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Influence of emissions, climate and the stratosphere on tropospheric ozone

Science Report: SC010040/SR

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This report is the result of research commissioned and funded by the Environment Agency's Science Programme.

Published by:

Environment Agency, Rio House, Waterside Drive, Aztec West,
Almondsbury, Bristol, BS32 4UD
Tel: 01454 624400 Fax: 01454 624409
www.environment-agency.gov.uk

ISBN: 978-1-84432-821-5

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September 2006

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Dissemination Status:

Publicly available / released to all regions

Keywords:

Tropospheric ozone, chemistry-climate modelling, climate-chemistry, air quality, trace gas emissions, global warming potentials, climate change, trends.

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Science Project Number:

SC010040

Product Code:

SCHO0907BNGN-E-P

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Steve Killeen

Head of Science

Executive Summary

Tropospheric ozone is a ubiquitous air pollutant and the third largest greenhouse gas contributor to radiative forcing of climate change. Global models provide insight into the past, present and possible future spatial and temporal distributions of tropospheric ozone. This study used global models to assess the influence on tropospheric ozone of changes in anthropogenic activities (in particular, emissions of ozone precursors), and the potential consequences of the associated human-driven climate change.

Model results suggest that over the time period up to 2030, anthropogenic ozone precursor emissions will be the dominant control on ozone concentrations, and that there are clear benefits for air quality and climate change to following an aggressive global emissions reduction strategy for NO_x , CH_4 , CO and VOCs . Policymakers should vigorously pursue such emissions reductions; they should also ensure that all emissions inventories are as accurate as possible.

Up to 2030, models suggest that climate change is likely to have relatively small (although highly uncertain) impacts on tropospheric ozone globally, but potentially important impacts in some regions. Higher water vapour concentrations, associated with a warmer atmosphere, will tend to reduce ozone across most of the remote troposphere, but will enhance ozone in more polluted regions. Models predict that climate change will drive increases in the downwards transport of stratospheric ozone, and in some parts of the troposphere, particularly the upper troposphere, this counteracts the reductions in tropospheric ozone from higher water vapour. These and other consequences of climate change, including changes in emission sources affected by climate, such as biogenic isoprene and lightning NO_x , need to be better understood.

Over Europe, if land-based anthropogenic emissions continue to be reduced, as seems likely, the relative importance of external influences on ozone will increase. More than half of the tropospheric ozone over the UK is estimated to originate outside the European troposphere, with significant contributions from North America, Asia, the stratosphere, and from ship emissions. Ship NO_x emissions are poorly characterised, but are growing rapidly and are largely unregulated. This should be a major concern for policymakers.

Ozone precursor emissions have wider impacts on tropospheric chemistry and climate. In addition to producing ozone, they also affect hydroxyl radical (OH) and hence the residence time and concentration of methane, the second most important greenhouse gas contributor to climate change. Global model experiments have estimated the global warming potentials (GWPs) of several ozone precursors, including NO_x . NO_x generally has a negative GWP, with the reduction in CH_4 larger than the increase in O_3 . GWPs for NO_x are strongly dependent on emission location and season, and there is some uncertainty in the GWP for aircraft NO_x , with model estimates ranging from large positive values to near zero. These discrepancies stem from uncertainties in model formulation, which need to be better understood before it is realistic for policymakers to incorporate ozone precursor emissions in future climate treaties.

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1 Introduction

1.1 Tropospheric ozone

Climate change and air pollution are issues that are closely linked, not least by the trace gas ozone (O₃). Ozone concentrations in the troposphere are thought to have increased significantly since pre-industrial times, as a direct result of human activities (Volz and Kley, 1988; Staehelin *et al.*, 1994; Lamarque *et al.*, 2005). Tropospheric O₃ now represents the third largest greenhouse gas contribution to radiative forcing of climate change, with a forcing equivalent to about 24 per cent of that from carbon dioxide (Ramaswamy *et al.*, 2001). Ozone is also a ubiquitous air pollutant, affecting the respiratory health of a large proportion of the world population (WHO, 2003), reducing the yields of staple agricultural crops such as wheat and soy-bean, and damaging natural ecosystems (Emberson *et al.*, 2003; Wang and Mauzerall, 2004).

There are two sources of tropospheric ozone: transport from the overlying stratosphere and *in situ* chemical production. Ozone production takes place when carbon monoxide (CO) and volatile organic compounds (VOCs) are photo-oxidised in the presence of nitrogen oxides (NO_x, that is, NO and NO₂) (Crutzen, 1974; Liu *et al.*, 1980; Atkinson, 2000). The ozone budget is closed by two loss processes: dry deposition to the Earth's surface, and chemical destruction. Ozone destruction occurs mainly via reactions with water vapour and peroxy and hydroxyl radicals (HO_x, that is, HO₂ and OH). Integrated over the whole troposphere, chemical production and loss rates are several times larger than the influx from the stratosphere and the surface deposition flux.

The main ozone precursors (CO, hydrocarbons and NO_x) are emitted as byproducts of human activities, as well as having significant natural sources. These anthropogenic and natural emissions also influence HO_x, affecting ozone destruction. The rapid growth in anthropogenic emissions following industrialization has been largely responsible for the increase in tropospheric ozone, and has been the subject of intense study (Levy *et al.*, 1985; Crutzen and Zimmerman, 1991; Lelieveld and Dentener, 2000; Lamarque *et al.*, 2005). Interest has more recently turned to the potential influence of climate change on future levels of ozone (Johnson *et al.*, 1999, 2001; Isaksen *et al.*, 2003; Stevenson *et al.*, 2005; Murazaki and Hess, 2006; Hauglustaine *et al.*, 2005). Warming will increase water vapour concentrations, and changes in temperature and water vapour will affect the reaction rates of many chemical conversions. Climate change may also alter global circulation dynamics, changing several processes that govern the distribution of tropospheric ozone, such as stratosphere-troposphere exchange, the distribution of convection, and ventilation of the boundary layer. Changes in climate could also affect many of the natural sources of trace gases, such as wetland CH₄ (Walter and Heimann, 2000; Gedney *et al.*, 2004; Shindell *et al.*, 2004), biogenic VOCs (Sanderson *et al.*, 2003a; Wiedinmyer *et al.*, 2005; Lathière *et al.*, 2005), lightning NO_x (Price *et al.*, 1997) and soil NO_x (Ganzeveld *et al.*, 2002).

Global atmospheric chemistry models currently generate our best estimates of tropospheric distributions of trace species such as ozone and the hydroxyl radical (OH). These models underpin our understanding of the oxidizing capacity of the atmosphere and radiative forcings arising from changes to methane (CH₄) and O₃ concentrations (for example, Prather *et al.*, 2001, 2003; Gauss *et al.*, 2003). As such, they are the best tools at our disposal for making quantitative future projections of the composition of the troposphere.

1.2 Report structure

This report summarises the major research activities carried out in a five-year fellowship (April 2001-March 2006) jointly sponsored by the Environment Agency and the Natural Environment Research Council (NERC).

Section 2 gives a brief outline of state-of-the-art in climate-chemistry modelling, along with a more detailed description of the main model used in this research. Typical experimental set-ups of the model are explained, to illustrate the capabilities and limitations of such experiments.

The main results of the modelling work are presented in Section 3, with a focus on understanding the various influences on tropospheric ozone, and, more generally, the oxidising capacity of the atmosphere (the key determinant of the lifetime of methane).

A significant subset of the work concerns background ozone levels in Europe, a major air quality issue. The report also offers some of the first estimates of global warming potentials (GWPs) for a variety of ozone precursors; GWPs are a necessary first step towards including ozone precursors in any future climate agreements.

In Section 4 the main conclusions are drawn together, while recommendations are presented in Section 5.

Five appendices are also included. The first lists all peer-reviewed publications resulting from the fellowship, together with their abstracts and, where available, a hyperlink to the full publication text. The second appendix lists conference, workshop and other scientific presentations made, again with links to the full presentations where available. Appendix 3 provides information on the teaching, supervision and public engagement carried out over the course of the fellowship. The fourth appendix lists other activities that have contributed to national and international atmospheric research, such as reviewing of papers and research proposals. Finally, the last appendix provides details of research proposals prepared over the last five years; two of these have resulted in funding.

2 Methods

2.1 Climate-chemistry models

Climate-chemistry models (CCMs) bring together the climate physics of general circulation models (GCMs) (such as radiative transfer, advection, mixing, cloud processes, surface-atmosphere interactions) with atmospheric chemistry (such as trace gas emissions, photochemical, gas-phase, aqueous-phase and heterogeneous reactions, and wet and dry deposition). CCMs are part of the progression in climate models towards a more complete representation of the climate system, and allow full spatial and temporal evolution of atmospheric composition based on prescribed emissions sources, rather than direct prescription of trace gas concentrations as boundary conditions (for example, from a simpler independent source). CCMs, in concert with new satellite observations of atmospheric composition (see Frankenberg *et al.*, 2005; van Noije *et al.*, 2006; Shindell *et al.*, 2006), provide us with the most accurate picture to date of the global distributions of key species. In addition, CCMs allow us to explore the factors that control past, present and future atmospheric composition, using idealised experiments.

2.2 Modelling approach

The model mainly used here was based on the Hadley Centre GCM, HadAM3, coupled to the tropospheric chemistry model STOCHEM. The HadAM3-STOCHEM climate-chemistry model (Sanderson *et al.*, 2003a, 2003b; Stevenson *et al.*, 2004, 2005) was used to carry out several series of numerical experiments. HadAM3 (Pope *et al.*, 2000) is a GCM describing the state of the atmosphere. Prescribed sea surface temperatures (SSTs) provide the lower boundary condition over the oceans. Over land, the MOSES2.2 surface exchange scheme is employed (Essery *et al.*, 2001), together with a prescribed seasonal vegetation distribution. HadAM3 was run at standard climate resolution: 3.75° longitude x 2.5° latitude, with 19 vertical levels, concentrated towards the surface, but extending upwards to around 10 hPa. The model time step was 30 minutes, with meteorological fields passed to STOCHEM every three hours. Some variants on this model have been used; for instance STOCHEM was used in conjunction with HadCM3, which includes an oceanic GCM sub-model, to simulate long-term (century-scale) climate evolution and variability (interannual to interdecadal) (see Johnson *et al.*, 2001, 2002; Doherty *et al.*, 2006; Derwent *et al.*, 2006a). Secondly, STOCHEM can be driven by meteorological analyses, rather than GCM output; this approach was used in Collins *et al.* (2002a, 2003) and Derwent *et al.* (2001; 2002a; 2004; 2006b; 2006c). Model versions with slightly different resolution and chemical schemes are also available (see below).

STOCHEM is a Lagrangian tropospheric chemistry-transport model, originally described by Collins *et al.* (1997), with subsequent major updates to chemistry (Collins *et al.*, 1999), convective mixing (Collins *et al.*, 2002b), vegetation emissions (Sanderson *et al.*, 2003a) and surface deposition (Sanderson *et al.*, 2003b). Tracers such as the short-lived radionuclide ²²²Rn are also included (see Robertson *et al.*, 2005). STOCHEM extends from the surface to around 100 hPa; this is approximately the level of the tropical tropopause, but is some way into the lower stratosphere at higher latitudes. The climate model that drives STOCHEM extends to higher levels (around 10 hPa for the 19-level version), to improve the representation of stratospheric circulation, and in particular stratosphere-troposphere exchange (STE). Versions with about 60 levels, usually used for stratospheric modelling, have also been coupled to STOCHEM, and these have been used in work where the

emphasis on the stratosphere is larger (Stevenson *et al.*, 2003b; also see Appendix 5, NERC-funded proposal on stratosphere-troposphere modelling).

The STOCHEM domain is typically divided into 50,000 equal mass air parcels, which are advected using winds from HadAM3, using a fourth order Runge-Kutta method. Every one-hour advection time step, winds are linearly interpolated to each parcel's position in the horizontal, using cubic interpolation in the vertical. A random walk component is added to simulate horizontal and vertical diffusion. Following each advection step, air parcels are mapped to a latitude-longitude grid of dimensions 5° x 5° with nine equally spaced vertical levels, of around 100 hPa in thickness. Each grid box contains, on average, two to three Lagrangian air parcels. To represent the deformation of air parcels, some inter-parcel mixing is implemented between air parcels within the same grid box. Air parcel concentrations are brought towards the mean value for the grid box. A more sophisticated inter-parcel mixing scheme, based on each parcel's nearest neighbours, has been developed but has not yet been fully implemented. Turbulent mixing in the boundary layer is achieved by randomly reassigning the vertical coordinates of air parcels over the depth of the layer. Convective mixing is described fully in Collins *et al.* (2002b), and uses 3-D convective mass fluxes.

The chemical scheme is described by Collins *et al.* (1999) and includes 70 species that take part in 174 photochemical, gas-phase and aqueous-phase reactions. The mechanism describes the tropospheric chemistry of CH₄, CO, NO_x, O₃, and 11 non-methane hydrocarbons (NMHC). There is no specific representation of stratospheric chemistry in this model version. Several aqueous-phase reactions and equilibria are included (see Stevenson *et al.*, 2003b); there is a simplified representation of heterogeneous reactions that convert NO_y (total oxidized nitrogen) to nitrate aerosol with a fixed global rate constant. The chemical time step is five minutes. The dry and wet deposition schemes are described in detail in Sanderson *et al.* (2003b) and Stevenson *et al.* (2003b), respectively. To represent the influx of stratospheric O₃ to the top of the model domain, we used vertical wind fields at 100 hPa coupled with an ozone climatology (Li and Shine, 1995). Similarly, we introduced an NO_y influx (as HNO₃), assuming a fixed mass ratio of N:O₃ of 1:1,000 (Murphy and Fahey, 1994). Model versions that include terpene emissions and a secondary organic aerosol chemistry scheme have also been developed (Derwent *et al.*, 2003), as have versions with ammonium nitrate aerosol (C. Johnson, personal communication, 2005), and methyl bromide (R. Derwent, personal communication, 2004).

Anthropogenic emissions typically use the EDGAR database (Olivier and Berdowski, 2001), whilst future scenarios are from IIASA (International Institute for Applied Systems Analysis; Dentener *et al.*, 2005) and IPCC SRES (Intergovernmental Panel on Climate Change Special Report on Emissions Scenarios; Nakicenovic *et al.*, 2000). Biogenic emissions of isoprene are distributed using spatial vegetation fields from the GCM land surface scheme, and are dependent upon temperature and photosynthetically available radiation (Sanderson *et al.*, 2003a). Interactive lightning NO_x emissions are based on the parameterizations of Price *et al.* (1997), and are distributed in the vertical using the profiles of Pickering *et al.* (1998).

The model is typically integrated for periods of between one and five years, preceded by a one-year spin-up. In a few studies, longer integrations of 40-110 years have been performed and used in multiple analyses (for example, Johnson *et al.*, 2001, 2002; Dentener *et al.*, 2005; Stevenson *et al.*, 2005; Derwent *et al.*, 2006a; Doherty *et al.*, 2006). One year of integration takes about 12 hours of real-time, using 36 processors on the massively parallel SGI Altix machine 'Newton' (<http://www.csar.cfs.ac.uk>). Computing resources for this project were provided as part of the NERC Universities Global Atmospheric Modelling Program (UGAMP), as the fellowship was half-funded by NERC. Part of the final year of the fellowship was devoted to exploring the possibility of running the model in a local computing environment, using powerful multi-processor Linux PCs. Initial results running HadAM3 suggest that this route should work and should approach the integration speeds of the supercomputing

environment, but without the attendant problems (queues, allocation of resources, changes in compilers and so on).

Many aspects of modelling have large uncertainties, especially those areas poorly constrained by observations. A single model may not exhibit the same sensitivities (for example, to changes in emissions or a climate variable such as temperature or humidity) as the real world. Modellers can go some way towards addressing these concerns by taking part in model intercomparisons, where several models perform essentially the same experiment, and results are then compared with both each other and observations. Much of the last two years of the fellowship were devoted to helping coordinate a major model intercomparison carried out under the umbrella of the EU network ACCENT (*Atmospheric Composition Change: a European Network of Excellence*). This project resulted in several important publications (Stevenson *et al.*, 2006; Dentener *et al.*, 2006a,b; van Noije *et al.*, 2006; Shindell *et al.*, 2006; Ellingsen *et al.*, 2006; Eyring *et al.*, 2007), with results fed directly into several chapters of the *Fourth Assessment Report of IPCC* (Forster *et al.*, 2007; Denman *et al.*, 2007; Meehl *et al.*, 2007).

3 Results and discussion

3.1 Response of ozone to emissions

The ACCENT exercise compared results from 26 differently formulated global models. Most of these models performed a year 2000 ‘base’ simulation and three 2030 scenarios, based on different emissions projections:

- a ‘central’ case (CLE: Current legislation, where current (year 2000) legislation on air quality in each of the world’s countries was assumed to be fully implemented);
- an ‘optimistic’ case (MFR: Maximum technologically feasible reductions, where all currently known emission reduction technologies would be brought to bear across the world);
- a ‘pessimistic’ case (SRES A2, where high economic growth and essentially no air quality legislation would be implemented).

The results allowed us to quantify inter-model and model-observation differences for the present-day atmosphere, together with inter-model and inter-scenario uncertainties for projections of tropospheric O₃ (Stevenson *et al.*, 2006; Ellingsen *et al.*, 2006), CO (Shindell *et al.*, 2006), NO₂ (van Noije *et al.*, 2006), as well as nitrogen and sulphur deposition (Dentener *et al.*, 2006a). Figure 3.1 presents multi-model ensemble mean and standard deviations for the annual mean simulated distribution of O₃ in the troposphere for year 2000. Typical inter-model uncertainties (expressed as one standard deviation) are about 15-30 per cent, with the largest values in the most remote parts of the troposphere (over the Antarctic and tropical oceans, and in the upper troposphere).

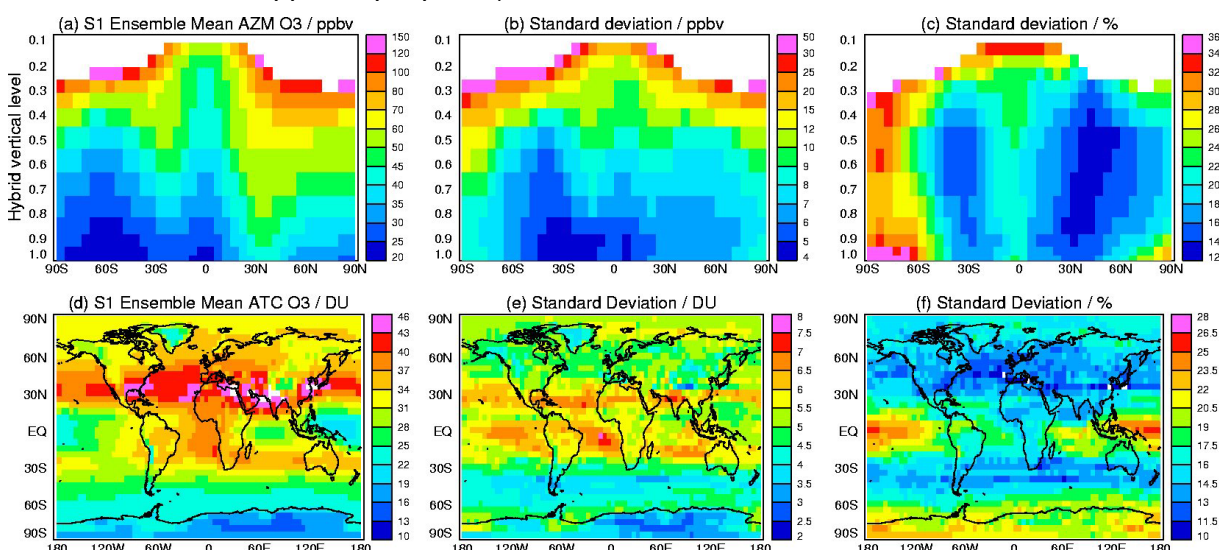


Figure 3.1: Modelled ozone from case S1 (year 2000): (a)-(c) are annual zonal means (AZM) (ppbv), (d)-(f) are annual tropospheric columns (ATC) (DU); (a) and (d) are ensemble means (all 26 models); (b) and (e) are absolute standard deviations; (c) and (f) are percentage standard deviations. To calculate the ensemble values, individual model results were interpolated to a common grid (5° x 5° x 19 levels) and masked at the chemical tropopause (O₃ = 150 ppbv). Taken from Stevenson *et al.* (2006).

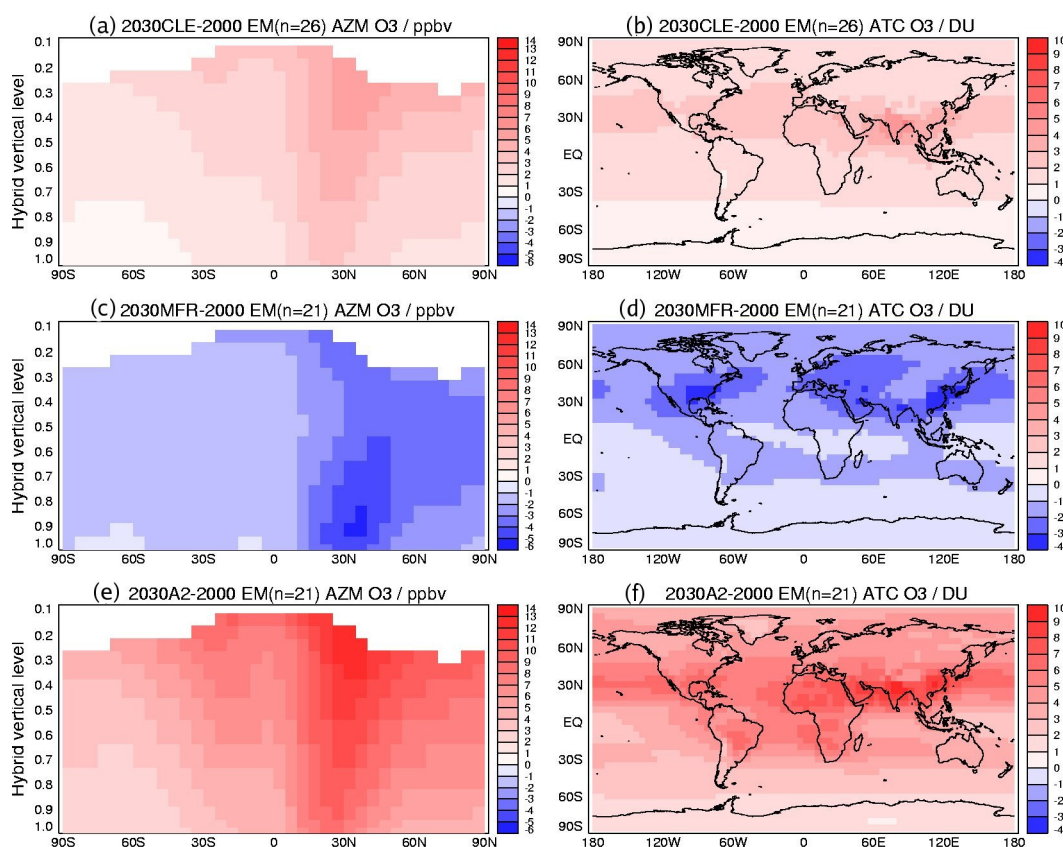


Figure 3.2: Changes in tropospheric ozone (2030-2000) under three emissions scenarios: (a, b) CLE, (c, d) MFR and (e, f) SRES A2. The first column (a, c, e) shows changes in annual zonal mean (AZM) ozone (ppbv); the second column (b, d, f) shows changes in annual tropospheric column (ATC) ozone (DU). Each plot is ensemble mean (EM) of several (n) models. Taken from Stevenson *et al.* (2006).

Figure 3.2 shows simulated changes in O_3 between 2000 and 2030 for the three scenarios. These changes also carry inter-model uncertainties of about 20-30 per cent. There are clear differences between the three scenarios, which have important policy implications for 2030 levels of background tropospheric O_3 .

Other work on emissions has focused on the roles of individual sectors (such as shipping, aviation) or regions (such as North America, Europe, Asia), and is reported briefly below.

3.2 Response of ozone to climate change and variability

It is crucial to place the projected O_3 changes related to changing anthropogenic emissions in their proper context. Ozone shows considerable variability, largely because its chemical lifetime is of the same order as the timescale of weather systems (days to weeks). We explored the influence of climate change on atmospheric composition in models by holding emissions constant in two simulations, but forcing climate in different ways (such as unforced versus the IS92a or SRES A2 scenarios, see Johnson *et al.*, 2001; Stevenson *et al.*, 2005). In addition, we looked at the influence of climate variability by analysing chemical variability in multi-decadal simulations (Johnson *et al.*, 2002; Stevenson *et al.*, 2005; Doherty *et al.*, 2006).

As part of the ACCENT intercomparison, 10 models conducted multi-annual (five to ten year) simulations of the central 2030 scenario (CLE) under both a year 2000 climate and a year

2030 climate (Stevenson *et al.*, 2006). By comparing these two cases, the influence of climate change could be determined. Figure 3.3 shows the multi-model ensemble mean results for O₃, together with results one standard deviation either side of this mean. From these results, it was not clear if climate change would have a positive or negative feedback on ozone. Two processes appeared to be dominant, but in opposite directions. Firstly, a warmer atmosphere would hold more water vapour and this, in general, would lead to enhanced O₃ destruction, particularly in the lower troposphere over the tropical oceans (STOCHEM-HadAM3 shows this effect strongly). Secondly, climate change would enhance the influx of O₃ from the stratosphere; this is most evident in the Northern Hemisphere mid-latitude upper troposphere. Other studies with STOCHEM-HadAM3 have also indicated important influences of climate on biogenic isoprene and lightning NO_x emissions (for example, Sanderson *et al.*, 2003a; Stevenson *et al.*, 2005). The modulation of O₃ induced by climate change in 2030, although rather uncertain, appears to be relatively small compared to the influence of changes in emissions (compare Figures 3.2 and 3.3).

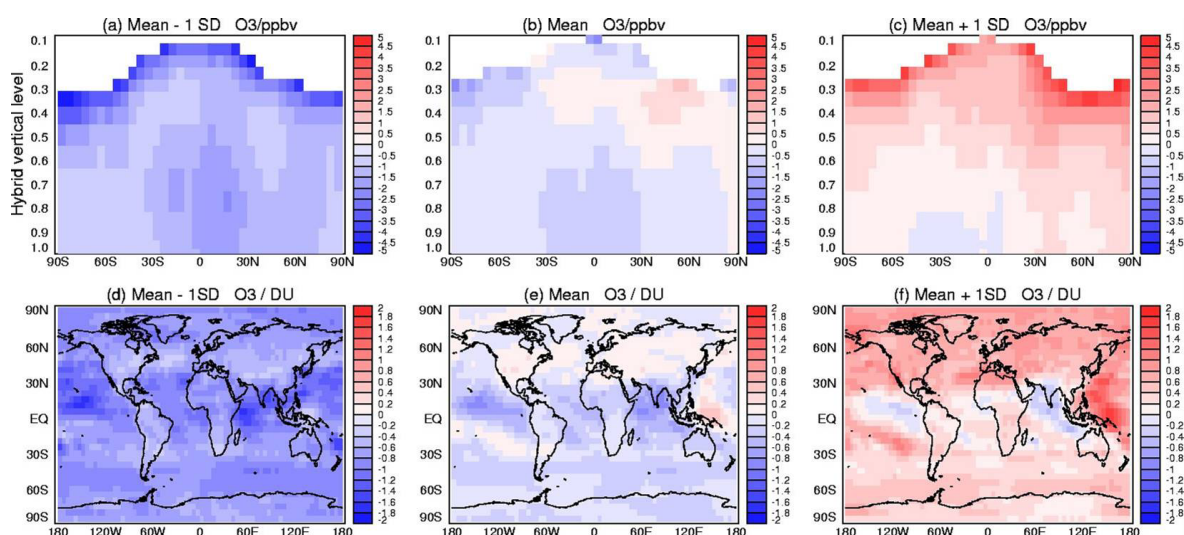


Figure 3.3: Ozone changes due to climate change: the difference between case S5 (CLE 2030, with a year 2030 climate) and S2 (CLE 2030, with a year 2000 climate). The standard deviations are large, and the modelled range of ozone change varies in sign. Panels (b) and (e) plot the ensemble mean change; (a) and (d) the ensemble mean minus one standard deviation; and (c) and (f) the ensemble mean plus one standard deviation. Taken from Stevenson *et al.* (2006).

The major mode of climate variability that influences global O₃ is El Niño-Southern Oscillation (ENSO) (Stevenson *et al.*, 2005; Doherty *et al.*, 2006). STOCHEM-HadAM3 simulated the observed pattern of variability (Ziemke and Chandra, 2003) quite well; however, the magnitude was somewhat underestimated. The mechanism through which climate would modulate O₃ would mainly be via changes in circulation and the associated distribution of convection. In addition, biospheric feedbacks such as biomass burning due to drought and changes in lightning NO_x emissions also exert local influences on O₃ (Doherty *et al.*, 2006).

3.3 European background ozone

The amount of ground-level O₃ at a given location is determined by a number of factors including local ones, such as nearby emission sources or the boundary layer height, but also much more distant processes such as long-range transport and stratosphere-troposphere

exchange. Global scale models, as described above, are useful tools to investigate the influence of these distant or ‘external’ influences on local air quality. For example, by labelling O₃ molecules with the region where they were formed in the model, we were able to attribute various sources to the background surface O₃ found over Europe (Derwent *et al.*, 2004). This study found that over the UK, of the typical annual mean 40 ppbv of ground-level O₃ observed, 15-20 ppbv was formed locally (over Europe), 8-9 ppbv came from the stratosphere, 10 ppbv from North America and 5 ppbv from Asia.

Derwent *et al.* (2005) assessed the contribution of ship emissions of NO_x and SO₂ to European O₃ and acid deposition. Ship emissions were found to be responsible for up to 6 ppbv O₃, with a maximum contribution on the Atlantic coastline of Western Europe (Figure 3.4). The current rapid and essentially unregulated rise in ship emissions, together with the likely implementation of land-based emission reductions, indicate that the proportion of O₃ derived from ship NO_x emissions is likely to rise significantly, and, as with other external influences, threatens to counteract local policies to improve air quality. Similar conclusions were found in a subsequent multi-model study (Eyring *et al.*, 2007).

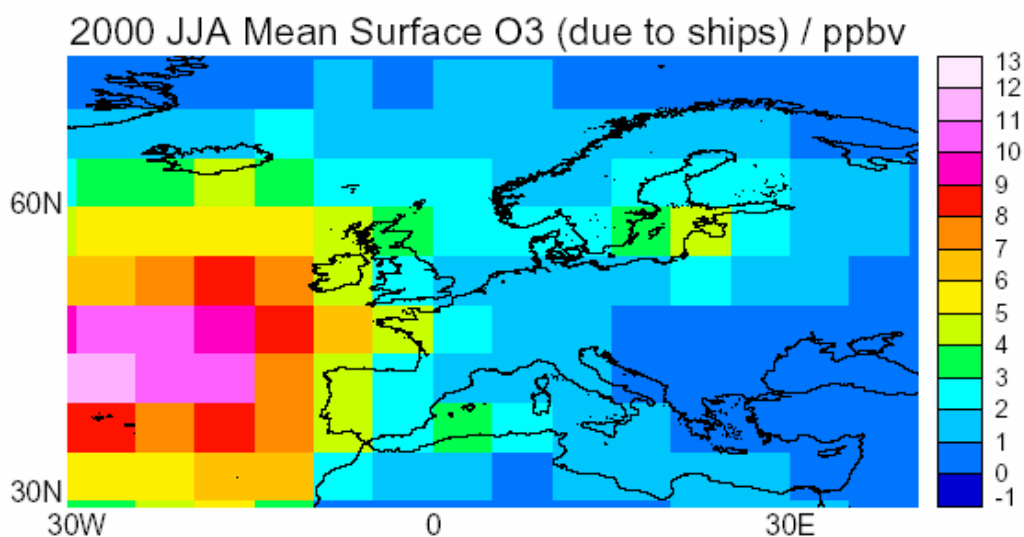


Figure 3.4: Contribution of ship emissions to surface level O₃ for the year 2000, in summer. Taken from Derwent *et al.* (2005).

Derwent *et al.* (2006a) used a range of model integrations to compare modelled O₃, CO and CH₄ at Mace Head with observed trends (Simmonds *et al.*, 2004), and to assess potential future trends under a variety of scenarios (Figure 3.5). In Figure 3.5, all the data have been smoothed with a 12-month running mean, to remove the seasonal cycle. Each scenario run was tuned to reasonably simulate the observed CH₄ growth rate in the time period 1990-1995; the fact that none of the scenarios convincingly simulated the next 10 years of observations shows that we do not fully understand the recent slowdown in the CH₄ growth rate; future projections of CH₄ may therefore be unreliable. Nevertheless, the model simulated interannual variations in CH₄ of approximately the correct magnitude (Johnson *et al.*, 2002). The model also had some difficulty in simulating observed CO, underestimating mean levels during the period 1990-2004 by about 20-30 ppbv (see also Dentener *et al.*, 2005).

In the ACCENT exercise, all models under-predicted Northern mid-latitude CO compared to MOPITT satellite observations (Shindell *et al.*, 2006), suggesting that CO emissions at these latitudes are underestimated, with the prime candidates being either Asian anthropogenic sources or boreal biomass burning. Figure 3.5 also shows that STOCHEM-HadAM3 underestimated interannual variations in CO, probably because it lacks year-to-year variations in biomass burning activity. Finally, the lower panel of Figure 3.5 shows observed and simulated trends in O₃. None of the simulations matched the large observed increase in O₃ partly because, as for CO, recent years have seen high levels of boreal biomass burning, and this is not accounted for by the model. However, even without interannual variations in burning, the model appears to show too much temporal variability. The high level of both observed and simulated variability makes it difficult to see clear trends in O₃. On the other hand, a coarse resolution global model, not driven by analysed meteorology, should perhaps not be expected to perfectly simulate observations at a single point.

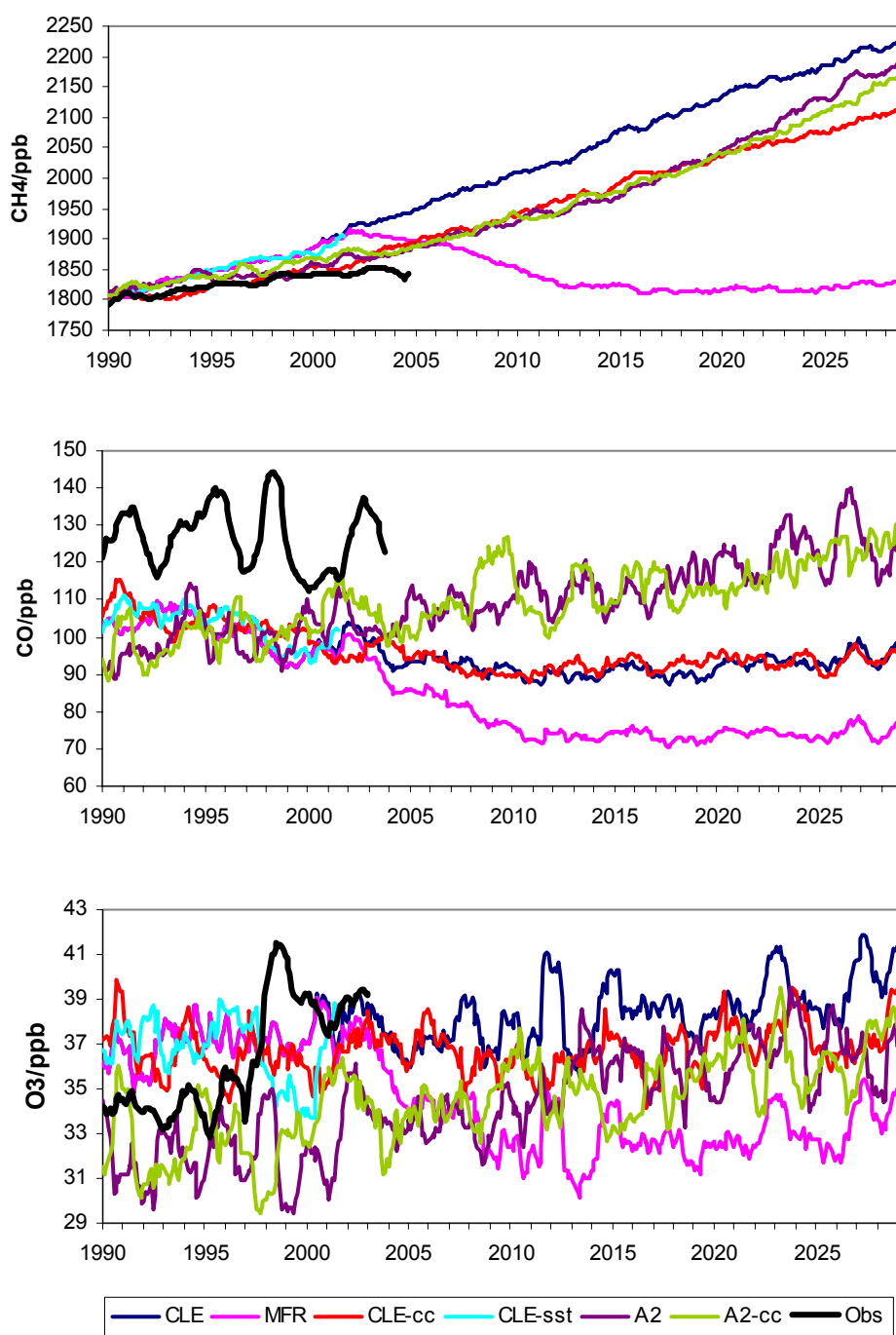


Figure 3.5: Twelve-month running mean of observed (1990-2003) and modelled (1990-2030; except CLE-sst: 1990-2002) for CH₄ (upper panel); CO (middle panel); and O₃ (lower panel) at Mace Head. From Derwent *et al.* (2006a).

3.4 Global warming potentials of ozone precursors

Models of atmospheric chemistry are the only available tools to estimate global warming potentials (GWPs) of O₃ precursors NO_x, CO, and VOCs. Emissions of these gases generate a complex array of effects that cascade through the tropospheric chemical system (Wild and Prather, 2000). Idealised pairs of numerical experiments were performed, with the first experiment acting as a control and the second being identical except for a short-lived pulse

of trace gas emissions. By differencing the results from these pairs of experiments, the chemical perturbation introduced by the emission pulse became apparent, as follows.

All of the O₃ precursors generated a short-lived O₃ enhancement, as might be expected. This extra O₃ generated a positive radiative forcing. However, the pulse also perturbed OH levels, disturbing CH₄ oxidation rates and hence CH₄ concentrations. Methane is the longest-lived component in the tropospheric chemical system, and CH₄ perturbations decay with a characteristic timescale of about 11 years (Wild *et al.*, 2001; Derwent *et al.*, 2001; Collins *et al.*, 2002a; Stevenson *et al.*, 2004; Derwent *et al.*, 2006c). This slow-decaying CH₄ perturbation maintains a long-lived perturbation to the whole system, including O₃. Calculation of the overall climate impact of an emission required the time-integrated effects of all the radiatively active gases, that is, the sum of the short-term O₃ increase, together with the long-term CH₄ perturbation, which also generates a long-lived O₃ perturbation.

Figure 3.6 illustrates these effects from four experiments that emitted month-long pulses of enhanced aircraft NO_x during four different months (Stevenson *et al.*, 2004). A pulse of NO_x generates a pulse of O₃ and OH, which reduces CH₄, which then decays with the 11-year timescale, which also generates a small long-lived negative O₃ anomaly. Table 3.1 gives GWPs for a variety of O₃ precursors (from Derwent *et al.* (2001) which contained some errors and was reworked using the methods of Stevenson *et al.*, 2004). These results illustrate the complex nature of GWPs of O₃ precursors; for example, the GWPs of surface NO_x are different in the Northern and Southern Hemispheres (NH and SH). Aircraft NO_x is significantly different to NO_x emitted at the surface (Table 3.2), and shows some seasonal dependence (Figure 3.6) (Stevenson *et al.*, 2004). There is also a lack of consistency between independent studies of aircraft NO_x, with its GWP ranging from a large positive value to near zero (Table 3.2). Most of this uncertainty stems from the O₃ contribution to the total GWP. Further recent work (Derwent *et al.*, 2006c) showed that GWPs for NO_x vary spatially between NH continental regions, and within Asia. This work clearly illustrates the difficulties in applying the GWP concept to NO_x.

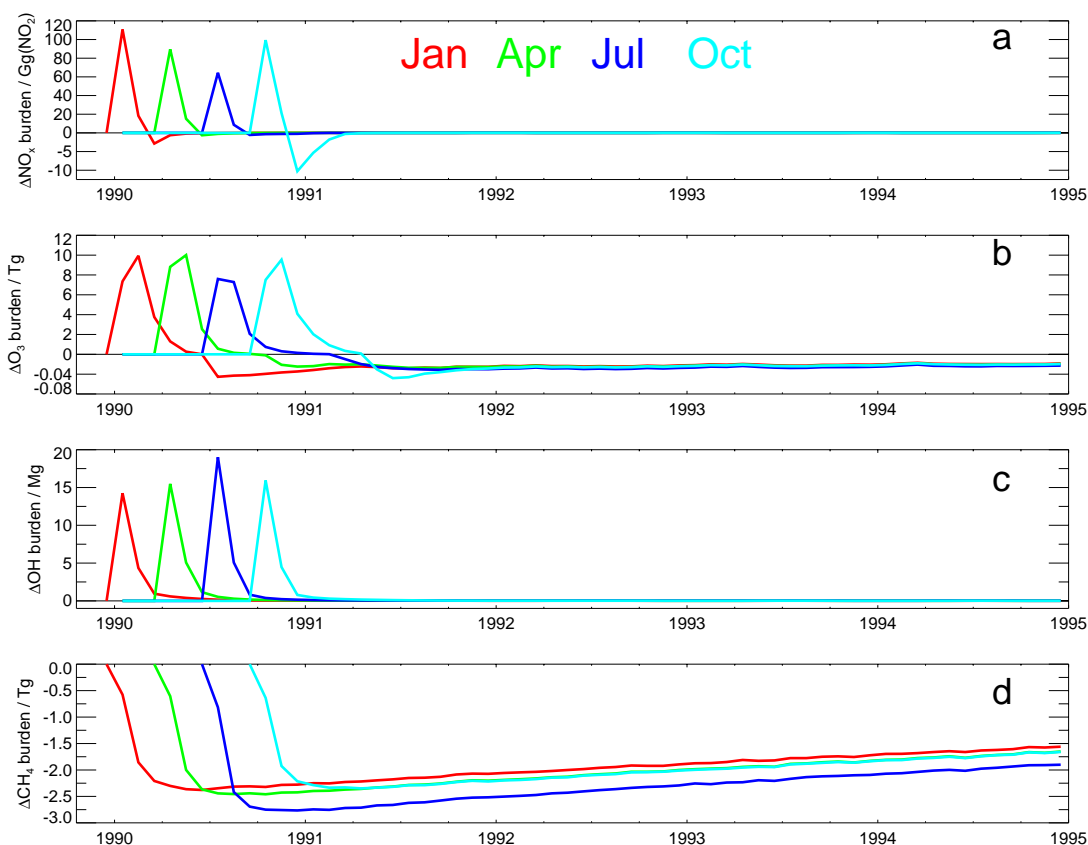


Figure 3.6: Time evolution of perturbations (relative to the control experiment) in the global burdens of (a) NO_x ; (b) O_3 ; (c) OH ; and (d) CH_4 , for four aircraft NO_x experiments (pulses emitted in January (red), April (green), July (dark blue), and October (light blue)). Note that the negative scales for NO_x and O_3 have been expanded for clarity. From Stevenson *et al.* (2004).

Emission	GWP- CH_4	GWP- O_3	GWP- CO_2	'Total' GWP
CH_4	16.9	4.1	2.4	+23.4
Surf NO_x in NH	-7.2	3.4		-3.8
Surf NO_x in SH	-19.4	10.0		-9.4
CO	0.85	0.63	1.6	+3.1
H_2	2.9	2.8		+5.7

Table 3.1: GWPs for a variety of emissions (Derwent *et al.*, 2001; Stevenson *et al.*, 2004)

Aircraft NO_x	GWP- CH_4	GWP- O_3	Total GWP
Derwent <i>et al.</i> (2001)	-44	+75	+31
Wild <i>et al.</i> (2001)	-44	+61	+17
Stevenson <i>et al.</i> (2004)	-40	+39	-0.9

Table 3.2: Three different estimates of aircraft NO_x GWPs

4 Conclusions

This project used global chemistry-climate models to investigate the characteristics of tropospheric O₃, in terms of its present day distribution and the effect of trends in emissions and climate change. Model simulations indicated the sources of European background ozone, and how these might evolve in future. Models were also used to estimate the GWPs of ozone precursor emissions.

Global CCMs, together with observational networks (such as ozone sondes, surface stations) and satellite datasets, provide us with our best overall picture of current distributions of ozone and its precursors. CCMs are reasonably good at simulating the observed atmosphere, and can also partly simulate the interannual variability of atmospheric composition, although some sources of variability (such as biomass burning) are only crudely represented. Perhaps the most stringent test of a CCM is its simulation of past trends, and to date, this has met with only partial success (although the necessary observational data are too sparse and too short for rigorous testing of the models). Hence the use of CCMs to investigate source apportionment, GWPs and future trends still has significant caveats, which should not be ignored. Nevertheless, CCMs give important insights into these areas of policy concern.

Results from a wide cross-section of state-of-the-art CCMs shows that future tropospheric O₃ levels will be strongly controlled by global O₃ precursor emissions whilst, at least in the near-term (2030), climate change is likely to have relatively modest (albeit highly uncertain) impacts (Stevenson *et al.*, 2006). These results have important implications for both air quality and climate policies. If the world follows a policy of implementing currently agreed (year 2000) legislation (the IIASA CLE scenario), we can expect modest rises (of the order of 5 ppbv; Figure 3.2) in tropospheric ozone by 2030. Alternatively, an aggressive policy of implementing the maximum currently technologically feasible reductions in global emissions (IIASA MFR) results in reductions of the order of 5 ppbv whereas lax emission controls (SRES A2) would yield much larger increases (greater than 10 ppbv; see Figure 3.2).

As the global climate warms, we can expect atmospheric water concentrations to increase. For most of the troposphere, this will enhance net ozone destruction; however, in the most polluted regions it will enhance net ozone production (Stevenson *et al.*, 2005; Murazaki and Hess, 2006). Downwards transport of stratospheric ozone is likely to increase, and some CCMs predict that this effect will outweigh ozone destruction, whilst others indicate it will not. There are several other poorly quantified climate-O₃ feedbacks, such as those associated with lightning NO_x, biogenic isoprene, boundary layer residence times, and other natural emissions.

Changing hemispheric emissions (currently driven by economic growth in Asia) and climate will affect background European ozone levels. We estimate that over half of the ozone found over the UK originates outside of the European troposphere, having sources in the stratosphere or elsewhere in the NH troposphere. A significant source of ozone, particularly on the western fringes of Europe, arises from ship NO_x emissions. This source is growing rapidly and is largely unregulated.

Global warming potentials for several ozone precursors were calculated in the CCM experiments. These suggest that NO_x generally has a negative GWP (a cooling impact on climate), as although it produces ozone, it also depletes CH₄, and this typically outweighs the first effect. GWPs for NO_x are highly dependent on the emission location, suggesting it will be

complex to include them in future climate treaties. Significant uncertainty still surrounds the GWP for aircraft NO_x, with estimates ranging from large positive values to near zero.

Future progress relies on further model development and validation, along with targeting of areas of high uncertainty, such as the tropics, and processes such as convection and stratosphere-troposphere exchange. Other significant sources of uncertainty stem from biomass burning and many other natural emission sources, and how these and other processes will respond to climate change.

4.1 Emissions

4.1.1 Key uncertainties and areas of further research

The largest uncertainty associated with projections of tropospheric ozone relates to the magnitude and distribution of future anthropogenic emissions, in particular NO_x sources. This clearly depends upon global, regional and local socio-economic and environmental policies, which are difficult to predict, hence the broad range of scenarios assessed here. Future scenarios have undergone major revision over the last five years or so (compare the IPCC SRES scenarios with the latest IIASA scenarios for gases such as NO_x and CO: see Dentener *et al.*, 2005), as changes in air quality legislation have been incorporated, and it is important that these scenarios remain as realistic as possible. Regionally, the highest uncertainties are associated with rapidly developing countries such as China and India; by sector, ship emissions are the least constrained. Biomass burning emissions, which are part natural and part anthropogenic, are also important and uncertain in terms of magnitude, seasonality, interannual variability and trends (both directly human-driven, via land use change, and climate-driven). Natural emissions are similarly uncertain, in particular biogenic sources of VOCs (including CH₄), and NO_x from lightning and soils.

4.1.2 Information/data gaps

Present-day (2001) global total ship NO_x emission estimates differ by a factor of two, depending on whether the inventory is based on bunker fuel supply (3.2 Tg N yr⁻¹) or on recorded ship journeys and engine statistics (6.5 Tg N yr⁻¹) (Eyring *et al.*, 2005). This discrepancy needs to be understood and minimised. The semi-natural sources from biomass burning (mainly CO and NO_x) and soils (NO_x), and wholly natural sources from vegetation (C₅H₈, CH₄ and other VOCs) and lightning (NO_x) all need to be better constrained, and process-based models linking these emissions to climate and ecosystem variables (such as temperature and vegetation type) need to be developed and validated. In some cases (for example, for lightning and wildfires), vertically resolved emissions are needed.

4.1.3 Risks to human/environmental health

Analysis of the 2030 scenarios clearly shows that the SRES A2 emissions pathway will considerably increase global mean surface ozone (4.3 ± 2.2 ppbv) and nitrogen deposition to areas of natural terrestrial vegetation (+ 25 per cent), whereas the CLE and MFR scenarios show smaller increases or decreases (1.5 ± 2.2 and -2.3 ± 1.1 ppbv for ozone, and + 15.8 and + 10.5 per cent for N-deposition, respectively) (Dentener *et al.*, 2006b). Higher mean ozone generally correlates with higher exposure indices (SOMO35 for human health, AOT40 for agriculture), as typical concentrations are close to threshold values.

4.1.4 Potential for collaboration

More tightly constrained estimates of shipping and developing world emissions could be developed, for example in collaboration with IIASA. A better understanding of climate-sensitive emissions is also required, particularly for the major natural sources of ozone precursors, such as biogenic VOCs and lightning NO_x.

4.1.5 Actions recommended for major industry regulators

Industry could aim to improve ship emissions inventories, both locally and at the European and global scale. Emissions accounting methods and control technologies could be exported to countries with less developed emissions recording systems.

4.2 Modelling

4.2.1 Key uncertainties and areas of further research

Key model uncertainties include parameterizations of mixing, including convection; boundary layer to free troposphere exchange; and stratosphere-troposphere exchange. Representation of such processes can be expected to improve as model resolution increases – this in turn is largely controlled by the availability of computer resources. Other uncertainties in modelling ozone include surface uptake, both stomatal and non-stomatal, which has implications for vegetation damage and boundary layer ozone concentrations. One of the most important atmospheric constituents for ozone is water vapour, a variable controlled by complex physical processes including cloud formation and precipitation, both typically sub-climate model grid scale and poorly constrained. In terms of chemistry, key areas include the oxidation of hydrocarbons such as biogenic VOCs, and heterogeneous processes such as interactions between gas and aerosol phases. Accurately representing model variability is also important, as it is often the extremes that have the most impact on the environment, such as heat waves and strong ENSO events. Analysis of observed variability may also be important in predicting impacts under future climate change.

4.2.2 Information/data gaps

There is a lack of information with which to validate models, although this is changing rapidly as satellite technology improves. Nevertheless, there remains a paucity of height-resolved data on tropospheric ozone and many of its precursors, including NO_x, CO and hydrocarbons. Most satellite products have poor vertical resolution or are just column amounts, but the vertical profiles are often key, as atmospheric lifetimes can vary by over an order of magnitude or more between the boundary layer and upper troposphere. Simulating long-term trends is also a key model validation statistic, but the number of datasets is limited. It is crucial that the few long-term datasets that do exist (such as Mace Head and other surface stations, and the MOZAIC commercial aircraft programme) continue to be supported.

4.2.3 Risks to human/environmental health

Models are an important route to determining large-scale exposure and exceedances, such as global distributions of nitrogen deposition or population exposure to ozone. Clearly, model uncertainties or inadequacies compromise our ability to accurately make such predictions and hence provide suitable advice on risks.

4.2.4 Potential for collaboration

All of the uncertainties identified above offer the potential for collaboration, with the aim of improving modelling capabilities. Perhaps the most obvious areas are those with the most direct impact on the UK: modelling of ozone episodes; ozone uptake by vegetation;

modelling of variabilities, especially those associated with extremes; use of satellite data in model validation; and the maintenance of long-term monitoring data.

4.2.5 Actions recommended for major industry regulators

It is obviously important that model simulations (of emission reductions, for example) are realistic, in order to provide useful data to regulators when they consider 'what if' scenarios. Industry regulators should therefore invest in model development to the required standards.

4.3 Impacts

4.3.1 Key uncertainties and areas of further research

Ozone impacts on human health and vegetation are difficult to ascertain, and rely on careful data analysis and laboratory experiment design. For human health, confounding factors have to be removed and this is often extremely difficult, requiring large data populations. For vegetation, large-scale and long-term field experiments are often required, where the responses of a wide variety of vegetation types down to species level is needed, for example to compare different crop varieties. Consequently, the parameter space remains poorly sampled. For vegetation, it is the actual uptake of ozone rather than the surrounding ambient concentration, that is the crucial parameter, and this is much more difficult to measure. West and Fiore (2005) estimated the global economic impacts of ozone exposure at \$4.4-10.7 billion yr⁻¹ ppb⁻¹; this clearly illustrates the economic argument for ozone reduction. Ozone, as a powerful oxidant, also affects the fabric of buildings and structures, reducing their useful lifetimes.

4.3.2 Information/data gaps

Further data is required on the response functions of human health and ecosystems to ozone exposure. The parameter space of ozone exposure needs to be systematically sampled, as high risk groups (both human and ecosystem) may currently remain unidentified.

4.3.3 Risks to human/environmental health

Modelling projections of the near future present a generally bleak picture, especially for the developing world, under scenarios of current legislation (CLE) or high growth with lax air quality legislation (SRES A2). The CLE scenario identifies India as the region most at risk of increased ozone exposure.

4.3.4 Potential for collaboration

Response functions of humans and ecosystems to ozone need to be better constrained, using a combination of *in situ* (such as station monitoring) data and related human/ecosystem health data. This data is most needed in the rapidly developing world.

4.3.5 Actions recommended for major industry regulators

Systems should be put in place to adequately monitor ozone exposure and its impacts, and the data gathered should be pooled, archived and fully analysed. Confounding factors (for example, co-exposure to other pollutants such as NO₂ or PM) need to be addressed and accounted for. Long-term (decadal) monitoring in areas likely to see an improvement or deterioration in air quality needs to be in place.

4.4 Implications for control of ozone

4.4.1 Key uncertainties and areas of further research

The ACCENT intercomparison showed that inter-model uncertainty in projections of ozone air quality, for a given emissions scenario, was of the order of ± 25 per cent. Much larger uncertainties derive from not knowing the future emissions pathways for different world regions, and when accounting for climate change. Globally, it is unclear if climate change will enhance or reduce ozone, but it appears likely to reduce ozone in remote regions whilst enhancing it in polluted regions. Models are currently not that good at simulating past trends in ozone (although the observational data with which to validate them are sparse), and this reduces confidence in future predictions. Models can also be used to quantify indirect climate forcing from ozone precursor emissions. By assessing both the air quality and climate implications of an emissions reduction (or increase), the benefits (and trade-offs) can be seen, which may have important implications for future climate and air quality legislation.

4.4.2 Information/data gaps

Clearly, improved emission inventories and accurate future (and past) scenarios are needed. In addition, climate change effects need to be fully understood and quantified, using process-based models of emissions and fully coupled chemistry-climate models to determine important interactions.

4.4.3 Risks to human/environmental health

Unless ozone precursor emission controls are put in place, significant risks are likely. Even under the highly optimistic MFR scenario, nitrogen deposition to natural vegetation still shows a significant increase, and globally ozone exposure falls only slightly from its current, often dangerous levels. The implications are that we should strive even further than the MFR scenario, and develop technologies to further reduce ozone precursor emissions, or learn to adapt to greater pollution.

4.4.4 Potential for collaboration

The implementation of emission controls with widespread dissemination (for example, to the developing world) seems a fruitful route for collaboration, especially in conjunction with modelling to illustrate the health and economic benefits of reduced emissions.

4.4.5 Actions that major industry regulators could undertake

Emission controls should be reviewed and wherever possible improved using the best available technology. The economic case, based on model results, can illustrate the cost-effectiveness of such actions.

5 Recommendations

The results presented in this report have a number of policy implications for the Environment Agency, the Department for Environment, Food and Rural Affairs (Defra) and the Kyoto process, and air quality and climate policy in general.

Background levels of ozone appear to be rising in some parts of the UK, counteracting the benefits from reductions in UK ozone precursor emissions. The underlying reasons behind this increase are related to long-range transport and the rise in Northern Hemisphere emissions of NO_x and VOCs, in particular from Asia, but also from North America. Rising levels of CH₄ may also contribute to higher background ozone in the future.

Global or hemispheric agreements to limit/reduce ozone precursor emissions, along with national and regional agreements, are required.

In addition to the Asian region as a major cause for concern, emissions from the shipping and aviation sectors are increasing rapidly, and are subject to much less regulation than many other sectors.

Emissions from ships and aircraft need to be more tightly regulated if they are not to counteract the benefits of emission reductions in other areas.

These recommendations mainly stem from results from one global-scale model, although other related studies back these findings.

Clearly defined metrics, together with experimental designs that quantify source-receptor relationships (such as the contribution of Asian or ship NO_x emissions to European O₃), need to be developed for several different models, to increase confidence in model-derived quantities.

Multiple models are also needed to estimate uncertainties. Multi-model analysis, like the ACCENT model intercomparison exercise, has started to address these issues.

Three global emission scenarios in 2030 were evaluated by 20 state-of-the-art models, allowing us to make strong, quantitative statements on air quality and climate forcing under these different scenarios. For example, ozone air quality over Central Europe, as expressed by a range of vegetation impact indices, showed little change between 2000 and 2030 under the 'current legislation' (CLE) scenario, but showed significant improvement under the 'maximum feasible reductions' (MFR) scenario, and significant deterioration under the SRES A2 scenario (Ellingsen *et al.*, 2007). These scenarios were also evaluated for their impacts on climate forcing, where the MFR, CLE and A2 scenarios yielded combined tropospheric ozone and methane radiative forcings over the period 2000-2030 of -50, 180 and 300 mW m⁻², compared to predicted forcing of about 1,050 mW m⁻² from CO₂ over the same period under the A2 scenario (Stevenson *et al.*, 2006).

It should be possible to extend this work, by exploring future economic scenarios, to provide cost-benefit analyses for policy-makers. This would require estimates of costs associated with changes in air quality and climate, from collaboration with impacts assessors and economists.

We also quantified the global warming potentials of ozone precursors. It was clear that CH₄ and CO, and most (although not all) VOCs would have positive GWPs (leading to climate warming).

Reductions in emissions of CH₄, CO and most VOCs would benefit ozone air quality and reduce climate forcing.

The GWP of surface NO_x emissions was found to be negative (climate cooling) and quite strongly dependent upon both season and location (Derwent *et al.*, 2006).

NO_x controls present something of a dilemma for policy-makers: they would improve air quality, but would also contribute to climate warming.

The GWP of aircraft NO_x was found to be close to zero in one study (Stevenson *et al.*, 2004), but positive in two others (see Table 3.2), showing that the altitude of emission is also crucial. This spatial and temporal heterogeneity in NO_x GWPs means that inclusion of NO_x in future climate treaties will be difficult.

The recommendation here is that these type of GWP experiments need to be performed by a much wider range of models, in order to increase the reproducibility and reliability of results and allow uncertainties to be quantified.

Climate change will also have impacts on air quality. Results from 10 CCMs show a range of responses in 2030, with two important processes: a decrease in background ozone associated with an increase in water vapour in the warmer atmosphere, and an increase in the stratospheric influx of ozone. It is not clear which of these two processes will dominate, so the likely climate feedback on ozone is currently not well known. The increase in water vapour is likely to enhance ozone production in polluted regions, with localised positive feedback in areas of already poor air quality. The effects of other important processes such as increased biogenic emissions, shifts in lightning and changes in the frequency of stagnation are not adequately known.

The identified feedbacks need to be better understood, and as coupled climate-chemistry models are developed (for example, to include further couplings with the biosphere), we need to re-assess the relative importance of all potentially important climate-chemistry feedback mechanisms.

Appendix 1: Abstracts of publications

In addition to published work, I have also included abstracts of those papers currently submitted for publication, and where available, abstracts of those in preparation. Abstracts are listed in alphabetical order of the lead author. Full copies of most publications are available via hyperlinks in the electronic version of this report.

[1. The oxidation of organic compounds in the troposphere and their global warming potentials](#)

Collins, W.J., R.G. Derwent, C.E. Johnson and D.S. Stevenson
Climatic Change, 52, 453-479, 2002a.

Oxidation by hydroxyl radicals is the main removal process for organic compounds in the troposphere. This oxidation acts as a source of ozone and as a removal process for hydroxyl and peroxy radicals, thereby reducing the efficiency of methane oxidation and promoting the build-up of methane. Emissions of organic compounds may therefore lead to the build-up of two important radiatively-active trace gases: methane and ozone. Emission pulses of 10 organic compounds were followed in a global 3-D Lagrangian chemistry-transport model to quantify their indirect greenhouse gas impacts through changes induced in the tropospheric distributions of methane and ozone. The main factors influencing the global warming potentials of the 10 organic compounds were found to be their spatial emission patterns, chemical reactivity and transport, molecular complexity and oxidation products formed. The indirect radiative forcing impacts of organic compounds may be large enough that ozone precursors should be considered in the basket of trace gases through which policy-makers aim to combat global climate change.

[2. A comparison of two schemes for the convective transport of chemical species in a Lagrangian global chemistry model](#)

Collins, W.J., R.G. Derwent, C.E. Johnson and D.S. Stevenson
Quarterly Journal of the Royal Meteorological Society, 128, 991-1009, 2002b.

We have developed a detailed parametrization scheme to represent the effects of subgrid-scale convective transport in a three-dimensional chemistry-transport model (CTM). The CTM utilizes the meteorological fields generated by a general-circulation model (GCM) to redistribute over 70 chemical species. The convective transport is implemented using the convective mass fluxes entrainment rates and detrainment rates from the GCM. We compare the modelled distributions of Rn-222 with observations. This shows that the vertical profile of this species is affected by the choice of convective-transport parametrization. The new parametrization is found to improve significantly the simulation of Rn-222 over the summertime continents.

[3. The effect of stratosphere-troposphere exchange on the future tropospheric ozone trend](#)

Collins, W.J., R.G. Derwent, B. Garnier, C.E. Johnson, M.G. Sanderson and D.S. Stevenson
Journal of Geophysical Research, 108 (D12), DOI: 10.1029/2002JD002617, 2003.

This paper investigates the impact of circulation changes in a changed climate on the exchange of ozone between the stratosphere and the troposphere. We have identified an increase in the net transport of ozone into the troposphere in the future climate of 37 per cent, although a decreased ozone lifetime means that the overall tropospheric burden decreases. There are regions in the mid-latitudes to high latitudes where surface ozone is predicted to increase in the spring. However, these increases are not significant. Significant ozone increases are predicted in regions of the upper troposphere. The general increase in

the stratospheric contribution (O_{3s} tracer) to tropospheric ozone in the climate changed scenario indicates that the stratosphere will play an even more significant role in the future.

4. The impact of air pollutant and methane emission controls on tropospheric ozone and radiative forcing: CTM calculations for the period 1990-2030

Dentener, F., D. Stevenson, J. Cofala, R. Mechler, M. Amann, P. Bergamaschi, F. Raes, R. Derwent

Atmospheric Chemistry and Physics, 5, 1731-1755, 2005.

To explore the relationship between tropospheric ozone and radiative forcing with changing emissions, we compiled two sets of global scenarios for the emissions of the ozone precursors methane (CH_4), carbon monoxide (CO), non-methane volatile organic compounds (NMVOC) and nitrogen oxides (NO_x) up to the year 2030 and implemented them in two global chemistry-transport models. The "current legislation" (CLE) scenario reflects the current perspectives of individual countries on future economic development and takes the anticipated effects of presently decided emission control legislation in the individual countries into account. In addition, we developed a "maximum technically feasible reduction" (MFR) scenario that outlines the scope for emission reductions offered by full implementation of the presently available emission control technologies, while maintaining the projected levels of anthropogenic activities. Whereas the resulting projections of methane emissions lie within the range suggested by other greenhouse gas projections, the recent pollution control legislation of many Asian countries, requiring introduction of catalytic converters for vehicles, leads to significantly lower growth in emissions of the air pollutants NO_x , NMVOC and CO than was suggested by the widely used and more pessimistic IPCC (Intergovernmental Panel on Climate Change) SRES (Special Report on Emission Scenarios) scenarios (Nakicenovic *et al.*, 2000), which made business-as-usual assumptions regarding emission control technology.

With the TM3 and STOCHEM models we performed several long-term integrations (1990-2030) to assess global, hemispheric and regional changes in CH_4 , CO, hydroxyl radicals, ozone and the radiative climate forcings resulting from these two emission scenarios. Both models reproduce broadly the observed trends in CO, and CH_4 concentrations from 1990 to 2002. For the "current legislation" case, both models indicate an increase of the annual average ozone levels in the Northern Hemisphere by 5 ppbv, and up to 15 ppbv over the Indian sub-continent, comparing the 2020s (2020-2030) with the 1990s (1990-2000). The corresponding higher ozone and methane burdens in the atmosphere increase radiative forcing by approximately 0.2 Wm^{-2} . Full application of today's emissions control technologies, however, would bring down ozone below the levels experienced in the 1990s and would reduce the radiative forcing of ozone and methane to approximately -0.1 Wm^{-2} . This can be compared to the $0.14\text{-}0.47 \text{ Wm}^{-2}$ increase of methane and ozone radiative forcings associated with the SRES scenarios. While methane reductions lead to lower ozone burdens and to less radiative forcing, further reductions of the air pollutants NO_x and NMVOC result in lower ozone, but at the same time increase the lifetime of methane. Control of methane emissions appears an efficient option to reduce tropospheric ozone as well as radiative forcing.

5. Nitrogen and sulphur deposition on regional and global scales: a multi-model evaluation

Dentener, F., *et al.*

Global Biogeochemical Cycles, 20, GB4003, DOI:10.1029/2005GB002672, 2006a.

We use 23 atmospheric chemistry-transport models to calculate current and future (2030) deposition of reactive nitrogen (NO_y and NH_x) and sulphate (SO_x) to land and ocean surfaces. For the year 2030, the models are driven by three different emission scenarios: the first reflects current air quality legislation (CLE) around the world; the second represents an optimistic case of the maximum emissions reductions currently technologically feasible (MFR); the contrasting third is the pessimistic IPCC SRES A2 scenario. We perform an extensive evaluation of the present-day deposition using nearly all information on wet

deposition available worldwide. Most models show a good agreement with observations in Europe and North America, whereas the model skills are poorer in India, South East Asia, Africa and South America. Nevertheless, in regions with quality controlled measurements 60-70% of the models calculate wet deposition rates that agree with measurements to within $\pm 50\%$. Models systematically overestimate NH_x deposition in South Asia, and underestimate NO_y deposition in East Asia. We show that there are substantial differences between models for the removal mechanisms of NO_y , NH_x , and SO_x , leading to \pm one standard variance in total deposition fluxes of about 30% in the anthropogenic emissions regions, and up to a factor of two outside. In all cases, the mean model constructed from the ensemble calculations is among the best when compared to measurements.

Our models indicate that currently, 43%, 36% and 51% of all NO_y , NH_x , and SO_x is deposited over the ocean, and 50-80% of the fraction of deposition on land falls on natural (non-agricultural) vegetation. We use a threshold of $1 \text{ g(N) m}^{-2} \text{ yr}^{-1}$ ("critical load") to indicate potential risk for ecosystems. Currently, 11% of the world's natural vegetation receives nitrogen deposition in excess of this threshold. The regions most concerned are the United States (20% of vegetation), Western Europe (30%), Eastern Europe (80%), South Asia (60%), East Asia (40%), South East Asia (30%), and Japan (50%). Deposition fluxes in the future are mainly driven by changes in emissions, and less importantly by changes in atmospheric chemistry and climate. Under CLE, NO_y deposition remains roughly constant in 2030 in most parts of the world, except for Asia where NO_y deposition further increases by 50% to 100%. In contrast, NO_y deposition for MFR could decrease worldwide by 50%. A2 would imply further increases of NO_y deposition in the polluted parts of the world by a factor of two. Following CLE, NH_x deposition goes down by 20% in Europe, and further increases by 40-100% in parts of Central and South America, Africa, and parts of Asia. NH_x deposition for A2 is rather similar to CLE. Deposition of SO_x varies strongly among the scenarios. For CLE SO_x deposition remains constant or decreases everywhere, except in Asia. Assuming MFR, large reductions in deposition can be realized throughout the world, whereas A2 implies large increases in deposition everywhere except for North America and Europe. These results have important implications for ecosystem nitrogen loads; the global fraction of vegetation exposed to nitrogen loads in excess of $1 \text{ g(N) m}^{-2} \text{ yr}^{-1}$ increases globally to 17% for CLE and 25% for A2. In MFR, the reductions in NO_y are off-set by further increases in NH_x deposition. The regions most concerned by exceedingly high nitrogen loads for CLE and A2 are Europe, and Asia, but also parts of Africa.

6. The global atmospheric environment for the next generation

Dentener, F., Stevenson, D.S., *et al.*

Environmental Science and Technology, DOI:10.1021/es0523845, 2006b.

Air quality, ecosystem exposure to nitrogen deposition, and climate change are intimately coupled problems: we assess changes in the global atmospheric environment between 2000 and 2030 using twenty-six state-of-the-art global atmospheric chemistry models and three different emissions scenarios. The first (CLE) scenario reflects implementation of current air quality legislation around the world, whilst the second (MFR) represents a more optimistic case in which all currently feasible technologies are applied to achieve maximum emission reductions. We contrast these scenarios with the more pessimistic IPCC SRES A2 scenario. Ensemble simulations for the year 2000 are consistent among models, and show a reasonable agreement with surface ozone, wet deposition and NO_2 satellite observations. Large parts of the world are currently exposed to high ozone concentrations, and high depositions of nitrogen to ecosystems. By 2030, global surface ozone is calculated to increase globally by 1.5 ± 1.2 ppb (CLE), and 4.3 ± 2.2 ppb (A2), using the ensemble mean of model results and associated $\pm 1 \sigma$ standard deviations. Only the progressive MFR scenario will reduce ozone by -2.3 ± 1.1 ppb. Climate change is expected to modify surface ozone by -0.8 ± 0.6 ppb, with larger decreases over sea than over land. Radiative forcing by ozone increases by 63 ± 15 , and $155 \pm 37 \text{ mWm}^{-2}$ for CLE and A2, respectively, and decreases by $-45 \pm 15 \text{ mWm}^{-2}$ for MFR. We compute that at present, 10.1 % of the global

natural terrestrial ecosystems receive nitrogen inputs above a critical load of $1 \text{ g N m}^{-2} \text{ yr}^{-1}$. These percentages increase by 2030 to 15.8% (CLE), 10.5% (MFR) and 25% (A2). This study shows the importance of enforcing current worldwide air quality legislation, and the major benefits of going further. Non-attainment of these air quality policy objectives, such as expressed by the SRES-A2 scenario, would further degrade the global atmospheric environment.

7. Global warming potentials for non-CO₂ greenhouse gases

Derwent, R.G., W.J. Collins, C.E. Johnson and D.S. Stevenson

In: *Non-CO₂ Greenhouse Gases*, Van Ham *et al.* (eds), 345-362, Millpress, Rotterdam, 2002a.

Policy responses to mitigate climate change rely on an understanding of the relative importance of the different non-CO₂ greenhouse gases. Global warming potentials (GWPs) represent one way of expressing the relative radiative impacts of a wide range of trace gases. The estimation of direct GWPs requires quantitative estimates of the radiative properties of each trace gas and their atmospheric lifetime. Indirect GWPs for the tropospheric ozone precursors requires an understanding of their impacts on the build-up of methane and ozone through changes that they induce in the oxidising capacity of the troposphere.

8. Global ozone concentrations and regional air quality

Derwent, R.G., W.J. Collins, C.E. Johnson and D.S. Stevenson

Environmental Science and Technology, 36(19), 379A-382A, 2002b.

This article had no abstract.

9. The global distribution of secondary particulate matter in a 3-D Lagrangian chemistry-transport model

Derwent, R.G., W.J. Collins, M.E. Jenkin, C.E. Johnson and D.S. Stevenson

Journal of Atmospheric Chemistry, 44, 57-95, 2003.

A global 3-D Lagrangian chemistry-transport model STOCHEM is used to describe the tropospheric distributions of four components of the secondary atmospheric aerosol: nitrate, sulphate, ammonium and organic compounds. The model describes the detailed chemistry of the formation of the acid precursors from the oxidation of SO₂, DMS, NO_x, NH₃ and terpenes and their uptake into the aerosol. Model results are compared in some detail with the available surface observations. Comparisons are made between the global budgets and burdens found in other modelling studies. The global distributions of the total mass of secondary aerosols have been estimated for the pre-industrial, present day and 2030 emissions and large changes have been estimated in the mass fractions of the different secondary aerosol components.

10. Intercontinental transport and the origins of the ozone observed at surface sites in Europe

Derwent, R.G., D.S. Stevenson, W.J. Collins and C.E. Johnson

Atmospheric Environment, 38, 1891-1901, 2004.

A global three-dimensional Lagrangian chemistry-transport model is used to describe the formation, transport and destruction of tropospheric ozone using 1990s global emissions and 1998 meteorological archives. Using a labelling technique, the geographical origins of the ozone formed within the troposphere have been revealed, showing whether the ozone found at the surface in Europe has had its origins above the continents of North America, Europe or Asia or elsewhere in the world. In this way, contributions to the ozone found at 21 surface monitoring sites across Europe can be attributed to production over North America and Asia, demonstrating that intercontinental ozone transport is an efficient process. Sensitivity tests to the global manmade sources of NO and carbon monoxide indicate that global ozone precursor emission controls may contribute towards reaching regional air quality policy goals for ozone in Europe.

11. The contribution from ship emissions to air quality and acid deposition in Europe

Derwent, R.G., D.S. Stevenson, R.M. Doherty, W.J. Collins, M.G. Sanderson, C.E.

Johnson, J. Cofala, R. Mechler, M. Amann and F.J. Dentener
Ambio, 34(1), 54-59, 2005.

A global three-dimensional Lagrangian chemistry-transport model STOCHEM is used to describe the European regional acid deposition and ozone air quality impacts along the Atlantic Ocean seaboard of Europe, from the SO₂, NO_x, VOCs and CO emissions from international shipping under conditions appropriate to the year 2000. Model-derived total sulfur deposition from international shipping reaches over 200 mg S m⁻² yr⁻¹ over the south-western approaches to the British Isles and Brittany. The contribution from international shipping to surface ozone concentrations during the summertime peaks at about 6 ppb over Ireland, Brittany and Portugal. Shipping emissions act as an external influence on acid deposition and ozone air quality within Europe and may require control actions in the future if strict deposition and air quality targets are to be met.

12. External influences on Europe's air quality: Baseline methane, carbon monoxide and ozone from 1990 to 2030 at Mace Head, Ireland

Derwent, R.G., P.G. Simmonds, S. O'Doherty, D.S. Stevenson, W.J. Collins, M.G. Sanderson, C.E. Johnson, F. Dentener, J. Cofala, R. Mechler and M. Amann
Atmospheric Environment, 40, 844-855, 2006a.

A global 3-D chemistry-transport model STOCHEM has been employed to study trends in the mole fractions of methane, carbon monoxide and ozone in baseline air masses entering Europe from the Atlantic Ocean over the period from 1990 to 2030. With a range of emission scenarios for manmade ozone precursor emission sources available, a wide range of model trends were predicted for the period up to 2030. In the scenario based on current planned air pollution controls, IIASA CLE, methane shows a strong upward trend, ozone shows a weaker upward trend, and carbon monoxide is approximately flat in baseline air masses. In one of the more pessimistic IPCC SRES scenarios, A2, all three gases show future increases. However, in the scenario based on maximum feasible emission reductions, IIASA MFR, all three trace gases decline. By 2030, projected climate change reduces the growth in CH₄, but has insignificant effects on baseline CO and O₃ in these simulations. Global or hemispheric ozone precursor emissions and their controls exert a potentially large external influence on Europe's air quality. This influence is currently not taken into account in future European air quality policy formulation.

13. Global environmental impacts of the hydrogen economy

Derwent, R.G., P.G. Simmonds, S. O'Doherty, A. Manning, W.J. Collins, C.E. Johnson, M.G. Sanderson and D.S. Stevenson
International Journal of Nuclear Hydrogen Production and Application, 1, 1, 57-67, 2006b.

Hydrogen-based energy systems appear to be an attractive proposition in providing a future replacement for the current fossil-fuel based energy systems. Hydrogen appears attractive because it is a clean fuel and because it offers efficiency improvements when it is utilised. The transport sector may provide some of the first applications of the novel hydrogen technologies. Hydrogen is an important, though little studied, trace component of the atmosphere. It is present at the mixing ratio of about 510 ppb currently and has important manmade and natural sources. Its atmospheric lifetime is about 2.5 years and there is a global burden of about 180 Tg in the atmosphere. Because hydrogen reacts with tropospheric hydroxyl radicals, emissions of hydrogen to the atmosphere perturb the distributions of methane and ozone, the second and third most important greenhouse gases after carbon dioxide. Hydrogen is therefore an indirect greenhouse gas with a global warming potential (GWP) of 5.8 over a 100-year time horizon. A future hydrogen economy would therefore have greenhouse consequences and would not be free from climate perturbations. If a global hydrogen economy replaced the current fossil fuel-based energy system and exhibited a leakage rate of one per cent, then it would produce a climate impact of 0.6% of the current fossil fuel based system. If the leakage rate were 10%, then the climate impact would be 6% of the current system. Careful attention must be given to reduce to a minimum

the leakage of hydrogen from the synthesis, storage and utilisation of hydrogen in a future global hydrogen economy if the full climate benefits are to be realised in comparison to fossil fuel-based energy systems.

14. Radiative forcing from surface NO_x emissions: spatial and seasonal variations

Derwent, R.G., D.S. Stevenson, R.M. Doherty, W.J. Collins, M.G. Sanderson and C.E. Johnson

Submitted to *Climatic Change*, 2006c.

The global three-dimensional Lagrangian chemistry-transport model STOCHEM has been used to follow changes in the tropospheric distributions of methane CH₄ and ozone O₃ following the emission of pulses of the oxides of nitrogen NO_x. Month-long emission pulses of NO_x produce deficits in CH₄ mixing ratios that bring about negative radiative forcing (climate cooling) and decay away with e-folding times of 10 to 15 years. They also produce short-term excesses in O₃ mixing ratios that bring about positive radiative forcing (climate warming) that decay over several months to produce deficits, with their attendant negative radiative forcing that decays away in step with the CH₄ deficits. Total time-integrated net radiative forcing is markedly influenced by cancellation between the negative CH₄ and long-term O₃ contributions and the positive short-term O₃ contribution to leave a small negative residual. Consequently, total net radiative forcing from NO_x emission pulses and the global warming potentials derived from them show a strong dependence on the magnitudes, locations and seasons of the emissions. These dependences are illustrated using the Asian continent as an example and demonstrate that there is no simple robust relationship between continental-scale NO_x emissions and globally integrated radiative forcing. Nevertheless, it seems inevitable that control of manmade surface NO_x emissions may lead to a positive radiative forcing (climate warming).

15. Influence of convective transport on tropospheric ozone and its precursors in a chemistry-climate model

Doherty, R.M., D.S. Stevenson, W.J. Collins and M.G. Sanderson

Atmospheric Chemistry and Physics, 5, 3205-3218, 2005.

The impact of convection on tropospheric O₃ and its precursors has been examined in a coupled chemistry-climate model. There are two ways that convection affects O₃. First, convection affects O₃ by vertical mixing of O₃ itself. Convection lifts lower tropospheric air to regions where the O₃ lifetime is longer, whilst mass-balance subsidence mixes O₃-rich upper tropospheric (UT) air downwards to regions where the O₃ lifetime is shorter. This tends to decrease UT O₃ and the overall tropospheric column of O₃. Secondly, convection affects O₃ by vertical mixing of O₃ precursors. This affects O₃ chemical production and destruction. Convection transports isoprene and its degradation products to the UT where they interact with lightning NO_x to produce PAN, at the expense of NO_x. In our model, we find that convection reduces UT NO_x through this mechanism; convective down-mixing also flattens our imposed profile of lightning emissions, further reducing UT NO_x. Over tropical land, which has large lightning NO_x emissions in the UT, we find convective lofting of NO_x from surface sources appears relatively unimportant. Despite UT NO_x decreases, UT O₃ production increases as a result of UT HO_x increases driven by isoprene oxidation chemistry. However, UT O₃ tends to decrease, as the effect of convective overturning of O₃ itself dominates over changes in O₃ chemistry. Convective transport also reduces UT O₃ in the mid-latitudes, resulting in a 13% decrease in the global tropospheric O₃ burden. These results contrast with an earlier study that uses a model of similar chemical complexity. Differences in convection schemes as well as chemistry schemes – in particular isoprene-driven changes – are the most likely causes of such discrepancies. Further modelling studies are needed to constrain this uncertainty range.

16. Tropospheric ozone and El Niño Southern Oscillation: Influence of atmospheric dynamics, biomass burning emissions and future climate change

Doherty, R.M., D.S. Stevenson, W.J. Collins, M.G. Sanderson and C.E. Johnson
Journal of Geophysical Research, 111, D19304, DOI:10.1029/2005JD006849, 2006.

We investigate how El Niño Southern Oscillation (ENSO) influences tropospheric ozone and its precursors in a coupled climate-chemistry model. As shown in similar studies, tropospheric column ozone (TCO) decreases in the Central and East Pacific and increases in the West Pacific/Indonesia in response to circulation and convective changes during El Niño conditions. Simulated changes in TCO for “peak” El Niño events in the Central and East Pacific are in good agreement but are underestimated in the West Pacific compared to previous observational and modelling studies for October 1997. Tropospheric column-average NO_x decreases over Indonesia and generally over South America as a result of suppressed convection and lightning over these land regions. NO_x and HO_x changes during El Niño modify ozone chemical production and destruction. When we include annually varying biomass burning emissions in our model simulations, we find that these emissions peak over Indonesia one to two months in advance of the peak elevated sea-surface temperatures (SSTs) and hence the “meteorological” El Niño. We underestimate the strength of the ozone increase due to El Niño-related dry conditions over Indonesia in October 1997 compared to observations. We also examine how future mean and variability changes in ENSO, as simulated in the HadCM3 climate model, impacts tropospheric ozone. A mean future El Niño like-state is simulated in the tropical Pacific in HadCM3, but this has no discernable impact on the future TCO trend in this region. However, we do simulate increased variability in precipitation and TCO related to ENSO in the future.

17. Ozone air quality in 2030: a multi-model assessment of risks for health and vegetation

Ellingsen, K. and 41 others, including D.S. Stevenson
In preparation, *Journal of Geophysical Research*, 2007.

Abstract not available yet.

18. Multi-model simulations of the impact of international shipping on atmospheric chemistry and climate in 2030

Eyring, V., D.S. Stevenson, *et al.*
Atmospheric Chemistry and Physics, 7, 1995-2035, 2007.

The global impact of shipping on atmospheric chemistry and radiative forcing, as well as the associated uncertainties, have been quantified using an ensemble of ten state-of-the-art atmospheric chemistry models and a pre-defined set of emission data. The analysis is performed for present-day conditions (year 2000) and for two future ship emission scenarios. In one scenario ship emissions stabilize at 2000 levels; in the other ship emissions increase with a constant annual growth rate of 2.2% up to 2030 (termed the ‘Constant Growth Scenario’ (CGS)). Most other anthropogenic emissions follow the IPCC (Intergovernmental Panel on Climate Change) SRES (Special Report on Emission Scenarios) A2 scenario, while biomass burning and natural emissions remain at year 2000 levels. The models show future increases in NO_x and O₃ burden which scale almost linearly with increases in NO_x emission totals. For the same ship emission totals but higher emissions from other sources a slightly smaller response is found. The most pronounced changes in annual mean tropospheric NO₂ and sulphate columns are simulated over the Baltic and North Seas; other significant changes occur over the N. Atlantic, the Gulf of Mexico and along the main shipping lane from Europe to Asia, across the Red and Arabian Seas. Maximum contributions from shipping to annual mean near-surface O₃ are found over the N. Atlantic (5-6 ppbv in 2000; up to 8 ppbv in 2030). Ship contributions to tropospheric O₃ column over the N. Atlantic and Indian Oceans reach 1 DU in 2000 and up to 1.8 DU in 2030. Tropospheric O₃ forcings due to shipping are $9.8 \pm 2.0 \text{ mW m}^{-2}$ in 2000 and $13.6 \pm 2.3 \text{ mW m}^{-2}$ in 2030. Whilst increasing O₃, ship NO_x simultaneously enhances OH over the remote ocean, reducing the global CH₄ lifetime by 0.13 yr in 2000, and by up to 0.17 yr in 2030, introducing a negative radiative

forcing. Over Europe, the increase in ship emissions under the A2/CGS enhances the positive trend in NO₂ over land up to 2030. In addition, efforts to lower European sulphate levels through reductions in SO₂ emissions from anthropogenic sources on land are partly counteracted by the rise in ship emissions. Globally, shipping contributes 3% to increases in O₃ burden between 2000 and 2030, and 4.5% to increases in sulphate under A2/CGS. However, if future ground based emissions follow a more stringent scenario, the relative importance of ship emissions will increase. Inter-model differences in the simulated O₃ contributions from ships are significantly smaller than estimated uncertainties stemming from the ship emission inventory, mainly the emission totals, the distribution of the emissions over the globe; and the neglect of ship plume dispersion.

19. Radiative forcing in the twenty-first century due to ozone changes in the troposphere and the lower stratosphere

Gauss, M. *et al.*

Journal of Geophysical Research, 108 (D9), DOI: 10.1029/2002JD002624, 2003.

Radiative forcing due to changes in ozone is expected for the twenty-first century. An assessment on changes in the tropospheric oxidative state through a model intercomparison ("OxComp") was conducted for the IPCC Third Assessment Report (IPCC-TAR). OxComp estimated tropospheric changes in ozone and other oxidants during the 21st century based on the "SRES" A2p emission scenario. In this study, we analyze the results of 11 chemistry-transport models (CTMs) that participated in OxComp and use them as input for detailed radiative forcing calculations. We also address future ozone recovery in the lower stratosphere and its impact on radiative forcing by applying two models that calculate both tropospheric and stratospheric changes. The results of OxComp suggest an increase in global-mean tropospheric ozone between 11.4 and 20.5 DU for the twenty-first century, representing the model uncertainty range for the A2p scenario. As the A2p scenario constitutes the worst case proposed in IPCC-TAR, we consider these results as an upper estimate. The radiative transfer model yields a positive radiative forcing ranging from 0.40 to 0.78 W m⁻² on a global and annual average. The lower stratosphere contributes an additional 7.5-9.3 DU to the calculated increase in the ozone column, increasing radiative forcing by 0.15-0.17 W m⁻². The modelled radiative forcing depends on the height distribution and geographical pattern of predicted ozone changes and shows a distinct seasonal variation. Despite the large variations between the 11 participating models, the calculated range for normalized radiative forcing is within 25 per cent, indicating the ability to scale radiative forcing to global-mean ozone column change.

20. Radiative forcing since pre-industrial times due to ozone change in the troposphere and the lower stratosphere

Gauss, M. *et al.*

Atmospheric Chemistry and Physics, 6, 575-599, 2006.

Changes in atmospheric ozone have occurred since the pre-industrial era as a result of increasing anthropogenic emissions. Within ACCENT, a European Network of Excellence, ozone changes between 1850 and 2000 are assessed for the troposphere and the lower stratosphere (up to 30 km) by a variety of seven chemistry-climate models and three chemistry-transport models. The modelled ozone changes are taken as input for detailed calculations of radiative forcing. When only changes in chemistry are considered (constant climate), the modelled global-mean tropospheric ozone column increase since pre-industrial times ranges from 7.9 DU to 13.8 DU among the ten participating models, while the stratospheric column reduction lies between 14.1 DU and 28.6 DU in the models considering stratospheric chemistry. The resulting radiative forcing is strongly dependent on the location and altitude of the modelled ozone change and varies between 0.25 Wm⁻² and 0.45 Wm⁻² due to ozone change in the troposphere and -0.123 Wm⁻² and +0.066 Wm⁻² due to the stratospheric ozone change. Changes in ozone and other greenhouse gases since pre-industrial times have altered climate. Six out of the ten participating models have performed

an additional calculation taking into account both chemical and climate change. In most models the isolated effect of climate change is an enhancement of the tropospheric ozone column increase, while the stratospheric reduction becomes slightly less severe. In the three climate-chemistry models with detailed tropospheric and stratospheric chemistry, the inclusion of climate change increases the resulting radiative forcing due to tropospheric ozone change by up to 0.10 Wm^{-2} , while the radiative forcing due to stratospheric ozone change is reduced by up to 0.034 Wm^{-2} . Considering tropospheric and stratospheric change combined, the total ozone column change is negative while the resulting net radiative forcing is positive.

21. Impact of climate change on tropospheric ozone

Granier, C., S. Bekki, F. Dentener, J.F. Muller, Y. Balkanski, I. Bey, W. Collins, L. Ganzeveld, S. Houweling, J. Olivier, R. Sander, M. Sanderson, M. Schultz, J. Sciare, D. Stevenson, W. Sturges and C. Zerefos

In: *Ozone-climate interactions*, I.S.A. Isaksen (ed), European Commission air pollution research report No.81, Brussels, 2003.

This report chapter had no abstract.

22. Atmospheric Impact of the 1783-84 Laki eruption: Part II Climatic effect of sulphate aerosol

Highwood, E.J. and D.S. Stevenson

Atmospheric Chemistry and Physics, 3, 1599-1629, 2003.

The long 1783-1784 eruption of Laki in southern Iceland was one of the first eruptions to have been linked to an observed climate anomaly, having been held responsible for cold temperatures over much of the Northern Hemisphere in the period 1783-1785. Results from the first climate model simulation of the impact of a similar eruption to that of 1783-1784 are presented. Using sulphate aerosol fields produced in a companion chemistry-transport model simulation by Stevenson *et al.* (2003), the radiative forcing and climate response due to the aerosol are calculated here using the Reading Intermediate General Circulation Model (IGCM). The peak Northern Hemisphere mean direct radiative forcing is -5.5 W m^{-2} in August 1783. The radiative forcing dies away quickly as the emissions from the volcano decrease; however, a small forcing remains over the Mediterranean until March 1784. There is little forcing in the Southern Hemisphere. There is shown to be an uncertainty of at least 50 per cent in the direct radiative forcing due to assumptions concerning relative humidity and the sophistication of the radiative transfer code used. The indirect effects of the Laki aerosol are potentially large but essentially unquantifiable at the present time. In the IGCM at least, the aerosol from the eruption produces a climate response that is spatially very variable. The Northern Hemisphere mean temperature anomaly averaged over the whole of the calendar year containing most of the eruption is -0.21 K , statistically significant at the 95 per cent level and in reasonable agreement with the available observations of the temperature during 1783.

23. Anthropogenic climate change for 1860 to 2100 simulated with the HadCM3 model under updated emissions scenarios

Johns, T.C., J.M. Gregory, W.J. Ingram, C.E. Johnson, A. Jones, J.A. Lowe, J.F.B. Mitchell, D.L. Roberts, D.M.H. Sexton, D.S. Stevenson, S.F.B. Tett and M.J. Woodage
Climate Dynamics, DOI 10.1007/s00382-002-0296-y, 2003.

In this study we examine the anthropogenically forced climate response over the historical period 1860 to present, and projected response to 2100, using updated emissions scenarios and an improved coupled model (HadCM3) that does not use flux adjustments. We concentrate on four new Special Report on Emission Scenarios (SRES), namely A1FI, A2, B2, B1 prepared for the Intergovernmental Panel on Climate Change Third Assessment Report, considered more self-consistent in their socio-economic and emissions structures, and therefore more policy relevant, than older scenarios like IS92a. We include an interactive model representation of the anthropogenic sulfur cycle and both direct and indirect forcings from sulfate aerosols, but omit the second indirect forcing effect through cloud lifetimes. The modelled first indirect forcing effect through cloud droplet size is near the centre of the IPCC

uncertainty range. We also model variations in tropospheric and stratospheric ozone. Greenhouse gas-forced climate change response in B2 resembles patterns in IS92a, but is smaller. Sulfate aerosol and ozone forcing substantially modulates the response, cooling the land, particularly northern mid-latitudes, and altering the monsoon structure. By 2100, global mean warming in SRES scenarios ranges from 2.6 to 5.3 K above 1900 and precipitation rises by one per cent per Kelvin through the twenty-first century (1.4 per cent per K omitting aerosol changes). Large-scale patterns of response broadly resemble those in an earlier model (HadCM2), but with important regional differences, particularly in the tropics. Some divergence in future response occurs across scenarios for the regions considered, but marked drying in the mid-USA and southern Europe and significantly wetter conditions for South Asia, in June-July-August, are robust and significant.

24. Interannual variability in methane growth rate simulated with a coupled ocean-atmosphere-chemistry model

Johnson, C.E., D.S. Stevenson, W.J. Collins and R.G. Derwent

Geophysical Research Letters, 29(19), 1903, DOI:10.1029/2002GL015269, 2002.

We assess the contribution made to the interannual variability of the global methane accumulation rate from its atmospheric sink using the STOCHEM tropospheric chemistry model coupled to the HadCM3 climate model. For both control and climate change scenarios, the standard deviation of the detrended accumulation rate was 1.4 ppbv per year for the period 1990-2009, compared with the measured standard deviation of 3.1 ppbv per year for the period 1984-1999. As the model emissions have no variability, the methane sink processes in the model are responsible for all the simulated variability of the methane accumulation rate. This appears to explain a significant fraction of the observed variability and was well correlated with simulated water vapour. The largest component of the model interannual variability is derived from the El-Nino Southern Oscillation cycle in the coupled ocean-atmosphere model, and this mode of variation is shown to be present in the methane accumulation rate.

25. Fresh air in the 21st Century?

Prather, M., *et al.*

Geophysical Research Letters, 30(2), 1100, DOI:10.1029/2002GL016285, 2003.

Ozone is an air quality problem today for much of the world's population. Regions can exceed the ozone air quality standards (AQS) through a combination of local emissions, meteorology favouring pollution episodes, and the clean-air baseline levels of ozone upon which pollution builds. The IPCC 2001 assessment studied a range of global emission scenarios and found that all but one projects increases in global tropospheric ozone during the twenty-first century. By 2030, near-surface increases over much of the northern hemisphere are estimated to be about 5 ppb (+2 to +7 ppb over the range of scenarios). By 2100, the two more extreme scenarios project baseline ozone increases of more than 20 ppb, while the other four scenarios give changes of -4 to +10 ppb. Even modest increases in the background abundance of tropospheric ozone might defeat current AQS strategies. The larger increases, however, would gravely threaten both urban and rural air quality over most of the northern hemisphere.

26. Test of a northwards-decreasing ²²²Rn source term by comparison of modelled and observed atmospheric ²²²Rn concentrations

Robertson, L.B., D.S. Stevenson and F. Conen

Tellus, 57B, 116-123, 2005.

Model-predicted atmospheric concentrations of Rn-222 based on two different Rn-222 source terms have been compared with observations in the lower troposphere. One simulation used a globally uniform Rn-222 source term from ice-free, the other assumed a northwards-decreasing source term (linear decrease from land surfaces of 1 atom cm⁻² s⁻¹ at 30 degrees N to 0.2 atom cm⁻² s⁻¹ at 70 degrees N). Zero emissions were assigned to oceans. The northwards-decreasing source term improved predictions at four out of six

stations north of 50 degrees N, reducing the mean prediction/observation ratio from 2.8 to 0.87. In the latitudinal band between 30 degrees N and 50 degrees N, the northwards-decreasing source term resulted in systematic under-prediction of atmospheric Rn-222, whereas the uniform source term provided predictions close to observations. Predictions based on the northwards-decreasing source term were significantly ($p < 0.01$) better than those based on the uniform source term for an averaged vertical Rn-222 profile around 44 degrees N, but were not for one around 38 degrees N. The results indicate that a northwards-decreasing source term could be a more realistic representation of actual Rn-222 emissions than a uniform $1 \text{ atom cm}^{-2} \text{ s}^{-1}$ source term. However, the decrease in Rn-222 source strength with increasing latitude might not begin at 30 degrees N, but somewhat further north. This hypothesis should be investigated through model-independent means.

27. Ozone trends according to ten dispersion models

M. Roemer and 17 others including D. Stevenson
EUROTRAC-2 Special Report, Munich, 2003.

Ten chemical dispersion models (nine European and one global) participated in an intercomparison study on the model's ozone response to an emission change. One meteorological year (1997) was chosen in combination with two emission cases: 1997 emissions, and "1987" emissions. The "1987" emissions are a rescaling of the 1997 emissions in such a way that the difference between the two implies a 40 per cent reduction in anthropogenic emissions of NMVOC and CO and a 30 per cent reduction of NO_x emissions all over Europe. Background and other emissions were kept constant. The model results of daily maximum ozone were compared with observations (EMEP) and an intercomparison of the responses (daily ozone maximum, daily maximum OH) was made. Concerning the comparison for the 1997 simulation, the following observations come forward. For the models that are driven by analysed meteorological fields, the correlation coefficient of the summer daily maximum ozone (model to observations) varies from 0.75 to 0.50. Most of the variations in daily maximum ozone are captured quite reasonably. It is also seen that the performance of the models is much better in the flat terrains of Benelux/Germany than in the mountainous areas elsewhere. An interesting feature in the model results is the elevated ozone values that are observed over the sea at the coastal areas of the Mediterranean and North Sea. This is related to the fact that dry deposition of ozone is very low over water surfaces, and it might be interesting to collect data on a regular basis in these offshore locations. The models display a north-south gradient in OH concentrations with (in the summer) approximately a factor of ten higher concentrations in the south than in the north. All models show similar patterns, but there is a large difference in absolute values: about a factor of three between models with the highest and lowest OH.

All models produced a reduction in the ozone peak values as a result of the emission reduction. But the magnitude of the reduction varied substantially from about -10 to about -25 per cent. There seems to be a fixed ranking order for the model's response, observed at nearly all sites. The response of a model depends on the ratio between controllable ozone versus uncontrollable ozone. Controllable ozone is the ozone chemically produced as a result of anthropogenic emissions. Ozone transported from outside the model domain and ozone resulting from biogenic emissions can be seen (in this experiment) as uncontrollable ozone. Potentially important elements determining this ratio are: boundary conditions, transport times, model domain, emission rates, VOC reactivity of emissions, chemistry and model resolution. Given the different characteristics of the models and the results of a few sensitivity runs performed by a few models, an explanation of differences in trends (model response) can be classified into likely and less likely candidates. Processes that are less likely to be important in explaining differences in the model's ozone response are: deposition, transport, model domain, and biogenic emissions. The latter is included based on previous experiments. Model resolution is an unresolved matter; exploration of the zooming options is required to elaborate on this further. Chemical schemes were not investigated in this study. An earlier study by Poppe concluded that ozone concentrations were relatively insensitive to

chemical schemes, although for polluted conditions the differences were not negligible. In a next phase, differences in chemical schemes deserve more attention. The VOC composition of the emissions emerged as an important candidate in explaining differences. The ozone response of one of the models increased substantially by substituting a fraction of the less reactive components by more reactive ones. This suggests that the VOC composition is important in determining the model's response of ozone to emission changes.

28. The atmospheric sulphur cycle and the role of volcanic SO₂

Stevenson, D.S., C.E. Johnson, W.J. Collins and R.G. Derwent

In: Oppenheimer, C., Pyle, D.M., and Barclay, J. (eds), *Volcanic degassing*, Geological Society, London, Special Publications, 213, 295-306, 2003a.

A global 3-D chemistry-transport model has been applied to study the atmospheric sulphur cycle, and in particular the volcanic component. The model is in general agreement with previous studies of the global S budget. We find that volcanic emissions constitute 10% of the present-day global SO₂ source to the atmosphere, but form 26% of the SO₂ burden, and 14% of the sulphate aerosol burden. Two previous modelling studies suggested the volcanic fraction of sulphate was 18% and 35%, from sources representing 7% and 14%, respectively, of the global total SO₂ emission. The results are dependent upon various assumptions about volcanic emissions (magnitude, geographical location, altitude), the global distribution of oxidants, and the physical processes of dry and wet deposition. Because of this dependence upon poorly constrained parameters, it is unclear which modelling study is closest to the truth.

29. Atmospheric impact of the 1783-84 Laki eruption: Part I. Chemistry modelling

Stevenson, D.S., C.E. Johnson, E.J. Highwood, V. Gauci, W.J. Collins and R.G. Derwent
Atmospheric Chemistry and Physics, 3, 487-507, 2003b.

Results from the first chemistry-transport model study of the impact of the 1783-1784 Laki fissure eruption (Iceland: 64 degrees N, 17 degrees W) upon atmospheric composition are presented. The eruption released an estimated 61 Tg(S) as SO₂ into the troposphere and lower stratosphere. The model has a high resolution tropopause region, and detailed sulphur chemistry. The simulated SO₂ plume spreads over much of the Northern Hemisphere, polewards of around 40 degrees N. About 70 per cent of the SO₂ gas is directly deposited to the surface before it can be oxidised to sulphuric acid aerosol. The main SO₂ oxidants, OH and H₂O₂, are depleted by up to 40 per cent zonally, and the lifetime of SO₂ consequently increases. Zonally averaged tropospheric SO₂ concentrations over the first three months of the eruption exceed 20 ppbv, and sulphuric acid aerosol reaches around 2 ppbv. These compare to modelled pre-industrial/present-day values of 0.1/0.5 ppbv SO₂ and 0.1/1.0 ppbv sulphate. A total sulphuric acid aerosol yield of 17-22 Tg(S) is produced. The mean aerosol lifetime is 6-10 days, and the peak aerosol loading of the atmosphere is 1.4-1.7 Tg(S) (equivalent to 5.9-7.1 Tg of hydrated sulphuric acid aerosol). These compare to modelled pre-industrial/present-day sulphate burdens of 0.28/0.81 Tg(S), and lifetimes of 6/5 days, respectively. Due to the relatively short atmospheric residence times of both SO₂ and sulphate, the aerosol loading approximately mirrors the temporal evolution of emissions associated with the eruption. The model produces a reasonable simulation of the acid deposition found in Greenland ice cores. These results appear to be relatively insensitive to the vertical profile of emissions assumed, although if more of the emissions reached higher levels (above 12 km), this would give longer lifetimes and larger aerosol yields. Introducing the emissions in episodes generates similar results to using monthly mean emissions, because the atmospheric lifetimes are similar to the repose periods between episodes. Most previous estimates of the global aerosol loading associated with Laki did not use atmospheric models; this study suggests that these earlier estimates have been generally too large in magnitude, and too long-lived. Environmental effects following the Laki eruption may have been dominated by the widespread deposition of SO₂ gas rather than sulphuric acid aerosol.

30. Radiative forcing from aircraft NO_x emissions: mechanisms and seasonal dependence

Stevenson, D.S., R.M. Doherty, M.G. Sanderson, W.J. Collins, C.E. Johnson and R. G. Derwent

Journal of Geophysical Research, 109, D17307, DOI:10.1029/2004JD004759, 2004.

[\(Correction to Table 4.\)](#)

A chemistry-climate model has been applied to study the radiative forcings generated by aircraft NO_x emissions through changes in ozone and methane. Four numerical experiments, where an extra pulse of aircraft NO_x was emitted into the model atmosphere for a single month (January, April, July, or October), were compared to a control experiment, allowing the aircraft impact to be isolated. The extra NO_x produces a short-lived (few months) pulse of ozone that generates a positive radiative forcing. However, the NO_x and O₃ both generate OH, which leads to a reduction in CH₄. A detailed analysis of the OH budget reveals the spatial structure and chemical reactions responsible for the generation of the OH perturbation. Methane's long lifetime means that the CH₄ anomaly decays slowly (perturbation lifetime of 11.1 years). The negative CH₄ anomaly also has an associated negative O₃ anomaly, and both of these introduce a negative radiative forcing. There are important seasonal differences in the response of O₃ and CH₄ to aircraft NO_x, related to the annual cycle in photochemistry; the O₃ radiative forcing calculations also have a seasonal dependence. The long-term globally integrated annual mean net forcing calculated here is approximately zero, although earlier work suggests a small net positive forcing. The model design (such as upper tropospheric chemistry, convection parameterization) and experimental setup (pulse magnitude and duration) may somewhat influence the results: further work with a range of models is required to confirm these results quantitatively.

31. Impacts of climate change and variability on tropospheric ozone and its precursors

Stevenson, D.S., R.M. Doherty, M.G. Sanderson, C.E. Johnson, W.J. Collins and R. G. Derwent

Faraday Discussions, 130, 41-57, DOI:10.1039/b417412g, 2005.

Two coupled climate-chemistry model experiments for the period 1990-2030 were conducted: one with a fixed climate and the other with a varying climate forced by the IS92a scenario. By comparing results from these experiments, we have attempted to identify changes and variations in physical climate that may have important influences upon tropospheric chemical composition. Climate variables considered include: temperature, humidity, convective mass fluxes, precipitation, and the large-scale circulation. Increases in humidity, directly related to increases in temperature, exert a major influence on the budgets of ozone and the hydroxyl radical: decreasing O₃ and increasing OH. Warming enhances decomposition of PAN, releasing NO_x, and increases the rate of methane oxidation. Surface warming enhances vegetation emissions of isoprene, an important ozone precursor. In the changed climate, tropical convection generally reduces, but penetrates to higher levels. Over northern continents, convection tends to increase. These changes in convection affect both vertical mixing and lightning NO_x emissions. We find no global trend in lightning emissions, but significant changes in its distribution. Changes in precipitation and the large-scale circulation are less important for composition, at least in these experiments. Higher levels of the oxidants OH and H₂O₂ lead to increases in aerosol formation and concentrations. These results indicate that climate-chemistry feedbacks are dominantly negative (less O₃, a shorter CH₄ lifetime, and more aerosol). The major mode of interannual variability in the IS92a climate experiment is ENSO. This strongly modulates isoprene emissions from vegetation via tropical land surface temperatures. ENSO is also clearly the dominant source of variability in tropical column ozone, mainly through changes in the distribution of convection. The magnitude of interannual variability in ozone is comparable to the changes brought about by emissions and climate changes between the 1990s and 2020s, suggesting that it will be difficult to disentangle the different components of near-future changes.

32. Multi-model ensemble simulations of present-day and near-future tropospheric ozone

Stevenson, D.S., *et al.*

Journal of Geophysical Research, 111, D08301, DOI:10.1029/2005JD006338, 2006.

Global tropospheric ozone distributions, budgets and radiative forcings from an ensemble of 26 state-of-the-art atmospheric chemistry models have been intercompared and synthesized as part of a wider study into both the air quality and climate roles of ozone. Results from three 2030 emissions scenarios, broadly representing 'optimistic', 'likely', and 'pessimistic' options, are compared to a base year 2000 simulation. This base case realistically represents the current global distribution of tropospheric ozone. A further set of simulations considers the influence of climate change over the same time period, by forcing the central emissions scenario with a surface warming of around 0.7K. The use of a large multi-model ensemble allows us to identify key areas of uncertainty, and improves the robustness of the results. Ensemble mean changes in tropospheric ozone burden between 2000 and 2030, for the three scenarios, range from a five per cent decrease, through a six per cent increase, to a 15 per cent increase. The inter-model uncertainty (\pm one standard deviation) associated with these values is about \pm 25 per cent. Model outliers have no significant influence on the ensemble mean results. Combining ozone and methane changes, the three scenarios produce radiative forcings of -50, 180 and 300 mW m⁻², compared to a CO₂ forcing over the same time period of 800 to 1,100 mW m⁻². These values indicate the importance of air pollution emissions in short-medium term climate forcing, and the potential for stringent/lax control measures to improve/worsen future climate forcing.

The model sensitivity of ozone to imposed climate change varies between models, but modulates zonal mean mixing ratios by \pm 5 ppbv via a variety of feedback mechanisms, in particular those involving water vapour and stratosphere-troposphere exchange. This level of climate change also reduces the methane lifetime by around four per cent. The ensemble mean year 2000 tropospheric ozone budget indicates chemical production, chemical destruction, dry deposition and stratospheric input fluxes of 5,100, 4,650, 1,000 and 550 Tg(O₃) yr⁻¹, respectively. These values are significantly different to the mean budget documented by the Intergovernmental Panel on Climate Change (IPCC) Third Assessment Report (TAR). The mean ozone burden (340 Tg(O₃)) is 10 per cent larger than the IPCC TAR estimate, whilst the mean ozone lifetime (22 days) is 10 per cent shorter. Results from individual models show a correlation between ozone burden and lifetime, and each model's ozone burden and lifetime respond in similar ways across the emissions scenarios. The response to climate change is much less consistent. Models show more variability in the tropics compared to mid-latitudes. Some of the most uncertain areas of the models include: treatments of deep tropical convection, including lightning NO_x production; isoprene emissions from vegetation and isoprene's degradation chemistry; stratosphere-troposphere exchange; biomass burning; and water vapour concentrations.

33. Multi-model simulations of carbon monoxide: Comparison with observations and projected near-future changes

Shindell, D.T., G. Faluvegi, D.S. Stevenson, *et al.*

Journal of Geophysical Research, 111, D19306, doi:10.1029/2006JD007100, 2006.

We have analyzed present-day and future simulations of carbon monoxide (CO) in 26 state-of-the-art atmospheric chemistry models run as part of a study of future air quality and climate change. In comparison with near-global satellite observations from the MOPITT instrument and local surface measurements, the models show large underestimates of CO in the Northern Hemisphere (NH) extra-tropics, especially over East Asia, while performing reasonably well over most other parts of the globe. These results suggest that year-round emissions, probably from fossil-fuel burning, in East Asia, and seasonal biomass burning emissions in South-Central Africa, are greatly underestimated in current inventories such as EDGAR3.2, consistent with results from inversion model studies. The variability between models is large, likely resulting primarily from differences in the models' hydrologic cycles

and their representations and emissions of non-methane volatile organic compounds (NMVOCs). In the future, the models' global mean projections of changes in CO in response to emissions changes are quite robust. For the high-emissions A2 scenario, global mean mid-tropospheric (500 hPa) CO increases by 12.6 ± 3.5 ppbv (16 per cent). For the low-emissions MFR scenario, 500 hPa CO decreases by 8.1 ± 2.3 ppbv (11 per cent), while for the mid-range CLE scenario the change is 1.7 ± 1.8 ppbv (two per cent).

The effect of projected climate change is to decrease global 500 hPa CO by 1.4 ± 1.4 ppbv. Local changes can be much larger, however. In response to climate change, substantial effects are seen in the tropics, but inter-model variability is quite large. In response to emissions changes, the CO responses are still robust across models at regional scales. In the CLE scenario, near-surface (850 hPa) CO increases by more than 10 ppbv over Indonesia during boreal fall and over South Africa year-round, while decreasing by 10-20 ppbv over much of the industrialized NH with largest changes during spring. In the MFR scenario, decreases occur nearly everywhere year-round, though these are small in the Southern Hemisphere (SH). In the NH, peak decreases are 30-50 ppbv over East Asia and the North East US and South East Canada, with decreases of 20-30 ppbv over most of Europe. In the A2 scenario, near-surface CO increases worldwide and year-round, with the largest changes over central Africa (20-30 ppbv), southern Brazil (20-35 ppbv) and South and East Asia (30-70 ppbv). The trajectory of future emissions thus has the potential to profoundly affect air quality over most of the world's populated areas.

[34. Climate impact of tropospheric ozone changes](#)

Stordal, F., S. Bekki, R. van Dorland, D. Hauglustaine, M. Millan, E. Schuepbach, R. Sausen, D. Stevenson and A. Volz-Thomas
In: *Ozone-climate interactions*, I.S.A. Isaksen (ed), European Commission air pollution research report No.81, Brussels, 2003.

This report chapter had no abstract.

[35. Multi-model simulations of tropospheric NO₂ compared with GOME retrievals for the year 2000](#)

van Noije, T.P.C., *et al.*,
Atmospheric Chemistry and Physics, 6, 2943-2979, 2006.

We present a systematic comparison of tropospheric NO₂ from 17 global atmospheric chemistry models with three state-of-the-art retrievals from the Global Ozone Monitoring Experiment (GOME) for the year 2000. The models used constant anthropogenic emissions from IIASA/EDGAR3.2 and monthly emissions from biomass burning based on the 1997-2002 average carbon emissions from the Global Fire Emissions Database (GFED). Model output was analyzed at 10:30 local time, close to the overpass time of the ERS-2 satellite, and co-located with the measurements to account for sampling biases due to incomplete spatiotemporal coverage of the instrument. We assessed the importance of different contributions to the sampling bias: correlations on seasonal time scale give rise to a positive bias of 30 to 50 per cent in the retrieved annual means over regions dominated by emissions from biomass burning. Over the industrial regions of the eastern United States, Europe and eastern China, the retrieved annual means have a negative bias with significant contributions (between -25 and +10 per cent of the NO₂ column) resulting from correlations on time scales from a day to a month.

We present global maps of modelled and retrieved annual mean NO₂ column densities, together with the corresponding ensemble means and standard deviations for models and retrievals. The spatial correlation between the individual models and retrievals are high, typically in the range 0.81-0.93 after smoothing the data to a common resolution. On average, the models underestimate the retrievals in industrial regions, especially over eastern China and over the Highveld region of South Africa, and overestimate the retrievals in regions dominated by biomass burning during the dry season. The discrepancy over South

America south of the Amazon disappears when we use the GFED emissions specific to the year 2000. The seasonal cycle is analyzed in detail for eight different continental regions. Over regions dominated by biomass burning, the timing of the seasonal cycle is generally well reproduced by the models. However, over Central Africa south of the Equator, the models peak one to two months earlier than the retrievals.

We further evaluate a recent proposal to reduce the NO_x emission factors for savanna fires by 40 per cent and find that this leads to an improvement in the amplitude of the seasonal cycle over the biomass burning regions of Northern and Central Africa. In these regions, the models tend to underestimate the retrievals during the wet season, suggesting that the soil emissions are higher than assumed in the models. In general, the discrepancies between models and retrievals cannot be explained by *a priori* profile assumptions made in the retrievals, nor by diurnal variations in anthropogenic emissions, which lead to a marginal reduction of the NO_2 abundance at 10:30 local time (by 2.5 to 4.1 per cent over Europe). Overall, there are significant differences among the various models and, in particular, among the three retrievals. The discrepancies among the retrievals (10-50 per cent in the annual mean over polluted regions) indicate that the previously estimated retrieval uncertainties have a large systematic component. Our findings imply that top-down estimations of NO_x emissions from satellite retrievals of tropospheric NO_2 are strongly dependent on the choice of model and retrieval.

Appendix 2: Conference, workshop and other presentations

Hyperlinks to most talks are available in the electronic version of this document (as Microsoft PowerPoint presentations, .ppt), along with posters (as Adobe Portable Document Files, .pdf).

2001:

RMS Atmospheric Chemistry Specialist Group meeting, University of Nottingham, April 10, poster: [‘Tropospheric ozone simulations from a global model’](#).

SPARC Workshop on the Tropopause, Bad Tölz, Germany, April 17-21, invited talk: ‘Future chemical/climate change in the upper troposphere’.

Eighth European Symposium on the Physico-Chemical Behaviour of Atmospheric Pollutants, ‘A Changing Atmosphere’, Torino, Italy, September 17-20, talk: [‘Global influences on future European surface ozone’](#).

‘Origins, Emissions and Impacts of Volcanic Gases’, meeting of the Volcanic and Magmatic Studies Group of the Geological Society, London, 25-26 October, poster: [‘The atmospheric sulphur budget and the contribution of passive and eruptive volcanic emissions’](#).

2002:

University of Oxford, talk to the Department of Atmospheric, Oceanic, and Planetary Physics, 21 February, [‘Modelling global tropospheric ozone: Implications for future air quality and climate’](#).

EUROTRAC2 Symposium ‘Transport and chemical transformation in the troposphere’, Garmisch-Partenkirchen, March 11-15, poster: [‘Comparison of surface ozone at TOR-2 sites with a global model’](#); talk: ‘Seasonal cycles of ozone across Europe’ (Lindskog *et al.*); talk: ‘Vertical uplifting and regional modelling’ (Beekmann *et al.*).

EGS 27th General Assembly, Nice, France, 21-26 April, talk: ‘Influences on 21st century European tropospheric ozone’; poster: [‘UGAMP-UKMO tropospheric chemistry model intercomparison’](#); poster: [‘Comparison of surface ozone at TOR-2 sites with a global model’](#).

‘Volcanism and the Earth’s Atmosphere’, AGU Chapman Conference, Santorini, Greece, 17-21 June, talk: [‘The 1783-1784 Laki eruption: Modelling the impact on atmospheric composition’](#).

Seventh Scientific Conference of IGAC, Heraklion, Greece, 18-25 September, poster: [‘Projected changes in global tropospheric ozone in 2030’](#).

UGAMP Tropical Workshop, University of Reading, 6 November, talk: ‘Tropical tropospheric chemistry’.

2003:

EGS-AGU-EUG Joint Assembly, Nice, France, April 6-11, talk: [‘Production and removal of volcanic aerosol at tropopause levels: A model study of the 1783-1784 Laki eruption’](#); talk (presenter R. Doherty): ‘The influence of present and future interannual climate variability on tropospheric ozone and methane as simulated by a coupled climate-chemistry model’; poster: ‘Volcanic influences on the atmospheric methane budget’; poster: ‘Use of a latitudinally-dependant ²²²Rn emission distribution for improving atmospheric transport model validations’.

RMS Conference 2003, UEA, Norwich, September 1-5, talk (presenter R. Doherty): ‘The influence of interannual climate variability on tropospheric ozone as simulated by a coupled climate-chemistry model’.

2004:

University of Leeds, talk to the School of Environment, 24 February: '[Atmospheric impact of the 1783-1784 Laki volcanic eruption](#)'.

University of Oxford, talk to the Department of Atmospheric, Oceanic, and Planetary Physics, 26 February, '[Modelling the atmospheric sulphur cycle and the impact of the 1783-1784 Laki \(Iceland\) volcanic eruption](#)'.

RMS Atmospheric Chemistry Specialist Group meeting, University of Leicester, April 1, talk: '[2020 Vision: Modelling the near future tropospheric composition](#)'.

Eighth IGAC Conference, Christchurch, New Zealand, 4-9 September, poster: '[Radiative forcing from aircraft NOx emissions: mechanisms and seasonal dependence](#)'; poster: '[Interaction between stratospheric and tropospheric chemistry in a coupled chemistry-climate model](#)'.

ACCENT Transport and Transformation of Pollutants Workshop, Barnsdale, Rutland, October, talk: '[Chemistry-climate modelling: impacts of climate change on tropospheric chemical composition](#)'.

2005:

ACCENT/IPCC-AR4 Workshop, Oslo, Norway, January 17-18, talk: 'Impacts of emissions and climate change on tropospheric ozone'.

Presentation to AQEG, Defra, London, January 20, '[Overlaps of air quality and climate policy: Global modelling perspectives](#)'.

APRIL workshop on regional air quality modelling, London, 16 March, talk: '[Influence of future climate change on air quality – global model results](#)'.

Faraday Discussion 130: Atmospheric Chemistry, University of Leeds, April 11-13, talk: '[Impacts of climate change and variability on tropospheric ozone and its precursors](#)'.

Global Change Seminar, UoE, 17 May, '[What will control future tropospheric ozone?](#)'

ACCENT Workshop 'Development of modelling facility (benchmark) for climate/chemistry and large scale pollution studies', Oslo, Norway, 27-29 June, talk: '[ACCENT intercomparison experiment two](#)'.

NERC UTLS Workshop 'Water vapour in the upper troposphere and lower stratosphere', Lancaster University, 5-7 July, talk: '[Climate feedbacks on tropospheric ozone](#)'.

First ACCENT Symposium, Urbino, Italy, 12-16 September, talk: '[Multi-model ensemble simulations of tropospheric ozone](#)'.

RMS biennial conference, Exeter, 12-16 September, poster: '[Multi-model ensemble simulations of tropospheric ozone](#)' (presented by W.J. Collins).

SPARC CCMval Workshop, Boulder, USA, October 17-19, Invited talk: '[Multi-model ensemble simulations of tropospheric ozone](#)'.

ACCENT Transport and Transformation of Pollutants Workshop, Barnsdale, Rutland, October 31- November 2, talk: '[Nitrogen deposition on regional and global scales: a multi-model evaluation](#)'.

2006:

Intercontinental Transport Modelling Intercomparison Organizational Workshop:

Washington D.C., USA, January 30-31, poster (presenter R. Doherty): '[Assessing long range transport of ozone in chemistry-transport models](#)'.

Appendix 3: Teaching, supervision and public engagement

2001:

Taught 9 x 2-hour course on 'Climate Change' for the Office of Lifelong Learning, UoE.
Supervised NERC-funded PDRA, Ruth Doherty.

2002:

Taught 9 x 2-hour course on 'Climate Change' for the Office of Lifelong Learning, UoE.
Supervised NERC-funded PDRA, Ruth Doherty.

2003:

Taught 4 x 1-hour lectures on 'Weather Forecasting and Climate Prediction' for first-year undergraduate course on Meteorology.
Supervised NERC-funded PDRA, Ruth Doherty.

2004:

Taught a 2.5-hour session on 'Hazards: Volcano and Fire' for the MSc 'Remote Sensing and Image Processing'.
Taught 2 x 1-hour lectures on 'Weather Forecasting and Climate Prediction' for first-year undergraduate course on Meteorology.
Supervised NERC-funded PDRA, Ruth Doherty.

2005:

Taught a 3-hour session on 'Climate Change' for the MSc module 'Air Quality and Global Change'; also set and marked exam question for this course.
Co-supervised two final-year undergraduate projects in Geophysics.
Supervised NERC-funded PDRA, Ruth Doherty.
Interview for Scottish TV on climate change and the Scottish skiing industry.
Interview for Scottish TV on climate change and severe weather in Scotland.

2006:

Co-supervised three final-year undergraduate projects in Geophysics.
Supervised UoE-funded PhD student, Lucia Viegas de Barros.
Supervised NERC-funded PDRA, Richard Damoah.
'Café Scientifique' talk to the Edinburgh public on climate change.
Interviews for Edinburgh Evening News on air quality in Edinburgh and climate change in Scotland.
Interview for Edinburgh University radio station on [Climate Change and Ozone](#).

Appendix 4: Contributions to atmospheric science research

Reviews of papers, reports and proposals; proposal review panels and thesis examinations.

2001: Two papers reviewed for *Journal of Geophysical Research*.

2002: Three papers reviewed for *Journal of Geophysical Research*;
Two papers reviewed for *Journal of the Geological Society of London*;
One paper reviewed for *Geophysical Research Letters*;
One paper reviewed for *Bulletin of Volcanology*;
One proposal reviewed for the Norwegian Research Council.

2003: One paper reviewed for *Journal of Geophysical Research*;
Reviewed Chapter 5 (*Volcano Monitoring and Public Safety*) of GMES-GATO (Global Monitoring for Environment and Security – Global Atmospheric Observations) report;
Examined M.Phil. thesis (Cambridge University);
Reviewed four NERC Standard Grant proposals;
Sat on NERC Standard Grant review panel (Environmental Chemistry).

2004: Two papers reviewed for *Journal of Geophysical Research*;
Reviewed six Standard Grant proposals for NERC;
Reviewed two Small Grant proposals for NERC;
Reviewed one Fellowship proposal for NERC;
Reviewed one Knowledge Transfer Grant proposal for NERC;
Sat on two NERC Standard Grant panels (Climate Change, Paleoclimatology and Meteorology; and Chemistry and Physics of the Environment);
Four proposals reviewed for the Norwegian Research Council.

2005: Three papers reviewed for *Journal of Geophysical Research*;
Two papers reviewed for *Atmospheric Chemistry and Physics Discussions*;
One paper reviewed for *Atmospheric Environment*;
One paper reviewed for *Geophysical Research Letters*;
Examined Ph.D. thesis (Edinburgh University);
Reviewed 10 Standard Grant proposals for NERC;
Reviewed three Small Grant proposals for NERC;
Reviewed three Fellowship proposals for NERC;
Sat on Masters Review Panel (MRes/GIS/Remote Sensing/Modelling) for NERC;
One proposal reviewed for the Norwegian Research Council.

2006: Two papers reviewed for *Journal of Geophysical Research*;
One paper reviewed for *Journal of Volcanology and Geothermal Research*;
Examined Ph.D. thesis (Edinburgh University);
Reviewed AQEG report on *Air Quality and Climate Change*.

In June 2003 I became a member of the newly formed NERC Peer Review College. In February 2006 I received a citation for being an 'outstanding referee' from the *Journal of Geophysical Research*.

Appendix 5: Research proposals

Over the course of the fellowship, several research proposals have been prepared.

Successful:

NERC Standard Grant, December 2004: 'Coupled stratosphere-troposphere chemistry-climate model studies' (PI M. Chipperfield, University of Leeds, Col D. Stevenson, UoE), total value £300,000.

NERC Standard Grant, December 2005: 'Transport processes and ozone budgets in the upper troposphere - a synthesis of EOS MLS measurements and chemistry transport model studies', total value £274,000

Unsuccessful:

NERC Standard Grant, December 2003: 'Biomass burning in tropospheric chemistry-climate models'; graded α_4 , but not funded.

NERC Standard Grant, July 2005: 'Quantifying uncertainties in climate-tropospheric chemistry interactions and feedbacks'; graded α_4 , but not funded.

NERC Small Grant, September 2005: 'Measuring and modelling vertical profiles of ^{222}Rn in the troposphere'; graded α_3 , not funded.

NERC Fellowship extension, November 2005: 'Inter-comparing, validating and applying chemistry-climate models'; not funded.

List of abbreviations

A1FI	An IPCC SRES high-growth, fossil fuel-intensive scenario
A2	An IPCC SRES high-growth scenario
ACCENT	Atmospheric Composition Change: a European Network of Excellence
AGU	American Geophysical Union
AOT40	'Accumulated Ozone over a Threshold of 40 ppbv', typically measured in ppbv-hrs, an index of vegetation exposure.
APRIL	Air Pollution Research In London
AQ	Air quality
AQEG	Air Quality Expert Group (Defra)
AQS	Air quality standards
ATC	Annual tropospheric column
AZM	Annual zonal mean
B1	An IPCC SRES low-growth scenario
B2	An IPCC SRES medium-growth scenario
C ₅ H ₈	Isoprene
CCM	Chemistry-climate model
CCMval	CCM validation (A SPARC activity)
CH ₄	Methane
CLE	Current legislation (IIASA scenario)
CO	Carbon monoxide
CTM	Chemistry-transport model
Defra	Department for Environment, Food and Rural Affairs
DMS	Dimethyl sulphide
DU	Dobson units
EDGAR	Emissions Database for Global Atmospheric Research
EGS	European Geophysical Society
EM	Ensemble mean
EMEP	Co-operative programme for monitoring and evaluation of the long-range transmissions of air pollutants in Europe
ENSO	El Niño Southern Oscillation
EU	European Union
EUREKA	A pan-European research co-ordination organisation
EUROTRAC-2	The EUREKA project on the <i>Transport and Chemical Transformation of Trace Constituents in the Troposphere over Europe</i> ; Second Phase
GATO	A subproject of GMES, Global Atmospheric Observations
GCM	General circulation model
GFED	Global Fire Emissions Database
GIS	Geographic information systems
GMES	Global Monitoring for Environment and Security
GWP	Global warming potential
H ₂ O ₂	Hydrogen peroxide
HadAM3	Hadley Centre Atmosphere-only Climate Model (version 3)
HadCM3	Hadley Centre Coupled Ocean-Atmosphere Climate Model (version 3)
HNO ₃	Nitric acid
HO ₂	Hydroperoxy radical
HO _x	The sum of OH and HO ₂
hPa	Hectopascal (100 Pascals)
IGAC	International Global Atmospheric Chemistry Project
IIASA	International Institute for Applied Systems Analysis
IPCC	Intergovernmental Panel on Climate Change

IPCC TAR	IPCC Third Assessment Report on Climate Change (published 2001)
IPCC-AR4	IPCC Fourth Assessment Report on Climate Change (due 2007)
IS92a	An IPCC 'business as usual' scenario
JJA	June-July-August
MOPITT	Measurements Of air Pollution In The Troposphere
MOSES2.2	Met Office Surface Exchange Scheme (version 2.2)
MOZAIC	Measurement of Ozone And water vapour by In-service airCRAFT
MFR	Maximum feasible reductions (IIASA scenario)
NERC	Natural Environment Research Council
NH	Northern Hemisphere
NH ₃	Ammonia
NH _x	Sum of all reactive reduced nitrogen compounds
NMHC	Non-methane hydrocarbons
NMVOG	Non-methane volatile organic compounds
NO	Nitric oxide
NO ₂	Nitrogen dioxide
NO _x	The sum of NO and NO ₂
NO _y	The sum of all oxidized nitrogen species
O ₃	Ozone
OH	Hydroxyl radical
RMS	Royal Meteorological Society
²²² Rn	The radionuclide Radon-222
SH	Southern Hemisphere
SO ₂	Sulphur dioxide
SO _x	Sum of all oxidised sulphur compounds
SOMO35	'Daily maximum eight-hour mean ozone over 35 ppbv', a health index, units: ppbv-days
SPARC	Stratospheric Processes and their Role in Climate
SRES	Special Report on Emissions Scenarios
STE	Stratosphere-troposphere exchange
STOCHEM	Stochastic Chemistry Model
TCO	Tropospheric column ozone
TOR-2	Tropospheric Ozone Research (phase 2) – a EUROTRAC-2 sub-project
UEA	University of East Anglia
UGAMP	Universities Global Atmospheric Modelling Program
UoE	The University of Edinburgh
UT	Upper troposphere
UTLS	Upper troposphere lower stratosphere
VOC	Volatile organic compound
WHO	World Health Organisation

References & Bibliography

The first section lists all peer-reviewed publications resulting from the fellowship (including those in press, submitted for publication, and those currently in preparation – see Appendix 1 for further details); the second section lists additional references used in the report.

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