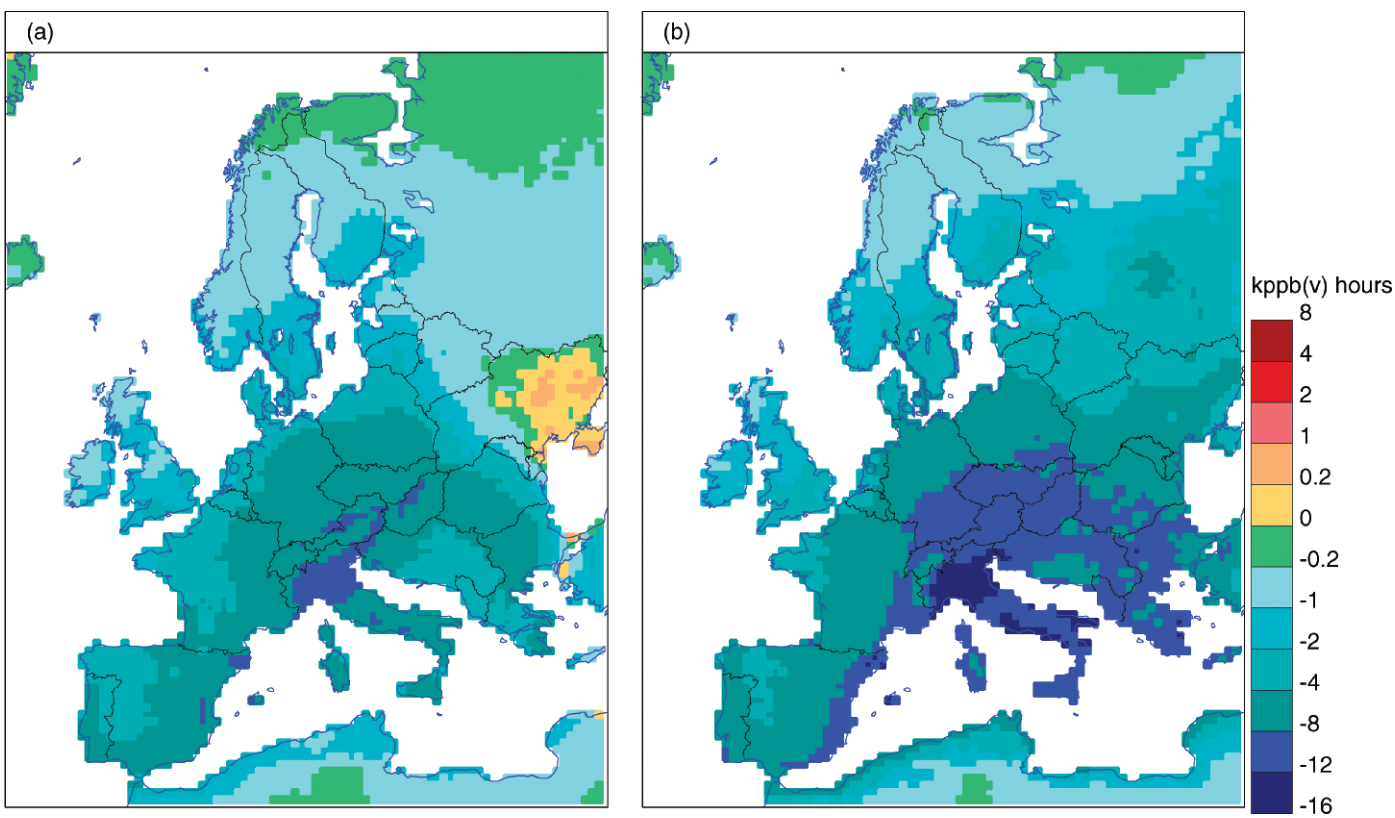


Figure 3. Change in AOT40c from 2004 to 2020 after changing the emission in Europe. (a) The average of the four D23A simulations minus the average of the four BASE_2004 simulations; (b) The average of the four MFR-DEEP simulations minus the average of the four BASE_2004 simulations. Units: kppb(v)·hours.

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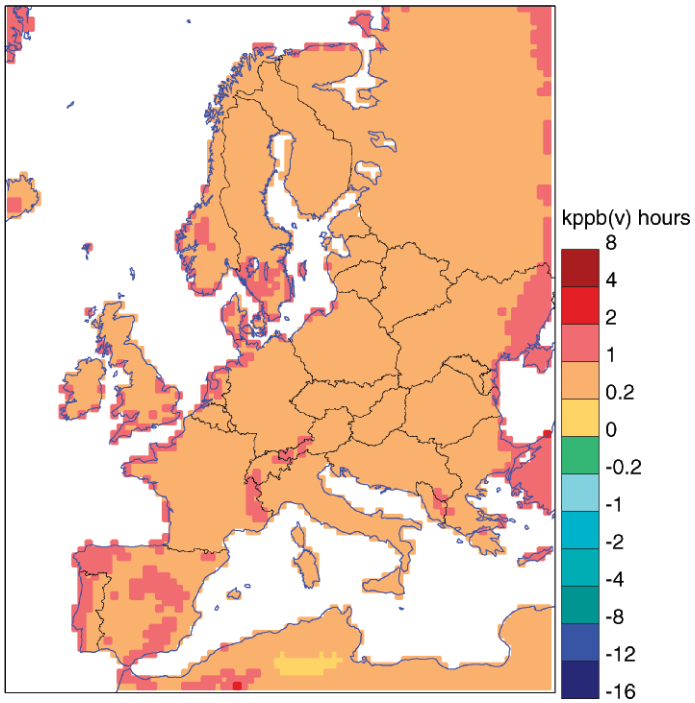


Figure 4. Change in AOT40c after adjusting the background concentrations from the 2004 values to an extrapolated value representative of 2020. The figure depicts the average value of the meteorological years 1999–2001, 2003. Units: kppb(v)·hours.

al. (5) discuss the spatial differences in areas at risk using the AOT40 index compared to the accumulated stomatal flux. The former measure showing a much higher exceedence in southern Europe compared to the flux-based measure, which displays more uniform exceedences across Europe. In this setup of MATCH we did not explicitly calculate and store the stomatal uptake of O_3 in different crops; our results are therefore depicting the estimated risk based on the accumulated dose of the concentrations of near-surface O_3 only.

Emission Reductions in Europe

Figure 3 shows the change in AOT40c from 2004 to 2020 when European precursor emissions evolve according to the emission scenarios D23A and MFR-DEEP, respectively. AOT40c will decrease substantially, all over Europe, in both scenarios. As expected from the definition and generation of the emission scenarios, the absolute decrease is largest where the highest AOT40c occur today (i.e., in southern and central Europe). The Nordic and Baltic countries will also experience significant decrease in AOT40c if Europe follows either of the two suggested emission paths. In the D23A scenario AOT40c is calculated to decrease 1–4 kppb(v)·h in southern Scandinavia, southern Finland, and the Baltic countries.

The difference between the two emission scenarios is substantial, meaning that strong commitments within Europe will substantially improve the air quality within Europe itself. Stronger commitments will clearly also have effects on AOT40c in Northern Europe, as evident from the difference between Figure 3a and Figure 3b.

Increased Hemispheric O_3 Background

Increased O_3 on the model boundaries, representing an elevated hemispheric burden of tropospheric O_3 in 2020 compared to 2004, causes increased AOT40c throughout the model domain (Fig. 4). The increase is not uniform due to nonlinear effects in the chemistry and deposition of O_3 and because AOT40c is a cumulative index based on exceedence of a threshold (which is not always reached). Depending on the ambient concentrations of other species (NO_x in particular) different parts of the domain will react differently to increased O_3. A general feature (not shown) is that the additional O_3 on the boundaries last longer over the oceans than over land due to the slow dry deposition of O_3 to water surfaces.

Southern Sweden, Denmark, and parts of Norway, as well as southwestern Europe (Spain and Portugal) and most grid cells along the Atlantic coast will clearly be affected by the increasing hemispheric background. These land areas will experience an increase of AOT40c of 1–2 kppb(v)·h. While this amount in southern Europe is small compared to the expected decrease in AOT40c following emission changes up to 2020, 1–2 kppb(v)·h is typically what is expected to be gained in the Nordic and Baltic countries following emission changes in Europe over the same period (Fig. 3a,b).

Climate Change

While most parts of southern and central Europe will experience a considerable increase in AOT40c due to climate change, the expected change in AOT40c from the late 20th century to the mid 21st century is small in the Nordic countries (Fig. 5a).

Andersson and Engardt (13) showed that O_3 is likely to increase in southwest Europe due to decreased dry deposition of O_3 in dryer climates and increased emissions of isoprene (which is an important biogenic O_3 precursor) in a future climate. The decreased AOT40c over the Alps mountainous region and in the far north in Figure 5 is connected to the decreasing snow cover, which is radically increasing the dry deposition of O_3.

The climate change calculations cover a period of 60 y, while the emission change and boundary values calculations discussed above span ca. 16 y. In order to compare the climate effect with the other effects, we assume the climate change signal to be linearly increasing from 1961–1990 to 2021–2050 and present in Figure 5b the climate change signal from 2004 to 2020. Over this time period the effect of climate change is similar to the effect of increasing hemispheric background (+0.2–1 kppb[v]·h) over most parts of southern and western Europe. The values are smaller than the anticipated effect of the changing emissions but, together with the increasing hemispheric concentrations of O_3, they act to lessen the benefits gained by reducing O_3 precursor emissions in Europe. Northern Europe is not expected to experience any significant changes in AOT40c following this climate change realization.

DISCUSSION

In this work, we investigated the relative importance of emission changes in Europe, increased hemispheric background concentrations of O_3, and climate change on AOT40 in Europe.

To study emission changes in Europe, we used two different O_3 precursor emission scenarios. One scenario describes a likely future situation; the second has been regarded as close to what is technically feasible. However, following the ambitious climate-mitigation commitments currently discussed in Europe, measures aimed at reducing European CO_2 emissions, may, in fact, simultaneously reduce O_3 precursor emissions below the MFR-DEEP scenario. Although there are many reports of increasing hemispheric background concentration of O_3, the magnitude of the change varies from location to location, which makes statements of future hemispheric background concentrations even more uncertain. The detailed change in climate in Europe over the coming years and its impact on near-surface O_3 is also very uncertain. This is both because of the imperfection of the models and tools we use to predict future climate and because climate is inherently dependent on actions and decisions by

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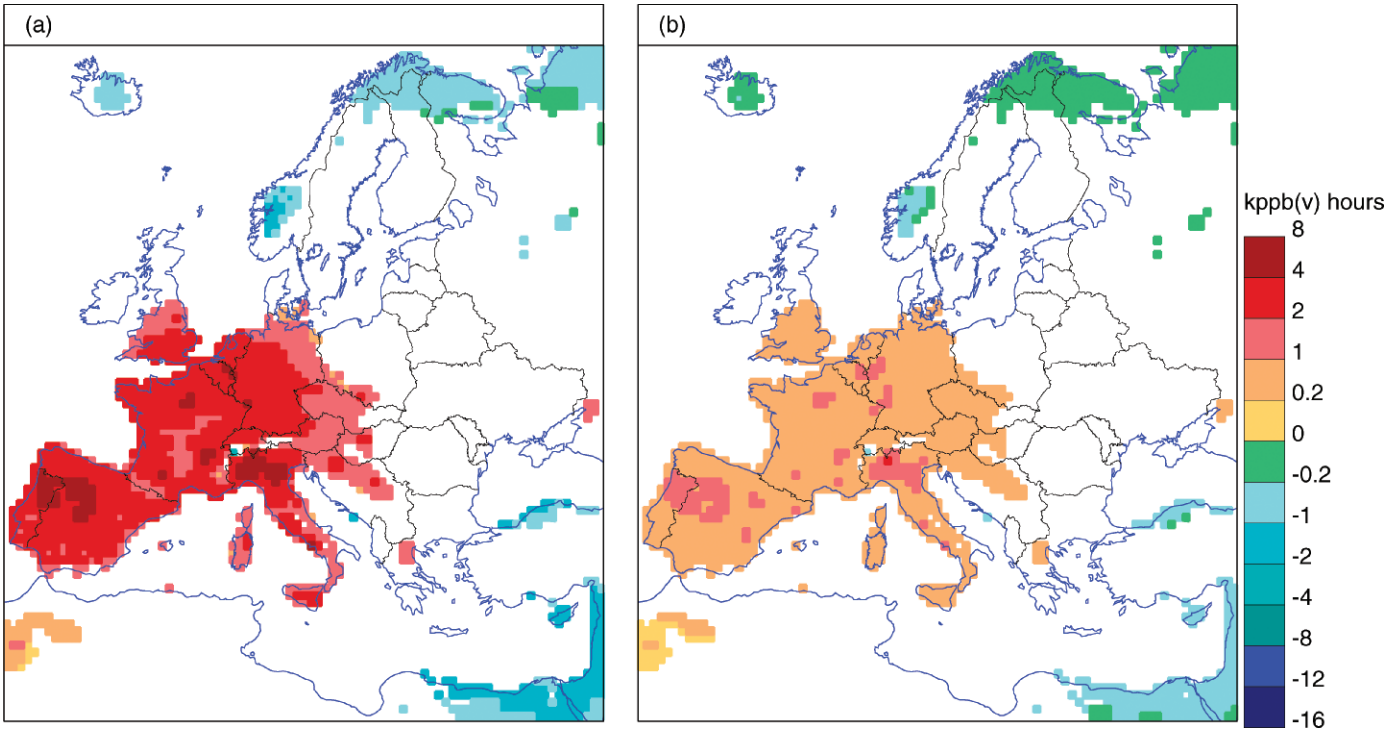


Figure 5. Change in AOT40c from control climate to future climate. (a) 1961–1990 to 2021–2050; (b) 2004 to 2020. Only grid cells with changes significant at the 99% level are shown. Units: kppb(v)·hours.

mankind during the coming decades. In our study we used one particular CTM operating on data from one regional climate model simulating one CO_2 emission scenario and forced by one particular global model on the boundaries. Using another CTM or climate model would give different details, but the overall effect of regional climate change on European O_3 in this study is similar to other recent studies (28–30).

CONCLUSIONS

Although the results presented above are not strictly additive due to nonlinearities in O_3 chemistry, the following conclusions can be drawn:

Anticipated reductions in European O_3 precursor emissions will have a large beneficial impact on near-surface O_3 across Europe. The resulting reduction in AOT40c will be most pronounced in southern and central Europe—where AOT40c is currently at its highest, but the effect will also be clearly evident in the Nordic countries. Increasing hemispheric background concentrations of O_3 will, however, to a large extent offset the improvements in AOT40c in the Nordic countries over the coming 16 y.

Climate change has the potential to greatly increase O_3 concentrations in most of continental Europe. Given that the committed climate change is already significant and irrevocable, we regard the long-term effect of climate change a very serious impediment in reaching and retaining acceptable O_3 levels in continental Europe during the 21st century. Over the 16-y period studied in the present work, climate change, together with the increasing hemispheric background of O_3, will to some extent counteract the gains in AOT40c achieved through reducing O_3 precursor emissions in Europe. In the Nordic countries climate change results in a small decrease of AOT40c, both over a 16-y perspective and in a 60-y perspective.

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