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Modelling the impacts of climate change on tropospheric ozone over three centuries

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Abstract

The ozone chemistry over three centuries has been simulated based on climate prediction from a global climate model and constant anthropogenic emissions in order to separate out the effects on air pollution from climate change. Four decades in different centuries has been simulated using the chemistry version of the atmospheric long-range transport model; the Danish Eulerian Hemispheric Model (DEHM) forced with meteorology predicted by the ECHAM5/MPI-OM coupled Atmosphere-Ocean General Circulation Model. The largest changes in both meteorology, ozone and its precursors is found in the 21st century, however, also significant changes are found in the 22nd century. At surface level the ozone concentration is predicted to increase due to climate change in the areas where substantial amounts of ozone precursors are emitted. Elsewhere a significant decrease is predicted at the surface. In the free troposphere a general increase is found in the entire Northern Hemisphere except in the tropics, where the ozone concentration is decreasing. In the Arctic the ozone concentration will increase in the entire air column, which most likely is due to changes in transport. The change in temperature, humidity and the naturally emitted Volatile Organic Compounds (VOCs) are governing with respect to changes in ozone both in the past, present and future century.

1 Introduction

Warming of the climate system is now an indisputable fact and it is very likely that most of the warming has an origin in the anthropogenic interference with the environment (IPCC, 2007). Since the atmospheric chemistry of most chemical compounds are highly dependent on temperature, the observed warming will inherently affect the chemical composition of the atmosphere. The climate system of the earth is very complex and intertwined. A temperature increase will lead to changes of other meteorological parameters like e.g. precipitation, pressure patterns, humidity and cloud cover. Common for all these parameters is that they influence the air pollution levels and distribution via physical and chemical processes. Many recent studies and reviews have investigated and discussed this interaction (e.g. Carmichael and Dentener, 2007; Giorgi and Meleux, 2007; Vataurd and Hauglustaine, 2007; Hedegaard et al., 2008; Hjorth and Raes, 2009; Jacob and Winner, 2009). The effect of changes in meteorology on air quality implies that even though we today decide to keep the anthropogenic emissions of air pollutants constant, the air pollution levels *will* change anyway. Evaluations and developments of new emission reduction strategies should include this additional effect and therefore sensitivity studies like the one presented here are necessary.

Since the industrialization in the end of the 19th century greenhouse gases have increased and our climate has been affected and thereby been under a continuous change into what we experience today. Due to the relative long lifetimes of some greenhouse gases it is well known that the climate will continue its ongoing change for the next 100 yr or more (Monastersky, 2009) with or without some accelerating or stagnating periods depending on the applied future air quality legislation (Raes and Seinfeld, 2009).

In this paper we focus on ozone and its related precursors. The state of the art within the discipline of modelling climate change and air quality interactions is sketched in the next section. Section 3 describes the model tools of this study and the current simulations carried out. In Sect. 4 the meteorological output is described and finally the chemistry results are outlined and discussed in Sect. 5. Section 6 highlights the conclusions and gives a brief view on ways to go within this research field in the future.

2 State of the art

Since about a decade ago a new branch of air quality modelling has developed within the international atmospheric chemistry transport modelling community. The observed climate change within the last century and an establishment of the anthropogenic origin of these changes by the publishing of the fourth IPCC Assessment Report in 2007 (IPCC, 2007) have initiated an urge to understand, how these changes in the global climate system affect future air quality.

The distribution and lifetimes of air pollutants depend strongly on many meteorological parameters. It is therefore very likely that the fate of the air pollutants will be altered under changed climate conditions even though the anthropogenic emissions are stagnated to levels of today. Both regional (Dawson et al., 2008; Hogrefe et al., 2004; Langner et al., 2005; Meleux et al., 2007), hemispheric (Hedegaard et al., 2008), and global (Brasseur et al., 2005; Liao et al., 2007; Murazaki and Hess, 2006; Stevenson et al., 2005; Wu et al., 2008a) chemistry transport models have been used to quantify the changes in air pollution levels due to climate change, and today more and more research groups all over the world develop coupled (in most cases one-way coupling) climate/air-quality-models in order to answer the growing amount of questions from policy makers.

There is a general belief that the size of the signal from climate change on air pollution levels and distributions is large enough to necessarily be accounted for when future emission legislation are developed. Wu et al. (2008b) defines "climate penalty" as the need for stronger emission controls in order to achieve a given air quality standard. As an example Wu et al. (2008b) concludes from their results that air quality control needed to reduce the NO_x levels 40% under current climate conditions will match a reduction of 50% under 2050 climate conditions. This 25% larger reduction by the authors defined as the "climate penalty" add to the fact that policy makers must take into account the effects from climate change when making new air quality control strategies.

Historically, one of the first studies published, with one-way coupling of climate model to a full chemistry transport model with an advanced chemistry scheme, was the regional study carried out by Johnson et al. (2001). They used the SRES A2 emission scenario for the period 1990–2100 and showed that the impacts of climate change leads to a decrease in the net production of tropospheric ozone combined with a smaller increase of stratospheric input ozone at the top of the model domain. However, the vertical resolution in the modelof the upper stratosphere is relatively low. The whole troposphere and the stratosphere up 100 hPa is only represented by nine layers. Therefore the model can not reproduce the stratosphere-troposphere exchange of ozone in cases of e.g. tropopause foldings where large amounts of stratospheric ozone is transported across the tropopause into the troposphere. Furthermore Johnson et al. (2001) did not include the feedback from natural emissions, which together with the relatively low model resolution makes the results very preliminary. Hogrefe et al. (2004) used a regional model centred over the eastern United States and showed that the effect from a changed climate may contribute to the air pollution levels in the future equal to the effects from changed emissions.

From 2005 and forth more and more research groups began to study the effects on climate change on air pollution levels and the model setups became more refined. Today, approximately half a decade later, a small climate change – air pollution community has developed within the chemistry transport modelling community.

Murazaki and Hess (2006) were the first[1] to publish results from a global simulation with one-way coupling of a climate model and a chemical transport model projecting the effects of climate change on air pollution levels throughout the 21st century. They used the global Model of OZone and Related Chemical Tracers, version 2 (MOZART-2) forced with future meteorology from the National Center for Atmospheric Research (NCAR) climate system model (CSM 1.0). Murazaki and Hess (2006) fixed both the anthropogenic and the biogenic emissions at a 1997 level in order to separate out the effects from climate change on air pollution. In general they found that background ozone will decrease in the future in contrast to the locally produced ozone. Especially, the ozone levels over the densely populated areas (high NO_x levels) will increase significantly since the predicted decrease in background ozone is far smaller than an expected increase in locally produced ozone.

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[1] Stevenson et al. (2005) have carried out similar simulation with a global model but only for the period 1990–2020.

This supports the results of Knowlton et al. (2004) who predict ozone-related health effects to be much more significant under future climate conditions. Hedegaard et al. (2008) published similar results as Murazaki and Hess (2006) but the simulation included variable biogenic emissions. The level of Biogenic Volatile Organic Compounds (BVOCs) increases significantly due to their temperature dependency and the temperature evolution during the 21st century. Besides ozone, Hedegaard et al. (2008) also examined the consequence of climate change on some particles. As an example they found that the sulphate levels over the already polluted areas would increase significantly due to an enhanced chemical production.

In 2005 Langner et al. (2005) published results including climate change-air pollution interaction from a regional study. They studied the tropospheric surface ozone over Europe, using the regional Multiscale Atmospheric Transport and Chemistry Model (MATCH) forced with climate data from the regional Rossby centre Atmospheric Climate model (RCA) version 1. Langner et al. (2005) found a general increase in surface ozone over southern and central Europe and a general decrease over northern Europe. They stated that, if the climate scenario used in the simulations is representative for the future climate, the increase in surface ozone in the future is comparable to the expected reductions resulting from emission reduction protocols in force by the time published.

Meleux et al. (2007) did similar experiments, which confirm the results found by Langner et al. (2005). Meleux et al. (2007) used the CHIMERE chemistry-transport model forced with climate predictions from the Regional Climate Model (RegCM). They also found a substantial increase in the daily peak ozone during the European summer periods which might pose a more serious threat to human health, agriculture and natural ecosystems in the future. Meleux et al. (2007) also conclude that the summer of 2003 very well could be a good example of future threats from air pollution and climate change on human, agriculture and nature.

All the models mentioned above are offline model setups. Only a very few groups have made simple online couplings of Climate GCM-CTM models (e.g. Forkel and

Knoche, 2006). The advantages from online coupling is the model ability to account for the radiative feedbacks from short-lived greenhouse gases like ozone and methane as well as aerosols between the large scale climate system and the small scale chemical regimes. However, online coupled climate-chemistry models demands very high scientific understanding of the individual feedbacks and physical processes and the process is currently limited by the computational constraints. The differences in the results of online and offline coupled models varies a lot depending on the complexities of the given models (Zhang, 2008).

Within online models, two conventional frameworks exist, – unified online coupling and separate online coupling. The first integrates an air quality model into a meteorological model and the second consists of two separated systems (meteorology model/air quality model), that exchange information at every time step. These types of coupling are with respect to climate change mostly used for regional climate/air quality modelling (Zhang, 2008).

On a global scale the online coupling is more focusing on earth system models, which is a large model system including atmosphere, ocean, biosphere, cryosphere and chemistry models. However, earth system models are still in their premature stages with respect to the inclusion of chemistry and biosphere. The large uncertainties in accurately describing the climate-aerosol-chemistry-cloud-radiation feedback processes and the large temporal and spatial demands in order to predict effects of climate change, justifies using the offline method.

3 Model setup and simulation

In this study the Danish Eulerian Hemispheric Model (DEHM), is driven on six-hourly meteorology input simulated by the coupled Atmosphere-Ocean General Circulation Model ECHAM5/MPI-OM and constant anthropogenic emission from the year 2000. The faith of 58 chemical species and 9 classes of particulate matter has been simulated in four decades (1890s, 1990s, 2090s and 2190s) to examine the evolution of air pollutants over three centuries. Here we focus on ozone and its precursors. Analysis of other species (particles) will be published elsewhere. The performance of the total model system with ECHAM5/MPI-OM model coupled to the DEHM model system has been thoroughly tested in earlier studies (Hedegaard, 2007; Hedegaard et al., 2008).

DEHM is an Eulerian Atmospheric Chemistry Transport Model (ACTM) with a two-way nesting capability to obtain higher resolution over limited areas (Christensen, 1997; Frohn et al., 2002a, 2002b; Frohn, 2004). In this study the model domain covers slightly more than the Northern Hemisphere (see Fig. 1). The horizontal grid has a resolution of 150 km × 150 km using a polar stereographic projection true at 60° N. Vertically the model consists of 20 unevenly distributed layers defined on terrain following σ-levels extending up to a height of approximately 16 km (100 hPa). Boundary conditions for the model domain depend on the direction of the wind, such that free boundary conditions are used for sections where wind flows out of the domain. Constant boundary conditions are used for sections of the boundary where wind is flowing into the domain; in this case, the boundary value is set to the annual average background concentration. For ozone these are taken from ozone soundings and are the same for all simulations in this study (Logan, 1999).

The chemistry scheme in DEHM is based on an extension of the European Monitoring and Evaluation Programme (EMEP) scheme (Simpson et al., 2003). The model describes concentration fields of 58 chemical species, including secondary inorganic particles and 9 species representing primarily emitted particulate matter ($PM_{2.5}$, PM_{10}, TSP, sea-salt $< 2.5\,\mu m$, sea-salt $> 2.5\,\mu m$, fresh black carbon, aged black carbon and organic carbon). A total of 122 chemical reactions are included. The DEHM model is based on the full 3-D advection-diffusion equation and is driven by meteorological data from a numerical general circulation models used for either weather or climate predictions. The advection is solved numerically using an Accurate Space Derivatives scheme with non-periodic boundary conditions for the horizontal advection and a finite elements scheme for the vertical advection. The diffusion is solved using the finite elements scheme. Wet and dry deposition is parameterized similar to the EMEP model

(Simpson et al., 2003) except for the dry deposition of species on water surfaces. In this case, the deposition depends on the solubility of the chemical specie and the wind speed (for further details see Asman et al., 1994 and Hertel et al., 1995). All applied numerical schemes have previously been carefully tested (Christensen, 1997; Frohn et al., 2001, 2002a, 2002b).

The anthropogenic emissions that feed into the chemical transport model DEHM have been fixed at a year 2000-level in order to isolate the signal from climate change on air pollution. The emissions of the primary pollutants consist of data from the Global Emissions Inventory Activity (GEIA) (Graedel et al., 1993), the Emission Database for Global Atmospheric Research (EDGAR) (Olivier et al., 1996) both with global coverage, and the EMEP emissions (Vestreng, 2001) covering Europe. The GEIA database includes natural emissions of NO_x from soil and lightning and Black Carbon, which mainly originates from biomass burning. These natural emissions have also been fixed on a 2000-level. On the contrary the emission of isoprene is calculated dynamically in the model according to the GEIA natural VOC emission model (Guenther et al., 1995). Other naturally emitted VOCs like for example terpenes are not yet included in the model.

The coupled atmosphere-ocean model ECHAM5/MPI-OM consists of an Atmospheric General Circulation Model ECHAM5 (Roeckner et al., 2003 and 2006) and the ocean-sea-ice model MPI-OM (Marsland et al., 2003). The atmospheric model ECHAM5 is horizontally defined in a spectral grid with truncation T63. Vertically the model is defined in a hybrid sigma-pressure system and divided into 31 layers with the top layer at 10 hPa. State-of-the-art parameterizations are used for shortwave and long wave radiation, stratiform clouds, boundary layer and land-surface processes and for describing gravity wave drag in the model. Details about the description of the aerosol effect in the model for these simulations can be found in May (2008).

The ocean-sea-ice model has a horizontal resolution of 1.5° × 1.5° and is vertically discretized into 40 z-levels. The model operates on the primitive equations for a hydrostatic Boussinesq fluid with a free surface. Along-isopycnical diffusion, horizontal tracer mixing by advection with unresolved eddies, vertical eddy mixing, near-surface wind stirring, convective overturning, and slope convection are all parameterized in the ocean model. Concentration and thickness of sea ice is treated interactively in the model by a dynamic and thermodynamic sea-ice model. For further details of the ocean-sea-ice model, see Marsland et al. (2003).

The atmosphere model ECHAM5 and the ocean-sea-ice model MPI-OM is interactively coupled and exchange information regarding sea-surface temperature, sea-ice concentration and thickness, wind stress, heat and freshwater once a day. Further details of the coupling can be found in May (2008) and Jungclaus (2006). The model does not employ flux adjustments. The coupling of the atmosphere and ocean model has been tested by Jungclaus et al. (2006) and is found to perform well with respect to sea surface temperatures, sea-ice conditions, meridional heat and transport of freshwater (May, 2008).

The climate simulation used in the current experiment is forced with emissions, based on realistic estimations until year 2000 and emissions according to the SRES A1B scenario in the period 2000–2100. In the final period 2101–2200 all the emissions have been fixed at a 2100-level. The SRES A1B scenario assumes a future world with very rapid economic growth and a rapid introduction of new and more efficient technologies balanced between both fossil – and non-fossil intensive energy sources. The population growth peaks in about 2050 and declines hereafter (Nakicenovic et al., 2000). It should be noted that this forcing from emission only applies to the projected meteorology. The anthropogenic emissions used for the DEHM model is different (see above) and in this study kept constant.

4 Meteorology by ECHAM5/MPI-OM (SRES A1B scenario)

Meteorological parameters are of great importance with respect to transport, deposition, chemical conversion and photochemistry of chemical compounds in the atmosphere. This study focuses on ozone and its precursor and here especially temperature, specific humidity and the amount of shortwave radiation influences the processes. Warmer air can contain more water vapour and speed up the chemical reaction rates. Increased specific humidity results in more H_2O molecules in the atmosphere, which contributes to an increased level of hydroxyl radicals (OH) which again speeds up the chemical reactions. The atmospheric photolysis processes are obviously controlled by the availability of sunlight and therefore the amount solar shortwave radiation is very important. In the following these three controlling meteorological parameters (temperature, specific humidity and solar radiation) are shown for the lowest model layer, however, in the analysis behind this study other parameters and layers have also been investigated thoroughly.

4.1 Temperature

The climate simulation used in this study where a part of the 4th IPCC Assessment Report (AR4) multi-model ensemble study. In the current simulation the global temperature is predicted to increase by 3.03 °C by the end of the 21st century and 4.27 °C by the end of the 22nd century both relative to the period 1971–2000 (May, 2008). This increase is a little higher than the average value (2.65 and 3.36, respectively) predicted by the multi-model ensemble in AR4 following the SRES A1B scenario (Meehl et al., 2007). However, it is well within the standard deviation of the IPCC AR4 multi-model ensemble by the end of the 21st century.

In Fig. 1 the mean temperature of the four decades considered in this study is plotted together with the absolute change between these decadal mean values and the significance of these changes using a student's t-test. Temporally the temperature is increasing significantly in the two future decades (2090s and 2190s). The ECHAM5/MPI-OM model simulation generally also predicts a temperature increase in the 20th century (represented by the difference between the 1990s and the 1890s), however, this increase is only significant in the tropics and a temperature decrease is predicted over the North Atlantic storm tracks.

The absolute largest temperature increase is found in the 21st century (see centre plot of Fig. 1 (2090s minus 1990s)). This is in line with results from May (2008) who found that the changes in the global annual mean near-surface temperature is largest around 2060 with a warming rate of more than 4.5 °C/century. This high warming rate is due to a strong increase in all greenhouse gases except methane and a marked reduction in the anthropogenic sulphur emissions according to the SRES A1B emission scenario. Focussing on the 21st century (2090s–1990s) geographically the temperature increase is largest in the Arctic region where it locally exceeds 9 degrees. Over land areas in general the temperature increase ranges from 3 to 6 degrees and over the ocean the increase is more modest in the range 1 to 4 degrees.

4.2 Specific humidity

In Fig. 2 the specific humidity is shown. The specific humidity is closely related to the temperature. At saturation the specific humidity is a quasi-exponential function of temperature according to the Clausius-Clapeyron equation (Goosse et al., 2009). This exponential dependency implies that the change in humidity is significantly largest at low latitudes (where the highest temperature is predicted). The specific humidity distribution follows the latitudes very closely both for the means of the four decades (upper panels of Fig. 2) and for the changes between these decades (middle panel of Fig. 2). As the temperature plot (Fig. 1) the specific humidity also shows the absolute largest changes within the 21st century (middle panel of Fig. 2). In the 20th century the predicted change in specific humidity is only significant in the tropical region. Vertically the largest change in specific humidity is confined to the lowest part of the troposphere (app. lowest 2 km) where most water vapour in the atmosphere is found, in contrast to the vertical profile of temperature change which is rather uniform.

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4.3 Shortwave radiation

The Shortwave radiation is latitudinal dependent and derived from the cloud cover calculated by the ECHAM5 according to the method used in Hedegaard (2007). In Fig. 3 the shortwave radiation of the four decades is shown together with the difference and the significance of these differences. The largest changes are predicted in 21st century, where the shortwave radiation is predicted to increase in the southern mid-latitudes and the subtropics. This is in line with the general idea that the earth in the future will experience longer and more persistent periods with drought and high temperatures in the subtropics (Christensen et al., 2007). In contrast to the temperature and specific humidity, the change in shortwave radiation is only significant in the 21st century (cf. lower panel, Fig. 3). The predicted change is highly dependent on latitude and increasing everywhere in the domain, except over the tropical Pacific.

Since the shortwave radiation is derived from latitude and ECHAM5 cloud cover, the area of decreasing shortwave radiation over the tropical Pacific most likely is due to an increase in cloud cover in the climate simulation. The ECHAM5/MPI-OM simulation has been part of the multi-model ensembles of 4th IPCC report (AR4), which confirms a general increase in cloud cover over the tropical Pacific (Meehl et al., 2007). In central and southern Europe the shortwave radiation is predicted to increase more than 20% which is in good agreement with the theory of longer and more persistent periods of dry warm summers in the future at these latitudes (Vautard and Hauglustaine, 2007; Stott et al., 2004).

5 Results and discussions

In this section the main chemistry processes regarding tropospheric ozone chemistry is discussed, together with a presentation of the results. This is followed by a discussion and a summary of the findings with regard to the photochemical processes in this model setup and the changes found in the distribution of the related chemical compounds due to climate change. The distribution, the differences and the significances of the ten-year average O_3 concentration in ppbV is plotted in Fig. 4. From the upper panel it can be seen that the O_3 concentration in the lowest model layer in all four decades is highest in the subtropics and the tropics over land, especially close to the anthropogenic precursor sources and downstream from these.

The life-time of ozone depends strongly on the latitude, time of year, solar radiation and vertical placement in the atmosphere. Because free oxygen and O_3 molecules (together called "odd oxygen") are rapidly interconverted an individual lifetime of O_3 is often irrelevant with respect to atmospheric transport. Considering atmospheric long-range transport this interconversion makes the lifetime of odd oxygen more interesting. The lifetime of odd oxygen is ranging from a few days in the lower troposphere and up a month or more in the middle and upper troposphere (Lollar, 2007). The relative longer lifetime of odd oxygen is the reason O_3 is considered as a long-range transported specie. O_3 is not directly emitted, but produced from its precursors, which is of both biogenic and anthropogenic origin. A local measured ozone concentration consists of a local, an inter-continental and a background contribution, which dependent on the distance to the precursor sources, the local photochemical conditions and the transport pathways. The concentration distribution plotted in Fig. 4 reflects these features very well.

The increase of the ozone concentration is due to a combined effect of increased temperature, solar radiation, humidity, isoprene chemistry and anthropogenic VOC pollution as well as influence from NO_x (Fig. 9). The trend in ozone concentration is the same in all three centuries. However, most pronounced in the 21st century with a general decrease over the ocean and very remote areas (as e.g. the dessert of Sahara and central Asia) and with an increase over the densely populated areas and areas where biomass burning relatively often occurs. The Arctic Ocean differs from the above pattern by experiencing a significant increase in the 21st and 22nd century (cf. Fig. 4 middle and lower panel).

The major contributor to tropospheric ozone is the termolecular recombination reaction:

$$O(^3P) + O_2 + M \rightarrow O_3 + M \tag{R1}$$

where M is an inert atom or molecule in the atmosphere primarily O_2 and N_2. $O(^3P)$ is formed in the photolysis of NO_2:

$$NO_2 + h\nu \rightarrow O(^3P) + NO \qquad \lambda \leq 424\ \text{nm} \tag{R2}$$

Some of the formed O_3 is destroyed by reactions with NO:

$$O_3 + NO \rightarrow NO_2 + O_2 \tag{R3}$$

It can be seen that Reactions (R1) to (R3) are a closed cycle where all the products also are reactants.

In the troposphere Reactions (R1) through (R3) illustrates the most important cycle for O_3, however since the cycle is closed, not all tropospheric O_3 stems from this cycle. The oxidation of VOC and CO plays a central role for this extra contribution to the O_3 budget, see Sect. 5.1. O_3 plays an important role by being the major source of hydroxyl radicals (OH), which determines the oxidation capacity of the atmosphere:

$$O_3 + h\nu \rightarrow O(^1D) + O_2 \qquad 290\ \text{nm} \leq \lambda \leq 319\ \text{nm} \tag{R4}$$

$$O(^1D) + H_2O \rightarrow 2OH \tag{R5}$$

The main source of OH formation is Reaction (R5). In relation to climate change it is important to note that $O(^1D)$ can either react as Reaction (R5) or collide with N_2 or O_2 and quenched to ground state oxygen ($O(^3P)$). As the temperature and water vapour increases in the future due to climate change (Figs. 1 and 2) Reaction (R5) is in more favourable than the formation of $O(^3P)$ which in the remote and clean areas leads to a decrease in the ozone concentration. This can be observed in Fig. 4, where a decrease of ozone in the 21st century over the ocean and in the Sahara is predicted.

The concentration of H_2O is increasing in this century (Fig. 2) and it is regions with low NO_x where the destruction of O_3 according to Reaction (R4) will be dominant.

Figure 5 shows the changes of OH concentrations. Since the hydroxyl radicals determine the oxidation capacity of the atmosphere, they are very important when considering pollution levels; e.g. fate of primary emitted pollutants, formation of ozone and secondary particles. In Fig. 5 (lower panel) there is an increasing tendency in the 21st century over the ocean, in large part of Europe, including Greenland, Arabia and central Asia. On the contrary the hydroxyl radical levels are decreasing elsewhere in the domain of interest.

5.1 Impact of Volatile Organic Compounds (VOCs)

Non-methane Volatile Organic Compounds (VOCs) can be split into anthropogenic VOCs (AVOCs) (~ 10% of total) and Biogenic VOCs (BVOCs) (~ 90% of total) (e.g. Goldstein and Galbally, 2007). We limit ourselves to discuss alkanes and alkenes ignoring the emission and chemistry of oxidized VOC's, alkynes and aromatics, though they are important VOC's in the troposphere. AVOC is dominated by saturated VOCs (alkanes) and BVOC is dominated by unsaturated VOCs (alkenes) but fractions play important roles in tropospheric chemistry. Alkanes can be transported over long distances due to their chemical stability. Hydroxyl radicals react via hydrogen abstraction of both a terminal and an internal carbon atom followed by addition of O_2 and thereby forming alkyl peroxy radicals (RO_2):

$$RH + OH + O_2 \rightarrow RO_2 + H_2O \tag{R6}$$

These alkyl peroxy radicals (RO_2) are highly reactive and can react with NO forming NO_2:

$$RO_2 + NO \rightarrow RO + NO_2 \tag{R7}$$

which through Reactions (R1) and (R2) produces extra O_3 compared to the photostationary state presented by Reactions (R1) to (R3).

Approximately 40% of BVOCs emitted is isoprene (Fall, 1999; Goldstein and Galbally, 2007). Isoprene emissions are in the DEHM model calculated based on the temperature and sunlight dependent parameterization from Guenther et al. (1995). The tropospheric lifetime of isoprene (C_5H_8) due to its reaction with OH, nitrate (NO_3) and O_3 is 1.4 h, 1.6 h and 1.3 days, respectively (summarized in Hedegaard, 2009). Due to this relatively low atmospheric lifetime the highest isoprene concentration (top panel of Fig. 6) is found close to the emission sources in especially the tropical areas. The changes in isoprene concentration are also confined to the emission areas and the increasing tendency is very similar to both the temperature and shortwave radiation pattern (Figs. 1 and 3)

The main degradation pathway of alkenes are OH addition to the double bond (e.g. isoprene has two double bonds) rather than hydrogen abstraction as is the case for alkanes. OH adds to one of the double bonds followed by addition of O_2 creating a peroxy radical. Since isoprene has two conjugated double bonds, the oxidation of isoprene to its peroxy radical can form eight different isomers in different ratios. There are therefore a close correlation between the concentration of isoprene and its peroxy radicals. It is beyond the scope of this article to discuss the reaction pathway further (see Seinfeld and Pandis, 2006; or for recent results Paulot et al., 2009).

Ozone can also attack the double bonds in unsaturated hydrocarbon and form carbonyl species and organic acids. It has been observed for the reaction between isoprene, ozone and hydrogen peroxide, that the product yield increase from 1 to 9% as the relative humidity increases (Sauer et al., 1999). Hence, the yields of methacrolein (C_4H_6O) and methyl vinyl ketone (C_4H_6O) will also increase since they are products formed in part from the decomposition as explained above.

NO_2 reacts with ozone to form nitrate radicals which photolysis fast during the day and thus is of little importance. However, during night time substantial concentrations can build up and affect the NO_x chemistry. Moreover it is suggested from field studies that NO_3 reactions can be a major contributor to isoprene loss at night (Rollins et al., 2009; Skov et al., 1992). NO_3 react by addition to isoprene at C_1 or C_4 carbon atom followed by addition of O_2 to make a 1,4 addition to form a nitrate peroxy radical isoprene adduct. In the model these peroxy radicals reacts with NO and thus will lead to ozone production at dawn trough the photolysis of NO_2 (R2). This contribution is however of minor importance. There is discussion in the literature of what will be the fate of these RO_2 radicals. They might react with one another, with another NO_3 radical or with OOH as discussed by Rollins et al. (2009).

However, in this study it is difficult to isolate the exact impact of NO_3, O_3 and OH respectively on isoprene because many other reactions also occurs simultaneously in the simulations and most of the areas with a high emission load of isoprene also to some extent are influenced by anthropogenic sources.

5.2 Production and loss reactions of hydro- and organic peroxy radicals

The main source of organic peroxy radicals is from AVOCs and BVOCs. The inorganic peroxy radical can be formed either from the simplest aldehyde:

$$HCHO + 2O_2 + h\nu \rightarrow 2HO_2 + CO \qquad 290\ \text{nm} \leq \lambda \leq 340\ \text{nm} \tag{R8}$$

$$HCHO + h\nu \rightarrow H_2 + CO \qquad 290\ \text{nm} \leq \lambda \leq 360\ \text{nm} \tag{R9}$$

$$HCHO + HO + O_2 \rightarrow HO_2 + H_2O + CO \tag{R10}$$

or from various reaction of OH with inorganic oxides (O_3, H_2O_2, SO_2 and CO) and the photolysis of carbonyl containing compounds.

HO_2 is removed either from the reaction with NO:

$$HO_2 + NO \rightarrow OH + NO_2 \tag{R11}$$

or by its self reaction:

$$HO_2 + HO_2(+H_2O) \rightarrow H_2O_2 + O_2 + H_2O \tag{R12}$$

Reactions (R11) and (R12) are competing reactions where the first dominate in NO_x rich areas exposed to high emission from combustion processes and the latter is dominating in the free troposphere and in marine environments. Reaction (R11) will be followed by Reaction (R1) and (R2) to form ozone whereas (R12) will lead to loss of odd oxygen and by that loss of O_3, see Fig. 4.

Figure 7 shows that the hydroperoxy radicals mainly increase in the 21st in the "semi-remote" areas which has a high fraction of vegetation and industry, and in the subtropical and tropical areas. The increase due to vegetation is explained above. The increased concentration of hydroperoxy radicals in the regions with high emissions from vegetation and regions with high emissions from anthropogenic sources, is caused by the increased level of water vapour and isoprene emissions in the future decades. When the water vapour content increases, the OH concentration increases, due to photolysis of ozone (Reaction R4). The hydroxyl radicals can then be converted to hydroperoxy radicals as described above.

In Fig. 8 the concentration and the difference of organic peroxy radicals between the three centuries are shown. From 1890s to 1990s the largest increase is found in the Tropics. However, the significance of this change is more scattered than in the 21st century where the concentration of the organic peroxy radicals is increased all over the domain. As in the 20th century the increases found in the 22nd century is more blurred with respect to significance, but in contrast to the 20th century more spread over the full domain. For example, a highly significant increase is predicted over the subtropic part of Europe, large parts of Asia and over the North Atlantic Ocean and the connecting part of the Arctic Ocean concerning the 22nd century. However, the increase in organic peroxy radicals can be explained by the same parameters as for hydroperoxy radicals.

NO_2 reacts with ozone to form nitrate radicals which photolysis fast during the day and thus is of little importance. However, during night time substantial concentrations can build up and affect the NO_x chemistry. NO_3 reacts fast with a series of alkenes among others isoprene and dimethyl sulphide. NO_3 reacts also with NO_2 to form N_2O_5 which hydrolyses heterogeneously with water to form nitric acid. This last reaction accounts for about the same removal as the reaction between OH and NO_2 at mid latitudes.

5.3 Development of ozone in the future related to its precursors

In the future, the radiation, temperature and water content of the atmosphere are predicted to increase according to the climate projection applied in this study. These parameters will increase the OH formation due to Reaction (R4) which initiates the cycle described in e.g. Reactions (R1)–(R6). Increasing amount of OH, HO_2 and RO_2 are also to some extend observed in the model runs (Figs. 7 and 8), and can explain the higher ozone concentrations observed in the simulations over the densely populated areas (US, central and southern Europe and parts of Asia) as observed in Fig. 4.

In a clean atmosphere, where the NO_x and VOC load is low, isoprene reduces the ozone concentration, which is in good agreement with the changes over the e.g. the oceans. However, in regions with moderate to high NO_x levels, the interaction between the emitted NO and the formed isoprene peroxides from OH, increases the concentration of HO_2 and NO_2, which then enhances the ozone formation. This can explain the higher ozone concentrations in Africa south of Sahara, Southeast Asia and South America. In addition these areas are covered with a large fraction of tropical plants and tropical rainforest, which in themselves emit a large fraction of the ozone precursor, isoprene. Moreover, the BVOC emission itself is expected to increase under changed climate conditions, which can further amplify the signal in the future ozone concentration.

In the Arctic the ozone concentration is predicted to increase throughout all three centuries, however the change is most significant in the 21st century. From analysis of each season it is found most significant during the winter season. In the Arctic region there is no sun in the winter months, and hence not any ozone degradation due to photolysis. Furthermore analysis of the ozone related species cannot explain the change over the Arctic region.

Over land the ozone dry deposition increases over land (not shown) in contrast to the concentration of O_3 (Fig. 4) and oppositely over the ocean the deposition decreases where as the concentration increases. Deposition to vegetative surfaces is much larger than to snow covered surfaces and it is found that the snow cover decreases significantly between the two periods (not shown), which can explain a decrease in the atmospheric ozone concentration over the land areas (Fig. 4).

Over the Arctic Ocean the temporal and spatial extent of sea ice decreases between the two decades (not shown). Ozone does dry deposit to water surfaces in the model (Asman et al., 1994; Hertel et al., 1995) and the deposition is larger for ice surfaces than for water, which results in a decrease in the dry deposition over ocean. This is good agreement with the results of the dry deposition (not shown) which the decreases over the ocean. However, there is also the possibility that the increased ozone levels could be due to increased horizontal transport from the source areas where it is shown that the ozone concentration will increase (Fig. 4). Furthermore vertical transport could also increase the concentration of the surface ozone concentration.

In Fig. 10 the change in ozone between the 1990s and 2090s is compared at three vertical levels; the surface layer, a layer situated at approximately 2 km altitude and one at approximately 5 km altitude. When entering the free troposphere the trend in ozone evolution becomes more zonal with a highly significant increase within and north of the subtropics and a highly significant decrease in the tropics. In the free troposphere the chemistry becomes simpler and less restricted to the emission areas. This study indicates that a given change in ozone concentration in the future depend on two competitive effects; Ozone destruction due increased water vapour in the atmosphere and ozone formation due to increased levels of ozone precursors. The increase in precursors is in this study dominated by an increase in biogenic isoprene emissions, since we keep the anthropogenic emissions constant. The latter effect is only significant in the presence of sufficient available NO_x. The ozone distribution in the free troposphere (last plate of Fig. 10) indicates that the effect from enhanced ozone precursors like isoprene exceeds the decrease in ozone concentration due to increased water vapour in and north of the subtropics and vice versa in the equatorial regions.

6 Summary, concluding remarks and future outlook

Surface ozone is of great importance since high ozone concentrations can have NO_xious effects on both humans and nature. This study has shown that at surface level the background ozone concentration will decrease during the 21st and the 22nd century. In contrast local ozone in the source areas of precursors (both anthropogenic and biogenic) will increase significantly due to climate change alone. In the free troposphere the ozone concentration will in general increase, except for the tropics where a significant decrease is found.

Over the Arctic Ocean a significant increase in surface ozone is found in the future, which can be explained by the predicted increase in the source areas and by long-range atmospheric transport. Also changes in dry deposition explain the observed in the simulation and finally changes in the vertical transport could contribute to the changes found over the Arctic ocean at surface level. To estimate the contributions from the above mentioned processes is beyond the scope of this paper and will be investigated further in the future.

Ozone in the atmosphere not only affects the human health and nature. It is also a significant short-lived greenhouse gas in the atmosphere. Today most atmospheric climate models have very simplified descriptions of atmospheric chemistry including ozone chemistry and therefore account poorly for the feedback from atmospheric chemistry to the climate system. The increase in ozone concentrations in large parts of the upper troposphere could have significant radiative effect, which not yet is accounted for in climate models. To include this and similar chemical feedback effects in the atmosphere, extensive Earth system models combined with an enormous computer capacity is necessary.

The scientific communities working on climate change and climate related subjects like atmospheric chemistry are currently merging and large model systems including various earth system processes are slowly developing. The study here is another example supporting the needs for the newly arisen concept called "Seamless Prediction", where the models merge into large Earth system models that can account for feedbacks between the atmosphere, hydrosphere, cryosphere, biosphere etc. Nevertheless, sensitivity studies like the current will still be necessary in future in order to understand and describe the individual (in this case chemical reactions and physical transport) processes.

The most import finding of this study is the fact that the contribution from climate change on ozone concentration in the future are largest in the 21st century. This change contributes significantly to the absolute ozone level and should therefore be accounted for when making new regulations for emissions of air pollutants in the future. The applied climate simulation is produced by ECHAM and based on the SRES A1B emission scenario, which only prescribe emissions until year 2100. From 2101 and forth the emissions in the climate model is kept constant, which to some extent can be verified as a reasonable choice by assuming that new technologies have taken over by then leading to radical changes in the global emission budget. This gives rise to some uncertainty, however not necessarily more than the one already cooperated into the method itself of using scenario data in general.

This study is a first step in the investigation of the chemical and physical processes governs the atmospheric photochemistry under changed climate conditions. The results are a first guess in the quantification of the signal from climate change on ozone chemistry over three centuries. Many improvements can be made, depending on the target of interest and the available computer and data resources. The first simulation for this study was made before the development and final release of the new improved emission scenarios called Representative Concentration Pathways (RCPs). These scenarios are going to replace the existing SRES scenarios for the coming 5th IPCC report in 2013. The RCPs are extended with a first guess on emissions until year 2200 and could be used similarly as the SRES A1B emission scenario in this study.

Another possible improvement of this experiment relates to the vertical resolution and domain of the DEHM model. Since vertical transport of ozone, through stratospheric-tropospheric exchange of air, might be a key parameter in explaining the changes in ozone concentration in the Arctic (and maybe everywhere), a higher upper model level and increased vertical resolution at the tropopause level could be another interesting feature to investigate in any future simulations.

Finally, the current experiment only deals with the impacts of climate change on ozone chemistry. The signal from emission change is obviously very significant too. And the interaction of these two signals would generate the best estimate of what to be expected in the future atmosphere with respect to ozone concentrations and the related photochemical processes. This work is in progress and will be published at a later stage.

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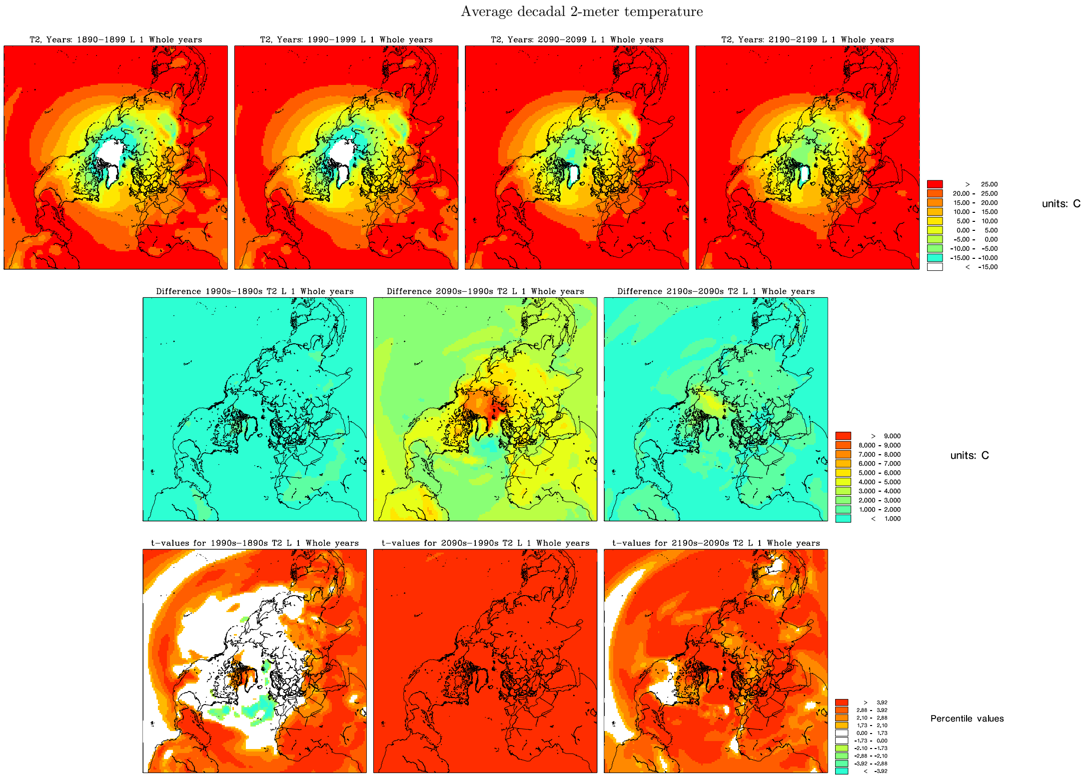

Fig. 1. Top panel: Decadal mean temperature of the lowest model layer (L1) in °Celsius for four decades (1890s, 1990s, 2090s and 2190s). Middle panel: The difference in °Celsius (1990s minus 1890s, 2090s minus 1990s and 2190s minus 2090s). Bottom panel: The significance of the differences according to the students t-test. Red colours indicate significant increase and green colours significant decrease. The threshold value for significance is chosen to 10% (white areas indicates no significant change).

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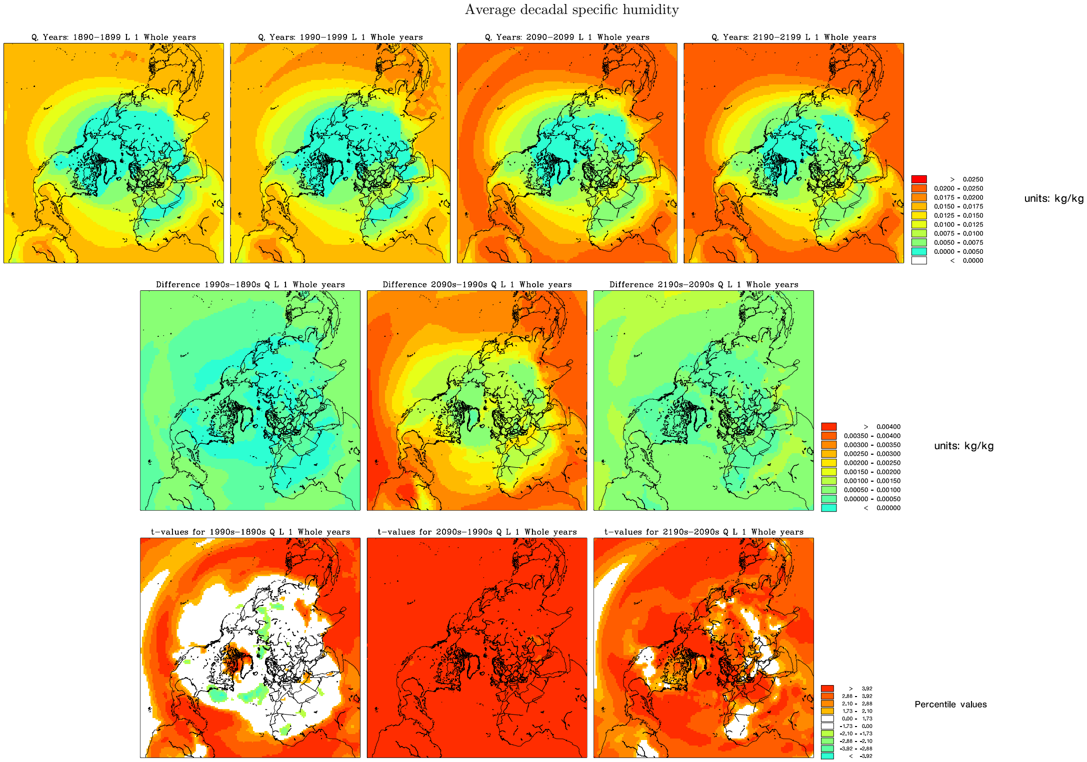

Fig. 2. Average decadal specific humidity in the lowest model layer in Kg Kg^{-1}. Setup the same as Fig. 1.

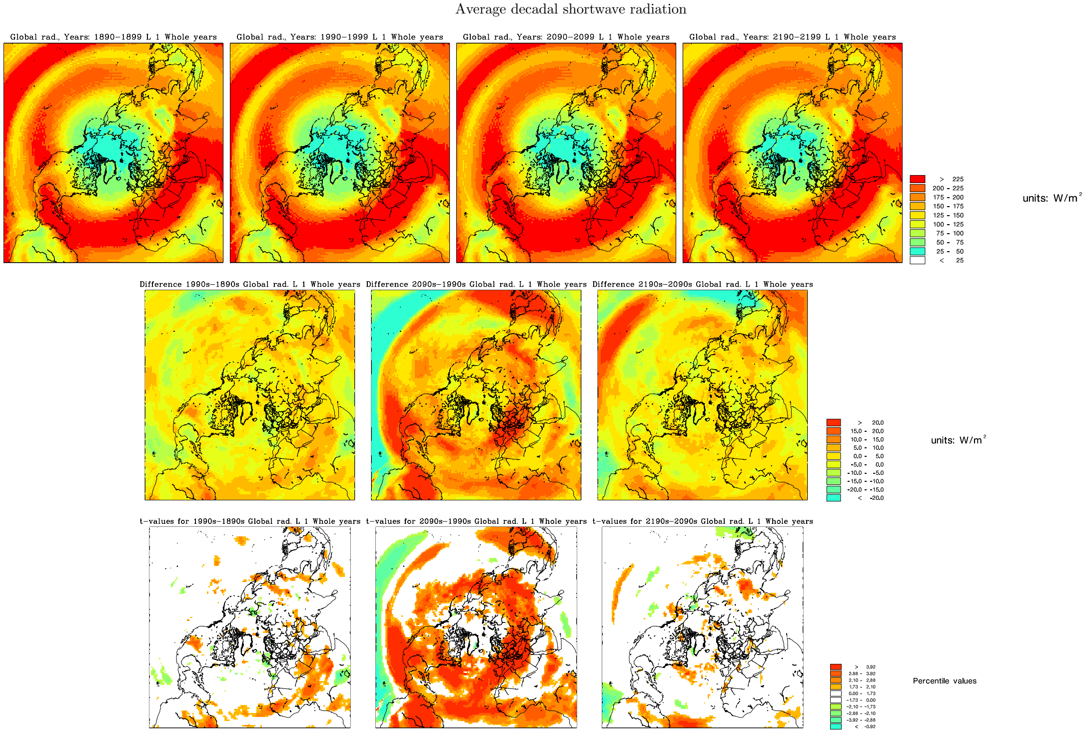

Fig. 3. Average decadal shortwave radiation in the lowest model layer in W m^{-2}. Setup same as Fig. 1.

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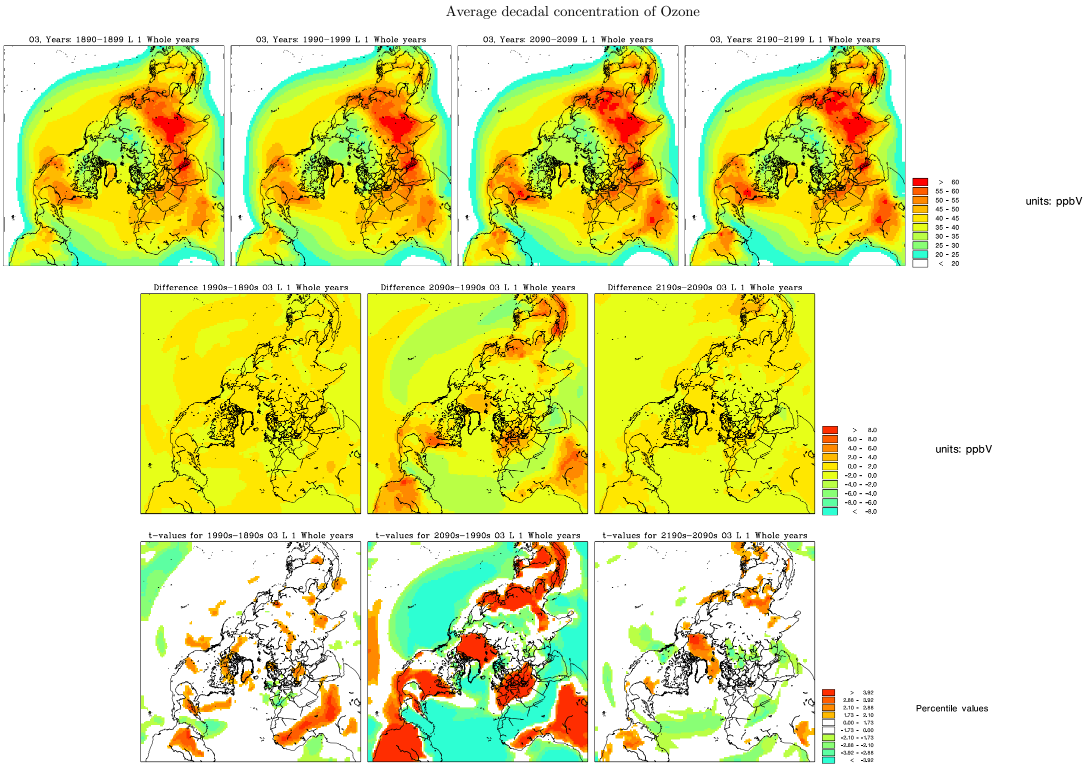

Fig. 4. Average decadal ozone (O_3) concentration in the lowest model layer in ppvV. Setup as in Fig. 1.

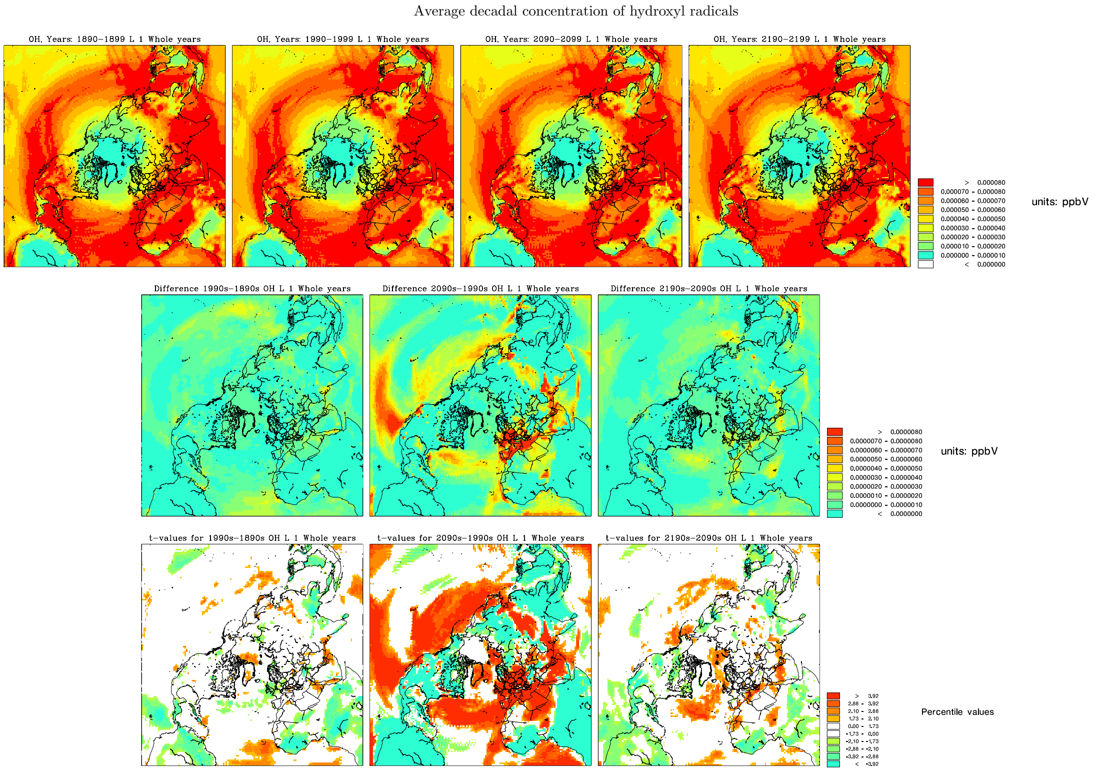

Fig. 5. Average decadal concentration of hydroxyl radical (OH) in the lowest model layer in ppbV. Setup the same as Fig. 1.

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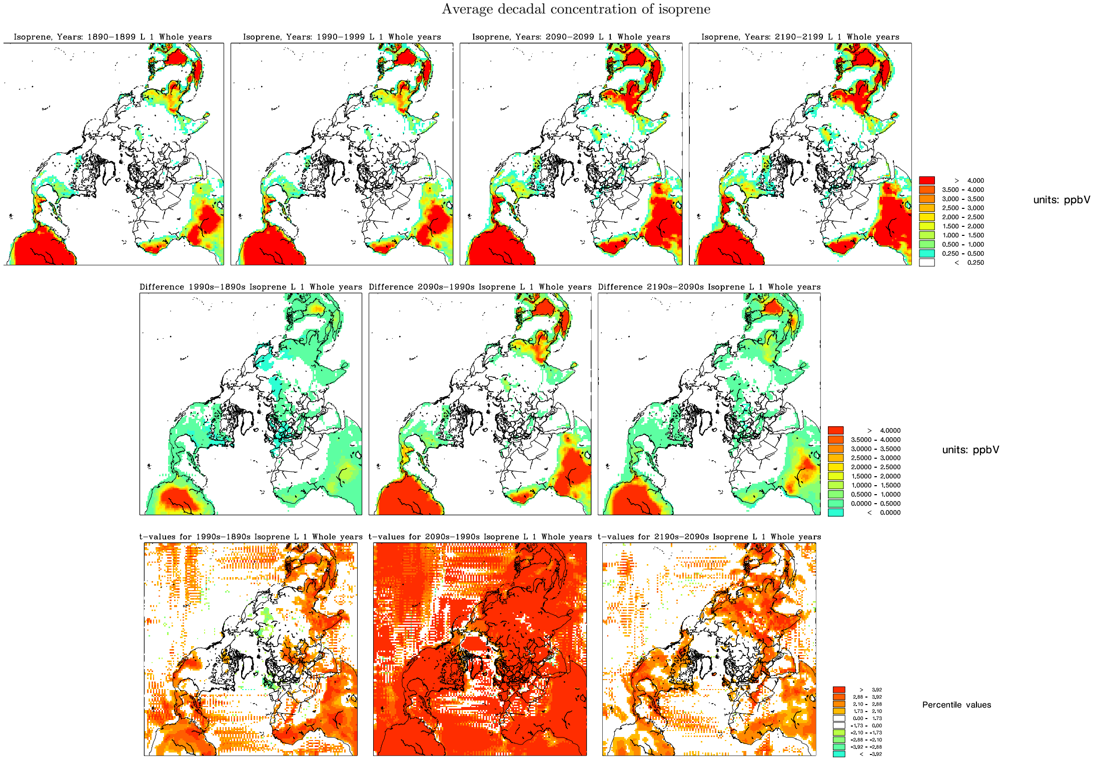

Fig. 6. Average decadal concentration of isoprene (C_5H_8) in the lowest model layer in ppbV. Setup the same as Fig. 1.

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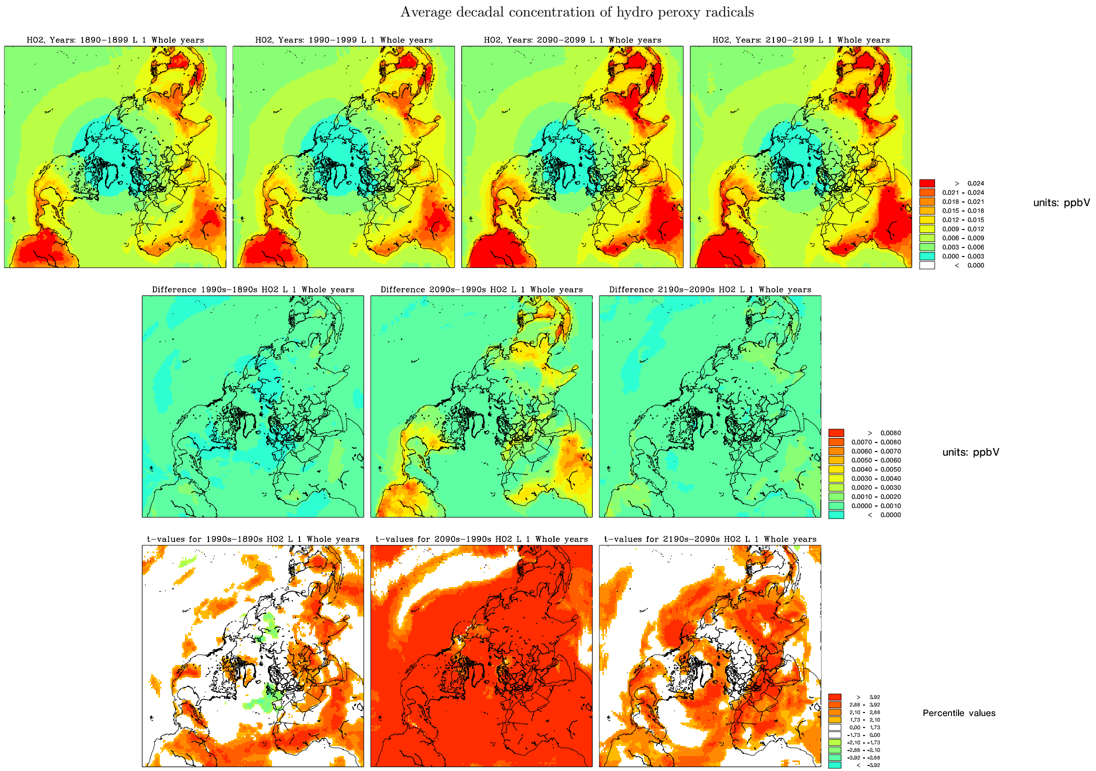

Fig. 7. Average decadal concentration of hydroperoxy radical (HO_2) in the lowest model layer in ppbV. Setup the same as Fig. 1.

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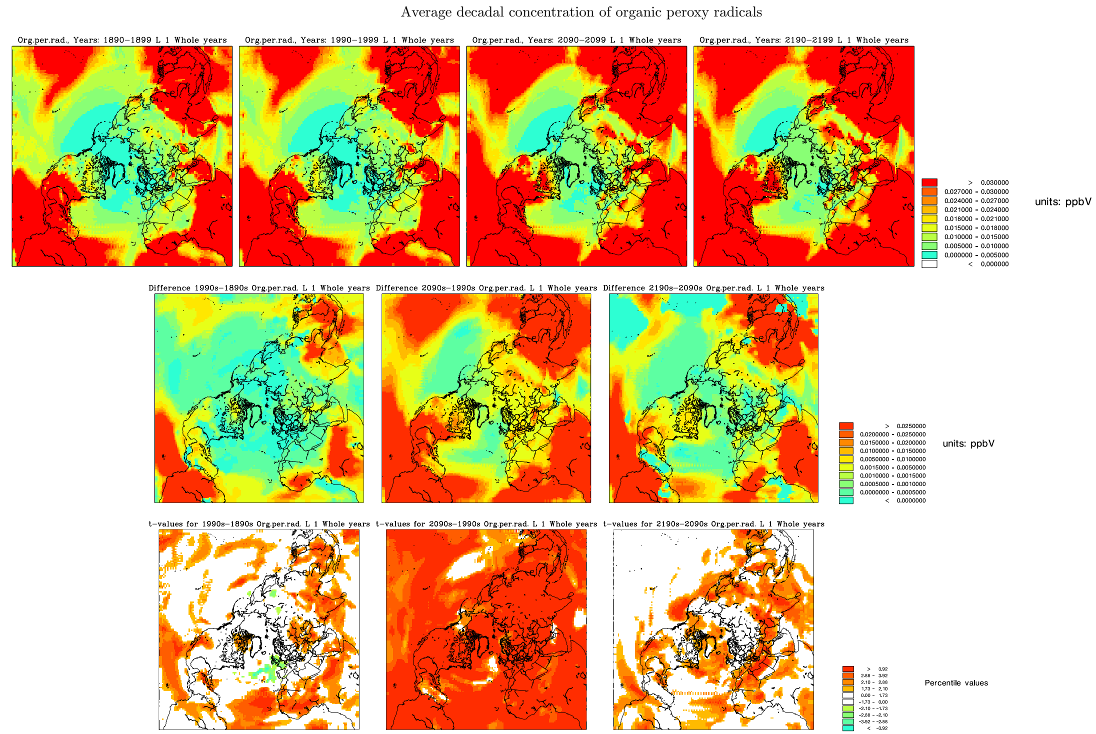

Fig. 8. Average decadal concentration of organic peroxy radicals in the lowest model layer in ppbV. Setup the same as Fig. 1.

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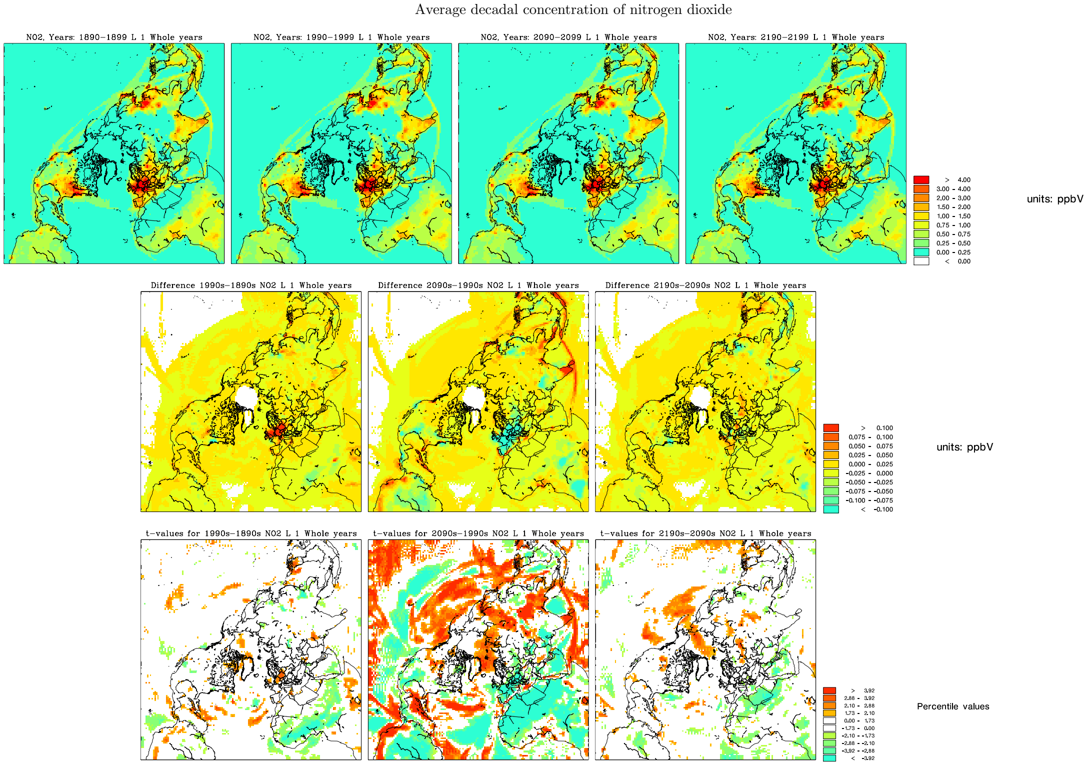

Fig. 9. Average decadal concentration of nitrogen dioxide (NO_2) in the lowest model layer in ppbV. Setup the same as Fig. 1.

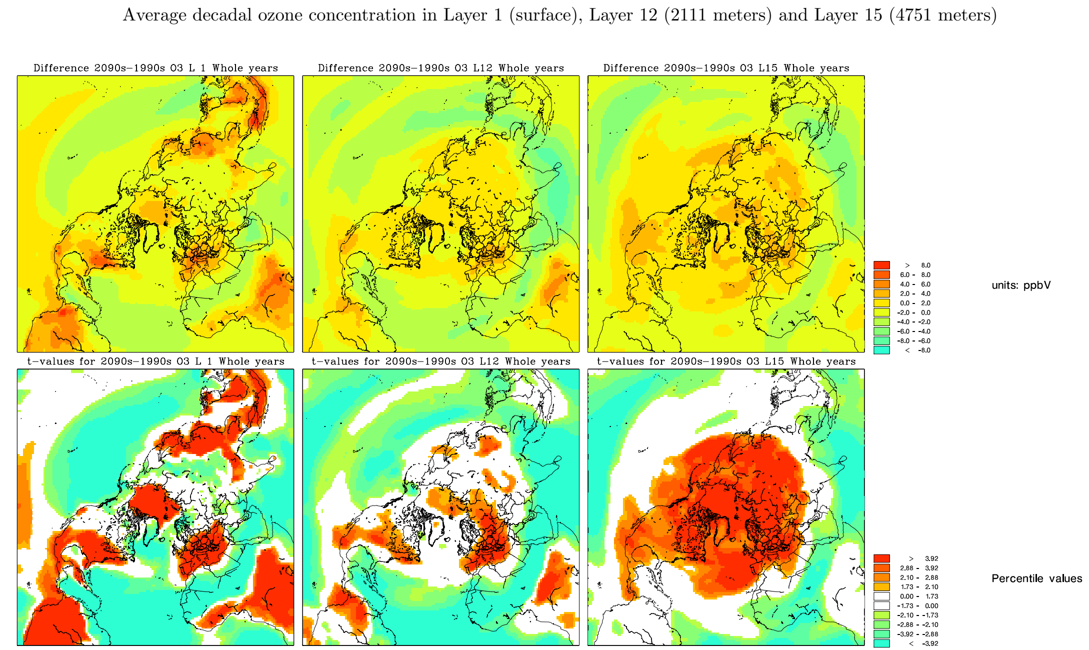

Fig. 10. Changes in ozone concentration in ppbv between the 1990s to the 2090s and the significance of these changes the surface layer, layer 12 (∼2100 m) and layer 15 (∼4750 m).