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Tropospheric ozone and its precursors: reanalysis over the past 45 years and projections for 2030

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Abstract
Within the European Union project RETRO a reconstruction is made of the chemical composition of the troposphere since 1960 with the focus on ozone and its precursors. Long-term simulations are currently being performed using several global chemistry-transport and chemistry-climate models. Meteorological input is provided by the 45-year reanalysis ERA-40, which has recently been completed by the European Centre for Medium-Range Weather Forecasts (ECMWF). Changes in anthropogenic emissions are taken into account by a new inventory covering the whole period; estimates of the interannual variability of biomass burning and biogenic emissions are also included in the models. In this paper we present some preliminary results of this chemical reanalysis, which eventually should include changes in tropospheric ozone and precursor gases as well as in the associated radiative forcing. The results of the reconstruction will also be discussed in the context of future changes to be expected on the basis of the '2030 Photcomp' experiment, which has recently been performed as a contribution to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change (IPCC). In this experiment, projections are made of the air quality and the tropospheric composition for the year 2030 based on the new International Institute for Applied Systems Analysis (IIASA) emission scenarios 'Current Legislation' and 'Maximum Technically Feasible Reduction' as well as the IPCC Special Report on Emission Scenarios (SRES) scenario A2.

Keywords: *Tropospheric composition, chemical reanalysis, projections*

1. Projections for 2030

In preparation for the Intergovernmental Panel on Climate Change (IPCC) Fourth Assessment Report (AR4), 25 state-of-the-art atmospheric chemistry models calculated changes in the tropospheric composition and resulting climate impacts between 2000 and 2030. For 2030, which is the policy-relevant intermediate future, the models applied a new set of emission scenarios that were recently developed at the International Institute for Applied Systems Analysis (IIASA) and described by Dentener et al. (2004). The Current Legislation (CLE) scenario reflects the current perspectives of individual countries on future economic development and takes into account the anticipated effects of presently decided emission control legislation by national and international governments; the Maximum Technically Feasible Reduction (MFR) scenario outlines the scope for emission reductions

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offered by full implementation of the presently available emission control technologies, while maintaining the projected levels of economic activity. Both scenarios differ substantially from the SRES scenarios. The IIASA scenarios for 2030 were therefore contrasted with the more pessimistic SRES A2 scenario. The offline chemistry-transport models (CTMs) performed four different simulations: three scenario runs for 2030 and one baseline run for the reference year 2000. In all cases the CTMs were driven with meteorological data representative of the year 2000. To quantify the impact of climate change on tropospheric composition by 2030, the chemistry-climate models (CCMs) in addition performed a simulation using the CLE emission scenario in combination with a climate appropriate for 2030. All of the CCMs were configured as atmosphere-only models, with prescribed sea surface temperatures (SSTs) providing the lower boundary condition over the oceans. Global annual mean surface warming between 2000 and 2030 varied from 0.3 K to 0.9 K. More details about the range of the applied climate scenarios are given by Stevenson et al. (2005).

Evaluation of the 2000 reference simulation and analysis of the 2030 scenario runs are investigated in a series of papers. A detailed comparison of tropospheric NO_2 column densities with satellite observations from the Global Ozone Monitoring Experiment (GOME) for the year 2000 shows general agreement of spatial patterns and seasonal cycles (T. van Noije et al., manuscript in preparation). On average the models tend to underestimate the column densities in the major industrialized regions of the world. This is especially evident over China, where the annual mean as well as the seasonal cycle are underestimated. Quantitative differences can also be observed over the biomass burning regions. In this exercise the biomass burning emissions are taken from the Global Fire Emissions Database (GFED) averaged over the years 1997–2002. Given the relatively large uncertainties in biomass burning emission inventories and the fact that a 5-year average was applied, the seasonality and patterns of NO_2 from biomass burning are reasonably well represented. An additional conclusion of this intercomparison study is that relatively large discrepancies exist between the different GOME retrievals (T. van Noije et al., paper presented at the European Geophysical Union General Assembly 2005). The evaluation for the year 2000 also includes a comparison with measurements of nitrogen deposition rates (F. Dentener et al., manuscript in preparation) as well as with tropospheric and surface ozone levels.

The tropospheric ozone budgets and atmospheric methane lifetimes from the various scenario simulations are discussed by Stevenson et al. (2005). The ensemble mean changes in tropospheric ozone burden between 2000 and 2030 range from a 5% decrease (MFR), through to a 6% increase (CLE), to a 15% increase (A2). The standard deviation between the individual models associated with these values is about $\pm 25\%$. As an example, we show in Figure 1 the corresponding zonal annual mean ozone levels calculated with the chemistry-transport model TM4. Combining both ozone and methane changes, the different scenarios give ensemble mean radiative forcings of -40, 180 and 300 mW m^{-2}, compared to a CO_2 forcing over the same period of 800–1100 mW m^{-2}. The model sensitivity of ozone to imposed climate change varies between models, but modulates zonal mean mixing ratios by ± 5 ppb via a variety of feedback mechanisms, in particular those involving water vapour and stratosphere–troposphere exchange. The projected changes in the annual mean surface ozone distribution calculated with TM4 for the different scenarios are displayed in Figure 2.

By 2030, the ensemble mean global surface ozone level is estimated to increase by 1.5 ± 1.2 ppbv (CLE) or 4.3 ± 2.2 ppbv (A2). Only the progressive MFR scenario will reduce surface ozone by 2.3 ± 1.1 ppbv. Surface ozone is analysed in detail by K. Ellingsen et al. (manuscript in preparation). Special attention is given to relate the projected future changes

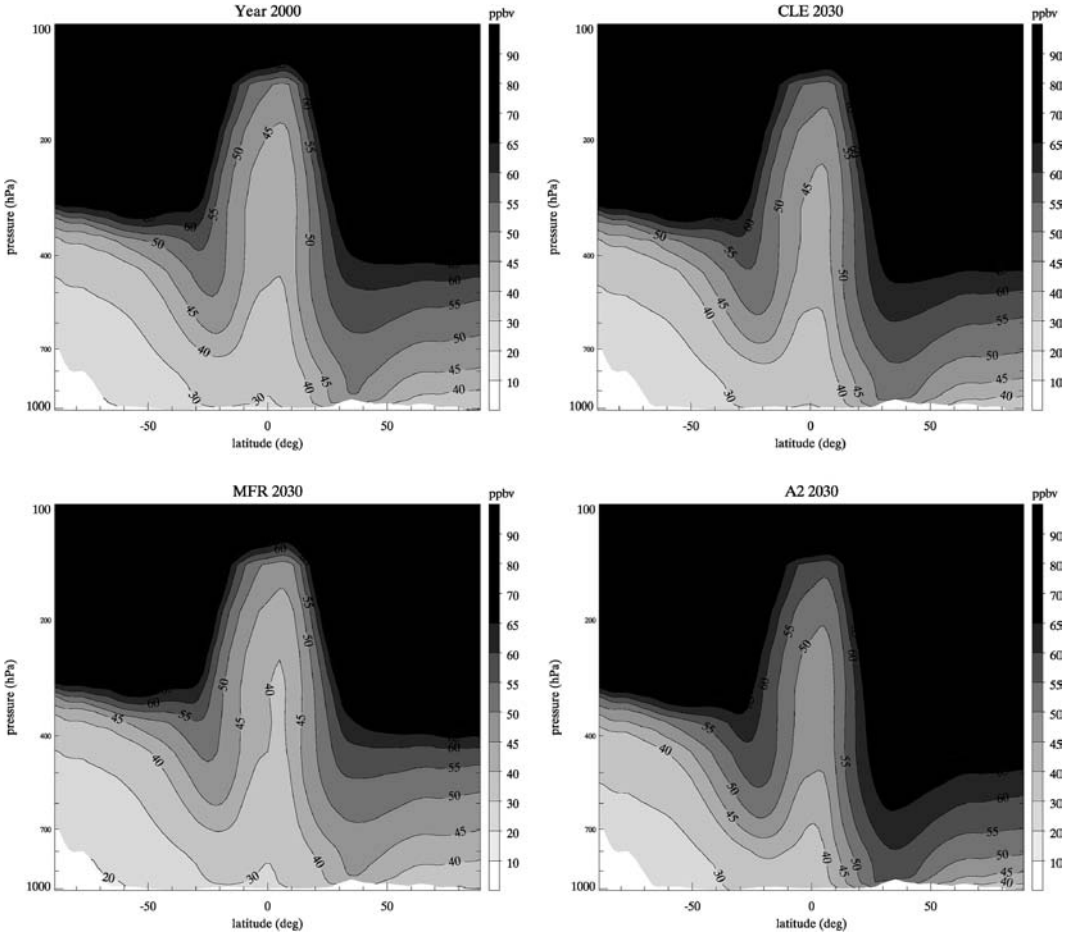

Figure 1. Annual zonal mean ozone (ppbv) for the year 2000 together with the projections for 2030 based on the Current Legislation (CLE), Maximum Technically Feasible Reduction (MFR) and SRES A2 emission scenarios.

in surface ozone to the different air quality standards established by the various national or international governments. The impact of increased ship emissions is currently under investigation by V. Eyring et al. (manuscript in preparation).

2. Chemical reanalysis

Some of the European models that have participated in the '2030 Photcomp' experiment described above, are also involved in a joint effort within the European Union project RETRO to make a reconstruction of the tropospheric composition for the period 1960–2000. Again chemistry-transport as well as chemistry-climate models are involved in the exercise. Meteorological input is provided by the ERA-40 reanalysis from the European Centre for Medium-Range Weather Forecasts (ECMWF). In many respects the ERA-40 reanalysis, covering the 45-year period from September 1957 to August 2002, has been shown to be of superior quality compared to earlier reanalysis datasets. However, serious problems of bias and inhomogeneities related to changes in the observing system have been

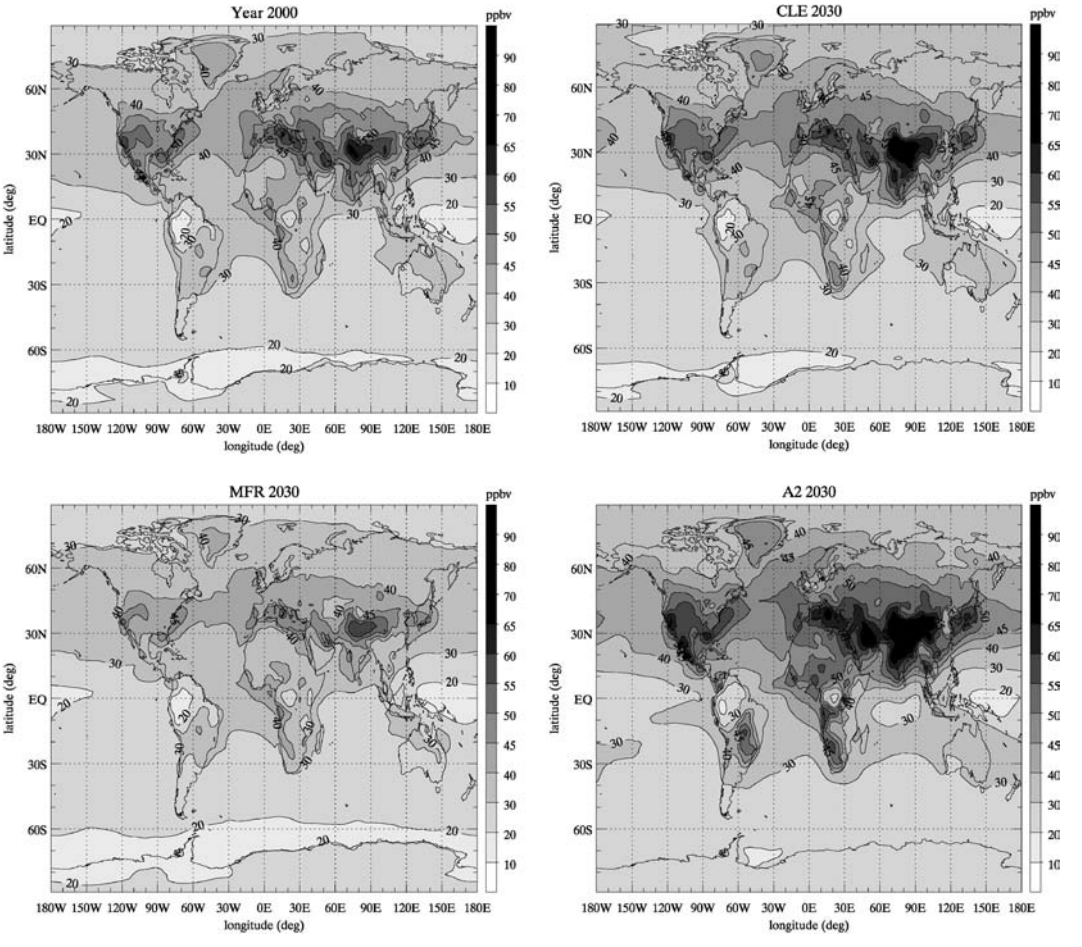

Figure 2. Annual mean surface ozone (ppbv) for the year 2000 together with the projections for 2030 based on the Current Legislation (CLE), Maximum Technically Feasible Reduction (MFR) and SRES A2 emission scenarios.

identified (van Noije et al. 2005; and references therein). In particular the Brewer–Dobson circulation in ERA-40 is strongly enhanced compared with other meteorological data, leading to an overestimation of stratosphere–troposphere exchange in atmospheric chemistry models (van Noije et al. 2004, 2005). For the chemical reanalysis calculations planned within RETRO this challenges the possibility of accurately representing the temporal variations and possible trends in the contribution of stratospheric ozone to the tropospheric ozone budget.

Concerning historic trends and variability in the emissions to be used in the long-term reanalysis calculations, we are in a much better position. Anthropogenic emissions covering the period 1960–2000 on a monthly basis have been compiled (Pulles et al. 2005); estimates of emissions from biomass burning covering the period of interest have also been produced within the project (Schultz et al. 2005). Long-term simulations have recently been started and the first results have been obtained. As an example Figure 3 shows the annual mean ozone field for the year 1960 calculated with the TM4 model. Extensive validation and analysis of the results will be presented in forthcoming publications.

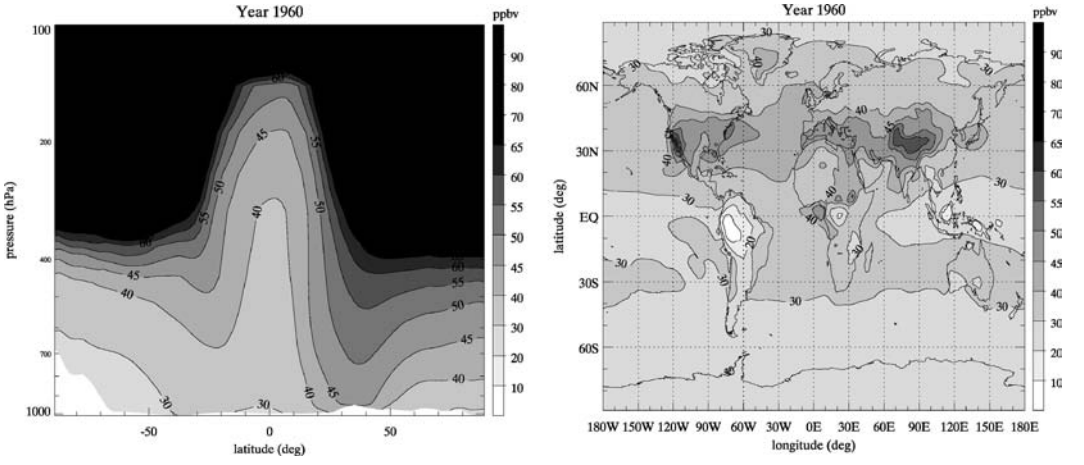

Figure 3. Zonal mean (left) and surface (right) ozone (ppbv) for the year 1960, calculated on the basis of the ERA-40 reanalysis.

3. Conclusion

New emission datasets have been compiled, which, together with the ERA-40 reanalysis data, provide the input for long-term simulations that aim to make a reconstruction of the tropospheric composition during the last four decades of the twentieth century. Projections of the global air quality for the next generation show the importance of enforcing worldwide air quality legislation. Non-attainment of these policy objectives would lead to adverse impacts on air quality as well as radiative forcing of climate.

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