

Effect of climate change on tropospheric oxidants



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In an Earth System Model, interactions between components such as the carbon cycle, chemistry, and aerosols are important. The UK Chemistry and Aerosols (UKCA) project aims to provide a state-of-the-art chemistry-aerosol model to enable these interactions to be investigated. Here, we describe the tropospheric chemistry component of UKCA, which has been coupled to the Hadley Centre's Earth System climate model, HadGEM. Earth System interactions include coupling between chemistry and vegetation, hydrology, radiation, and aerosols. Here, we present an evaluation of UKCA against observations. Results from the coupled model showing the role of climate change on tropospheric oxidants will also be shown.

1. HadGEM/UKCA Model Description

- 3.75 x 2.75 horizontal resolution and 38 vertical levels up to 40 km.
- Semi-Lagrangian transport scheme with a dynamical time step of 30 minutes.
- Tropospheric chemistry scheme involving 26 tracers and 46 species (NO_x-HO_x-CH_x-O_x chemistry with some non-methane hydrocarbons) and a chemical time step of 5 minutes. Stratospheric O₃ prescribed. Includes wet and dry deposition schemes.
- Interactive/prescribed CH₄ emissions from wetlands and interactive lightning emissions.
- Prescribed aircraft NO_x emissions and other surface emissions.
- Tabulated photolysis rates or interactive photolysis scheme (Fast- λ).
- Prescribed SSTs from AMIP-II or output from a 2100 SRES A2 simulation.

2. Model Evaluation in Present-Day Climate: Surface Comparison

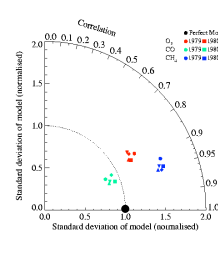
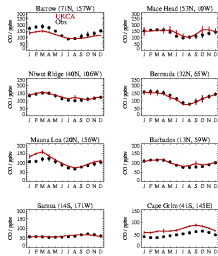


Figure 1: Comparison of climatological modelled (red) and observed (black) surface concentrations of carbon monoxide at a subset of CMDL monitoring sites.

Figure 2: Taylor Diagram showing the overall performance of UKCA against observed surface concentrations of CO, CH₄, and O₃ at 12 CMDL sites.

Figure 1 shows a comparison of the seasonal cycle of modelled and observed monthly mean surface concentrations of CO at a subset of CMDL monitoring sites. It indicates that UKCA captures both the seasonal cycle and the absolute concentrations very well although there is a tendency to overestimate Southern Hemisphere concentrations. In order to make the comparison more quantitative, the use of Taylor Diagrams (Taylor 2001) is adopted. Figure 2 shows such a plot, where each point represents the comparison of one year's model output against the monthly mean observations of O₃, CO, and CH₄ from 10 CMDL monitoring sites in terms of their correlation, their root-mean-square difference, and the ratio of their variances. The points for all 3 species lie reasonably close to the 'perfect model' position, indicating that UKCA is simulating surface concentrations quite well.

3. Model Evaluation in Present-Day Climate: Ozonesonde Comparison

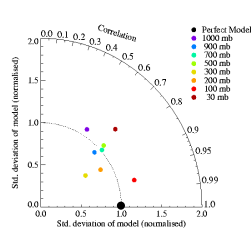
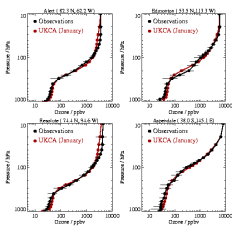


Figure 3: Comparison of climatological modelled (red) and observed (black) ozone profiles at a number of ozonesonde stations from Logan et al. 1999

Figure 4: Taylor Diagram showing the overall performance of UKCA against observed ozone profiles at ~40 worldwide sites from Logan et al. 1999

Figure 3 shows a comparison of modelled and observed vertical profiles of ozone for January at a subset of worldwide sites from Logan et al. 1999. It indicates that UKCA can model the shape and absolute concentrations of the vertical profiles very well when stratospheric ozone is prescribed 3 km above the tropopause. A more quantitative comparison of modelled and observed ozone on different pressure surfaces is represented by the Taylor diagram in Figure 4. It suggests that the best performing pressure surface (shown in red) is 100 mbar. However, all pressure surfaces perform well with correlation coefficients above 0.6 with standard deviations from the model close to that of the observations.

References:

- Logan, J. A. et al., Trends in the vertical distribution of ozone: A comparison of two analyses of ozonesonde data, *J. Geophys. Res.*, 104 (D21): 26373-26399, 1999.
 Taylor, K.E., Summarizing multiple aspects of model performance in a single diagram, *J. Geophys. Res.*, 106 (D7): 7183-7192, 2001.

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4. Impact of Climate Change on Tropospheric Ozone and Hydroxyl

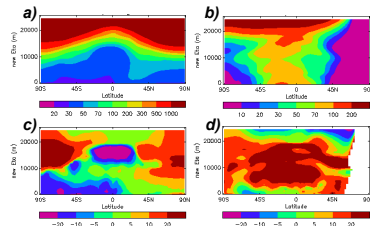


Figure 5: Zonal mean distribution of a) ozone and b) hydroxyl concentrations in Dec, Jan, and Feb from the present-day simulation. Relative difference in c) ozone and d) hydroxyl due to climate change (2100 SRES A2).

Figures 5a and 5b show the zonal mean distribution of ozone and hydroxyl for December, January, and February (DJF) from a present-day simulation. Figures 5c and 5d show the relative difference due to a change in climate to 2100 using the SRES A2 scenario. The climate change experiment did not include any changes in surface or aircraft emissions but did include a change in the prescribed stratospheric ozone concentrations.

Ozone in the lower stratosphere during both seasons shows an increase of over 20% in both hemispheres, but particularly in the Southern Hemisphere; this increase is attributed to changes in the prescribed stratospheric concentrations in line with decreasing concentrations of CFCs. In the troposphere, the global annual tropospheric ozone burden is unchanged between the two simulations. However, significant changes occur both seasonally and regionally. For example, there is a marked decrease in Southern Hemisphere concentrations at the surface and in the lower troposphere during DJF. A budget analysis suggests that the decrease is due to greater net chemical loss of ozone because of increased water vapour concentrations. In the Northern Hemisphere, ozone increases are evident at the surface but the greatest increase is in the mid and upper troposphere during DJF; this can probably be attributed to the higher stratospheric ozone concentrations and the increased mass exchange between the troposphere and stratosphere expected in the 2100 model simulation. In the tropics, the most significant change is in the UTLS region, where ozone concentrations decrease by greater than 20% during the DJF time period.

Where surface and lower tropospheric ozone concentrations increase in the Northern Hemisphere during DJF in the 2100 simulation, concentrations decrease in the JJA time period (not shown). Similarly, the change is reversed in the Southern Hemisphere, where ozone increases slightly at these latitudes during JJA in the 2100 simulation relative to the present day.

For the hydroxyl radical, concentrations increase almost globally and in both seasons in the 2100 atmosphere. This is due to increased production of OH from the water vapour increases associated with warmer temperatures. There is a corresponding decrease in the global annual methane lifetime against tropospheric OH loss from 9.55±0.02 years to 7.67±0.08 years.

A tropospheric chemistry scheme, developed as part of the UKCA project, has successfully been coupled to the Hadley Centre's Earth System Climate Model, HadGEM. Evaluation of modelled present-day concentrations has been carried out against climatological surface and profile observations and with the use of Taylor Diagrams. This indicates that the tropospheric version of UKCA is performing well. In addition, a climate change experiment has been performed for the year 2100, using the SRES A2 scenario. Strong regional and seasonal differences in ozone and hydroxyl are evident and further analysis is required to attribute these changes to changes in temperature, specific humidity, CO₂ concentrations, lightning emissions and/or stratosphere-troposphere exchange of ozone.