AIR QUALITY EXPERT GROUP

Ozone in the United Kingdom

Prepared for:
Department for Environment, Food and Rural Affairs;
Scottish Executive; Welsh Assembly Government; and
Department of the Environment in Northern Ireland

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Terms of reference

The Air Quality Expert Group (AQEG) was set up in 2001 to provide independent scientific advice on air quality, in particular on the air pollutants contained in the Air Quality Strategy for England, Scotland, Wales and Northern Ireland and those covered by the EU Directive on ambient air quality assessment and management (the Air Quality Framework Directive).

AQEG reports to the Secretary of State for Environment, Food and Rural Affairs, Scottish Ministers, the National Assembly for Wales and the Department of the Environment in Northern Ireland (the Government and Devolved Administrations). AQEG is an advisory non-departmental public body in England, Wales and Northern Ireland. In terms of the Scotland Act 1998, the Group is a jointly-established body.

AQEG's main functions are:

- to give advice to ministers on levels, sources and characteristics of air pollutants in the UK;
- to assess the extent of exceedences of Air Quality Strategy objectives and proposed objectives, EU limit values and proposed or possible objectives and limit values, where monitoring data is not available;
- to analyse trends in pollutant concentrations;
- to assess current and future ambient concentrations of air pollutants in the UK; and
- to suggest potential priority areas for future research aimed at providing a better understanding of the issues that need to be addressed in setting air quality objectives.

The Group will not give approval for products or equipment.

Further information on AQEG can be found on the Group's website at:
http://www.defra.gov.uk/environment/airquality/panels/aqeg/index.htm.

Information on these pages includes the dates, agendas, and minutes of meetings as they become available, a list of the members, the Register of Interests, and draft and final reports as they become available.

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Table of contents

Executive summary

High ozone concentrations in the atmosphere near the ground are of concern because of potential effects on human health and damage to vegetation. Air quality strategies in Europe and elsewhere in the world have been directed towards measures to limit ozone levels. However, ozone presents a difficult control problem because it is a gas created in the atmosphere and not one directly emitted from processes that can be regulated, and its creation can take place over a wide range of time and distance scales. This report by the Air Quality Expert Group (AQEG) provides scientific evidence to inform UK control strategies for ozone.

A network of ozone monitors exists in the UK and has been used to measure ozone continuously since 1986. The measurements indicate variability from hour to hour, day to day and from season to season. This report recognises that there are a number of ways of summarising ozone concentrations. However, policy imperatives to cover health and ecological impacts mean that there is no preferred summary ozone statistic (known as an ozone metric) with which to describe ozone effects. A number of ozone metrics are therefore used in this report. Both long-term and short-term average ozone concentrations (for example annual and hourly averages) need to be considered, so ozone metrics broadly fall into these two categories. AQEG was not able to propose a method of simplifying the analysis and interpretation of ozone metrics.

The ozone metrics used in this report relate to standards for protecting health; they indicate that current ozone levels can exceed internationally accepted guidelines. The focus in this report has been on interpreting recent ozone trends and predicting future trends, rather than looking at exceedences. Changes in ozone concentrations are subject to influences related to ozone-producing sources, meteorology and chemical reactions over urban, regional and hemispherical distances.

A detailed analysis of measurements over the last few years shows there is some variability from year to year because of fluctuations in the occurrence of hot summer weather conditions, associated with episodes of high ozone lasting a few days. However, there is evidence of an increase in hemispheric background ozone concentrations, expressed as an annual average trend, as a result of increases in global emissions. At the same time, control of nitrogen oxides (NO_X) emissions in the UK has led to an increase in ozone in urban areas, although controls of NO_X and volatile organic compound (VOC) emissions in the UK and Europe have led to decreases in the intensity of summer ozone episodes. In the future, up to 2030, these trends may continue. Most uncertainty is attached to the trend in hemispheric background ozone. It depends on whether global emissions of ozone precursors will increase or decrease. If the latter occurred it could lead to a decrease in the annual mean background concentration.

The complexity and spatial scale of processes leading to ozone production means that interpretation and forecasting of ozone has depended on calculations. Some of these calculations have made use of both measurements and mathematical models in combination to predict future ozone levels at urban monitoring sites in the UK. Other theoretical models rely on the best current understanding of chemical and physical processes, leading to predictions of ozone concentrations over regions and globally. These process models can be very complex, making full use of available computing power, especially when aspects of climate change are included. A full evaluation of these models has not been made by AQEG. However, confidence in models is such that AQEG finds the use of a number of well-evaluated models, of differing complexity, acceptable, when they are used to identify the magnitude of changes in ozone as a result of moderate changes in emissions (that is, changes within foreseeable emission scenarios). There has been encouraging agreement between different types of model.

In urban areas where most of the UK population lives, AQEG concludes that over the next two decades ozone will rise and tend towards levels in the surrounding rural areas, since suppression of urban ozone by nitric oxide (NO) will be reduced as nitrogen oxide emissions fall. This effect will be superimposed on any regional and global trends in rural ozone.

Complex global models provide boundary conditions for models of ozone behaviour on a regional scale. They also help to identify climate change processes leading to changes in future global ozone concentrations, but the models cannot incorporate all known processes, including feedback. For predictions to 2030, the influence of climate change is uncertain, but its effect on mean surface ozone is thought to be small compared to the direct effect of changes in regional and global emissions of ozone-producing gases. Climate change may have relatively greater influence on future episodes of peak ozone in particular geographic areas. Care must be taken with regional and global models that results are not applied where they are inappropriate. For example, they may not be applied without further corrections to urban areas, because of the small distance scales involved in NO_X-ozone interactions in such environments. Further development of more complex urban, regional and global models is likely in future, but extensive, documented evaluation is necessary before a model should be used to determine an emissions control strategy.

Chemical transport models can be used to determine the impact of a source in one location on the concentration of ozone in another. However, for ozone such relationships can be very complex, because of the way different chemical species interact and the way ozone metrics are applied. Instead, this report has looked at the most effective control options to reduce ozone using a number of metrics, where the control option is a broad measure, that is, a reduction of all emissions in one sector, rather than control of a specified source. It turns out that for some of the ozone metrics considered, levels are unlikely to show much change as a result of foreseeable emission reductions. This is partly because these ozone metrics depend on influences over urban, regional and global scales. Regional influences are dependent on combined natural and man-made VOC emissions and the implementation of measures to reduce natural VOC emissions would be difficult to apply. Studies of the impact of foreseeable measures on UK emissions to reduce ozone within the UK also indicate that there is little scope to easily make large ozone reductions. AQEG considers that it is important to understand the factors which influence an ozone metric before making emission control choices.

An annex to the report shows a range of possible future UK and global emission scenarios. AQEG has considered the additional measures needed, beyond foreseeable measures, to achieve compliance with a range of ozone metrics, including the scenario involving the maximum technically feasible reduction of VOC and NO_X emissions in the UK and Europe. For some of the emission scenarios considered there would be a worsening of the health-related ozone metrics caused by the increase in urban ozone. There is firm evidence from both modelling and measurement that local action has only limited impact on air pollution episodes of high ozone in the UK. Large-scale reductions of 60% or more, in both the UK and Europe, of VOC and NO_X emissions throughout the UK and Europe would be necessary to reduce ozone concentrations in urban areas, given the increases that will result from the decreased suppression of ozone by NO, referred to above, and the possible future increases in background ozone. Only some of the scenarios approach the scale of reduction needed to bring about the reduction of ozone in urban areas, so that further careful assessment of practical policy options is needed. In addition to local and regional reductions of NO_X and man-made VOCs, these should take into account the impact of future global methane and carbon monoxide (CO) emissions, emissions of NO_X from shipping worldwide and natural emissions of VOCs in Europe.

This report necessarily confines its discussion to ozone and the likely future dependence of ozone levels on precursor emissions and a number of other factors, such as weather and climate. NO_X and VOC emissions not only lead to ozone formation, but also influence levels of other health-related pollutants, especially particulate matter and nitrogen dioxide. In addition, these emissions affect climate through their impact on ozone, methane and secondary aerosol. AQEG continues to argue that a holistic approach is essential, with policy taking a full and combined account of changes in precursor emissions on all of these pollutants and of human exposure to them, together with the effects on climate.

The main chapters of this report answer eight questions posed by the Department for Environment, Food and Rural Affairs (Defra). Each chapter takes the form of a short answer, followed by a more detailed answer and then by supporting evidence. The following box contains the questions and short answers.

Question A: *A large quantity of urban and rural monitoring data has been collected since the last Photochemical Oxidants Review Group (PORG) report, by Defra's own networks, local authority stations and others. What does this reveal in terms of trends (using metrics considered relevant to effects) and spatial concentration patterns?*

Annual mean ozone concentrations have generally increased over the last ten years or so in urban areas, while the changes in concentrations in rural areas are less marked and show variations with location and time. Ozone concentrations are generally lower in urban areas than in the surrounding rural areas due to reaction with NO_X (mostly NO) emissions, which are greatest in urban areas. The main cause of the increase in urban areas is the reduction in NO_X emissions, which has led to a decrease in this "urban decrement", reducing the differences between urban and rural concentrations.

Reductions in precursor emissions in the European region have led to reductions in peak ozone concentrations at rural sites, although there are significant variations from year to year due to the weather, with higher concentrations generally measured in years with hotter summers (such as 1995, 2003 and 2006). The trends in some of the health-based ozone metrics are influenced by both changes in mean and peak concentrations.

Question B: *Observations since the 1970s have shown that global background ozone concentrations have been rising throughout this period. What is the strength of these data, and what is the evidence concerning the trends and likely projections of precursor emissions, and the resultant ozone concentrations?*

An international policy review has concluded that there is strong evidence that background ozone concentrations in the northern hemisphere have increased by up to 10 μg m^{-3} per decade over the last 20-30 years (Raes and Hjorth, 2006). This increase has been attributed to the growth in man-made ozone precursor emissions from industry, road, air and ship transport, homes and agriculture. Future ozone concentrations depend on which of the possible future emission scenarios is followed. Future annual mean surface ozone concentrations in the southern half of the United Kingdom are modelled to increase by about 6 μg m^{-3} in a "current legislation" (IIASA CLE) scenario and to decrease by about 4 μg m^{-3} in a "maximum technically feasible reduction" (IIASA MFR) scenario between 2000 and 2030. Observed background ozone concentrations in air masses entering north-west Europe over the 2000-2006 period have remained level and have shown no overall trend.

Question C: *What is the likely impact of climate change on future ozone levels in Europe, over the next two decades? What is the significance of such impacts compared to other influences, such as inter-annual variability or (global and regional) emission trends?*

The net impact of climate change on mean surface ozone levels over Europe on the 2030 time horizon is not known with any confidence but is likely to be small compared with the most important influence. This is the change in anthropogenic emissions in Europe and throughout the whole northern hemisphere of the important precursor gases to ozone formation: NO_X, methane (CH_4) and non-methane VOCs, in particular, and CO. Climate change may have relatively greater influence on future peak episodic ozone in particular geographic areas through a number of different mechanisms such as changes in precursor emissions, ozone loss by deposition and meteorology. Inter-annual variability in annual mean surface ozone at a given location is large compared with the likely magnitude of net ozone change by 2030, so multi-year data series are necessary for unravelling the competing influences on ozone concentration at different locations.

Question D: *What are the likely future trends in urban ozone concentrations over the next two decades and what is driving them?*

Urban ozone concentrations are expected to rise over the next two decades and to tend towards the concentrations found in the rural areas that surround them. These increases in urban ozone concentrations are largely driven by vehicle emission controls that have brought about a reduction in NO_X emissions in urban areas. Road traffic NO_X emissions have previously depressed urban ozone levels and although this scavenging effect is being diminished by pollution controls, many urban areas in the UK are still expected to have lower ozone concentrations in 2020 than those in the surrounding rural areas. Urban ozone concentrations will also respond to the changes occurring to ozone in the surrounding rural areas, largely driven by changes on the hemispheric/global scale. Depending on the strength of these trends, these could also cause increases in urban ozone, which will be in addition to the NO_X-scavenging effect.

Question E: *Ozone is currently modelled on a number of spatial and temporal scales. What are the main uncertainties associated with such work, and what research is required to reduce these uncertainties?*

Although a number of models address ozone on a range of temporal and spatial scales across the UK, there is no consistent and comprehensive understanding of model performance and the uncertainties that affect them. Research is required to understand and intercompare the influence of different spatial and temporal resolutions, chemical mechanisms and parameterisations upon predicted concentrations and their policy implications. This process would involve harmonising model performance evaluation and collecting information on uncertainties of the various model formulations. Research is also required to evaluate the relative importance of man-made and natural biogenic sources of ozone precursors.

Question F: *Integrated assessment modelling to support the European Commission's Thematic Strategy for Air Quality suggests that regional ozone levels in the UK are likely to remain relatively steady regardless of foreseeable emission reductions across Europe. Does the Group agree with this analysis and what is the explanation for this lack of response to reductions in precursor emissions?*

AQEG agrees that under the specific emission scenarios considered for the European Commission's Thematic Strategy, regional ozone levels in the UK (based on the SOMO35 metric) would be likely to remain steady in the foreseeable future. However, this does not indicate that regional ozone levels in the UK are insensitive to precursor emissions in European countries and the surrounding seas, especially for episodes of high concentrations.

Question G: *What are likely to be the most effective control options to reduce UK population exposure to ozone (in terms of precursors to be targeted) and on what scale should they operate? The Group may include discussion of the types of controls they consider to be feasible, but do not need to consider the policy implications of such measures.*

The ozone precursor compounds of relevance are methane, non-methane volatile organic compounds (VOC), oxides of nitrogen and carbon monoxide. While UK action can be beneficial, effective control of ozone concentrations in the UK will require emission reductions to be implemented throughout Europe and increasingly the entire northern hemisphere. Local actions, especially those of a short-term nature to address episodes of high ozone concentrations, have generally had or been simulated to have limited benefits.

Control of VOC emissions will almost always lead to an improvement in ozone air quality and a reduction in population exposure. Additional benefits result from concerted international action and from focussing the emission control on those source sectors making the largest contributions to ozone formation. Methane mitigation is seen as a cost-effective strategy on the global scale, bringing multiple benefits for air quality, public health, agriculture and the climate system. Less attention has been paid to global carbon monoxide emissions but reduction of these emissions also has the potential to reduce ozone exposure. The picture is more complicated for control of NO_x emissions; large emission reductions are generally needed in urban areas to overcome the initial ozone disbenefit. Control of the rising emissions of NO_x from shipping would also be beneficial to annual and summer-time mean ozone in western Europe.

Question H: *Of the recommendations made in the Fourth Report of the Photochemical Oxidants Review Group in 1997, which remain to be implemented, to what extent do they remain valid and which have been superseded by scientific understanding?*

AQEG has reviewed progress on the recommendations made by the Photochemical Oxidants Review Group (PORG) for the topics *ozone monitoring, NO_x and chemistry, hydrocarbons* and *synthesis and interpretation.* Recommendations related to impacts on *human health effects, vegetation* and *materials* have not been reviewed, because these topics are not discussed in the present AQEG report. The progress on 21 relevant recommendations is outlined in Chapter 9.

Chapter 1

Introduction

1. The Department for Environment, Food and Rural Affairs (Defra) and the Devolved Administrations commissioned this, the fifth report from the Air Quality Expert Group (AQEG), in order to inform ozone policy considerations. The report investigates the recent historic trends in, current status of and likely future changes to tropospheric ozone concentrations in the UK. The main focus is on human exposure to ozone pollution, particularly in urban areas, and it largely excludes damage to vegetation and ecosystems. By necessity, AQEG has also considered trends, considered changes in and changes to ozone precursor emissions on the European and global scales as a background to the main focus of the report. These aspects are also relevant to ozone's role as an important greenhouse gas, which was discussed in detail in the third AQEG report *Air Quality and Climate Change: A UK Perspective.* Defra requested that the report should be structured as a series of short answers to eight questions, plus supporting evidence where necessary. The questions and short answers are listed in the executive summary.

2. Much of the background material needed for this report may be found in the fourth report of the Photochemical Oxidants Review Group (PORG) published in 1997. PORG was an official body of experts, set up by the-then Department of Environment, to review current knowledge on the physical and chemical aspects of photochemical oxidants and associated precursors.

3. Ozone is one of the more important photochemical oxidants. It is a secondary pollutant formed photochemically from the sunlight-initiated oxidation of volatile organic compounds (VOCs) in the presence of nitrogen oxides, NO_X (= nitric oxide (NO) + nitrogen dioxide (NO_2)). The chemistry of ozone formation is discussed in some detail in Chapter 2 of the fourth PORG report. The timescale of VOC oxidation, and hence of ozone formation, is quite long – hours, days or even longer – so that ozone is formed many kilometres downwind of the emissions of its precursors. Ozone formation is a transboundary process and international agreements are required for its control. The human health impacts of ozone derive from its irritant properties and its induction of an inflammatory response in the lung. Ozone also has adverse effects on crop yields, on tree growth and on the composition of natural plant communities.

4. Vigorous abatement actions by European countries have led to appreciable reductions in both NO_X and VOC emissions in western Europe, as shown by the national emissions inventories and confirmed through measurements of airborne concentrations (EUROTRAC, 2003). As a result, ozone peak concentrations have decreased significantly over recent years, as illustrated for example by annual maximum 8-hour concentrations and number of days above 50 ppb. On the other hand, the lower percentiles in ozone concentrations have increased in polluted areas, in particular during winter. This is a result of an increasing tropospheric ozone background and a decrease in ozone scavenging by NO_X emissions (EUROTRAC, 2003).

5. Episodes of high ozone concentrations typically occur in summer when the solar intensity is higher, leading to increased rates of photochemical oxidation of VOCs. Continental sources of ozone precursor emissions are important under easterly airflows. There is a substantial year-to-year variability in summer ozone concentrations because of the variability in the weather. There has also been a decrease in UK peak ozone concentrations over the last 20 to 30 years because of reductions in emissions of precursor species in Europe.

6. Ozone concentrations are generally lower in urban regions than in surrounding rural regions because of the effects of emissions of NO_X which are greatest in urban regions. NO reacts rapidly with ozone to form NO_2, leading to an urban decrement in ozone levels. Reductions in NO_X emissions over the last 10 to 15 years have led to a reduction in this decrement, so that ozone concentrations in urban areas have generally increased.

7. Intercontinental transport of ozone and of ozone precursors has an important impact on ozone concentrations at the regional and local scales. Ozone is formed in the background troposphere, especially in the northern hemisphere, mainly by the oxidation of methane (CH_4) and also carbon monoxide (CO). NO_X is again essential for ozone formation and derives from anthropogenic emissions, but also from natural sources such as lightning and soil emissions. Transport of ozone from the stratosphere, where mixing ratios are higher, provides a further source of ground-level ozone.

8. There has been more than a doubling of the tropospheric background ozone concentration since 1850, reflecting the anthropogenic influence (Volz and Kley, 1988). An increase was seen in the background ozone concentration in the latter part of the 20th century monitored at suitably placed stations both in Europe and elsewhere. This increase is ascribed to imports of ozone from across the North Atlantic. It has a direct influence on the ozone budget, which is further augmented by regional ozone formation. An increase in background ozone reduces the magnitude of the regional contribution required for exceedence of ozone standards.

9. The UK Air Quality Strategy (Defra, 2007) confirmed an ozone air quality objective, which applied from the end of 2005, of 100 $\mu g\ m^{-3}$, measured as the daily maximum of a running 8-hour mean ozone concentration, not to be exceeded more than 10 times a year. The European Union (EU) has a less stringent target of a daily maximum of a running 8-hour mean of 120 $\mu g\ m^{-3}$, not to be exceeded more than 25 times a year, averaged over three years. The date for achievement of this target is 31st December 2010. Target values for the protection of vegetation and ecosystems are based on critical levels and cumulative exposure. The AOT40 index is based on exposure over 40 ppb (80 $\mu g\ m^{-3}$) during daylight hours in the growing season. The current Air Quality Strategy and EU targets for AOT40 are 18,000 $\mu g\ m^{-3}$ h, calculated from one hour values from May to July and to be achieved, so far as possible, by 2010.

10. The National Emission Ceilings Directive (NECD) (2001/81/EC) sets ceilings for each Member State of the EU for emissions, *inter alia*, of the ozone precursors NO_X and non-methane VOCs, to be met by 2010. Member States are obliged to report each year their national emissions inventories and projections for

2010. A consultation draft on the updated UK national programme to meet the NECD was published in October 2006. Wider international agreements on emissions of ozone precursors are negotiated through the United Nations Economic Commission for Europe (UNECE) Convention on Long-Range Transboundary Air Pollution (CLRTAP). The 1999 Gothenburg Protocol sets emissions ceilings for NO_X and VOCs for 2010. The percentage reductions required vary from country to country, but, once fully implemented, they should cut European emissions of NO_X by 41% and of VOCs by 40% compared to 1990 levels.

11. This report is concerned exclusively with recent and projected concentrations of ozone and their dependence on precursor emissions. Those same precursors also influence the concentrations of other secondary air quality pollutants, especially particulate matter (PM) and NO_2. Measures to reduce human exposure to ozone may conflict with the level of exposure to these other pollutants and policy necessarily takes account of such conflicts through, for example, cost-benefit analysis. Such considerations lie outside AQEG's remit but will be used in policy developments related to the conclusions drawn in this report. In addition, these same precursor emissions influence climate through their impact on ozone, methane and secondary aerosol, and a holistic approach to policy in which all of these effects are considered together would be preferred.

12. Three other reports on ozone have recently been published or are in preparation:

    - Defra has commissioned a report on ozone impacts on vegetation and ecosystems. This report will update the report of the National Expert Group on Transboundary Air Pollution (NEGTAP, 2001) and is likely to be published in 2009.

    - The Royal Society's report *Ozone in the 21st century* was published in October 2008 (Royal Society, 2008) and examines likely changes in ozone on a 50 to 100 year timeframe and specifically addresses the effects of climate change.

    - UNECE has set up a Task Force on Hemispheric Transport of Air Pollution (HTAP) which will report by 2010 to inform CLRTAP on hemispheric air pollution and, in particular, on source-receptor relationships for intercontinental transport of air pollution. An interim report was published in 2007 to inform the review of the Gothenburg Protocol.

13. This report from AQEG addresses the eight questions (A-H) posed by Defra and laid out in the executive summary. It examines a shorter timeframe than the Royal Society report, and is limited to the next two decades. The chapters of the report each address a specific question. A short answer is provided, together with a fuller answer, followed by the supporting evidence. The executive summary includes the short answer to each question. The supporting evidence is substantial for Chapter 2 and Chapter 8 because of the need to analyse and assess monitoring data and to examine model results.

14. In discussing global and regional emission projections, this report refers, in a number of places, to the Intergovernmental Panel on Climate Change (IPCC) Special Report on Emissions Scenarios (SRES) and the scenarios developed by the International Institute for Applied Systems Analysis (IIASA) for the Regional Air Pollution Information and Simulation (RAINS) model. A brief summary of these emission projection scenarios is given in Box 1.1. When comparing and evaluating emission projections from different sources, it is important to understand the differences in scenarios and assumptions used. Versions of emission projections are periodically updated and changed by the organisation that first developed them as the basic input parameters are better refined, so it is important to note which version of emission projections are used in any analysis. Annex 1 provides recent versions of global, European and UK emission projections for ozone precursor gases for future scenarios used in studies referred to in this report and gives references to the sources of emissions data.

1.1 Units

15. Concentration units of $\mu g\ m^{-3}$ (micrograms per cubic metre), where the volume of air is standardised to 20°C and a pressure of 101.3 kPa, are used where possible in this report, as these are the units used in current legislation. On occasion, and especially in the supporting evidence for the answer to question C, ozone mixing ratio, in units of ppb (parts per billion, i.e. the number of ozone molecules within a sample of air containing 1,000,000,000 molecules in total) has been used. Much of the evidence for this answer relies on global atmospheric models of trace species concentrations. Modelling studies use mixing ratio to describe trace species abundance because the chemical processes, for example, reactions with NO and NO_2, are much clearer with ppb than with $\mu g\ m^{-3}$ units. The ppb unit also features in the ozone metric SOMO35.

16. There is a simple conversion between the two units as defined here, with

$$1.0 \text{ ppb of ozone} \equiv 2.0\ \mu g\ m^{-3} \text{ at } 290\ K \text{ and } 101.3\ kPa.$$

1.2 Ozone metrics of relevance to human health

17. A number of ozone metrics have been used in this report and those key metrics relevant to human health are shown in Table 1-1.

18. Ambient concentrations of primary air pollutants (that are emitted directly into the atmosphere) typically have highly skewed distributions of hourly concentrations with the mode at low concentrations and a small number of hours with high concentrations. Ozone is a secondary pollutant with a hemispheric background concentration much greater than zero and frequency distributions are therefore very different from those found for primary pollutants. Identical annual mean concentrations, as might be measured at different monitoring sites can be made up from very different frequency distributions and the annual mean does not capture all of the features of ambient concentrations that may be of concern in terms of the impact on human health, such as short periods of high concentrations. A range of different metrics are therefore used for ozone and these metrics reflect different features of the frequency distribution of hourly concentrations.

Box 1.1 The IPCC SRES and IIASA RAINS Global Emission Projection Scenarios

The Intergovernmental Panel on Climate Change (IPCC) Special Report on Emissions Scenarios (SRES) scenarios (IPCC, 2001) provide global emission projections of ozone precursor gases for a wide range of scenarios covering the main emission driving forces, from demographic to technical and economic development. The terms of reference for the SRES scenarios did not require consideration of any future policies that explicitly address climate change. The SRES scenarios are broadly grouped into four families following different "storylines", each assuming a distinctly different direction for future developments.

The A1 storyline is for a future world with very rapid economic growth, a global population that peaks mid-century and declines thereafter, the rapid introduction of new and more efficient technologies and with a substantial reduction in regional differences in per capita income. Within this family are three sub-scenarios with different technological emphasis:

A1FI – A1, fossil fuel intensive
A1T – A1, with non-fossil energy source emphasis
A1B – A1, with a balance across energy sources.

The A2 storyline is a more pessimistic scenario, describing a very heterogeneous world based on self-reliance, regional differences in economic and technological development, and continuous increase in global population.

The B1 storyline describes a convergent world like A1, with global population peaking in mid-century, but with rapid changes in economic structures, introduction of clean and resource-efficient technologies and an emphasis on global solutions to social and environmental sustainability.

The B2 storyline describes a world with emphasis on local solutions to social and environmental sustainability, less rapid and more diverse than in B1 and A1, with continuously increasing global population, but at a lower rate than A2.

Emission projections for the SRES scenarios and further details can be found at http://www.grida.no/climate/ipcc_tar/wg1/519.htm

Two main emission projection scenarios have been developed by the International Institute for Applied Systems Analysis (IIASA): the "Current Legislation" (CLE) scenario and the "maximum technically feasible reduction" (MFR) scenario. These have been developed for a global version of the Regional Air Pollution Information and Simulation (RAINS) model (Schöpp *et al*., 1999) and the model used to forecast ozone precursor emissions by region and source sector to 2030.

The CLE scenario reflects the current perspectives of individual countries on future economic development and takes into account the anticipated effects of presently decided emission control legislation in the individual countries. It considers the state of national emissions legislation in each country as of the end of 2002 and the evolution of emission controls in the coming years as laid down in the legislation. International and national fuel quality and emission standards currently in force in Europe and North America were considered, and information and standards were collected for other countries and world regions. Country-, sector- and technology-specific impacts of emission control measures were considered. These included combustion modification and secondary measures like Selective Catalytic Reduction (SCR) for reduction of NO_X emissions from stationary combustion; lower sulphur fuels and flue gas desulphurisation for reduction of sulphur dioxide (SO_2) emissions; and engine modification and catalyst- and filter-based exhaust after-treatment systems on mobile sources. Emission controls for methane include modified practices in agriculture, technologies for waste treatment (e.g. landfill gas recovery) and energy production, and reducing natural gas leakage.

The MFR scenario is a more optimistic emission reduction scenario based on the full implementation of all presently available best technical emission control measures while maintaining the same projected level of anthropogenic activities worldwide. The focus is on the theoretical potential of today's most advanced emission control technologies and the MFR scenario does not take into account their practical limitations and the high costs associated with their global penetration. But it also does not take account of the potential benefits of non-technical measures that modify energy demand or human behaviour such as increased energy efficiency measures, fuel substitution and reduced transport demand.

Further details of the IIASA RAINS model and the CLE and MFR emission scenarios and their application to tropospheric ozone modelling can be found in Amann *et al*. (2004), Cofala *et al*. (2006), Dentener *et al*. (2005) and Schöpp *et al*. (1999).

These frequency distributions in turn vary with location and reflect the differing influences of local, regional and global factors determining ozone concentrations. Examples of these different frequency distributions are given in the supporting evidence for Chapter 2, which draws heavily on monitoring data from the UK Automatic Urban and Rural Network (AURN). Box 1.2 outlines measurement and calibration techniques used in the AURN.

19. Several of the metrics listed in Table 1-1 are based on the annual average of the daily maximum of the running 8-hour mean concentration. (The running 8-hour mean is assigned the date of the last hour of the running mean using GMT.) The metrics are calculated using cut-off concentrations of zero (that is, including all days), 70 µg m^{-3} and 100 µg m^{-3}. These metrics have been recommended and used for health impact quantification by the UK Interdepartmental Group on Costs and Benefits (IGCB, 2007). For the metrics with cut-offs, the cut-off concentration is subtracted from the daily maximum of the running 8-hour mean concentration and the value set to zero if the result is zero or negative. The average across all of the days in the year is then calculated. These metrics have been recommended as appropriate for the assessment of the impact of the daily variation in ozone concentration on human health. The range of cut-offs reflects uncertainty as to whether there is a threshold for ozone (COMEAP, 1998). The World Health Organization (WHO) concluded that there was evidence that associations existed below the current guideline value (120 µg m^{-3}), but its confidence in the existence of associations with health outcomes decreased as the concentrations decreased (WHO, 2004). The cut-off at 70 µg m^{-3} is not based on direct evidence of a threshold for health effects at this value. It was recommended (UNECE/WHO, 2004) for use in cost-benefit analysis and integrated assessment modelling on the basis of a combination of the uncertainty in the shape of the concentration response function at low ozone concentrations, the seasonal cycle and geographical distribution of ozone concentrations and the range of concentrations for which European-scale ozone modelling was able to provide reliable estimates (IGCB, 2007).

20. The metric of the annual average of the daily maximum of the running 8-hour mean with a cut-off at 70 µg m^{-3} is closely related to the SOMO35 (sum of the daily maximum of running 8-hour means over 35 ppb) metric adopted for European-scale integrated assessment modelling. It can be calculated by multiplying the 70 µg m^{-3} cut-off metric by the number of days in the year and then applying a factor to take account of the different units used. We have calculated the 70 µg m^{-3} cut-off metric in µg m^{-3}, SOMO35 is typically quoted in ppb.days or ppm.days. The 70 µg m^{-3} cut-off metric has been preferred over SOMO35 in this report because the units are easier to interpret and compare with other metrics and the magnitude of the metric is not unduly influenced by low data capture in a year.

21. The metric most sensitive to the magnitude of regionally-generated photochemical episodes is the maximum 1-hour average during the year. This metric is thus likely to show a response to reductions in relevant precursor emissions. It is, however, highly variable from year to year and from site to site and is particularly subject to instrument malfunction or interference. High percentiles of the hourly concentration, such as 99.9th or 99th percentile are therefore sometimes preferred for data analysis.

Table 1-1 The key ozone metrics of relevance to human health considered in this report.

Metric	Relevance	Key influences on the values of this metric at urban locations
Annual average	Basic metric used to show long-term trends	Includes all of the hours in the year. Strongly influenced by the magnitude of local NO_x emissions and by topography through nocturnal depletion
Annual average of the daily maximum of the running 8-hour mean	Used as "basic metric" for many of the health metrics. Also used as Defra's air quality indicator	Strongly influenced by the magnitude of local NO_x emissions
Annual average of the daily maximum of the running 8-hour mean with a 70 µg m^{-3} cut-off	Health impact, related to SOMO35	Influenced by the magnitude of local NO_x emissions and by photochemical episodes
Annual average of the daily maximum of the running 8-hour mean with a 100 µg m^{-3} cut-off	Health impact	Strongly influenced by photochemical episodes and to a lesser extent by the magnitude of local NO_x emissions
Maximum 1-hour average (peak hour in the year)	Used as the basis for some epidemiological studies, although it has been suggested that the 8-hour metric is more representative. Also an indicator of short-term peaks, but note low statistical power, since it is the value for one single hour	The metric most sensitive the magnitude of regionally-generated photochemical episodes and thus likely to show a response to reductions in relevant precursor emissions
Number of days with daily maximum of running 8-hour mean exceeding 100 µg m^{-3}	Equates to the number of exceedences of the UK ozone standard (the Air Quality Strategy objective is no more than 10 exceedences per year)	Strongly influenced by photochemical episodes and to a lesser extent by the magnitude of local NO_x emissions
Number of days with daily maximum of running 8-hour mean exceeding 120 µg m^{-3}	Equates to the number of exceedences of the EU Target Value (no more than 25 days, averaged over 3 years) and Long Term Objective (no exceedences) from the 3rd Daughter Directive	Strongly influenced by photochemical episodes and to a lesser extent by the magnitude of local NO_x emissions
SOMO35 (sum of means over 35 ppb)	Used as a metric by IIASA, for Clean Air for Europe (CAFE) and NECD revision, related to annual average of the daily maximum of the running 8-hour mean with a 70 µg m^{-3} cut-off	Influenced by the magnitude of local NO_x emissions and by photochemical episodes

Box 1.2 Measurement and calibration techniques for ambient ozone

Ozone concentrations in ambient air are calculated from the absorption of ultraviolet (UV) light at 254 nanometres (nm) wavelength. The air sample passes through a cell tube of length (l), and the absorption is measured using a UV detector. An ozone-removing scrubber is used to provide a zero reference intensity. The analyser alternately measures the absorption of the air path with no ozone present (I_0) and the absorption of the ambient sample (I_1). The concentration (c) is calculated using the Beer-Lambert equation:

$$I_1 = I_0 \, e^{-alc}$$

where a = absorption coefficient at 254 nm.

Modern automatic ozone analysers perform this measurement on a continuous basis. Data from ozone monitors in the UK Automatic Urban and Rural Network (AURN) are collected from all monitoring stations every hour and are available, also on an hourly basis, at http://www.airquality.co.uk.

This method of measurement could potentially suffer interference from any substance optically active at a wavelength of 254 nm. However, interference is minimised as the built-in ozone-removing scrubber almost specifically removes only ozone, thus any potential interfering species are present in both parts of the measurement cycle. A number of compounds (including mercury vapour, water, phthalates and certain particulate materials) are, however, suspected to still show interference on occasions.

This methodology has been in use for at least the last 20 years in the UK. Prior to this, ozone was determined with analysers using the principle of ethylene chemiluminescence.

All UK ozone measurements in the AURN have been subject to rigorous and consistent quality assurance methodologies for many years. This ensures that reliable trend analysis can be interpreted from the data available. All ozone analysers in the AURN are regularly calibrated on site with standard ozone photometers. These ozone photometers are calibrated against the UK ozone standard, which in turn undergoes regular international comparison with primary ozone standards held by other national measurement institutes.

Ozone analysers in the AURN are calibrated with an uncertainty of ±3.5% relative uncertainty (at the 95% confidence level). The standard ozone photometers are calibrated against the UK ozone standard with an uncertainty of ±3.0%.

Clearly, between calibrations, the uncertainty of the data increases due to analyser drift, etc. This is carefully controlled by regular analyser span checks and detailed analysis and adjustment of the data with sophisticated software tools in the data ratification process.

The EU 3rd Daughter Directive now requires ozone measurement to be compliant with the European Committee for Standardisation (CEN) standard BS EN 14625:2005 *Ambient air quality – Standard method for the measurement of the concentration of ozone by ultraviolet photometry.* The effect of this on UK AURN measurements is unlikely to be significant, as most of the requirements have been network practice for many years, but it will increase confidence in the data, and in their comparability with other European data.

With these procedures in place and using CEN-compliant ozone analysers, the maximum uncertainty of the UK ozone dataset is ±12% (at the hourly limit value) and hence meets the EU Directive Data Quality Objective of 15% (at the 95% confidence level).

Chapter 2

Temporal trends and spatial distributions in ozone concentrations determined from monitoring data

Question A: *A large quantity of urban and rural monitoring data has been collected since the last Photochemical Oxidants Review Group (PORG) report, by Defra's own networks, local authority stations and others. What does this reveal in terms of trends (using metrics considered relevant to effects) and spatial concentration patterns?*

Short answer to question A

22. **Annual mean ozone concentrations have generally increased over the last 10 years or so in urban areas, while the changes in concentrations in rural areas are less marked and show variations with location and time. Ozone concentrations are generally lower in urban areas than in the surrounding rural areas due to reaction with nitrogen oxides (NO_X) (mostly nitric oxide, NO) emissions, which are greatest in urban areas. The main cause of the increase in urban areas is the reduction in NO_X emissions, which has led to a decrease in this "urban decrement" (see Box 2.1 below), reducing the differences between urban and rural concentrations.**

23. **Reductions in precursor emissions in the European region have led to reductions in peak ozone concentrations at rural sites, although there are significant variations from year to year due to the weather, with higher concentrations generally measured in years with hotter summers (such as 1995, 2003 and 2006). The trends in some of the health-based ozone metrics are influenced by both changes in mean and peak concentrations.**

Box 2.1 Urban decrement

The focus of this report is on ozone concentrations in urban areas. Ozone concentrations in urban areas are typically lower than those in the surrounding countryside as a result of its removal by reaction with nitric oxide (NO), for which emissions are greatest in urban areas. We have used the term "urban decrement" to describe the amount by which ozone concentrations are lower in an urban area than in surrounding rural locations. An urban decrement can be defined for the annual mean ozone concentration or any of the other human health-related ozone metrics that we have considered.

Detailed answer to question A

2.1 Introduction

24. The main influences on urban ozone concentrations have been described in the introduction. They are:

- Regional photochemical ozone production
- An urban decrement due to local emissions of NO
- The hemispheric background.

25. Changes in hemispheric background ozone concentrations are discussed in detail in Chapter 3.

2.2 Temporal trends since 1990

2.2.1 Annual means

26. Annual mean ozone concentrations at urban sites generally show an increase in concentration over the last 10 years or so due to the reduction in local emissions of NO in urban areas. This is generally the strongest influence on annual means in urban areas. This increase is due to the change in the partitioning of the total oxidant between nitrogen dioxide (NO_2) and ozone as NO emissions have reduced. The magnitude of the increase is generally greatest at the locations with the largest reduction in NO concentrations. These are typically the urban background locations with the highest initial local nitrogen oxides (NO_X) emissions. Roadside locations with very high emissions generally do not show as large an increase in ozone concentrations because the NO_X source is very close and NO concentrations remain high relative to oxidant concentrations. A site-specific model combining emission inventories for NO_X and the oxidant partitioning model of Jenkin (2004) can explain the trends in measured annual mean ozone concentrations at urban sites with considerable success.

27. Annual mean ozone concentrations are also influenced by the magnitude of the regional photochemical generation of ozone. The years with higher annual means, such as 1995, 2003 and 2006, correspond to warmer summers with more frequent photochemical episodes.

28. The trends in annual mean ozone concentration at rural sites are generally less steep and are variable from site to site. This is because the influences of the changes in hemispheric background concentrations, regional photochemistry and the reduction in NO scavenging vary from site to site. Remote sites in the north-west of the UK are most likely to show the influence of changes in hemispheric background concentrations in terms of overall trends and year-to-year variability, although these influences can also be detected at sites in the south-east and urban sites using modelling methods to isolate the different influences. The influence of regional photochemistry leading to an increase in annual mean ozone is generally greatest in the south-east of the UK because this is the region closest to the major sources of photochemical ozone precursors.

29. The influences on ozone concentrations also vary according to season. The seasonal variation is much more pronounced for air masses that are influenced by regional emissions. This results in ozone concentrations lower than the hemispheric background in the winter due to scavenging by NO and higher than the background in the summer due to photochemical ozone production.

2.2.2 Peak concentrations

30. The trend in the annual maximum of 1-hour ozone concentrations (peak ozone concentrations) is generally downwards at rural sites as a result of the control of regional anthropogenic volatile organic compound (VOC) and NO_X emissions, although there is considerable year-to-year variability due to the weather. The trends in high percentile 1-hour ozone concentrations (99.9th percentile, for example) and in maximum and high percentile 8-hour concentrations are similar. This is consistent with ambient measurement data for VOCs, which show a consistent downward trend in the concentrations of the more reactive anthropogenic VOCs associated with ozone formation at monitoring sites in the UK. The recent downward trends in the emissions and ambient concentrations of NO_X have been described in the Air Quality Expert Group's (AQEG's) reports on NO_2 (AQEG, 2004, 2007a). Observations of the reactive hydrocarbon isoprene in the UK are indicative of elevated inputs of biogenic hydrocarbons during summertime, particularly during heat-waves (Lee *et al.,* 2006).

31. The trends in peak ozone concentrations at urban sites are generally flatter because the reduction in local NO_X emissions has tended to decrease the urban decrement during photochemical ozone episodes at the same time as the peak ozone concentrations at rural sites have declined.

32. A comparison of measured peak ozone concentrations at rural sites and those in central London over the last 15 years or so illustrates this well (see Figure 2.19).

2.2.3 Exceedence metrics

33. The trends in the number of days with the daily maximum of running 8-hour mean ozone concentration above 100 $\mu g\ m^{-3}$ and 120 $\mu g\ m^{-3}$ are generally strongly influenced by photochemical episodes and to a lesser extent by the magnitude of local NO_X emissions. The number of exceedence days is highly variable from year to year due to variations in meteorology but is generally downwards at rural sites, although to a lesser extent than the peak ozone concentration. A reduction in the urban decrement in the number of exceedences per year at urban sites compared with that at nearby rural sites might have been expected as NO_X emissions have declined, although such a trend is not readily apparent from the measurement data.

2.2.4 Health-based metrics based on the annual average of the daily maximum running 8-hour concentration with various cut-offs

34. The trends in the annual average of the daily maximum of the running 8-hour mean concentration generally follow the trends in annual mean concentration, although the value of this metric for a given year is higher, since it is based on the daily maximum concentration.

35. Trends in the annual average of the daily maximum of the running 8-hour mean with a 100 µg m^{-3} cut-off are sensitive to both photochemical episodes and the magnitude of local NO_X emissions, especially in urban areas. This metric generally shows a decline at rural sites due to the reductions in the emissions of precursor species but with considerable year-to-year variability. The trends at urban sites are unclear with competing influences of the reduction in regional photochemical ozone and the reduction in local NO_X emissions. The trends in annual average of the daily maximum of the running 8-hour mean with a 70 µg m^{-3} cut-off, and thus SOMO35, are somewhat clearer at urban sites, with many sites showing an increase as local NO_X emissions have reduced. The trend in this metric at rural sites is less clear with considerable year-to-year variability, since the cut-off for this metric is very close to typical hemispheric background concentrations.

2.3 Spatial patterns

36. The spatial pattern of ozone concentrations across the UK varies depending on the metric illustrated and from year to year due to variation in the weather and changes in emissions. Maps of a range of ozone metrics have been calculated using empirical measurement-based pollution climate models (PCMs) for the years 1995, 2003 and 2005. These years have been chosen to illustrate two recent years with high and low photochemical ozone contributions (2003 and 2005 respectively) and a year with high photochemical ozone contributions combined with high urban NO_X emissions (1995).

2.3.1 Annual means

37. The hemispheric background is a major contributor to annual mean ozone concentration across the UK. Upland areas tend to have the highest annual mean ozone concentrations due to topographic effects, as has been described by PORG (1997). There is also a significant decrement in urban areas, as discussed above. Annual mean concentrations in rural areas were highest in 2003 and lowest in 2005 at most locations due to the photochemical episodes during 2003 and perhaps a higher hemispheric background in 2003. Urban concentrations were lowest in 1995 due to the greater local NO_X emissions at the time, as illustrated by the results of a transect across London (see Figure 2.33).

2.3.2 Exceedence metrics

38. The spatial distribution of the number of days with running 8-hour mean ozone concentrations greater than 100 µg m^{-3} and greater than 120 µg m^{-3} is dominated by the contribution from photochemical episodes. This contribution is generally greatest in the south-east of the UK, which is the region closest to the major continental sources of the precursors of photochemical ozone episodes. This contribution is also highly variable from year to year depending on the weather conditions. The number of days above 100 µg m^{-3} was highest in the south, south-west and Wales in 1995, highest in the south east in 2003 (and also high in the north of Scotland in 2003 due to the higher background) and highest in East Anglia in 2005, although values were generally much lower. There are also clear urban decrements for these metrics.

2.3.3 Health-based metrics based on the annual average of the daily maximum running 8-hour concentration with various cut-offs

39. The maps of the annual average of the daily maximum of the running 8-hour mean concentration have been calculated from the annual mean maps using a non-linear function and thus show a similar spatial pattern.

40. Values of the annual average of the daily maximum of the running 8-hour mean concentration with a cut-off of 100 µg m^{-3} metric were much higher in 2003 than in 2005. Values were higher in south and south-west England and Wales in 1995 than in 2003 for this metric. This is in contrast to the annual mean, which was higher in 2003 in these areas. The contribution from photochemical episodes and an urban decrement are the most important factors influencing the spatial distribution for this metric.

41. Maps of the annual average of the daily maximum of the running 8-hour mean concentration with a cut-off of 70 µg m^{-3} show highest concentrations in the south of the UK, where the contribution from photochemically-generated ozone is greatest, and in northern Scotland, where the impact of the hemispheric background is most pronounced due to the low regional NO_X emissions in this area. There is also an urban decrement for this metric. Concentrations were generally highest in 2003 and lowest in 2005 due to the larger contribution from photochemical ozone episodes in 2003 and 1995. Urban concentrations were lowest in 1995 due to the greater local NO_X emissions at the time.

2.3.4 Transects

42. A comparison of the modelled transects of ozone concentrations for the annual mean and other health-based metrics across London with data from monitoring sites close to the transect confirms that the empirically-generated maps include a reasonably realistic description of the urban decrements for this city.

2.4 Concluding remarks

43. It is clear that the temporal trends and spatial patterns are different for the different ozone metrics and respond in different ways to changes in the key influences on ozone concentrations. Any consideration of the impact of possible future measures on urban ozone concentrations should therefore consider a range of metrics, and this is the approach that we have adopted in answering question G.

Supporting evidence for question A

2.5 Overview

44. Data from the UK automatic monitoring network and elsewhere indicate that the concentration of ozone at a given location in the UK can be influenced by a combination of global (hemispheric), regional and local scale effects. As a result, the observed trend in ozone concentrations, concentration distributions and related metrics is determined from the net trend of these three influences, the relative contributions of which can vary both spatially and temporally. Specifically, the data demonstrate the following three major influences:

I. The hemispheric background ozone concentration has been gradually increasing as a result of global-scale effects, thereby influencing the background or background levels of ozone brought into the UK from the Atlantic Ocean.

II. Substantial short-term elevations in ozone concentrations during summertime episodes are a consequence of the formation of additional ozone from regional-scale photochemical processing of emitted VOC and NO_X, with such events tending to be more frequent and intense towards the south of the UK. Their severity has progressively decreased since about 1990 as a result of EU controls of anthropogenic VOC and NO_X emissions.

III. The control of NO_X emissions in the UK has reduced local-scale removal of ozone by reaction with emitted NO, contributing to a general increase in ozone concentrations since about 1990. This has most relevance to urban areas, where NO_X emissions are higher.

45. In the following sections, the effects of the above influences are illustrated by a systematic consideration of ozone observations at remote, rural and urban locations. Trends in the observed concentrations of hydrocarbons and NO_X are also presented, with specific reference to trends in VOC and NO_X emissions. Additional information is given in Annex 2.

2.6 Ozone observations at remote network locations

46. Figure 2.1 demonstrates the trend in the hourly mean ozone distributions (illustrated by the annual maximum, annual minimum and selected percentiles of the hourly mean ozone concentrations) at Strath Vaich, a remote site in northern Scotland, for the period 1990-2006. This site is characterised by very low NO_X levels, the annual mean typically being 1 $\mu g\ m^{-3}$ or less. There has been little change in the maximum ozone concentration at this site although, as with other locations described below, it displays substantial scatter. However, the 95th through to the 5th percentiles of the ozone concentrations all display statistically significant upward trends in the range 0.3-0.5 $\mu g\ m^{-3}\ yr^{-1}$, with a small upward trend also observed in the minimum. The reasonably similar upward trend across the majority of the distribution is consistent with observations at this remote location being dominated by the trend in the hemispheric ozone background, which is described and discussed in more detail in Chapter 3. The seasonal cycle of ozone concentrations at Strath Vaich thus typically displays the springtime maximum, characteristic of background air. To illustrate this, Figure 2.2 shows daily ozone concentrations (as the maximum of the running 8-hour mean) for the example year of 2005.

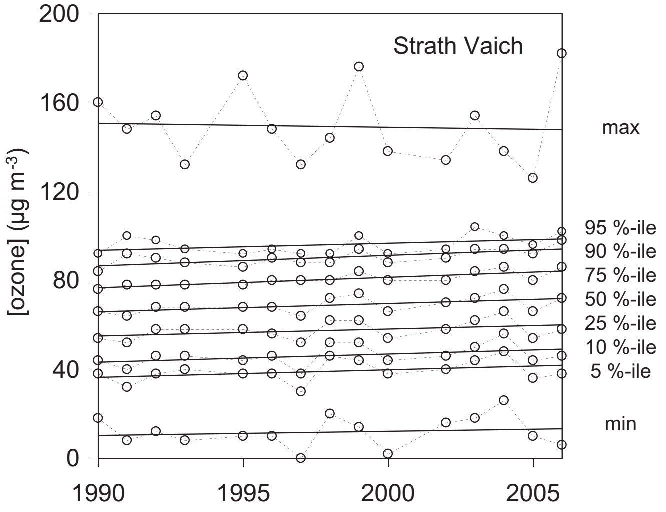


Figure 2.1 Trend in hourly mean ozone distributions at Strath Vaich (a remote site in northern Scotland) based on data over the period 1990-2006. Solid lines are linear regressions of data indicating the average trend over the period. [Ozone] denotes the concentration of ozone.

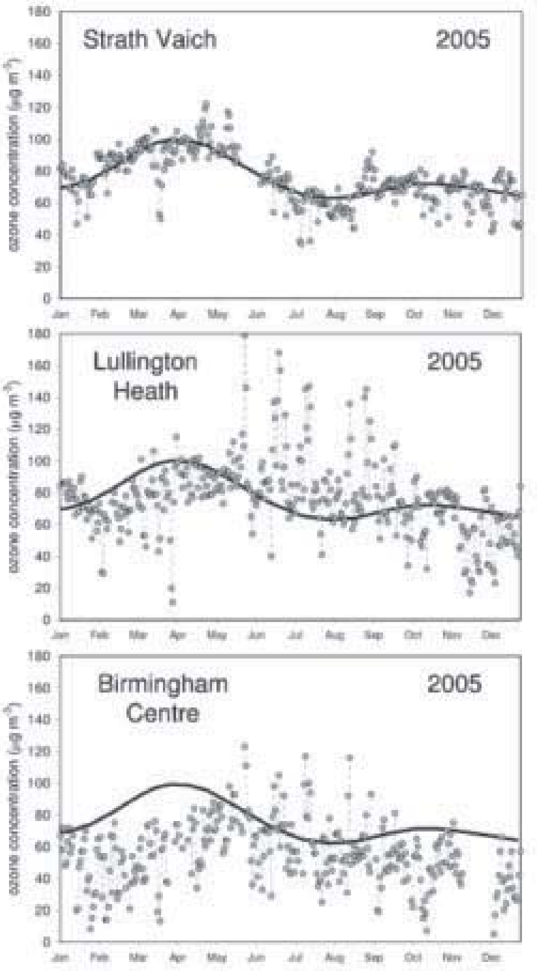


Figure 2.2 Daily maximum of the running 8-hour mean ozone concentrations in 2005 at Strath Vaich (a remote site in northern Scotland), Lullington Heath (a rural site in southern England) and Birmingham Centre. The curve is an approximate fit to the Strath Vaich data, and is reproduced in the panels for the other sites to facilitate comparison.

2.7 Ozone observations at UK rural network locations

2.7.1 Temporal and spatial trends in elevated ozone events at rural locations

47. The hemispheric background influences concentrations of ozone throughout the UK, but the observed concentrations are further modified by processes occurring on regional and local scales which can both increase and decrease ozone. Data from rural sites, in particular, show substantial short-term elevations in ozone concentrations during summertime episodes, which are a consequence of the formation of additional ozone from regional-scale photochemical processing of emitted VOC and NO_X over north-west Europe. This is illustrated in Figure 2.2 for Lullington Heath, a rural site in southern England, for the example year of 2005. Such events are characterised by stable anticyclonic conditions, when slow-moving air resides in the boundary layer for a period of up to several days. Under such conditions, the air mass circulates slowly over north-west Europe, receiving emissions of the ozone precursors when both temperature and solar intensity are elevated, thereby promoting efficient photochemical processing. This general picture of the conditions associated with photochemical ozone episodes in the UK has been supported, for example, by an analysis of air mass back trajectories associated with events when hourly mean ozone concentrations have reached or exceeded the public information threshold of 180 µg m^{-3} (Jenkin *et al*., 2002).

48. General information on the temporal and spatial trends in regional-scale ozone formation is also apparent from consideration of such events. Figure 2.3 (upper panel) shows data from 13 long-running rural and remote sites since 1990 which provide reasonable geographical coverage over the UK. The number of hours with mean ozone concentrations ≥ 180 µg m^{-3} at these sites combined (and individually) shows year-on-year variability due to the requirement for appropriate meteorological conditions, but with a general decreasing trend over the period. This is apparent from considering only the "heat-wave" years of 1990, 1995, 2003 and 2006, in which meteorological conditions particularly conducive to regional-scale photochemical ozone formation were experienced. The information also demonstrates that, although no two years are identical, the number of hours of exceedences tends to decrease towards the north and west of the UK. Locations towards the south and east are more prone to elevated photochemical ozone because trajectories during anticyclones tend to arrive from continental Europe with greater probability of passing over regions of high ozone precursor emissions. This is apparent from satellite measurements of NO_2 (Figure 2.4), which clearly shows the region of elevated anthropogenic emissions in north-west Europe.

49. The spatial variation is illustrated further in Figure 2.3 (lower panel) which shows the mean number of hours ≥ 180 µg m^{-3} annually, based on the average of the data over the period 1990-2006 for the complete series of rural sites. The data are presented in relation to a north-westerly co-ordinate, starting from Lullington Heath in the south-east. The data show a general decreasing trend with distance north-west, but also display a degree of scatter. As indicated in the figure, this scatter can be broadly related to the altitude of the site, with higher altitude sites at a given distance north-west showing a tendency towards a greater number of hours exceedence. As discussed

previously by PORG (1997), the lower altitude sites are more likely to become decoupled from the air aloft when a shallow night-time inversion layer forms, and are therefore more influenced by ozone removal via deposition. Consequently, elevated ozone concentrations during photochemical events tend to persist for a smaller proportion of the diurnal cycle at such locations.

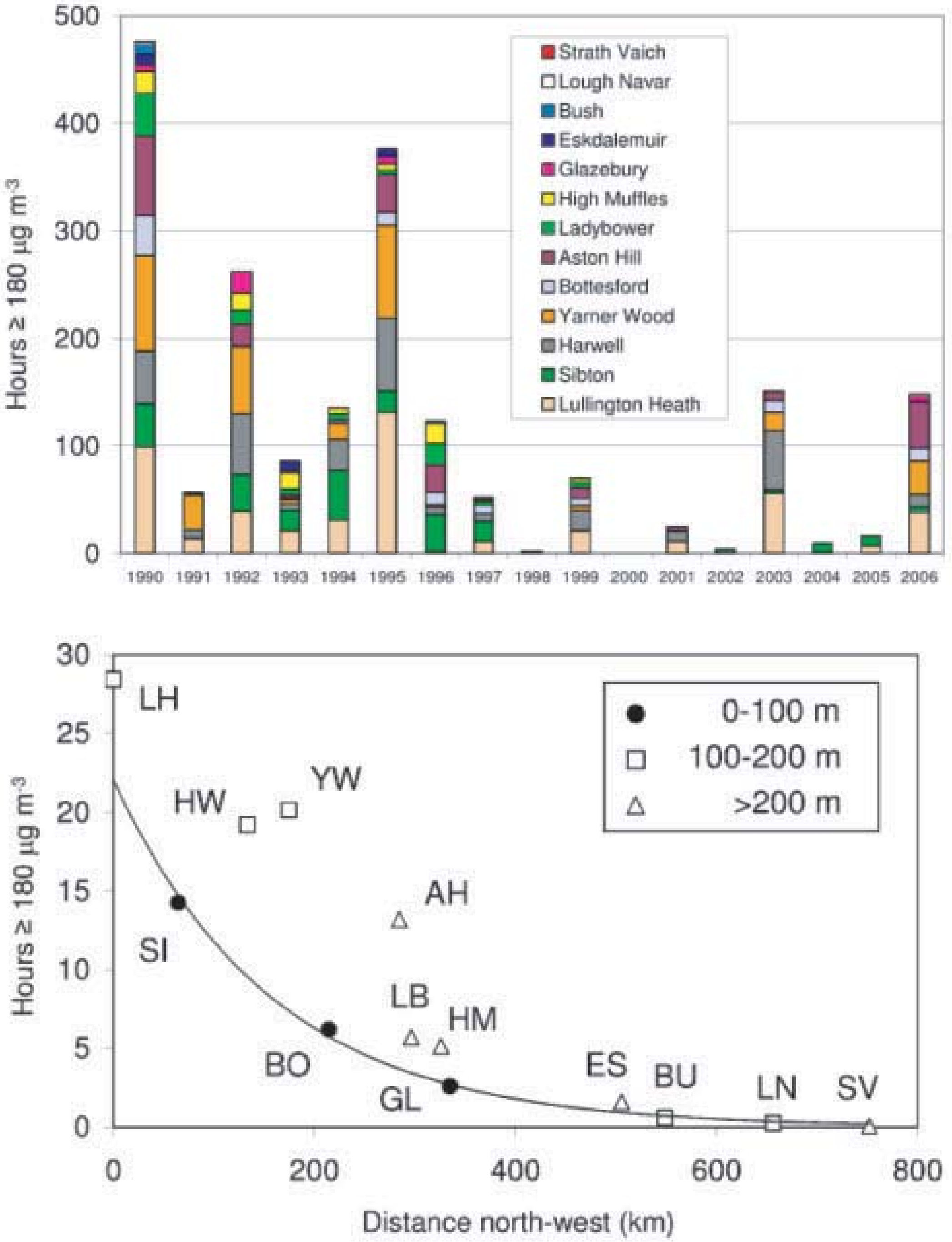


Figure 2.3 (Upper panel) Number of hours with [ozone] ≥ 180 µg m^{-3} at 13 UK rural and remote sites in each year over the period 1990-2006. (Lower panel) Annual number of hours with [ozone] ≥ 180 µg m^{-3} at UK rural sites (based on data averaged over the period 1990-2006) as a function of distance along a north-westerly co-ordinate. Sites are also classified in terms of altitude intervals, with the displayed line being an exponential fit to the three low altitude sites.

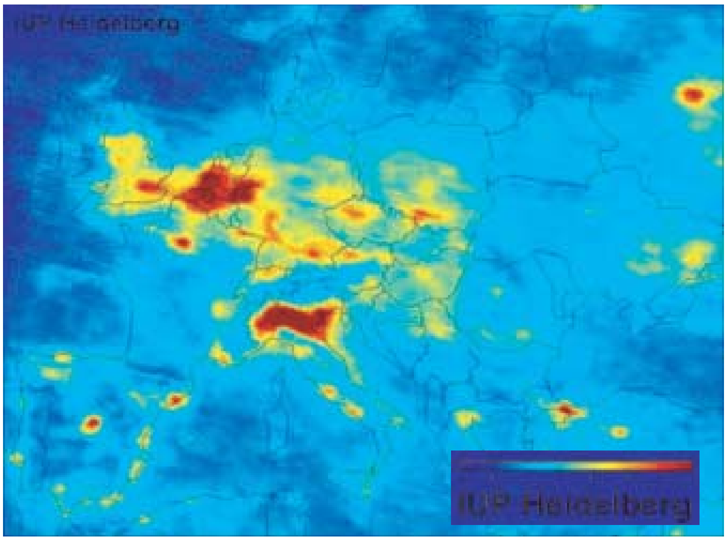


Figure 2.4 European mean tropospheric nitrogen dioxide vertical column density (VCD) between January 2003 and June 2004, as measured by the SCIAMACHY instrument on ESA's Envisat. The scale is in 10^{15} molecules.cm^{-2}. S. Beirle, U. Platt and T. Wagner, Institute for Environmental Physics, University of Heidelberg.

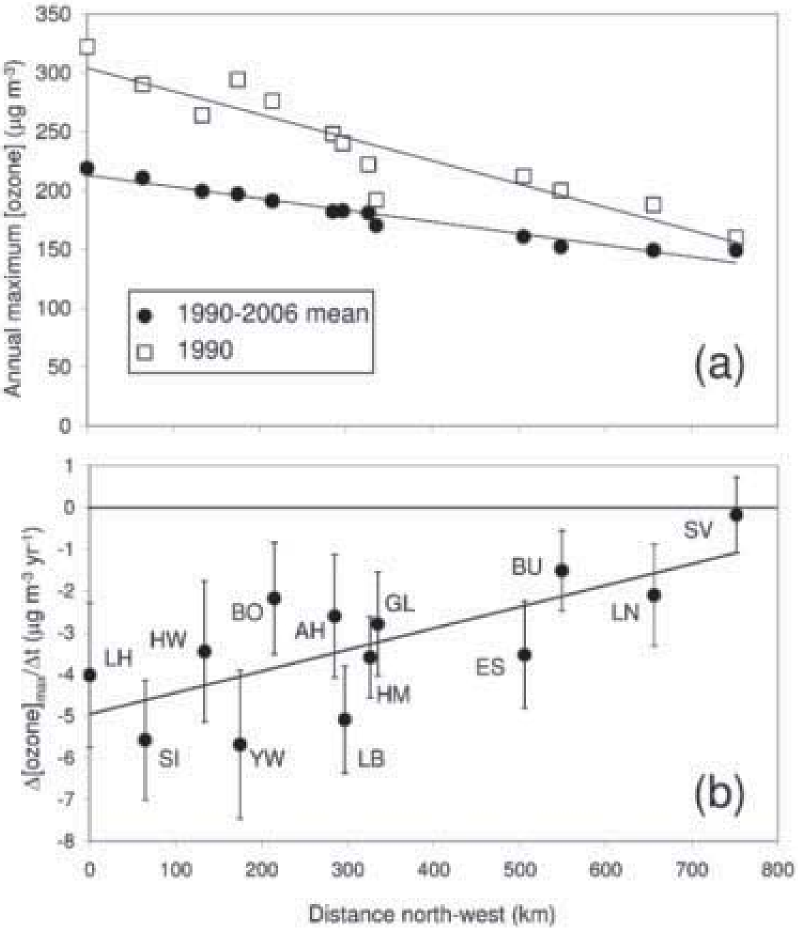


Figure 2.5 (a) Annual maximum hourly mean [ozone] at 13 UK rural and remote sites as a function of distance along a north-westerly co-ordinate (sites are identified in Figure 2.3). Data are shown for 1990 and averaged over the period 1990-2006. Lines are regressions of the data. (b) Corresponding average rate of change in annual maximum [ozone] over the period 1990-2006. Displayed error bars are 1σ, and the line is a regression of the data.

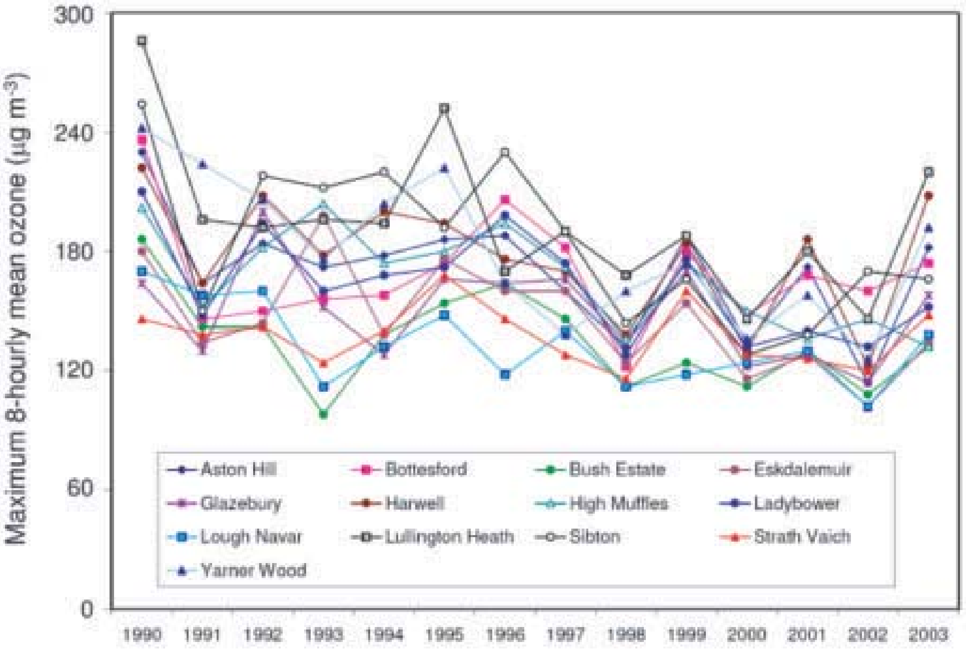


Figure 2.6 Time series of the maximum 8-hourly mean ozone concentrations monitored at a selection of long-running rural EMEP background sites between 1990 and 2003.

50. Further information on the temporal and spatial trends in elevated ozone concentrations is apparent from consideration of the annual maximum hourly-mean ozone concentrations recorded at the same set of 13 long-running sites over the period 1990-2006. Figure 2.5a shows that the maximum concentrations show an approximately linear decline with distance north-west, as illustrated for 1990 and for the average of all years in the time series. A significant decreasing trend in the annual maximum ozone concentration is apparent over the period at all sites except Strath Vaich (Figure 2.5b), indicative of a decreasing intensity of regional-scale ozone pollution episodes. This observed decrease in the frequency and severity of photochemical ozone events in the UK, is consistent with that expected from reductions in the emissions of anthropogenic VOC and NO_X in the EU since the early 1990s (Derwent *et al.*, 2003), as will be discussed further below. Figure 2.5b also shows that the absolute magnitude of the decreasing trend diminishes with distance north-west, as the sites become less impacted by regional-scale processes, as discussed above. Figure 2.6 shows that the decreasing intensity of the regional-scale ozone pollution episodes can also be illustrated using the annual trends in the maximum 8-hour mean ozone concentrations monitored during each year at the selection of long-running rural sites, with the majority showing downwards trends that are statistically significant.

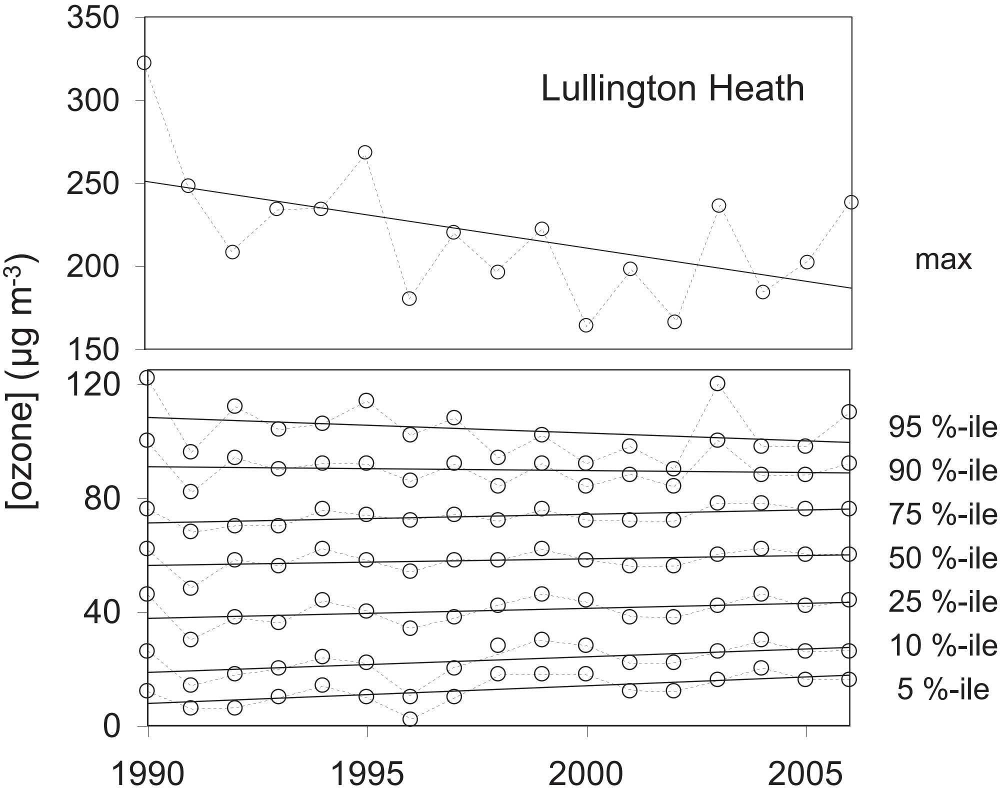


Figure 2.7 Trend in hourly mean ozone distribution at Lullington Heath (a rural site in southern England) based on data over the period 1990-2006. Solid lines are linear regressions of data indicating the average trend over the period.

2.7.2 Temporal trends in ozone distributions at rural locations

51. Figure 2.7 demonstrates the trend in the hourly-mean ozone distribution at Lullington Heath over the period 1990-2006. Figure 2.8 presents the associated rates of change in the annual maximum, annual minimum and selected percentiles of the hourly-mean ozone concentrations, in comparison with those observed at a number of other locations, including the remote site at Strath Vaich discussed above (section 2.6). The progressive decreases in the maximum and the 95th and 90th percentiles at Lullington Heath reflect the general decrease in regional-scale ozone formation, these statistics corresponding to elevated ozone concentrations during summertime photochemical episode events, as discussed above. As indicated in Figure 2.8, similar trends have also been observed at other long-running rural sites in southern UK. As a result of this decreasing regional component, the trends in the percentiles at these sites remain more negative than those observed at the remote site at Strath Vaich down to about the 50th percentile.

52. The upward trends in the lower percentiles at the rural sites tend to slightly exceed those observed at Strath Vaich, which can be explained by an additional contribution (i.e. over and above the increasing hemispheric background) resulting from a decreasing trend in removal by reaction with locally-emitted NO, these statistics probably corresponding to wintertime minima when a shallow inversion layer can cause elevated NO_X concentrations even at such rural sites (for example, see ozone depletion events in Figure 2.2 for Lullington Heath). The annual minimum hourly-mean ozone concentration shows no trend because the concentration is essentially zero in each of the years.

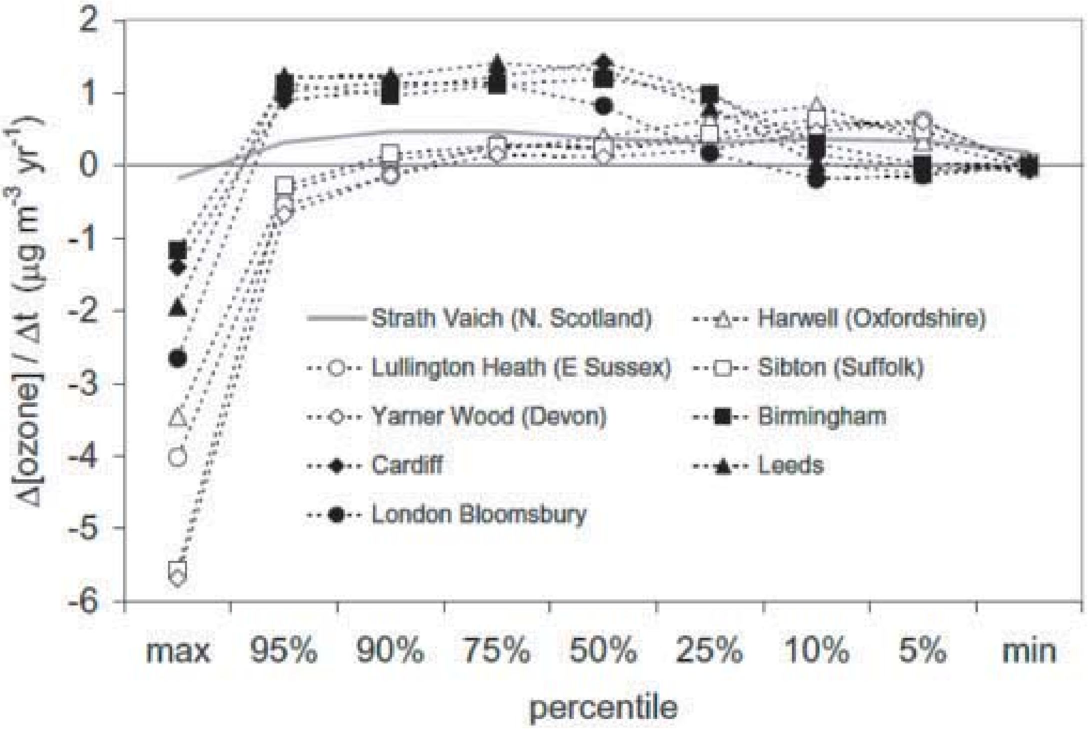


Figure 2.8 Rate of change in hourly mean ozone concentration at a number of locations based on annual maximum, annual minimum and selected percentiles. The grey line represents observations from the remote site at Strath Vaich (1990-2006). Open symbols are selected rural sites in southern England (1990-2006) and closed symbols are selected urban sites (1993-2006).

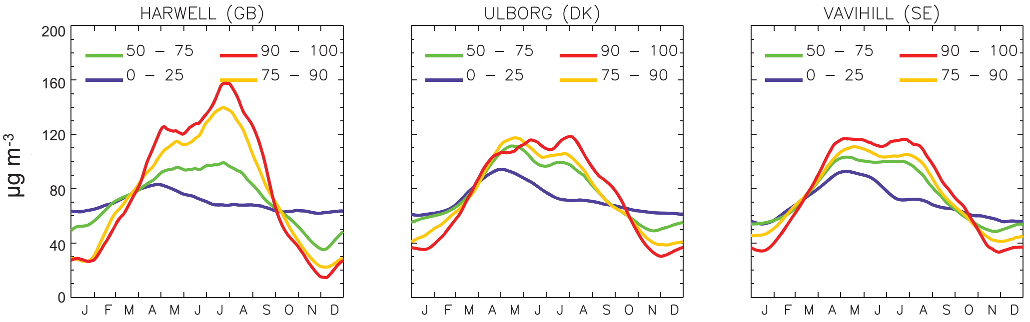


Figure 2.9 Smoothed seasonal cycles of ozone concentrations (based on 1988-1996 data) at three surface sites in Europe as a function of the NO_X emissions integrated along back trajectories from the time of measurement. The different curves represent seasonal cycles for the indicated percentile ranges of the trajectory integrated NO_X emissions. Adapted from the synthesis and integration report of the TROTREP project (Monks *et al.*, 2003).

53. The distribution trend at Lullington Heath (Figure 2.7), and other similar sites, is thus clearly influenced by the local-, regional- and global-scale effects indicated above, with a general narrowing of the distribution with time. Some additional evidence for the role of chemical processes in elevating ozone concentrations in the summer (through photochemistry involving VOC and NO_X) and removing ozone in the winter (through direct reaction with emitted NO) comes from the observations of seasonal ozone concentrations at rural sites in relation to pollutant input. Figure 2.9 shows smoothed data for Harwell (Oxfordshire) and for rural sites in Denmark and Sweden averaged over the period 1988-1996. The data have been categorised in terms of integrated NO_X emissions along trajectories arriving at the given location, and demonstrate that polluted air masses showed a summer surplus and a winter deficit in ozone with increasing seasonal amplitude associated with increased anthropogenic emissions. The difference between the "polluted" and "clean" seasonal cycles therefore quantifies the impact of the chemical modification which, over the period considered at Harwell, peaked at about -40 µg m^{-3} in winter and +80 µg m^{-3} in summer. The time series in Figure 2.7 shows that the magnitude of these modifications at Lullington Heath (and other similar rural UK sites) has progressively diminished since the early 1990s, in response to EU VOC and NO_X emissions controls, resulting in the narrowing of the ozone distribution referred to above.

54. Figure 2.5 and Figure 2.8 show that the average trends in the maximum ozone concentrations at the rural southern UK sites over the period 1990-2006 are typically in the region of -4 to -6 µg m^{-3} yr^{-1}. The trends in the 99.9th percentile of the hourly mean ozone concentrations at a larger series of 18 rural and remote sites in the UK have also been analysed, based on data from all available years up to 2005. The results are summarised in the histogram in Figure 2.10, with the detailed site-specific information presented in Table A2-1 and Figure A2.1 (in Annex 2). The 99.9th percentile at a given site typically shows comparable, but slightly smaller, rates of decrease compared with the maxima. The average trend of the longer-running rural and remote sites (located throughout the UK) is -2.4 µg m^{-3} yr^{-1}, with eight sites showing statistically significant downward trends. The data for these sites are presented geographically in Figure 2.11. Consistent with the data for the annual maxima discussed in the previous section, these tend to show a north-south gradient in the rate of decline.

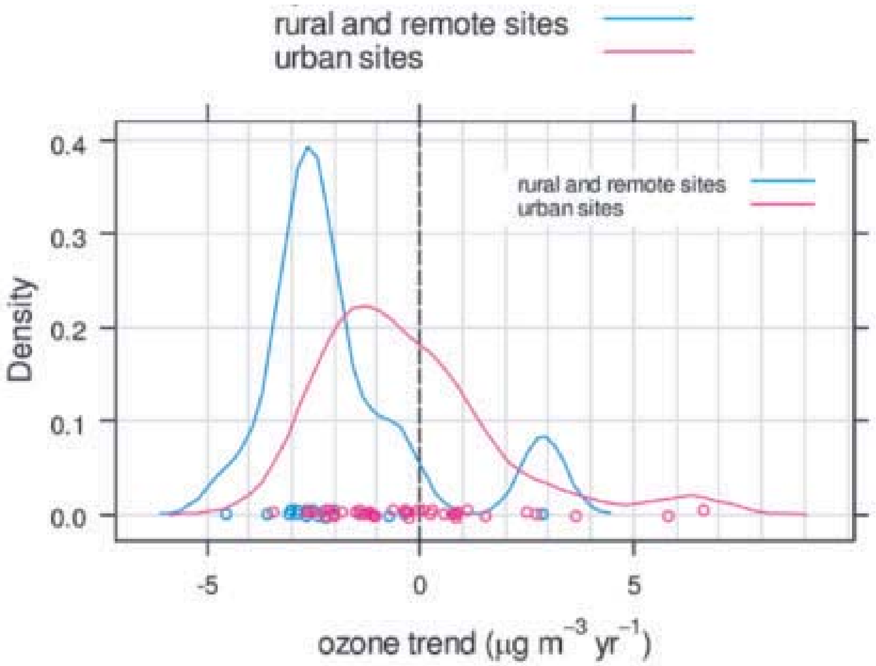


Figure 2.10 Distribution of trends in the 99.9th percentile of the hourly-mean ozone concentration at 18 rural/remote sites (blue) and 45 urban sites (pink) in the UK, based on data up to 2005. The circles show the value of the trend at individual sites and the area under each curve is one. The site-specific information is presented in Tables A2-1 and A2-2 (in Annex 2).

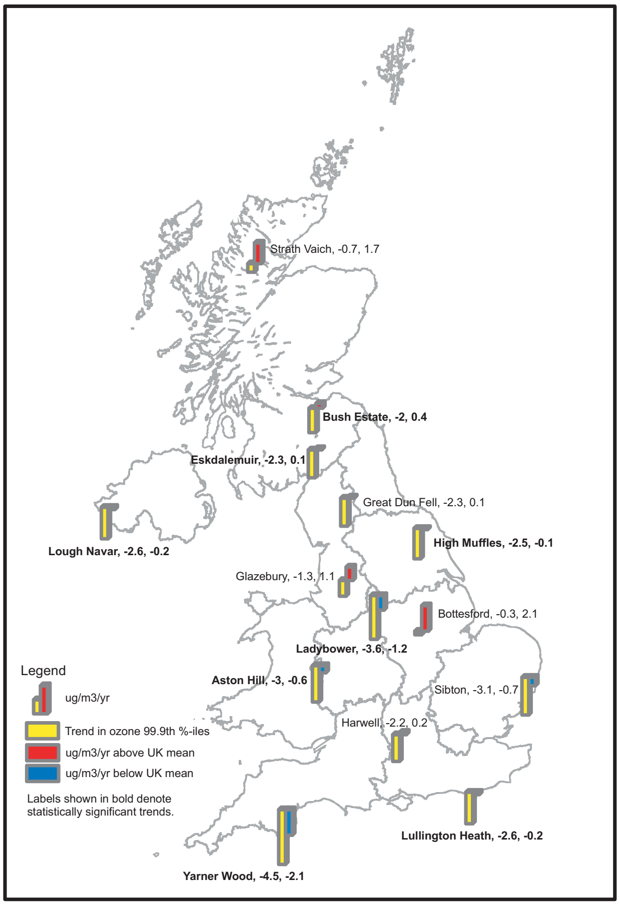


Figure 2.11 Spatial patterns in the average trends in 99.9th percentile hourly-mean ozone concentration at rural and remote sites (yellow bar) and their relationship to the UK mean of -2.4 μg m^{-3} yr^{-1}, based on data up to 2005.

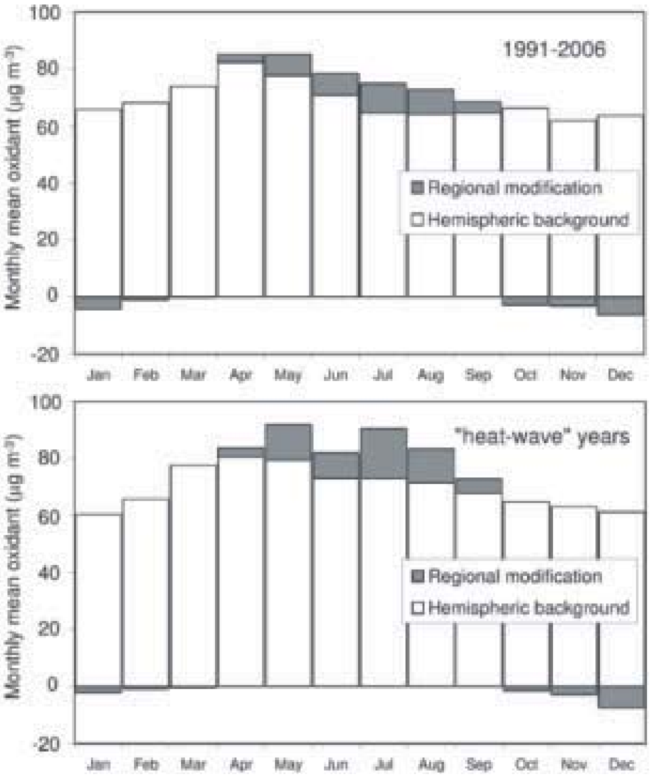


Figure 2.12 Seasonal variation of monthly mean background oxidant (i.e. ozone) contributions at Lullington Heath, based on data averaged over the period 1991-2006 (upper panel) and for the average of the heat-wave years, 1995, 2003 and 2006 (lower panel).

2.7.3 Temporal trends in background oxidant sources at Lullington Heath

55. Data from the rural site at Lullington Heath, for the period 1991-2006, have been analysed to estimate the relative contributions of sources of oxidant. As described in detail elsewhere (e.g. AQEG, 2004), the oxidant concentration is defined as the sum of the concentrations of ozone and NO_2. This quantity has been shown to be made up of a combination of a background (NO_X-independent) source and a local (NO_X-dependent) source (Clapp and Jenkin, 2001). The former effectively equates to the background ozone concentration, and the latter is derived from primary NO_2 emissions. The background contribution thus provides an estimate of the ozone concentration which would exist at the given location in the notional absence of NO_X, i.e. when local removal by reaction with emitted NO and (less significantly) local production from emitted NO_2 have not occurred.

56. Figures 2.12 and 2.13 show the seasonal variation and long-term trend in the background oxidant (i.e. ozone) concentration at Lullington Heath, with this background being further separated into estimated "global (hemispheric)" and "regional" contributions. These quantities have been determined by, first, removing the "local" contribution from the measured oxidant, based on the observed concentration of NO_X and an inferred average fractional contribution of NO_2 to NO_X emissions of 9.3% (Jenkin, 2004). This allowed the background oxidant (i.e. ozone) concentration to be determined. The background was then separated into the hemispheric background and a regional modification to this background, on the basis of air mass histories described by four-day back

trajectories for each day of the 16-year time period (those arriving from the west being used to define the background, with the regional modification being obtained by difference). These quantities thus provide an estimate of the background ozone concentration, upon which the regional modification is superimposed.

57. Figure 2.13 shows the seasonal variation as monthly mean values averaged over the whole period 1991-2006 (upper panel) and for the average of the heat-wave years, 1995, 2003 and 2006 (lower panel). The regional modification results from a combination of regional-scale photochemical ozone formation and increased removal of ozone through deposition when air masses have travelled over the continent prior to arrival. The regional modification is thus notably positive in the summer months, when photochemical formation is the dominant influence, but negative in the wintertime when net removal through deposition occurs. The summertime regional enhancement is, on average, greatest in July and August when most photochemical episodes occur, with average monthly mean contributions over the whole period of *ca.* 10 µg m^{-3}. The seasonal variation of the inferred hemispheric background shows the springtime maximum typically observed for ozone at remote sites, as discussed above and in chapter 3.

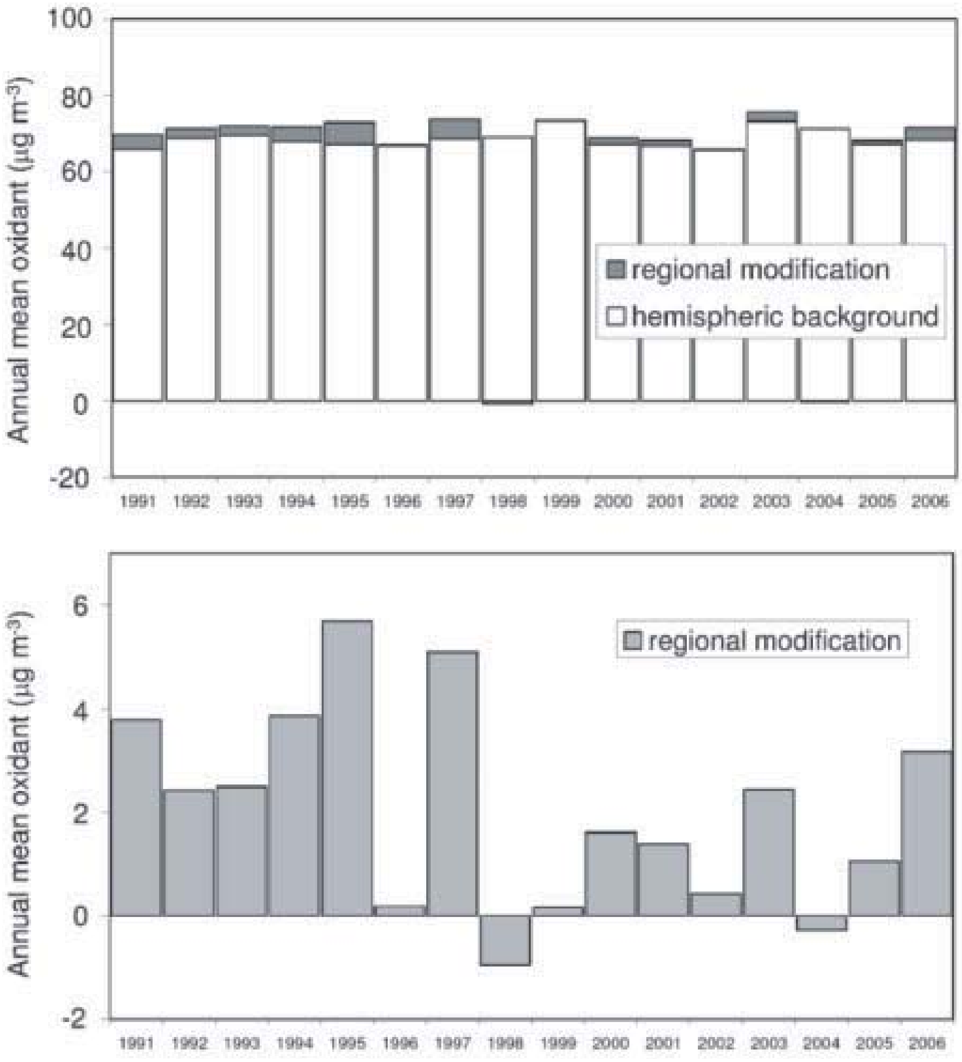


Figure 2.13 Time series of the annual mean background oxidant (i.e. ozone) contributions at Lullington Heath. The regional contribution is shown on an expanded scale in the lower panel.

58. Figure 2.13 shows the time series of the oxidant components on an annual mean basis over the 16-year period. The regional modification shows year-on-year variability owing to variation in the meteorological conditions experienced, but with net regional-scale ozone formation occurring in most years. A general decreasing trend in the regional oxidant contribution is apparent, consistent with the impact of EU controls on VOC and NO_X emissions. The heat-wave year of 1995 shows the largest regional contribution in the early part of the time series, with 2003 and 2006 having the largest contributions in the later years. The inferred annual mean hemispheric background ozone concentration has a barely significant upward trend of 0.08 $\mu g\ m^{-3}\ yr^{-1}$. However, consideration of data over the period 1991-1999 yields a more notable upward trend of 0.4 $\mu g\ m^{-3}\ yr^{-1}$ with no clear trend subsequently. The variation in the inferred hemispheric background thus shows some of the features observed at Strath Vaich (see section 2.7) and at the Mace Head site on the west coast of Ireland, as discussed in Chapter 3

Box 2.2 Daily metrics of ozone exposure

Hourly mean ozone concentrations show characteristic diurnal cycles with lowest levels during the early morning and highest levels during the mid-afternoon. Daily metrics are single numbers chosen to represent this wide dynamic range in concentrations. The daily metrics most commonly used in ozone air quality studies are: maximum hourly mean concentration, maximum 8-hourly mean concentration and daily mean concentration.

Each of these commonly used metrics can describe the observed dynamic range in ozone concentrations. This is because they are strongly correlated with each other, though the detailed nature of these correlations may vary from site to site and from year to year.

For example, at a typical urban background monitoring site in central London during 1993, there were 350 days with valid daily ozone metrics and these were correlated as follows:

Max 1-hour mean = 1.195 x max 8-hour mean $r^2 = 0.93$
Daily mean = 0.56 x max 8-hour mean $r^2 = 0.85$

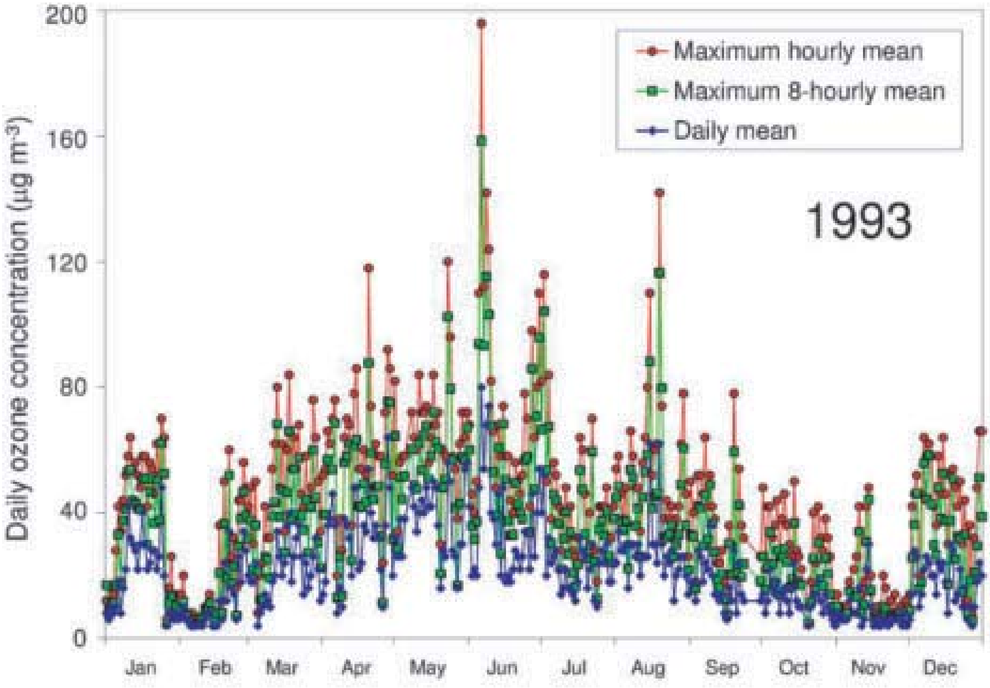


Figure 2.14 The time series of three daily ozone metrics for the urban background site in central London during 1993.

2.7.4 Temporal trends in human health ozone metrics at rural locations

59. Making the assumption that there is no threshold for ozone and human health effects, there remains an important question concerning the relevant ozone metric against which ozone monitoring data should be compared to derive policy-relevant conclusions concerning recent air quality trends. Box 2.2 describes some of the more commonly used daily ozone metrics, with Figure 2.14 illustrating the close correlation observed between maximum hourly mean, maximum 8-hourly mean and daily mean concentrations at a typical urban background site in central London. Figure 2.15 shows that the annual mean of the daily maximum running 8-hour mean concentration is closely correlated with the annual mean ozone concentration for a series of site types, and this is illustrated further using trend data for 18 rural and remote UK sites up to 2005 in Figure A2.2 (in Annex 2).

60. In the following discussion, we consider that the annual average of the daily maximum 8-hour mean ozone concentrations is an appropriate ozone metric, and the EMEP ozone database has been recompiled on this basis. There are 46 EMEP rural sites, located across Europe at elevations below 500 m, that have long enough observational records to enable robust trend analysis. An analysis of the trends in the annual averages of daily maximum 8-hour mean ozone concentrations for the period from 1990 through to 2002 has been carried out, the detailed results of this analysis being presented in Table A2-3 (in Annex 2). Of the 46 EMEP sites, 17 showed downwards trends and 29 showed upwards trends. Of these, 11 sites showed highly statistically significant upwards trends and two showed similarly significant downwards trends. Taking all the sites together, the average trend was (0.3 ± 0.6) µg m^{-3} yr^{-1}. For comparison, an analysis of the trends in the annual mean concentrations at 18 rural and remote sites in the UK, based on data from all available years up to 2005, has also been carried out. The results are summarised in the histogram in Figure 2.16, with the detailed site-specific information presented in Table A2-1 and Figure A2.2 (in Annex 2). On average there has been an increase in annual mean ozone at these rural and remote sites (located throughout the UK) of 0.4 µg m^{-3} yr^{-1}, with six of the 18 sites showing statistically significant increases in the trend.

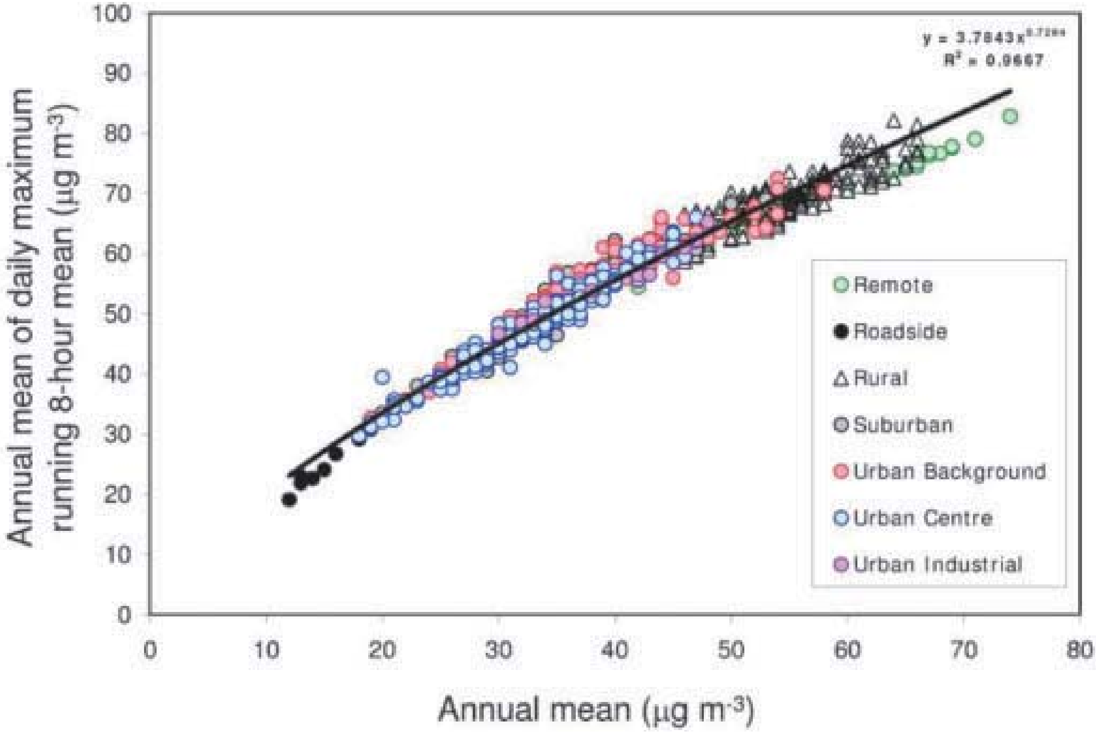


Figure 2.15 Relationship between measured annual mean of the maximum daily running 8-hour mean ozone concentration and the annual mean ozone concentration, for a series of site classifications in the UK.

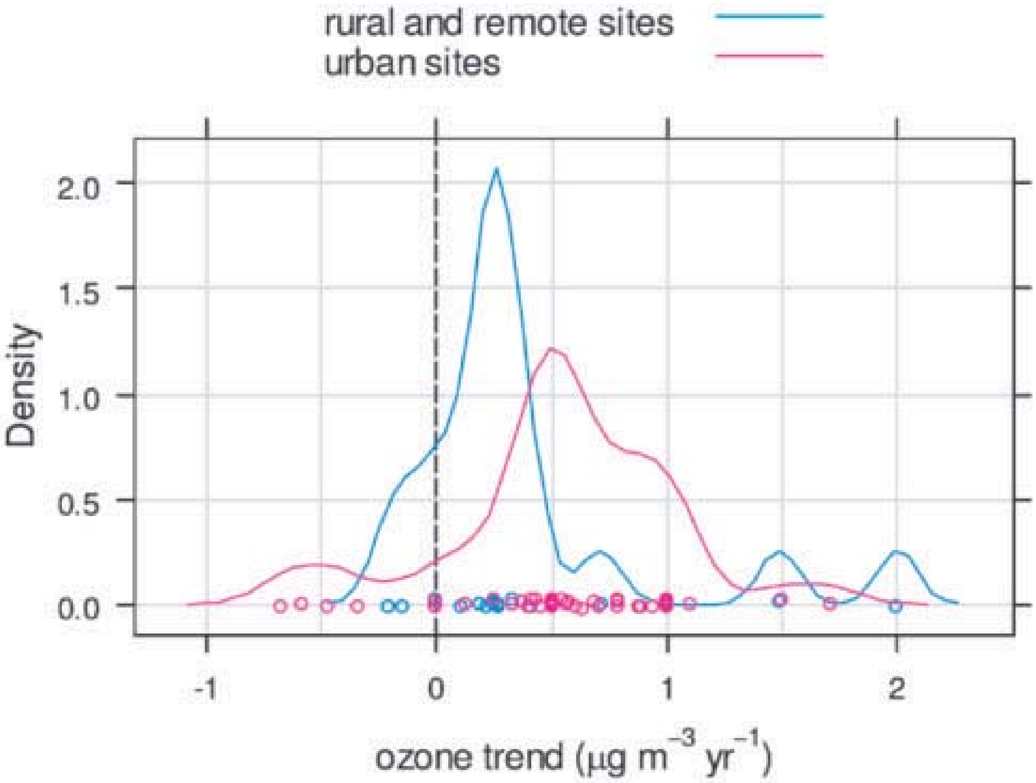


Figure 2.16 Distribution of trends in the annual mean ozone concentration at 18 rural/remote sites and 45 urban sites in the UK based on data up to 2005. The circles show the value of the trend at individual sites and the area under each curve is one. The site-specific information is presented in Tables A2-1 and A2-2 (in Annex 2).

61. To sort out what have been the major influences on the observed long-term trends in the annual average daily maximum 8-hour mean ozone concentrations at the 46 EMEP rural sites, Figure 2.17 plots out the observed trends for the sites identified in Table A2-3 (in Annex 2) over the 1990-2002 period, against the initial 1990 value of the ozone metric. This shows that there is a correlation between the magnitude of the observed ozone trend and the initial 1990 value of the ozone metric. Those EMEP sites with low initial values of the ozone metric, such as those shown in Figure 2.17 in Germany and Belgium, show strong upwards trends. These are sites which were influenced by traffic emissions initially. As NO_X emissions have decreased across Europe, NO_X-driven depletion of ozone has steadily reduced and levels of the ozone metric have increased. At sites heavily influenced by long-range transboundary transport, such as the Netherlands site in Figure 2.17, initial levels of the ozone metric were high because the sites were located away from traffic. Europe-wide measures to reduce ozone precursor VOC and NO_X emissions have steadily reduced levels of the ozone metric, leading to the observed downwards trends over the 1990-2002 period. Sites such as Mace Head, see Figure 2.17, that are on the Atlantic Ocean seaboard of Europe, are strongly influenced by the increasing hemispheric and global ozone background and show increasing trends over the 1990-2002 period.

62. The observations of rural ozone levels in Europe can therefore be rationalised in terms of the net effect of the three major influences on ozone concentrations identified above. Overall, an approximate balance has been maintained between these influences over the 1990-2002 period at the rural EMEP sites.

63. An analysis of the trends in the annual average of the daily maximum of the running 8-hour mean concentration with a cut-off at 70 µg m^{-3} at rural and remote sites in the UK has also been carried out since this metric is closely related to the SOMO35 metric that has been used in European-scale integrated assessment modelling (see Table 1-1). The results are summarised in the histogram in Figure 2.18 with the detailed site-specific information presented in Table A2-1 and Figure A2.5 (in Annex 2). Overall there is little evidence of a consistent trend in this metric at these sites, with most sites showing considerable year-to-year variability with some indications of a small decrease but no clear trend. This suggests that the different influences on ozone concentration have approximately cancelled each other out, resulting in no clear trend. This is in contrast to the general upward trend in annual mean concentration at rural and remote sites illustrated in Figure 2.16.

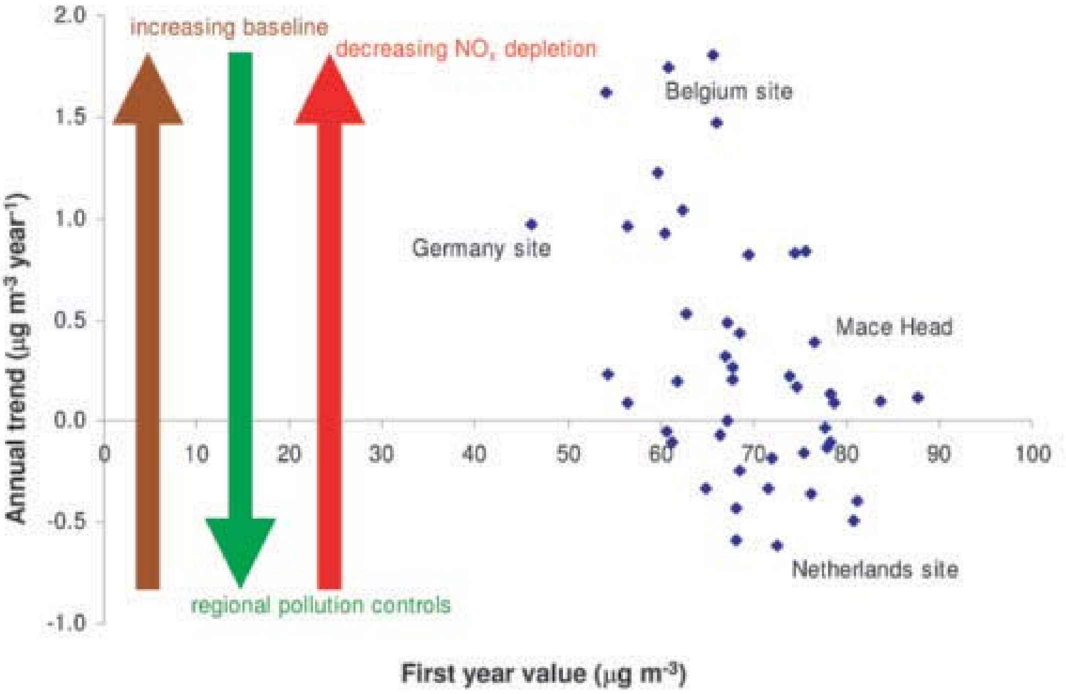


Figure 2.17 A scatter plot of the trends in the annual average daily maximum 8-hour mean ozone concentrations observed at 46 EMEP rural sites over the period 1990-2002 plotted against the initial 1990 value of the ozone metric.

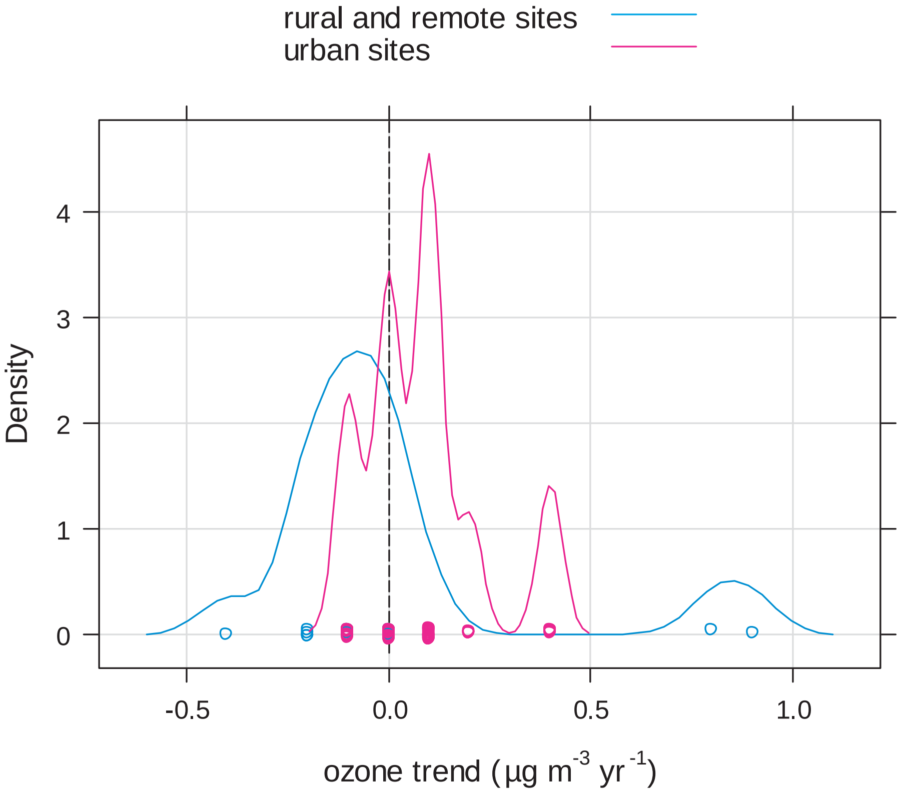


Figure 2.18 Distribution of trends in the annual average of the daily maximum of the running 8-hour mean with a 70 µg m^{-3} cut-off at 18 rural/remote sites (blue) and 45 urban sites (pink) in the UK, based on data up to 2005. The circles show the value of the trend at individual sites and the area under each curve is one. The site-specific information is presented in Tables A2-1 and A2-2 (in Annex 2).

2.8 Ozone observations at UK urban network locations

2.8.1 Temporal trends in elevated ozone events at urban locations

64. Urban locations are typically characterised by higher concentrations of NO_X than surrounding rural areas, and the balance between ozone production and loss (compared with the hemispheric background) is shifted as a result of the increased impact of local scavenging of ozone by reaction with emitted NO. This is apparent in Figure 2.2 for the Birmingham Centre site, for which the daily ozone concentrations are generally lower than those observed at the Lullington Heath rural site throughout the year. Despite this, the short-term elevations in ozone concentrations during regional-scale episodes in the summertime are still apparent, such that the levels on such occasions are typically observed to be greater at urban locations within the UK than those observed at remote north-westerly sites such as Strath Vaich, which are less impacted by regional-scale European pollution.

65. Figure 2.19 shows the maximum hourly-mean ozone concentration recorded at London sites over the period 1989-2006, compared with the trend at long-running rural sites. A notable urban decrement is apparent in the early part of the time series, but with the maxima at the rural sites and the London sites being comparable in more recent years. This demonstrates the reducing impact of local ozone scavenging by reaction with NO as NO_X emissions have declined over the period. As a result, the effect of EU-wide reductions in VOC and NO_X emissions in reducing peak ozone levels is almost cancelled out by the increasing impact of local NO_X reduction at urban sites, such that the decline in the maximum values at urban sites (if any) is much more subtle than that observed at rural locations.

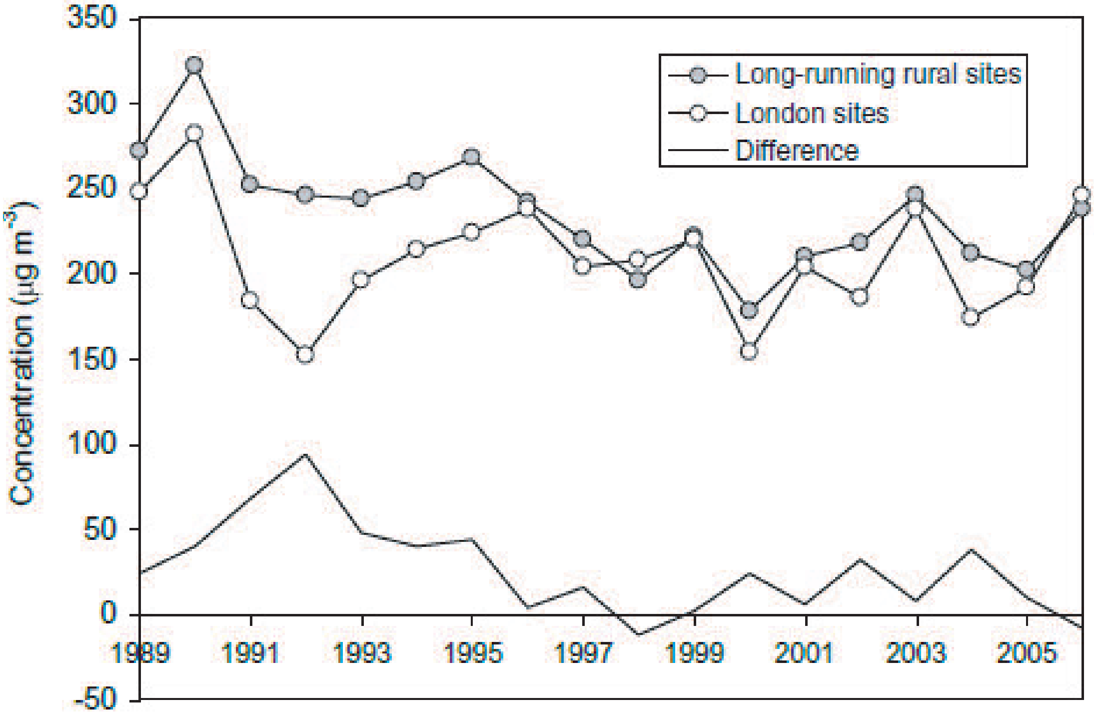


Figure 2.19 Maximum hourly mean ozone concentrations in each year at the long-running rural sites identified in Figure 2.3 and at London sites over the period 1989-2006. The difference between the rural and London sites for the given ozone metric is termed the "urban decrement".

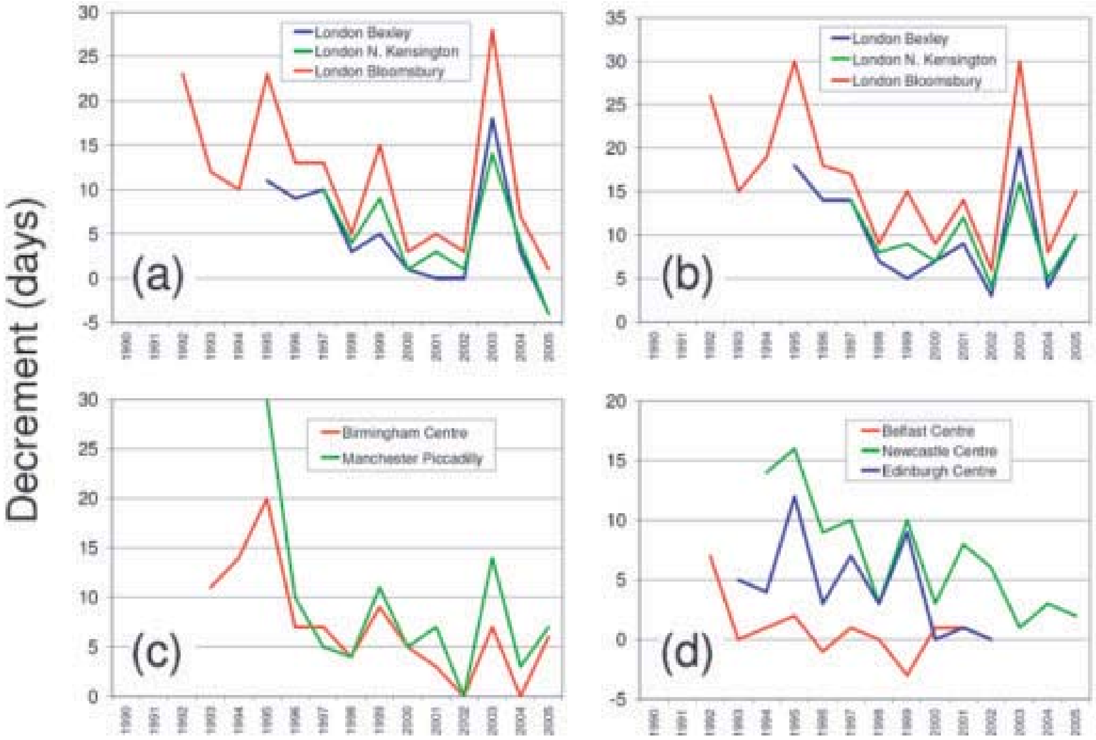


Figure 2.20 The decrement in the number of days the daily maximum running 8-hour mean concentration exceeds 120 µg m^{-3} at urban sites compared with paired rural sites, since the early 1990s. (a) Selected London sites, relative to Harwell; (b) Selected London sites, relative to Lullington Heath; (c) Birmingham Centre and Manchester Piccadilly, relative to Aston Hill; (d) Belfast Centre, relative to Lough Navar, Newcastle Centre, relative to High Muffles, and Edinburgh Centre, relative to Eskdalemuir.

66. The general reducing trend in the urban ozone decrement is also apparent from an analysis of daily maximum running 8-hour mean concentrations. Figure 2.20 shows that the number of days the daily maximum running 8-hour mean concentration exceeds 120 µg m^{-3} at selected urban sites in a given year has tended towards the number observed at paired rural sites in the same region since the early 1990s.

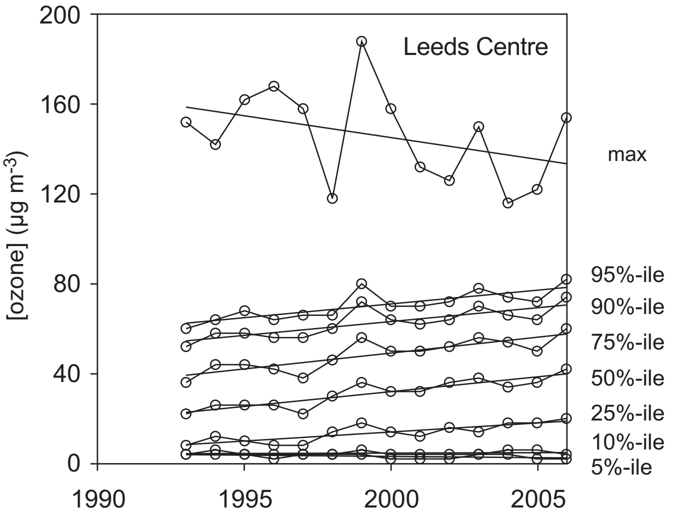


Figure 2.21 Trend in the hourly mean ozone distributions at the Leeds Centre urban site based on data over the period 1993-2006. Solid lines are linear regressions of data indicating the average trend over the period.

2.8.2 Temporal trends in ozone distributions at urban locations

67. Figure 2.21 demonstrates the trend in the hourly mean ozone distribution at an example urban location, Leeds Centre, over the period 1993-2006. Figure 2.8 presents the associated rates of change in the annual maximum, annual minimum and selected percentiles of the hourly mean ozone concentrations, in comparison with those observed at a number of other urban locations, and at the remote and rural sites discussed above in sections 2.6 and 2.7. At Leeds Centre (Figure 2.21), a general decrease in the annual maximum hourly mean ozone concentration is discernable. This indicates that the effects of regional-scale ozone precursor controls can be observed at comparatively polluted urban locations. As shown in Figure 2.8, similar observations are apparent at other long-running urban sites in England and Wales (Birmingham, Cardiff and London Bloomsbury). However, the lower percentiles at all these sites show an increasing trend, with the influences of decreasing removal by reaction with NO locally having the overriding effect on the distribution as a whole. As shown in Figure 2.8, the upward trends in the 95th through to the 25th percentiles at Leeds, Birmingham and Cardiff are of the order of 1 $\mu g\ m^{-3}\ yr^{-1}$, notably greater than observed at the rural sites. The low end of the percentile range ($\leq$ 10th percentile) are concentrations which are at, or close to, zero and show very little trend. These correspond to conditions where the NO_X concentration is sufficiently high that there is enough NO effectively to remove all ozone, so that even decreasing emissions does not lead to a notable increase in ozone concentration (i.e. the distribution is truncated because it hits zero). At the Bloomsbury site in central London, the maximum upward trend is also of the order of 1 $\mu g\ m^{-3}\ yr^{-1}$, but the trend collapses to zero at a higher percentile because the NO_X concentrations are generally greater, compared with the other urban centre sites.

68. Further evidence for the generally increasing trend across the distribution is shown in Figure 2.22. This presents the frequency distributions of the 8760 hourly ozone mean concentrations measured at an urban background site in central London during 1991 and 1998. These data also show a marked shift in the frequency distribution of ozone concentrations over this period, bringing a much reduced frequency of low ozone concentrations < 10 $\mu g\ m^{-3}$ and a much increased frequency of ozone concentrations in the 20-80 $\mu g\ m^{-3}$ range. This is likely to be due mainly to reduced NO_X emissions which deplete urban ozone, but will also reflect the steadily increasing ozone background, especially during wintertime. Similar behaviour is anticipated in most towns and cities in north-west Europe.

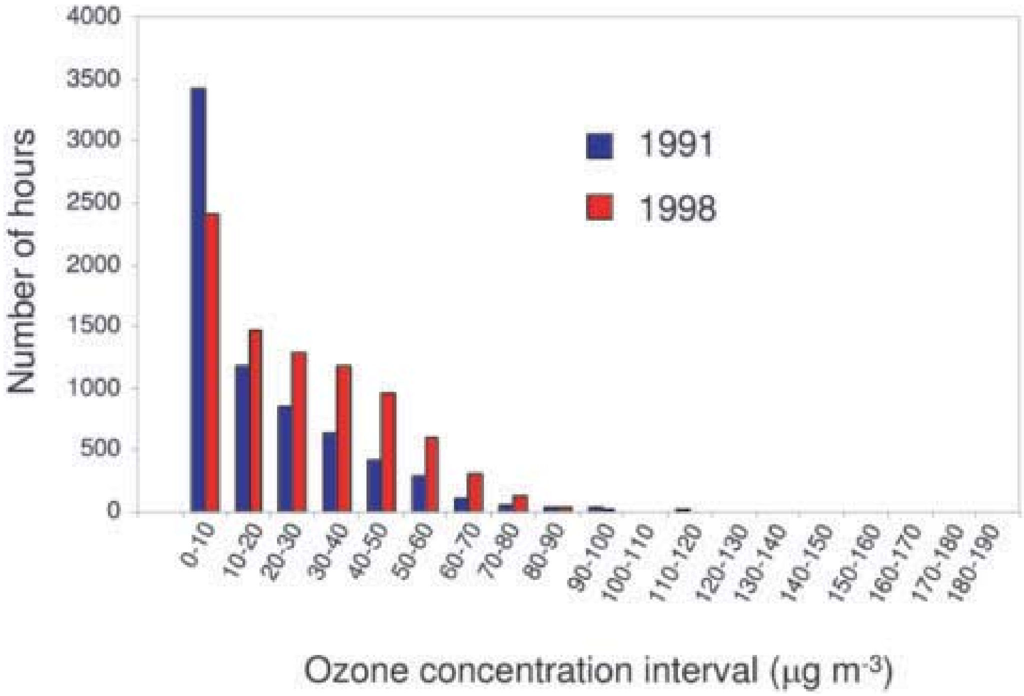


Figure 2.22 The frequency distributions of the hourly ozone concentrations measured during 1991 and 1998 at a typical urban background site in central London.

69. The data in Figure 2.8 demonstrate that maximum ozone concentrations at the selected urban UK sites have tended to show some decline since the early 1990s, although the rate of decrease at individual sites is generally not statistically significant. Analysis of a wider dataset also shows a range of average trends in the maximum, partly because summertime regional-scale ozone formation has less of an impact at urban sites to the north and west of the UK, such that the decreasing regional component is expected to be less apparent. Figure 2.10 shows a summary of the trends in the 99.9th percentile of the hourly-mean ozone concentrations at 45 urban sites in the UK, based on data from all available years up to 2005, with the detailed site-specific information presented in Table A2-2 and Figure A2.4 (in Annex 2). Although none of the individual sites shows a significant trend in the 99.9th percentile, the average trend is -0.3 $\mu g\ m^{-3}\ yr^{-1}$. As shown in Figure 2.10, the absolute rate of decline in these top-end ozone concentrations at urban sites is systematically lower than those observed at rural and remote locations, consistent with the discussion in the previous section.

70. Although the distributions of ozone concentrations at urban locations have displayed logical trends, primarily in response to the general decreasing trend in local- and regional-scale emissions, it is also apparent that some degree of year-to-year variability results from the different meteorological conditions experienced. Figure 2.23 shows the ozone distributions and annual mean concentrations for the London Bloomsbury and Leeds Centre sites for the heat-wave years of 1995 and 2003, and for the more typical meteorological year of 2005. The distributions for the Lullington Heath (rural) and Strath Vaich (remote) sites are also shown for comparison. The data for 2003 at the urban sites show a general shift to higher concentrations when compared with the 1995 data, owing to the reducing impact of local ozone removal by reaction with NO. However, the data for 2005 are generally shifted back to lower

concentrations than observed in 2003, because of the lower frequency of regional-scale photochemical ozone episodes in 2005, compared with both 2003 and 1995. The year-to-year variability in the hemispheric background (discussed in Chapter 3) also has an underlying effect. This is particularly apparent for the annual mean concentrations at the rural and remote sites, which show a clear elevation in 2003 compared with the other years. The data in Figure 2.23 therefore demonstrate once again that ozone concentrations at UK locations are influenced by trends and variability in processes on local, regional and global scales.

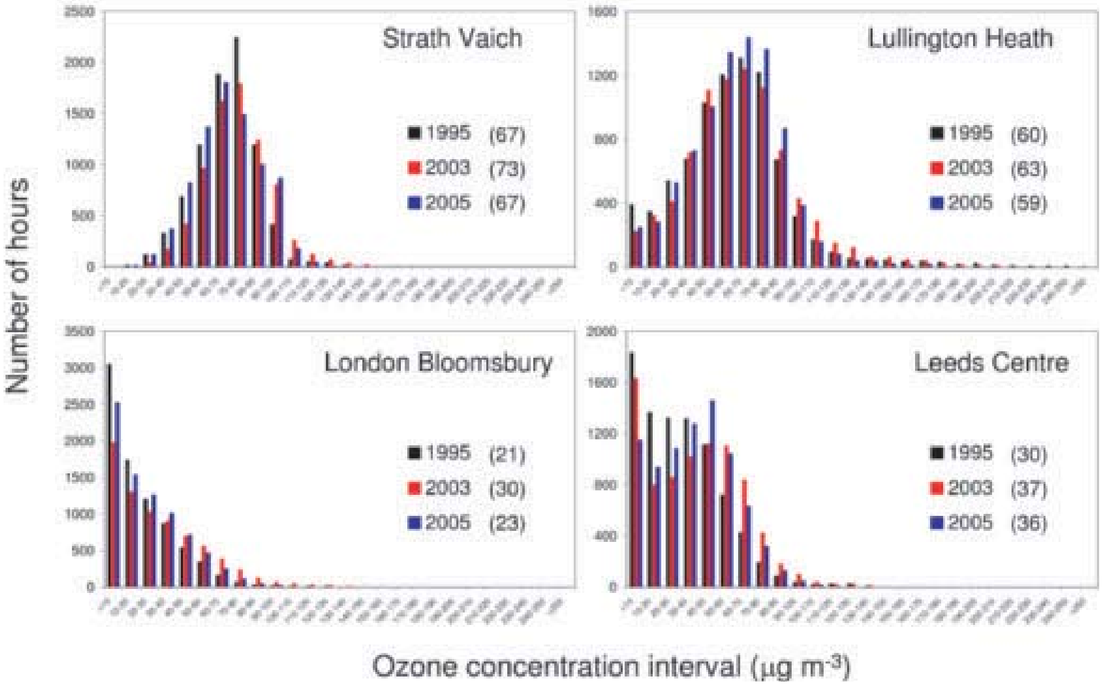


Figure 2.23 Frequency distributions of the hourly-mean ozone concentrations measured during 1995, 2003 and 2005 at selected urban, rural and remote locations in the UK. Annual mean concentrations (in μg m^{-3}) are also given in brackets.

2.8.3 Temporal trends in human health ozone metrics at urban locations

71. Following on from the analysis presented for rural sites in section 2.7.4, here we consider trends in urban ozone levels in terms of the human health metrics, the annual mean of the daily maximum running 8-hour mean concentration, the annual mean concentration and also the annual mean of the daily maximum running 8-hour mean concentration with a cut-off at 70 μg m^{-3}. Similarly to the conclusion for rural sites, Figure 2.15 shows that the first two of these metrics are closely correlated, and this is illustrated further using trend data for 45 UK sites in Annex 2 (section A2.1).

72. Although monitoring of the ozone levels in rural locations in Europe began during the 1970s, monitoring in urban locations is a relatively recent activity. There are over 1000 urban ozone monitoring sites contributing to the AIRBASE database of the EU in the 2000s. With this large increase in urban ozone monitoring, it is clear that urban ozone levels are substantially reduced below rural levels by NO_X-driven depletion processes. This depletion is increasingly more apparent when comparing suburban with urban background and traffic-influenced sites.

73. In view of the importance of urban exposure levels, a clear understanding of urban ozone trends is required. As an example, trends in the urban ozone metrics are examined for the UK over the period from 1994-2003. It is anticipated that the ozone behaviour found is typical of that experienced in most European towns and cities. The trends in the annual average daily maximum 8-hour mean ozone concentration at the UK urban sites are shown in Table 2-1. Upwards trends are found at the vast majority of the urban sites, 47 out of 49, and downwards trends at only two sites. A total of 19 sites showed trends that are highly statistically significant (> 90% confidence). The London Marylebone Road site is the most heavily trafficked and most heavily polluted site of all in Table 2-1 and the site which has the lowest annual mean daily maximum 8-hour mean ozone concentrations. The low ozone levels at this site are caused by NO_X depletion and the strong upwards trend of 1.4 $\mu g\ m^{-3}\ yr^{-1}$ observed at this site has been caused by the diminution of this NO_X depletion by the reduction of NO_X emissions from petrol-engined vehicles due to the fitting of exhaust gas catalysts. This same influence is apparent across all the other urban sites in Table 2-1.

74. The strong upwards trends in the ozone metric observed at almost all of the UK urban sites have resulted primarily from the reduction in NO_X emissions. At these urban sites there appears to have been little influence of the decrease in intensity of regional pollution episodes on annual mean daily maximum 8-hour mean ozone concentrations. Similar behaviour is anticipated in most European towns and cities during the 1990s and 2000s.

75. Trends in annual mean ozone concentrations at 45 urban background, urban centre and suburban sites, based on data from all available years up to 2005, have also been analysed. The results are summarised in the histogram in Figure 2.16, with the detailed site-specific information presented in Table A2-2 and Figure A2.5 (in Annex 2). The majority of these show upwards trends, with 17 showing statistically significant increases. The mean trend of the 45 sites is an increase in ozone of 0.3 $\mu g\ m^{-3}\ yr^{-1}$. As also shown in Figure 2.16, the rate of increase in the annual mean concentrations at urban sites is systematically greater than those observed at rural and remote locations, due to decreasing scavenging from locally emitted NO. As a result of decreasing NO_X emissions since the early 1990s, the less polluted urban sites are, in some respects, now tending towards "rural" character, at least in terms of their NO_X levels. This is illustrated in Figure 2.24, which shows a comparison of mapped annual mean NO_X concentrations in the UK in 1996 and 2005, based on empirical modelling activities (Kent *et al.,* 2007). This comparison clearly demonstrates the shrinkage of the high NO_X zones, which is accompanied by greater infiltration of ozone into urban areas.

76. An analysis of the trends in the annual average of the daily maximum of the running 8-hour mean concentration with a cut-off at 70 $\mu g\ m^{-3}$ at 45 urban background, urban centre and suburban sites, based on data from all available years up to 2005 has also been carried out. The results are summarised in the histogram in Figure 2.18 with the detailed site-specific information presented in Table A2-2 and Figure A2.6 (in Annex 2). The trend in this metric is much less clear than the upward trend in annual means at these sites. The balance of the different influences on this metric typically results in a small increase over the period. This is in contrast to the small decrease for this metric at rural sites

shown in Figure 2.18. The impact of the decrease in local NO_X emissions is greater at urban sites, with many sites showing a small increase although there is considerable year-to-year variation in the value of this metric.

Table 2-1 Trends and their statistical significance in the annual mean daily maximum 8-hour mean ozone concentrations observed at urban and roadside sites in the United Kingdom during the period from 1990 onwards.

Urban sites 1994-2003	**Statistical significance**	**Trend $\mu g\ m^{-3}\ yr^{-1}$**	**Urban sites 1997-2003**	**Statistical significance**	**Trend $\mu g\ m^{-3}\ yr^{-1}$**
Belfast Centre		+0.62	Barnsley Gawber		+1.10
Birmingham Centre	*	+0.90	Bolton	+	+1.76
Birmingham East		+0.34	Bradford Centre		+1.70
Bristol Centre		+0.36	Derry		+1.58
Cardiff Centre	+	+0.90	Glasgow Centre	*	+1.16
Edinburgh Centre		+0.64	Leamington Spa		+1.44
Hull Centre		+0.18	London Brent		+1.54
Leeds Centre	*	+1.14	London Eltham		+0.42
Leicester Centre		+0.78	London Hackney		+0.96
Liverpool Centre	+	+1.50	London Haringey		+1.70
London Bexley	**	+0.98	London Hillingdon	+	+1.98
London Bloomsbury	*	+0.80	London N. Kensington		+2.02
Middlesbrough	**	+1.62	London Southwark		+1.36
Southampton Centre		+0.88	London Sutton		+1.08
Swansea	*	+1.08	London Teddington	*	+1.66
Wolverhampton Centre	+	+0.96	London Wandsworth	+	+1.62
			Manchester Piccadilly		+0.68
average	+	+0.70	Manchester South	+	+1.30
			Newcastle Centre		+1.08
Roadside sites 1997-2003			Norwich Centre		+3.72
			Nottingham Centre		+0.74
Bury Roadside	+	+1.10	Plymouth Centre		+1.90
London Marylebone Road	**	+1.42	Port Talbot		+1.14
			Reading		-0.80
			Redcar	*	+2.18
			Rotherham Centre		+1.32
			Salford Eccles		+0.62
			Sheffield Centre		+0.68
			Stoke-on-Trent Centre		0
			Swansea		+0.74
			Thurrock		+1.48
			Average		+1.52

Notes: Statistical significance is based on the non-parametric Mann-Kendall test and Sen's slope estimates and is indicated by: ** at the 0.01 level of significance, * at the 0.05 level of significance, + at the 0.1 level of significance and blank means less than the 0.1 level.

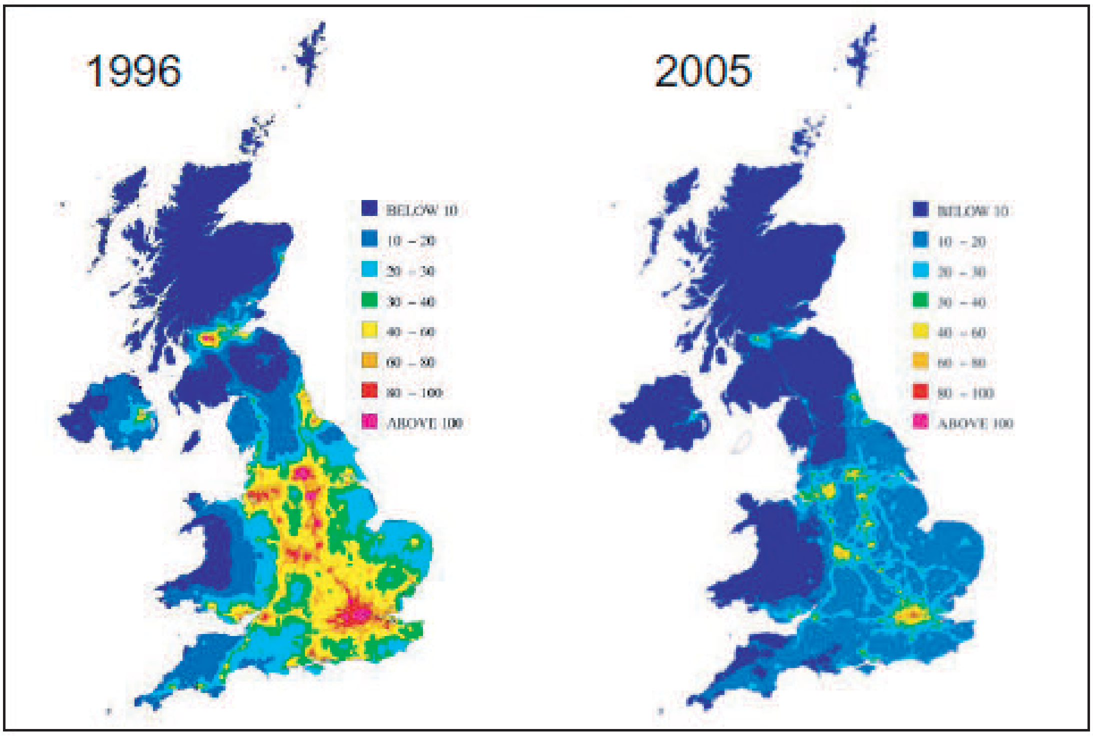


Figure 2.24 Estimated annual mean background concentrations of NO_x in the UK in 1996 and 2005, using GIS-based dispersion models calibrated using measurements (Kent *et al.*, 2007)

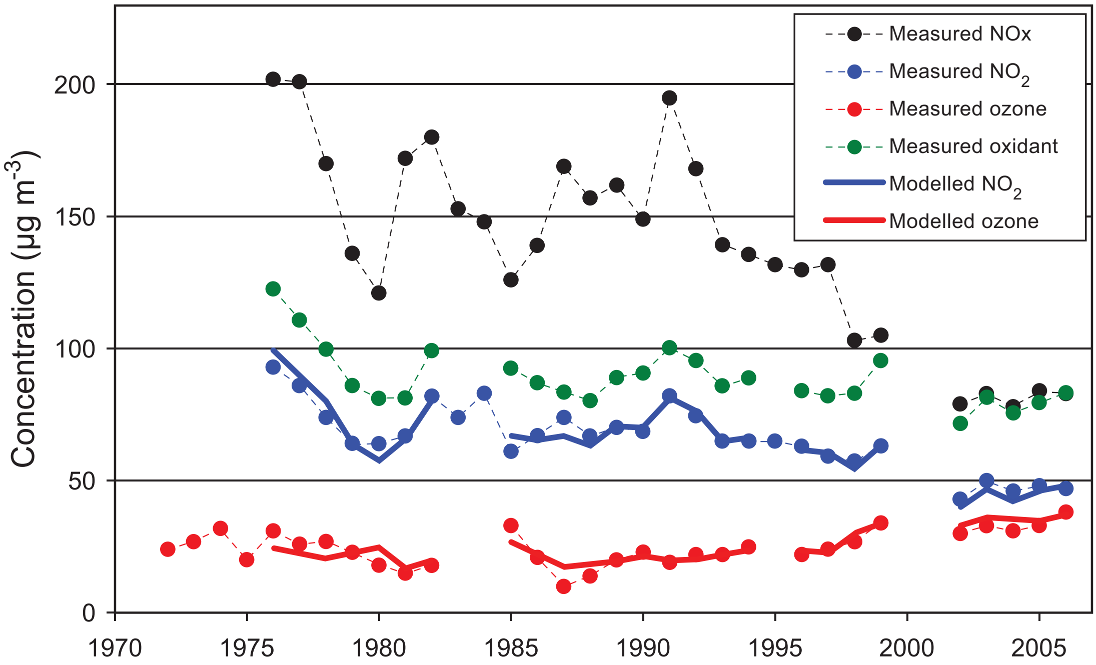


Figure 2.25 Measured annual mean concentrations of ozone, NO_2 and NO_x at Central London, London Bridge Place and London Westminster along with predictions from the oxidant partitioning model.

2.8.4 Site-specific projections of annual mean ozone concentrations at urban sites in the national monitoring network

77. The trends in annual mean ozone concentrations in the urban environment have been explored in terms of the partitioning of oxidant between its component forms of ozone and NO_2, and how this varies with trends in the availability of NO_X. Figure 2.25 shows measured annual mean concentrations of ozone, NO_2, total oxidant (OX) and NO_X in central London from 1972 to 2006 (OX is expressed as $\mu g\ m^{-3}$, as NO_2). A simplified version of the oxidant partitioning model (Jenkin, 2004) has been applied here to partition the OX concentration between ozone and NO_2 as a function of the measured NO_X concentration. The OX concentration includes contributions from background oxidant and local primary NO_2. OX concentrations were highest in the mid-1970s when photochemical ozone concentrations were likely to have peaked and measured NO_X concentrations were high. There was an additional peak in OX in 1991, which coincided with the high NO_X concentrations and a major NO_2 episode (Bower *et al.*, 1994) in that year. The fit of the predicted and measured ozone and NO_2 concentrations is very good, suggesting that the function within the oxidant partitioning model provides a good fit to the data. Annual mean ozone concentrations declined from the mid-1970s to a minimum in the late 1980s, followed by an increase to current levels as NO_X emissions and concentrations have declined considerably.

78. The more recent trends in annual mean urban ozone concentrations have been examined at a range of monitoring sites using the site-specific projections model (Stedman *et al.,* 2001). In this model we have tried to explain the trends in measured ozone resulting from a combination of the changes in NO_X described by emission inventories and the resulting changes in the partitioning of OX. The site-specific projections of NO_X have been calculated both backwards and forwards in time from a base year of 2003 using sector-specific emission projections from the National Atmospheric Emissions Inventory (NAEI) and a source apportionment of local sources derived from the mapped inventory. NO_2 and ozone projections have been calculated from a combination of these NO_X projections and estimates of regional and local oxidant concentrations, using the oxidant partitioning model to assign OX between NO_2 and ozone. The background oxidant concentration has been assumed to remain constant in all years. The local oxidant has been assumed to be a constant proportion of the total NO_X concentration, and the value of the primary NO_2 emissions fraction (f-NO_2) has been held constant at the values derived by Jenkin (2004) from monitoring data up to and including 2001 (14% in central London and about 9% elsewhere).

79. Figure 2.26(a) shows the results of this assessment for the London Bloomsbury site. The predicted increase in annual mean ozone concentration is in good agreement with the measurements at this site. Emission inventory projections to 2020 suggest that NO_X concentrations are likely to remain sufficiently high that NO_2 concentrations will remain higher than those of ozone.

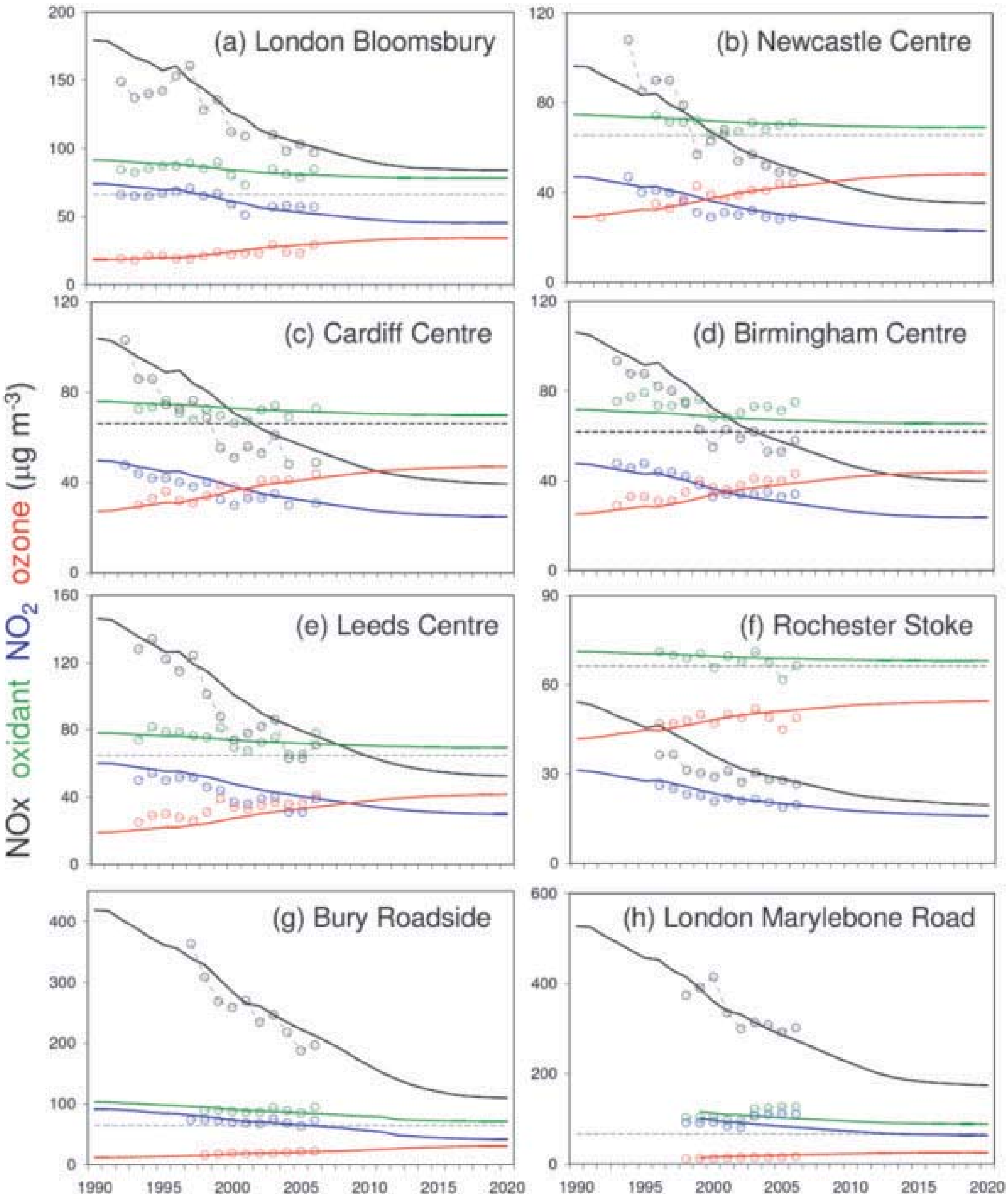


Figure 2.26 Site-specific predictions of NO_X, NO_2, ozone and oxidant at selected urban centre sites, (a)-(e), a rural site, (f), and roadside sites, (g)-(h). The lines are the modelled projections and the points are measured annual mean data. The broken line in each panel indicates the level of background oxidant specified in the model.

80. Figures 2.26(b)-(e) show similar plots for the Newcastle Centre, Cardiff Centre, Birmingham Centre and Leeds Centre sites. Once again there is reasonably good agreement between the predictions and the measured trends in NO_x, NO_2 and ozone at these sites. NO_x concentrations have declined sufficiently that annual mean ozone concentrations are now higher than annual NO_2 concentrations at a number of these urban monitoring sites. It is also interesting to note that about three-quarters of the predicted increase in annual mean ozone concentrations between the early 1990s and 2015 had already taken place by 2006.

81. Figure 2.26(f) shows the results of a similar analysis for the rural site at Rochester Stoke. While the trend in annual mean NO_x is somewhat less steep than implied by the model the predicted trends in ozone and NO_2 are well explained.

82. Figures 2.26(g)-(h) show the trends in ozone, NO_2 and NO_x at the roadside sites Bury Roadside and London Marylebone Road. The measured ozone concentrations are lower at these roadside locations because NO_x concentrations are much higher. The predicted concentrations and trends in ozone concentrations are in good agreement with the measurements at these two sites. NO_2 concentrations are less well predicted and this is discussed in the AQEG report on trends in primary NO_2 (AQEG, 2007a). Primary NO_2 emission fractions have been assumed to remain unchanged, as have the levels of regional oxidant in the simple analysis shown here. The main message from this analysis is that the broad patterns of increase in annual mean ozone concentrations at urban sites can be explained by a combination of changes in NO_x emissions and the partitioning of oxidant, such that a progressively greater fraction is in the form of ozone and the NO_x concentration decreases.

2.9 Observations of trends in concentrations of ozone precursors

83. The observed trends in ozone concentrations, concentration distributions and related metrics described in the previous sections have been interpreted in terms of the modification of a background ozone level by chemical processes occurring on local and regional scales. These processes involve either the production of ozone from the regional-scale photochemical processing of emitted VOC and NO_x, or the removal of ozone by local reaction with emitted NO. The observed ozone trends have thus been rationalised in terms of substantial reductions in the emissions of anthropogenic VOC and NO_x since the early 1990s, which have been driven, in particular, by Europe-wide controls on the emissions from petrol-engined motor vehicles through the fitting of three-way gas catalysts to reduce exhaust emissions, and canisters to reduce petrol evaporation emissions.

84. The site-specific analysis and interpretation of annual mean ozone trends presented in the previous section demonstrates the progressive reduction in NO_x concentrations at a selection of UK sites, and confirms that these observations are fully consistent with those predicted on the basis of the NAEI (e.g. as presented in AQEG, 2007b). In this section, trends in the concentrations of hydrocarbons at UK sites are examined, allowing assessment of the effectiveness of EU control measures implemented to reduce VOC emissions and how well they are represented in the NAEI.

85. Dollard *et al*. (2007) have recently carried out a comprehensive analysis of concentrations of up to 26 C_2 to C_8 hydrocarbons monitored for varying subsets of the period 1993-2004 at 11 urban background sites, and one rural and one kerbside site within the UK hydrocarbon network. The results demonstrate significant and sustained reductions in the concentrations of the majority of species over this period, with the more reactive VOC implicated in ozone formation typically declining at rates between about 15% and 25% per year, as illustrated for selected species and sites in Figure 2.27, for the period 1993-2004. As discussed by Dollard *et al*. (2007), the magnitude of these reductions is compatible with the declines in VOC emissions from relevant sources, as represented in the NAEI for the same period, and it is reasonable to infer that similar trends have occurred for other emitted, but unmeasured, VOCs which contribute to the same sectors. This therefore confirms the assessment of the impact of control measures designed to limit the emissions of anthropogenic VOC in the EU, and the contribution this has made to the decline in regional-scale ozone formation described in previous sections.

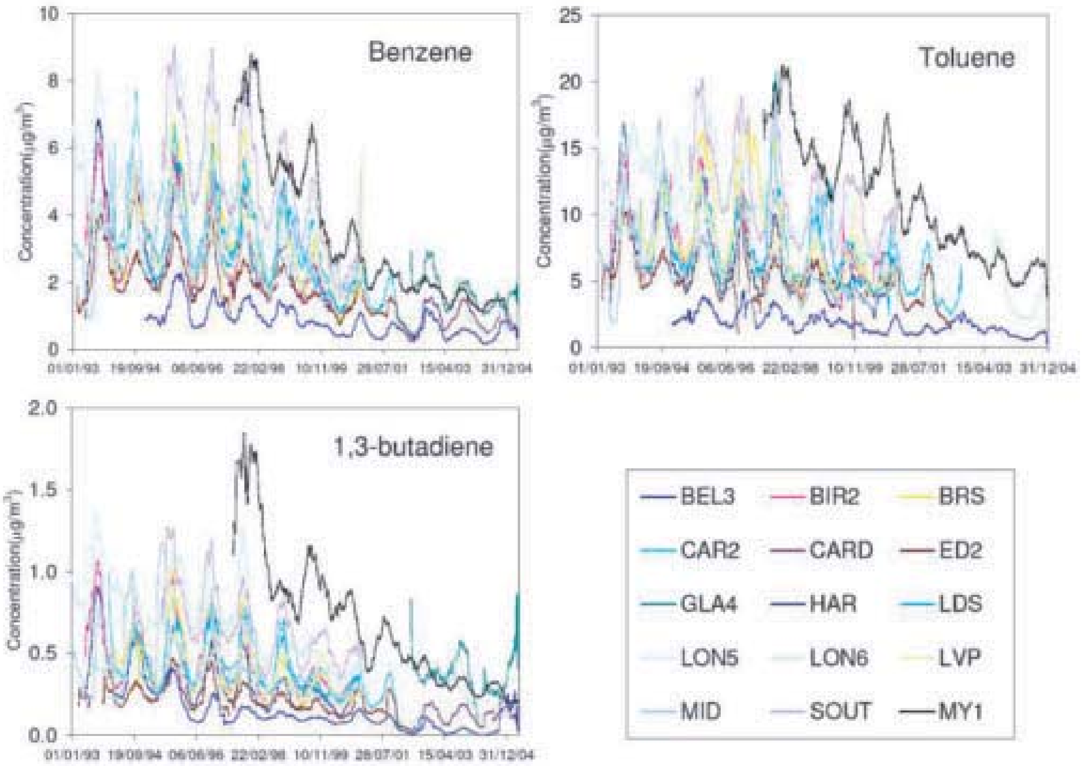


Figure 2.27 Ninety-day running mean concentrations of benzene, toluene and 1,3-butadiene at 15 UK hydrocarbon network sites over the period 1993-2004. The concentrations for Marylebone Road (MY1) have been divided by two to facilitate presentation of the data.

86. The measured series of hydrocarbons includes isoprene (2-methyl-1,3-butadiene), which is known to be emitted from both anthropogenic and biogenic sources, and has therefore been used to provide a marker for biogenic emissions activity. Previous studies have demonstrated that its anthropogenic source (dominated by exhaust emissions) is well correlated with that of the structurally similar hydrocarbon 1,3-butadiene, with isoprene/1,3-butadiene emissions ratios of *ca*. 0.4-0.5 on a µg/µgbasis (e.g. Derwent *et al.,* 1995; Reimann *et al.,* 2000). Figure 2.28 shows a correlation of daily mean isoprene and 1,3-butadiene

concentrations at Marylebone Road over the period 2000-2005, which confirms a limiting anthropogenic ratio of this magnitude, such that the variable excess contribution can be attributed to background biogenic sources. These observations provide a basis for separating isoprene into its anthropogenic and biogenic contributions, as shown in Figure 2.28 and Figure 2.29. Figure 2.29 confirms the steady decline in the anthropogenic contribution, which is 23% per year on average over this six-year period, with the biogenic contribution displaying a characteristic seasonal cycle resulting primarily from the light and temperature dependence of isoprene emissions. This shows that the inferred summertime monthly mean biogenic contribution at the end of the time series is comparable to the anthropogenic contribution, and also similar to that inferred previously for the Eltham and Bloomsbury (UCL) sites, as reported in AQEG (2007b). Closer inspection of the daily mean data for August 2003, and hourly mean data for 10th August 2003 (Figure 2.30), the highest temperature day on record, demonstrates a dominance of the background biogenic contribution under these favourable conditions, even at a busy roadside location. This suggests that biogenic emissions of isoprene (and probably other unmeasured biogenic hydrocarbons) could be playing an increasingly important role in urban-scale photo-oxidation processes under such conditions as the emissions of anthropogenic VOC decline. The existence of important anthropogenic and biogenic contributions to isoprene concentrations at urban locations is also supported by a principal component analysis of long-term observations at sites in Lille, northern France, reported by Borbon *et al.* (2003). They concluded that isoprene was derived significantly from exhaust emissions, and from a second unique (biogenic) source displaying a dependence on temperature and insolation, not observed for any of the other 40 measured C_2-C_9 hydrocarbons.

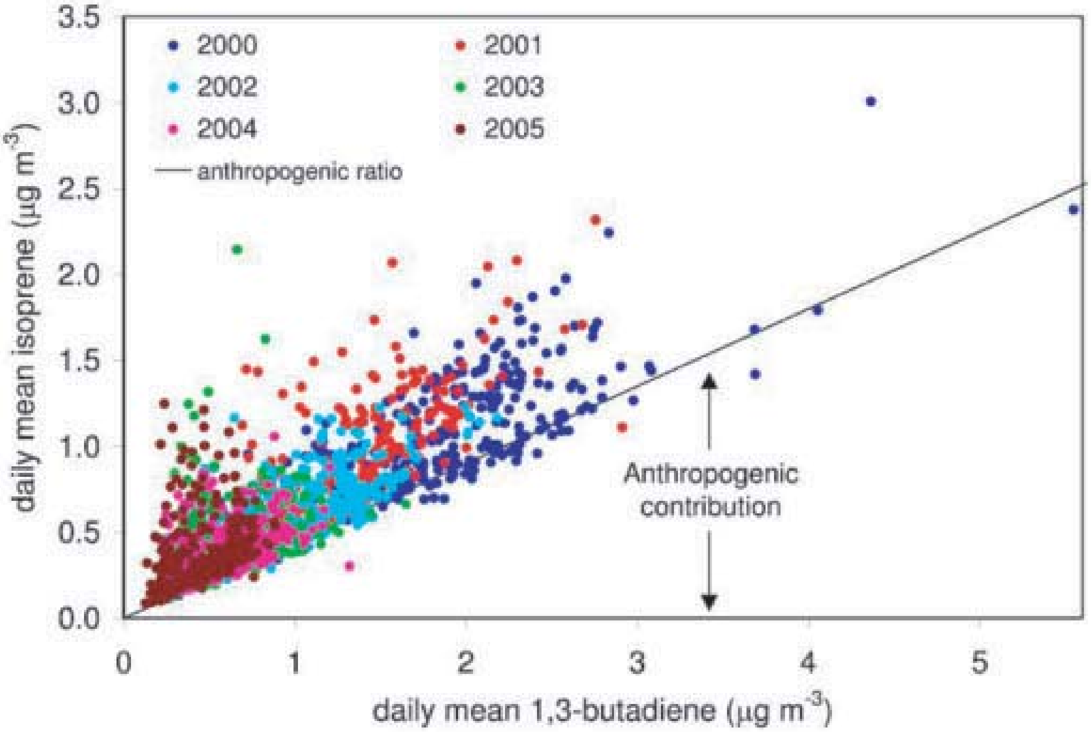


Figure 2.28 Correlation of daily mean concentrations of isoprene and 1,3-butadiene at Marylebone Road over the period 2000-2005. The line indicates a concentration ratio [isoprene]/[1,3-butadiene] = 0.45 which is representative of the limiting anthropogenic ratio.

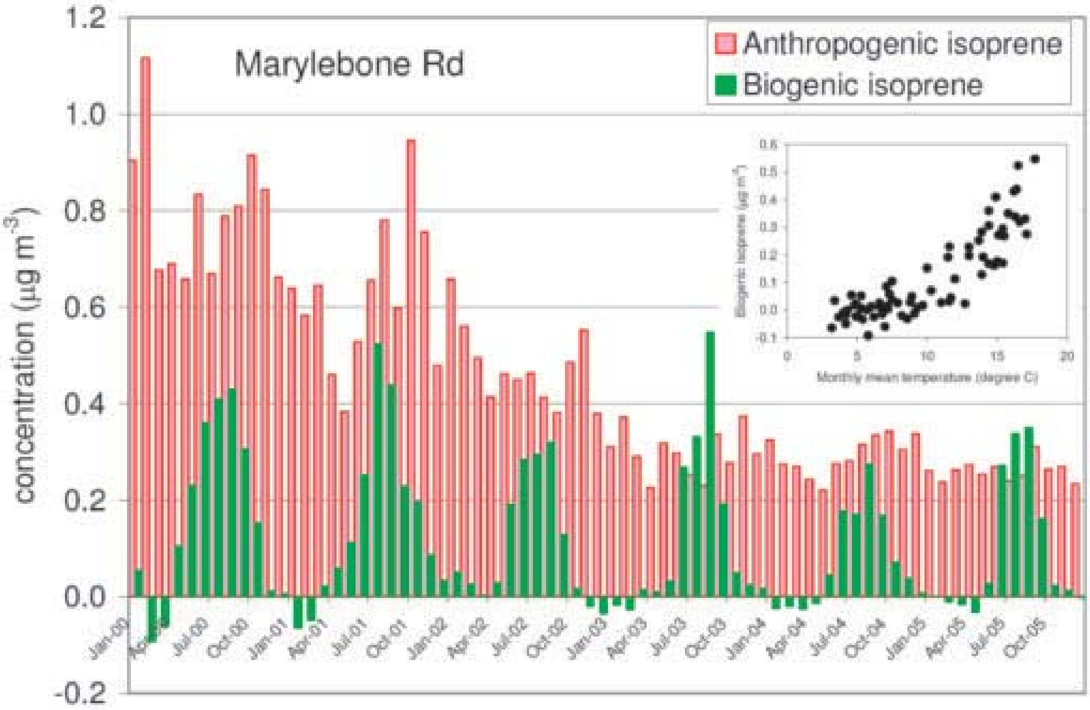


Figure 2.29 Monthly mean anthropogenic and biogenic isoprene concentrations over the period 2000-2005 at Marylebone Road. The anthropogenic contribution to the total was inferred from 1,3-butadiene concentrations, using limiting isoprene/1,3-butadiene ratios in the range ca. 0.45 – 0.55 derived from wintertime (Jan, Feb, Nov and Dec) data in each year. The inset shows a correlation of the biogenic isoprene concentration with monthly mean temperature for England and Wales (source Met Office).

87. It is important to note, however, that the inferred elevated biogenic isoprene concentrations at Marylebone Road, and the other similar levels measured at rural locations in southern England during the August 2003 heat-wave at Harwell, Oxfordshire, and at Writtle, Essex (see AQEG, 2007b and Lee *et al.,* 2006), are currently not straightforwardly reconciled with assessments of biogenic isoprene emissions in the UK (see for example, Stewart *et al.,* 2003). This is partly because estimates of biogenic isoprene source strengths which form the basis of representations in models are reported to be highly uncertain by a factor of about 4 up and down (Stewart *et al.,* 2003). In addition, close-to-surface observations of a highly reactive trace gas with a surface source are difficult to compare with Lagrangian model predictions averaged across the entire atmospheric boundary layer depth and with Eulerian model predictions that are averages over lowest level grid boxes with vertical depths of the order of 100 m, such that vertical concentration gradients almost certainly contribute to the shortfalls which have typically been simulated. The possibility that elevated anthropogenic evaporative emissions may contribute to the inferred biogenic contribution has therefore been considered by AQEG, but the low total diene content of gasoline (which actually has contributions from both 1,3-butadiene and isoprene) are not consistent with a major input of isoprene (or 1,3-butadiene) from this source. Further work therefore appears necessary to characterise the biogenic sources of isoprene and other more complex hydrocarbons in the UK, so that their impact on ozone formation can be fully assessed.

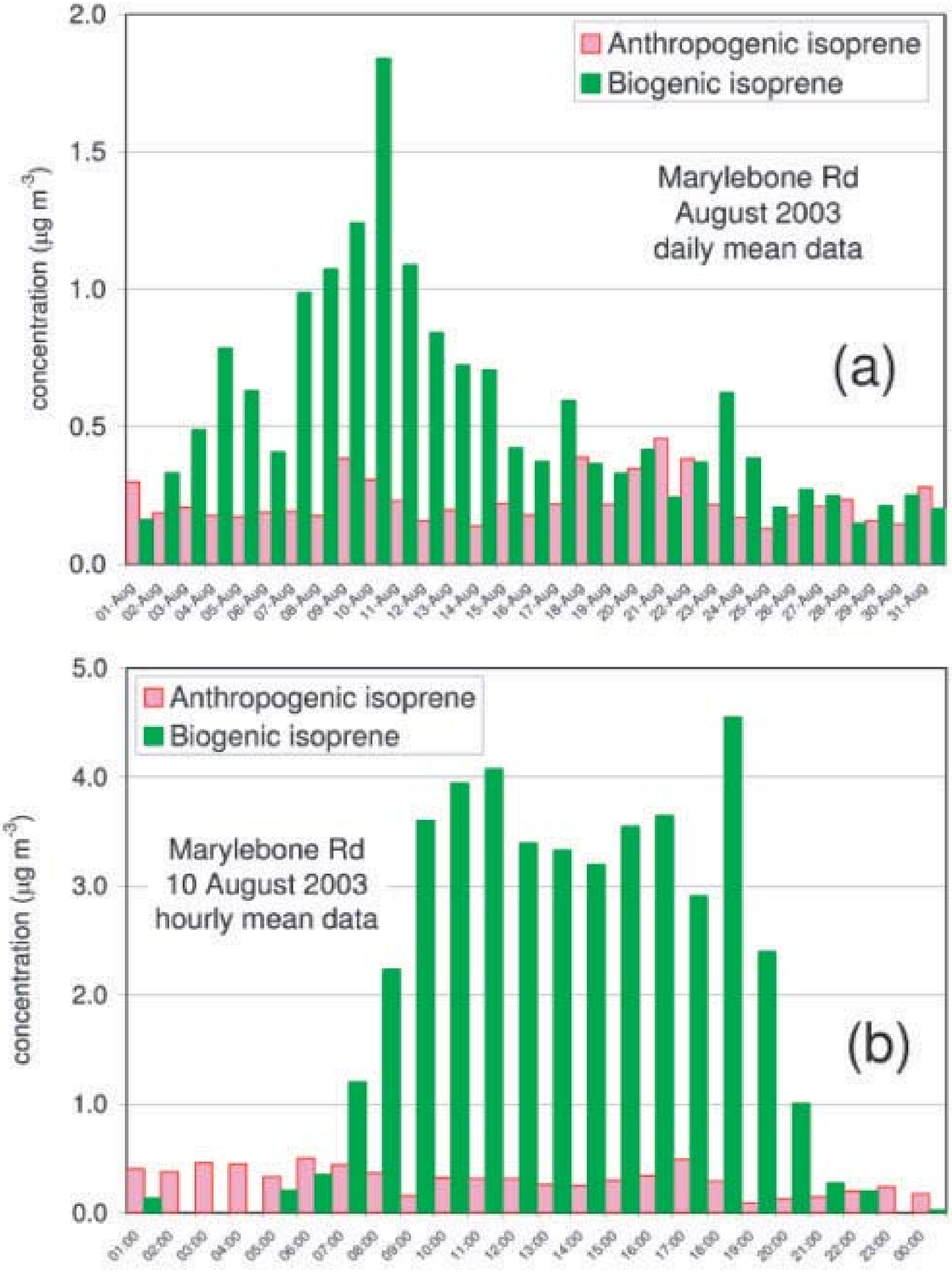


Figure 2.30 Estimated anthropogenic and biogenic isoprene concentrations at Marylebone Road as (a) daily means in August 2003, and (b) hourly-means on 10th August 2003. The anthropogenic contribution to the total was inferred from 1,3-butadiene concentrations, using a limiting isoprene/1,3-butadiene ratio of 0.463 derived from wintertime (Jan, Feb, Nov and Dec) data in 2003.

2.10 Spatial concentration patterns of ozone in the UK

2.10.1 Empirical maps

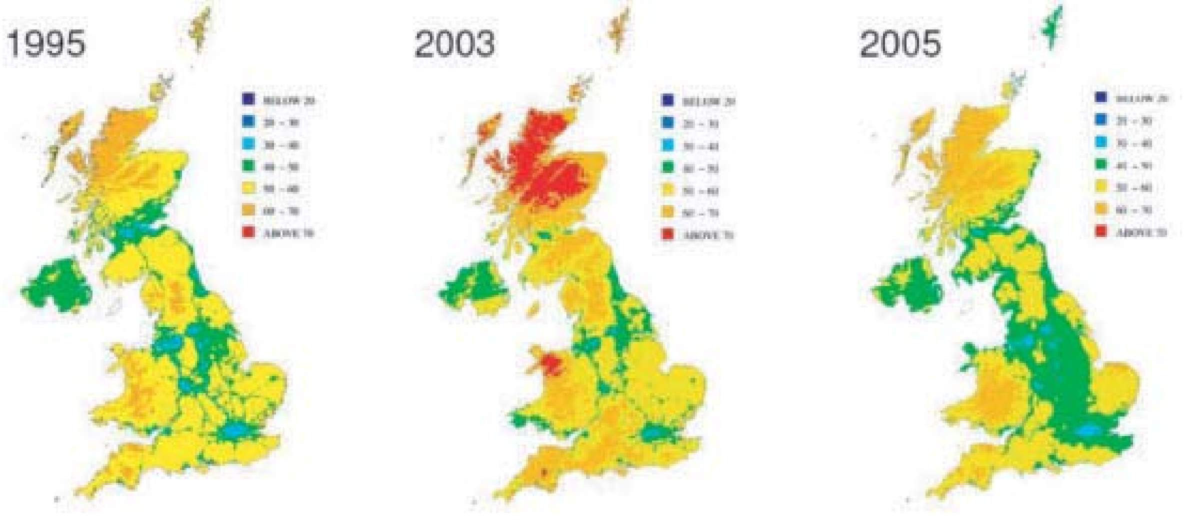
88. Maps of a range of ozone metrics have been calculated using empirical measurements-based Pollution Climate Models (PCMs) for the years 1995, 2003 and 2005 (Stedman and Kent, 2008). These years have been chosen to illustrate two recent years with higher and lower photochemical ozone

contributions (2003 and 2005) and a year with higher photochemical ozone contributions combined with higher urban NO emissions (1995). These maps are shown in Figure 2.31.

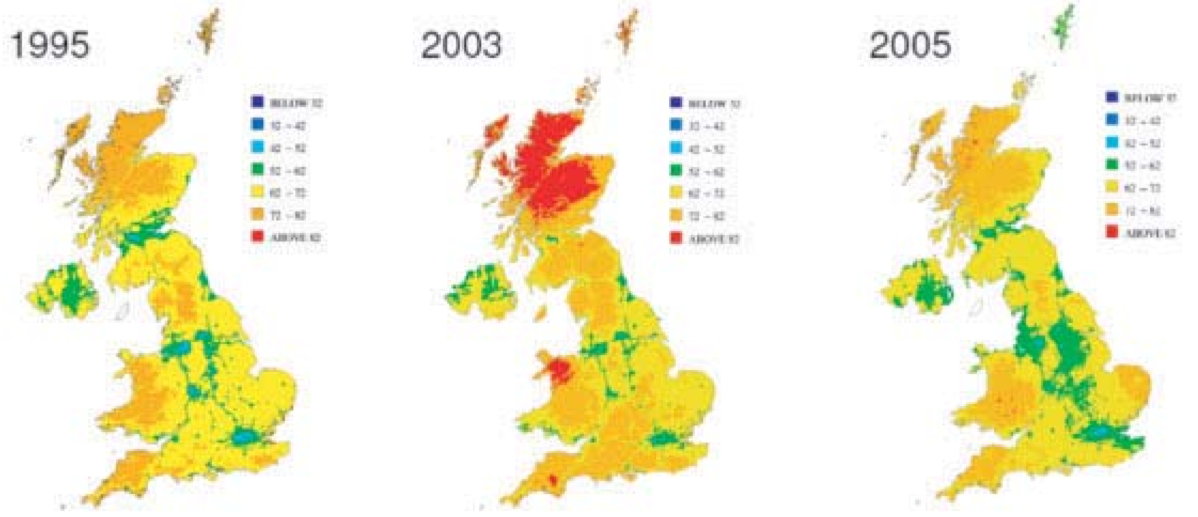
89. The empirical PCM mapping methods used to calculate the maps presented here have been described by Bush *et al*. (2005), Coyle *et al*. (2002) and Kent *et al*. (2006). The maps of annual mean concentration have been calculated by interpolation of monitoring data from rural monitoring sites for the well-mixed period in the afternoon. Two corrections were then applied. Firstly, an altitude correction was applied to take account of the effects of topography (Coyle *et al.,* 2002). Topographic effects are important for some ozone metrics, such as the annual mean, because of the disconnection of a shallow boundary layer from air aloft during the night at lowland locations. Surface ozone concentrations are lower at night in these locations due to a combination of dry deposition and scavenging with NO emissions. This effect is much less marked at higher altitudes and at coastal locations, where wind is generally stronger and a shallow boundary layer does not form. An urban decrement was then calculated using the oxidant partitioning model of Jenkin (2004). Maps of regional oxidant were calculated as the sum of altitude-corrected ozone and rural NO_2 as interpolated from measurements at rural sites. The partitioning of oxidant between ozone and NO_2 was then calculated as a function of modelled local NO_X concentrations (the sum of the contributions to modelled NO_X concentration for the appropriate year from local area and point sources from Kent *et al.,* 2007). The maps of annual average of the daily maximum of the running 8-hour mean ozone concentration were calculated from the annual mean using a non-linear function derived from monitoring data (see Figure 2.15). We recognise that this metric would not be expected to have a significant altitude dependence as has been implied by our methods in which values have been estimated from the annual mean. The focus of this report is on ozone impacts in urban areas. We therefore chose to base our estimates on the annual mean for which the urban decrement can be calculated using the oxidant partitioning model, rather than the more empirical relationships with NO_X concentrations which we have had to use for the remaining metrics.

90. The maps for the remaining health-based metrics were calculated by the interpolation of measurement at rural sites in rural areas followed by the calculation of an urban decrement. The urban decrement has been applied based on empirical linear relationships between the decrement and modelled local NO_X concentrations, which is described in Annex 2.

91. The hemispheric background is a major contributor to annual mean ozone concentration across the UK (Figure 2.31(a)). Upland areas tend to have the highest annual mean ozone concentrations due to topographic effects, as has been described by PORG (1997). There is also a significant decrement in urban areas, as discussed above. Annual mean concentrations in rural areas were highest in 2003 and lowest in 2005 at most locations due to the photochemical episodes during 2003 and perhaps a higher hemispheric background in 2003. Urban concentrations were lowest in 1995 due to the greater local NO emissions at the time. The maps of the annual average of the daily maximum of the running 8-hour mean concentration (Figure 2.31(b)) have been calculated from the annual mean maps using a non-linear function and thus show a similar spatial pattern.



(a) Annual mean ozone concentration ($\mu g\ m^{-3}$)



(b) Annual mean of the daily maximum of the running 8-hour mean ozone concentration ($\mu g\ m^{-3}$)

(c) Annual mean of the daily maximum of the running 8-hour mean with a 70 $\mu g\ m^{-3}$ cut-off ($\mu g\ m^{-3}$)

Figure 2.31 Maps for a range of ozone metrics, calculated using empirical measurements-based PCMs for the years 1995, 2003 and 2005.

(d) Annual mean of the daily maximum of the running 8-hour mean with a 100 μg m^{-3} cut-off (μg m^{-3})

(e) Number of days with maximum 8-hour running mean ozone concentration greater than 100 μg m^{-3}

(f) Number of days with maximum 8-hour running mean ozone concentration greater than 120 μg m^{-3}

Figure 2.31 (cont.) Maps for a range of ozone metrics, calculated using empirical measurements based PCMs for the years 1995, 2003 and 2005. The darkest blue area in Figure 2.31(e) identifies where the UK Air Quality Strategy objective of less than 10 days >100 μgm^{-3} is achieved.

92. Maps of the annual average of the daily maximum of the running 8-hour mean concentration with a cut-off of 70 µg m^{-3} (Figure 2.31(c)) show highest concentrations in the south of the UK, where the contribution from photochemically-generated ozone is greatest, and in northern Scotland, where the impact of the hemispheric background is most pronounced due to the low regional NO_X emissions in this area. There is also an urban decrement for this metric. Concentrations were generally highest in 2003 and lowest in 2005 due to the larger contribution from photochemical ozone episodes in 2003 and 1995. Urban concentrations were lowest in 1995 due to the greater local NO emissions at the time.

93. Values of the annual average of the daily maximum of the running 8-hour mean concentration with a cut-off of 100 µg m^{-3} metric (Figure 2.31 (d)) were much higher in 2003 than in 2005. Values were higher in south and south-west England and Wales in 1995 than in 2003 for this metric. This is in contrast to the annual mean, which was higher in 2003 in these areas. The contribution from photochemical episodes and an urban decrement are the most important factors influencing the spatial distribution for this metric.

94. The spatial distribution of the number of days with the maximum running 8-hour mean ozone concentrations greater than 100 µg m^{-3} and greater than 120 µg m^{-3} (Figures 2.31(e) and (f)) is dominated by the contribution from photochemical episodes and is therefore highly variable from year to year. The number of days above 100 µg m^{-3} was highest in the south and south-west and Wales in 1995, highest in the south-east in 2003 (and also high in the north of Scotland in 2003 due to the higher background) and highest in East Anglia in 2005, although values were generally much lower. There are also clear urban decrements for these metrics.

95. The results of this mapping exercise are summarised in Table 2-2 in terms of the population-weighted mean values of each of the metrics. The population-weighted mean has been calculated by multiplying the 1 km x 1 km background maps by 1 km x 1 km population statistics from the 2001 census. The values for all of the grid squares are summed and then divided by the total population to calculate the population-weighted mean. This is a useful summary statistic, which for the metrics derived as the annual mean of the daily maximum of running 8-hour means is related to human health impacts if the dose-response function is assumed to be linear. The health impact can be estimated by multiplying the population-weighted mean by the dose-response coefficient (expressed as a percentage change in impact per µg m^{-3}) and by a background rate of the health impact in the absence of the air pollutant (e.g. IGCB, 2006). Thus the maps calculated here would enable health effects to be estimated if the dose-response function is assumed to apply from either 0, 70 or 100 µg m^{-3}. Population-weighted means have been calculated for the UK as a whole, for each of the Devolved Administrations, and for London and the rest of England.

96. Figure 2.32 shows the UK population-weighted mean for each metric compared to the value in 1995. The metrics fall into three groups. The number of days with running 8-hour mean ozone concentrations greater than 120 µg m^{-3} and annual mean of the daily maximum of the running 8-hour mean concentration with a cut-off of 100 µg m^{-3} were both highest in 1995

and lowest in 2005. These are the metrics most strongly influenced by regional photochemical ozone production. The annual mean and the annual mean of the daily maximum of the running 8-hour mean concentration were both highest in 2003 and slightly higher in 2005 than in 1995. These metrics are strongly influenced by the hemispheric background (which was high in 2003) and the urban decrement (which was greatest in 1995). The number of days with running 8-hour mean ozone concentrations greater than 100 µg m^{-3} and the annual mean of the daily maximum of the running 8-hour mean concentration with a cut-off of 70 µg m^{-3} were highest in 2003 and lowest in 2005. These metrics are influenced by both regional photochemical ozone production (high in 1995 and 2003) and the urban decrement (which was greatest in 1995).

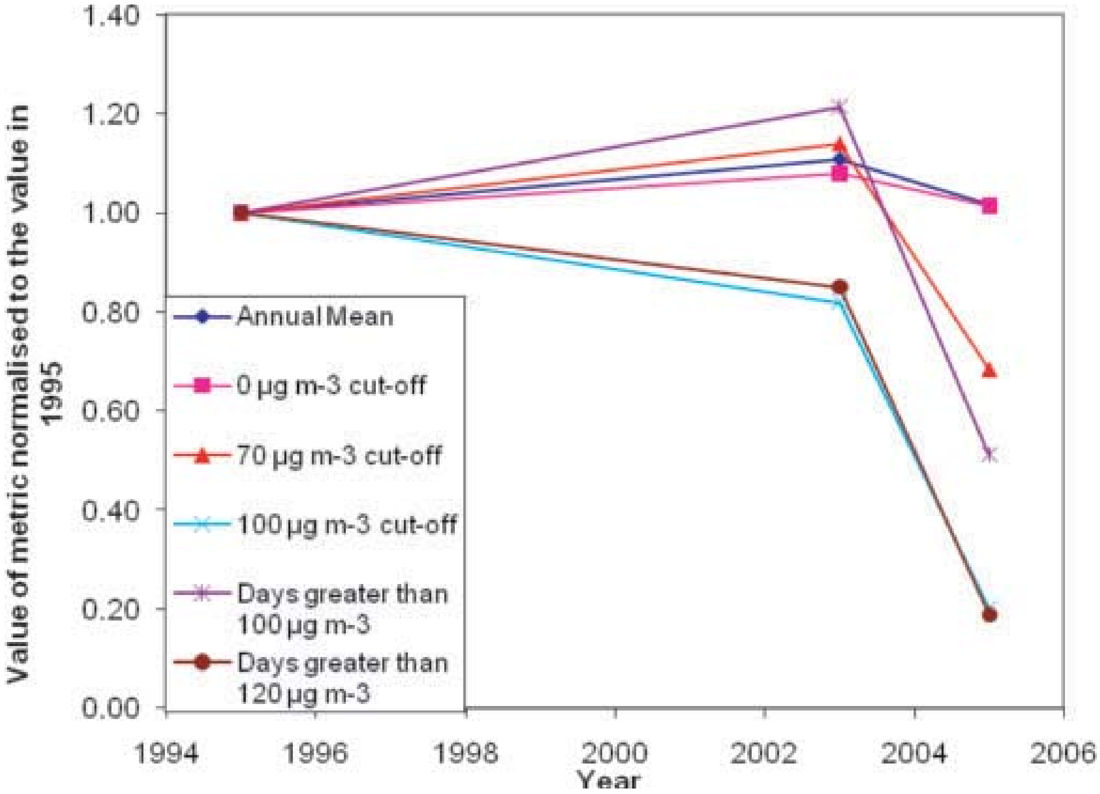


Figure 2.32 Changes in UK population-weighted mean values of the different ozone metrics (see Table 2.2 caption).

Table 2-2 Population-weighted mean values calculated from the maps of the following ozone metrics: annual mean, annual mean of the daily maximum of the running 8-hour mean concentration (with cut-off thresholds of 0, 70 and 100 µg m^{-3}), and number of days with running mean concentrations greater than 100 and 120 µg m^{-3}.

a) 1995

	Annual mean (µg m^{-3})	**0 µg m^{-3} cut-off metric (µg m^{-3})**	**70 µg m^{-3} cut-off metric (µg m^{-3})**	**100 µg m^{-3} cut-off metric (µg m^{-3})**	**Days greater than 100 µg m^{-3} (days)**	**Days greater than 120 µg m^{-3} (days)**
Scotland	40.9	56.3	4.6	0.7	11.6	4.3
Wales	49.7	65.0	11.2	4.3	36.0	25.7
Northern Ireland	42.8	58.3	5.2	1.1	18.9	6.7
Inner London	31.9	47.0	5.3	2.0	22.7	13.0
Outer London	36.4	51.8	7.3	2.8	27.7	16.5
Rest of England	44.5	59.9	8.5	3.1	29.6	18.6
UK	43.2	58.6	7.9	2.8	27.6	17

b) 2003

	Annual mean (µg m^{-3})	**0 µg m^{-3} cut-off metric (µg m^{-3})**	**70 µg m^{-3} cut-off metric (µg m^{-3})**	**100 µg m^{-3} cut-off metric (µg m^{-3})**	**Days greater than 100 µg m^{-3} (days)**	**Days greater than 120 µg m^{-3} (days)**
Scotland	51.1	66.3	8.1	1.0	2.4.5	7.9
Wales	51.3	66.5	9.2	2.3	33.9	15.2
Northern Ireland	45.2	60.7	4.7	0.5	16.5	7.1
Inner London	38.1	53.6	7.6	2.1	31.4	11.8
Outer London	42.2	57.8	9.5	2.7	38.1	15.1
Rest of England	48.6	64.0	9.4	2.5	34.9	15.6
UK	47.9	63.2	9.0	2.3	33.5	14.4

c) 2005

	Annual mean (µg m^{-3})	**0 µg m^{-3} cut-off metric (µg m^{-3})**	**70 µg m^{-3} cut-off metric (µg m^{-3})**	**100 µg m^{-3} cut-off metric (µg m^{-3})**	**Days greater than 100 µg m^{-3} (days)**	**Days greater than 120 µg m^{-3} (days)**
Scotland	43.6	59.1	3.7	0.1	4.5	0.1
Wales	51.2	66.5	8.1	0.7	23.3	3.8
Northern Ireland	42.5	58.0	3.8	0.1	3.0	0.2
Inner London	33.4	48.7	2.6	0.3	5.9	2.5
Outer London	37.5	53.0	3.8	0.5	9.6	3.6
Rest of England	44.9	60.4	5.9	0.7	16.1	3.7
UK	44.0	59.4	5.4	0.6	14.1	3.2

2.10.2 Transects across the London conurbation

97. The results of this mapping exercise for 1995, 2003 and 2005 have been further examined by plotting a transect of modelled ozone concentrations across the London area from west to east through central London. The annual means for the three years (Figure 2.33(a)) were reasonably similar and illustrate the urban decrement very well. The annual mean was generally highest in 2003 across London. The annual means in suburban areas were lowest in 2005 but the annual means in central London were lowest in 1995 due to the greater NO_X emission density in the city centre in 1995.

98. Monitoring data for non-roadside sites close to the transect (which is at Northing 183500) are also included in the figure. Data from the Automatic Urban and Rural Network (AURN) have been included for all three years (only two sites available in 1995). For 2003 we have also included a wider set of data from the London Air Quality Network (LAQN), and data from Ascot Rural (also LAQN) have been included for 2005 to illustrate measurements from a rural location to the west of London. Figure 2.34 shows the location of the transect and the monitoring sites for both the AURN and the LAQN. The agreement between the modelled transect and the measurements is very good. Differences could be caused by uncertainties in the modelled and measured values, and the fact that the monitoring sites do not lie directly on the transect. Both the ranking of the years and the magnitude of the urban decrement are confirmed by the measurements.

99. Figure 2.33(b) shows a similar plot for the daily maximum of the running 8-hour mean concentration with a cut-off of 70 µg m^{-3}. Values of this metric were highest in 2003 across the whole of London and only a little lower in 1995. Values of this metric were much lower in 2005 when there was less photochemically-generated ozone. Once again the monitoring data confirm the results of the modelling assessment. Data from the AURN are presented for this metric and for the metric with a cut-off of 100 µg m^{-3} (Figure 2.33(c)), with the Ascot Rural data also included for 2005.

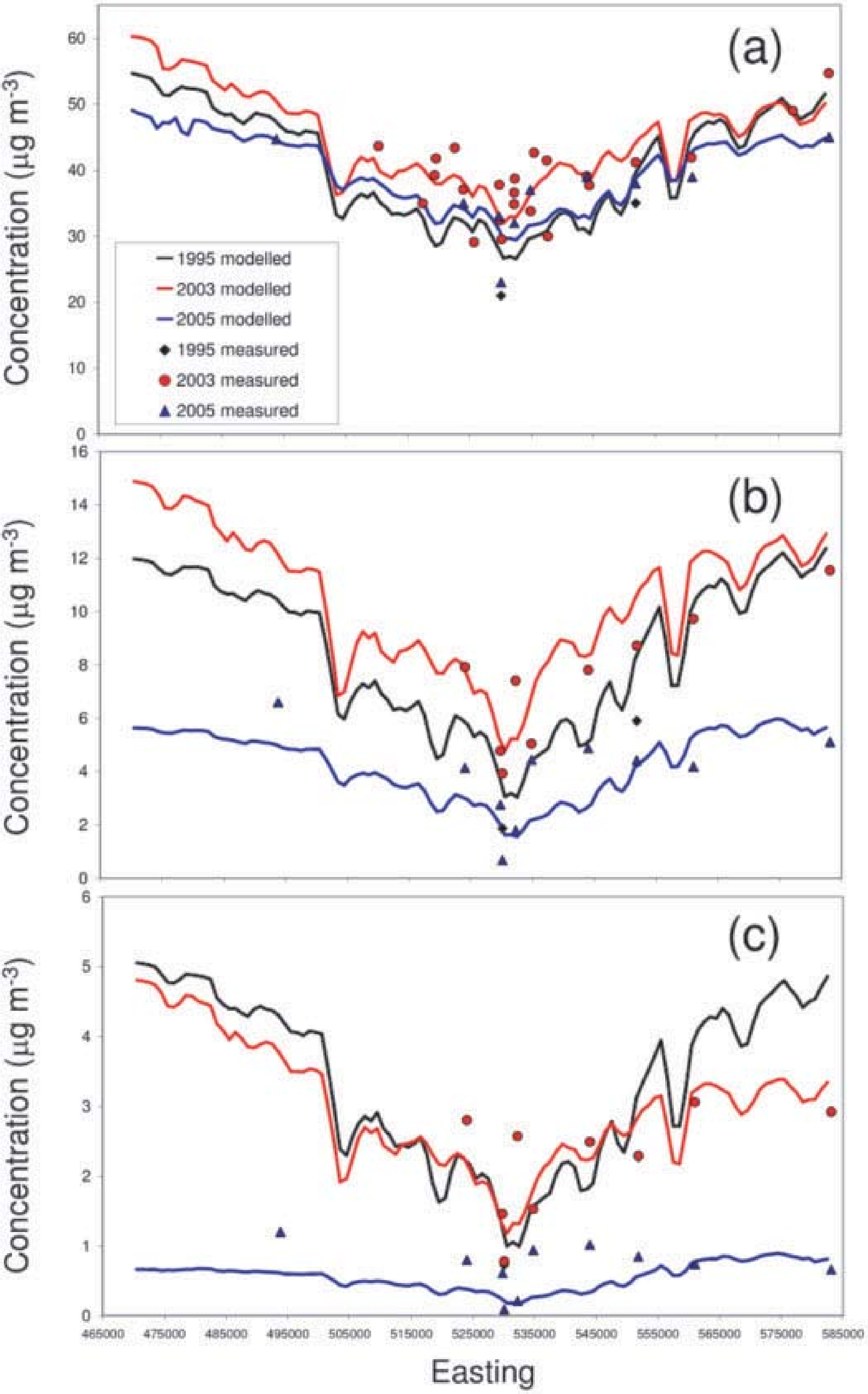
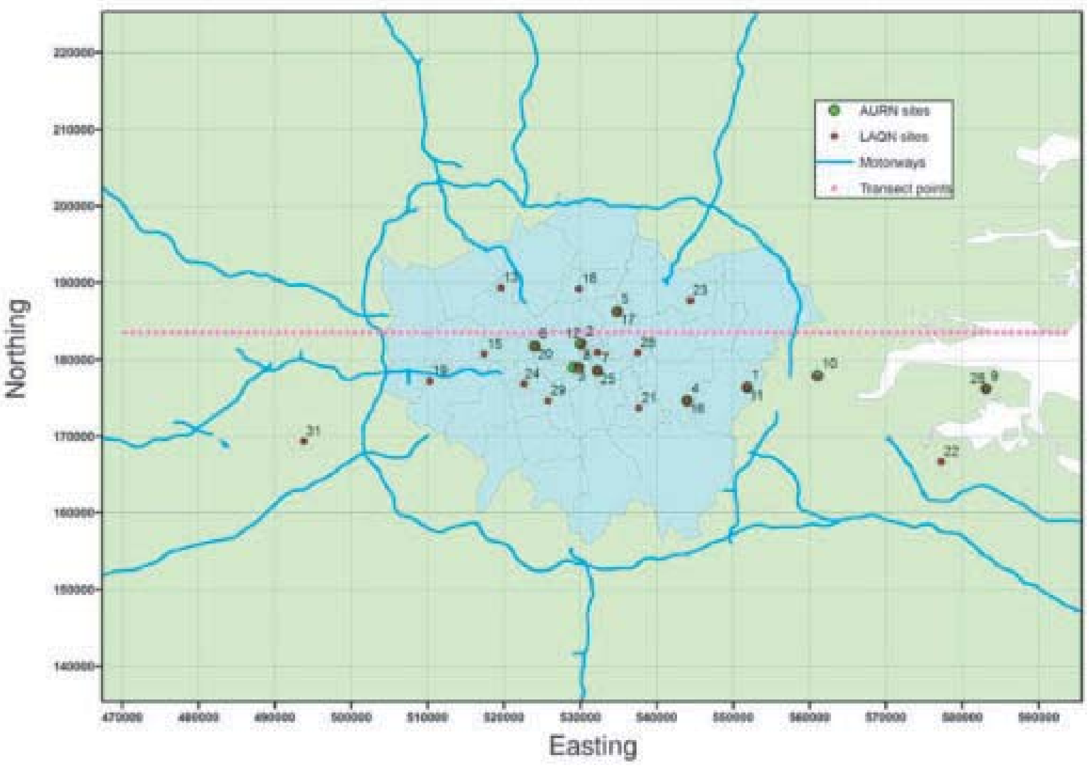


Figure 2.33 Transects of ozone metrics across London: (a) Annual mean ozone concentration; (b) Annual mean of the daily maximum of the running 8-hour mean ozone concentration with a 70 µg m^{-3} cut-off; and (c) Annual mean of the daily maximum of the running 8-hour mean ozone concentration with a 100 µg m^{-3} cut-off.



Site	Network	Code
London Bexley	AURN	1
London Bloomsbury	AURN	2
London Bridge Place	AURN	3
London Eltham	AURN	4
London Hackney	AURN	5
London N. Kensington	AURN	6
London Southwark	AURN	7
London Westminster	AURN	8
Rochester Stoke	AURN	9
Thurrock	AURN	10
Bexley 1 – Slade Green (AURN)	LAQN	11
Bloomsbury – AURN	LAQN	12
Brent 1 – Kingsbury (AURN)	LAQN	13
City of London 1 – Senator House	LAQN	14
Ealing 1 – Ealing Town Hall	LAQN	15
Greenwich 4 – Eltham	LAQN	16

Site	Network	Code
Hackney 4 – Clapton	LAQN	17
Haringey 2 – Priory Park	LAQN	18
Hounslow 2 – Cranford	LAQN	19
Kensington and Chelsea 1 – North Kensington	LAQN	20
Lewisham 1 – Catford	LAQN	21
Luton Background	LAQN	22
Redbridge 1 – Perth Terrace	LAQN	23
Richmond 2 – Barnes Wetlands	LAQN	24
Southwark 1 – Elephant and Castle	LAQN	25
Rochester Stoke (AURN)	LAQN	26
Thurrock	LAQN	27
Tower Hamlets 1 – Poplar	LAQN	28
Wandsworth 2 – Town Hall	LAQN	29
Westminster (AURN)	LAQN	30
Ascot Rural	LAQN	31

Figure 2.34 The locations of the London transect and monitoring sites included in the analysis.

100. The transect for the daily maximum of the running 8-hour mean concentration with a cut-off of 100 µg m^{-3} is shown in Figure 2.33(c). This figure also shows the impact of the urban decrement superimposed on the impact of photochemically-generated ozone. Values of this metric were very low in 2005 and were somewhat higher in 1995 than in 2003 in suburban areas. The greater urban decrement in 1995 led to values of this metric being similar in 1995 and 2003 in the centre of London. Once again there is reasonably good agreement between the model and available measurements.

101. Figure 2.35 shows the results of a more detailed assessment of data from the LAQN for 2003. This figure shows the transects for the annual mean, monthly mean for August 2003, mean for the 9th August 2003 and the values for 15:00 GMT on this day (which was the peak of a major photochemical episode). The maximum urban decrement is about 25 µg m^{-3} for the annual mean, 35 µg m^{-3} for the monthly mean, 50 µg m^{-3} for the daily mean and 85 µg m^{-3} for the 15:00 value. Thus the maximum urban decrement is about 30% to 40% of the rural values.

102. The analysis of the monitoring data transect for London confirms that the empirically-generated maps include a reasonably realistic description of urban decrements.

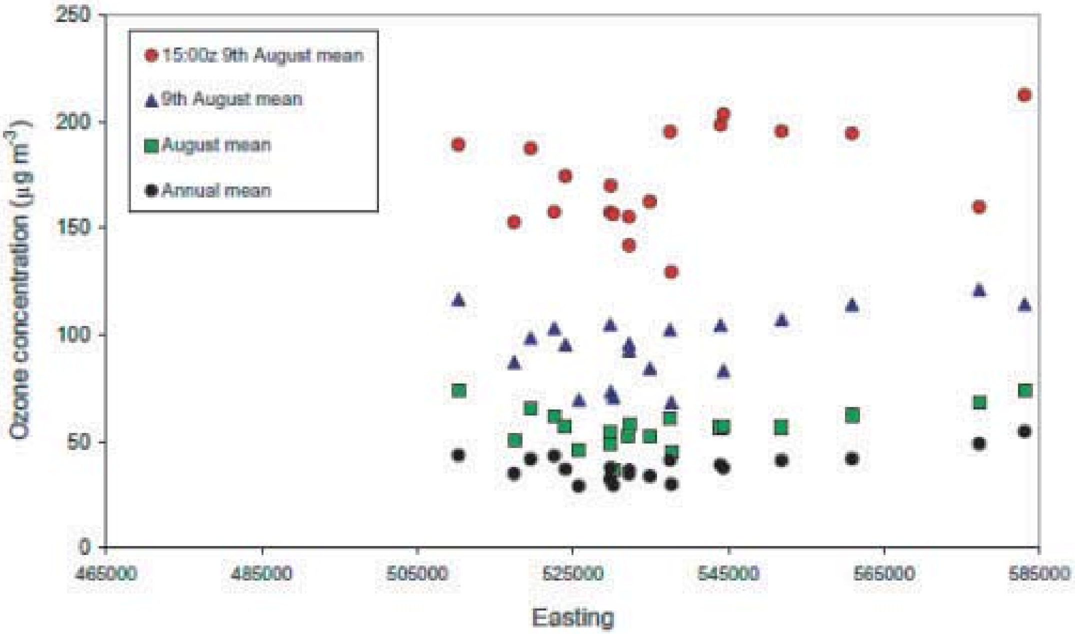


Figure 2.35 Ozone transect across London in 2003 (µg m^{-3}).

2.11 Recommendations

103. AQEG make the following recommendations:

- The current monitoring of ozone, NO_X and volatile organic compounds at urban and rural network locations provides an excellent resource for both policy purposes and for the UK research community. Mechanisms to analyse and interpret these measurements should be maintained.

- Urban and rural sites with co-located measurements of ozone and NO_X should be maintained to produce the long-term datasets required for the continued assessment of the temporal and spatial oxidant climatology in the UK, and its response to local, regional and global-scale precursor emissions trends.

- Further work is required to evaluate biogenic source strengths of isoprene and other complex hydrocarbons and oxygenated VOCs. Additional closure studies are required for VOC emissions in general at a variety of locations, to allow further assessment of the UK speciated inventory.

Chapter 3

Trends in background ozone concentrations

Question B: *Observations since the 1970s have shown that global background ozone concentrations have been rising throughout this period. What is the strength of these data, and what is the evidence concerning the trends and likely projections of precursor emissions, and the resultant ozone concentrations?*

Short answer to question B

104. An international policy review has concluded that there is strong evidence that background ozone concentrations in the northern hemisphere have increased by up to 10 μg m^{-3} per decade over the last 20-30 years (Raes and Hjorth, 2006). This increase has been attributed to the growth in man-made ozone precursor emissions from industry, road, air and ship transport, homes and agriculture. Future ozone concentrations depend on which of the possible future emission scenarios is followed. Future annual mean surface ozone concentrations in the southern half of the United Kingdom are modelled to increase by about 6 μg m^{-3} in a "current legislation" (IIASA CLE) scenario and to decrease by about 4 μg m^{-3} in a "maximum technically feasible reduction" (IIASA MFR) scenario between 2000 and 2030. Observed background ozone concentrations in air masses entering north-west Europe over the 2000-2006 period have remained level and have shown no overall trend.

Detailed answer to question B

105. There is a substantial body of evidence that points to a more than doubling in surface ozone levels in the northern hemisphere since pre-industrial times. Modern ozone measurements began in the 1960s and many background and remote northern hemisphere monitoring stations have recorded upwards trends over the main continental regions. At some sites, these upward trends slowed and levelled off during the 1980s, some during the 1990s and almost all by the 2000s. The trends revealed by ozone sondes for the middle and upper troposphere broadly agree with those from the surface observations. These trends since pre-industrial times have been driven by increasing emissions of man-made tropospheric ozone precursor gases, particularly methane, non-methane volatile organic compounds (VOCs), carbon monoxide and nitrogen oxides (NO_X).

106. There is, however, no agreed picture of the growth in global ozone background concentrations from the 1970s onwards that covers all the surface sites and all the main continents. With photochemistry producing local ozone lifetimes as short as a few days in the boundary layer, local measurement of tropospheric ozone does not reflect the abundance over the same continent, and a surface measurement is not representative of the bulk troposphere above. Thus, it is not contradictory for ozone trends in different atmospheric regions to be different because there may well be different trends in regional pollutants driven particularly by changes in NO_X emissions.

The continuity of some of the ozone records has been compromised by changes in measurement and calibration techniques, relocation of sites and by changes in local influences from road traffic and development at the stations.

107. The ozone record at Mace Head, on the west coast of Ireland, for the period from 1987-2007 is unique, however, in that it has been collected from only two instruments, quality assurance has been maintained consistently to a high standard and data capture has been excellent. Furthermore, the hourly measurements have been sorted by air mass origins to separate the data record into a dataset for unpolluted northern hemisphere air masses and one for European regionally-polluted air masses. The unsorted data show little overall trend from 1987-2006, in common with most remote European monitoring stations. This is because the observed downwards concentration trend in the polluted air masses have more or less compensated for the observed upwards trends in background data, leaving little overall trend. Hemispheric background ozone levels as recorded at Mace Head are of some policy significance because they characterise the background levels in the British Isles upon which regional-scale pollution episodes are superimposed.

108. The observed rise in the ozone mixing ratios in northern hemispheric background air masses at Mace Head, Ireland is statistically significant over the 1987-2006 period and amounts to +0.7 $\mu g\ m^{-3}\ yr^{-1}$. The trend has been significantly higher in winter and spring, compared with the summer months. The seasonal cycle in ozone mixing ratios has therefore increased in amplitude over this period. Observations at Pacific coast sites in North America show similar upwards trends to this Atlantic coast site in Europe (Jaffe and Ray, 2007). It is possible that such inflow sites reveal the influence of intercontinental ozone transport much more effectively compared with surface sites in continental regions.

109. Examination of the 1995-2006 period shows that monthly mean background ozone concentrations at Mace Head rose steadily to unprecedented levels of over 100 $\mu g\ m^{-3}$ during the winter of 1998/99 before falling somewhat thereafter, interrupted temporarily by a secondary peak in 2002/03. During 2006, 12-month running mean background ozone concentrations showed evidence of levelling out and re-establishing the former upwards trend. Over the 1995-2006 period, annual mean background ozone mixing concentrations showed an upwards trend of +0.4 $\mu g\ m^{-3}\ yr^{-1}$. The peaks in 1998/99 and 2002/03 have been identified as being due to boreal biomass burning in Alaska, Yukon and Siberia from an examination of simultaneous changes in the annual growth rates of carbon dioxide, methane and carbon monoxide (Simmonds *et al.,* 2005). Over recent years since 2000, 12-month rolling mean monthly mean background ozone abundances have remained relatively constant and have shown little evidence of significant trends either positive or negative. Over this same period, background methane abundances have also levelled off from their previous extended period of rising levels. Observations therefore show that the period of steadily rising ozone and methane background levels has given way to a period of relatively constant methane and ozone background levels, although predictions of future trends are highly uncertain.

110. Global chemical transport models have been used to verify the general picture described above for the growth in northern hemisphere background ozone levels, in general (Dentener *et al.,* 2005), and at Mace Head in particular (Derwent *et al.,* 2006). These studies have benefited from the significant improvements that have been made in the gridded global emission inventories of man-made tropospheric ozone precursor gases as a result of activities carried out under the aegis of the United Nations Framework Convention on Climate Change (UNFCCC) and the International Institute for Applied Systems Analysis (IIASA). Further details of the emission inventories and emission scenarios used here are given in Annex 1. Models are able to account for much of the observed growth in observed background ozone and methane levels up to the present day. Model-predicted trends for surface ozone up to 2030 vary spatially across Europe and are scenario dependent. Future annual mean ozone abundances for the southern United Kingdom in the STOCHEM model increase from 66 µg m^{-3} in 2000 to 72 µg m^{-3} in 2030 in the IIASA current legislation (CLE) scenario corresponding to an annual increase of +0.3 % per year.
In contrast, in the IIASA maximum technically feasible reduction (MFR) scenario, rolling mean ozone abundances fall to 63 µg m^{-3} in 2030, corresponding to a decline of -0.2% per year. Observed background ozone abundances over the 2000-2006 period have remained level and shown no overall trend, in the middle of the range of the STOCHEM model simulations. STOCHEM is, however, one of many global models from which results are available for Europe as a whole. A summary of these Europe-wide results is provided in Chapter 4.

Supporting evidence for question B

3.1 Overview

111. This chapter addresses the trends in global background ozone, the strengths of these data and the likely projections in future ozone concentrations as they impact on air quality in the United Kingdom. The supporting evidence assembled here deals with ozone trends since the pre-industrial era, the growth in the global ozone background, observations of the ozone background trend at Mace Head, Ireland, modelling the global background trend and projections of future background ozone concentrations in the British Isles.

112. Ozone is present in surface air at every monitoring site across Europe, the United Kingdom included, on almost every hour of the day. As an illustration, Figure 3.1 presents the frequency distribution of the observed ozone concentrations at the rural Harwell, Oxfordshire, site during 2006. The most frequent hourly concentrations lie in the range from about 50-75 µg m^{-3}.

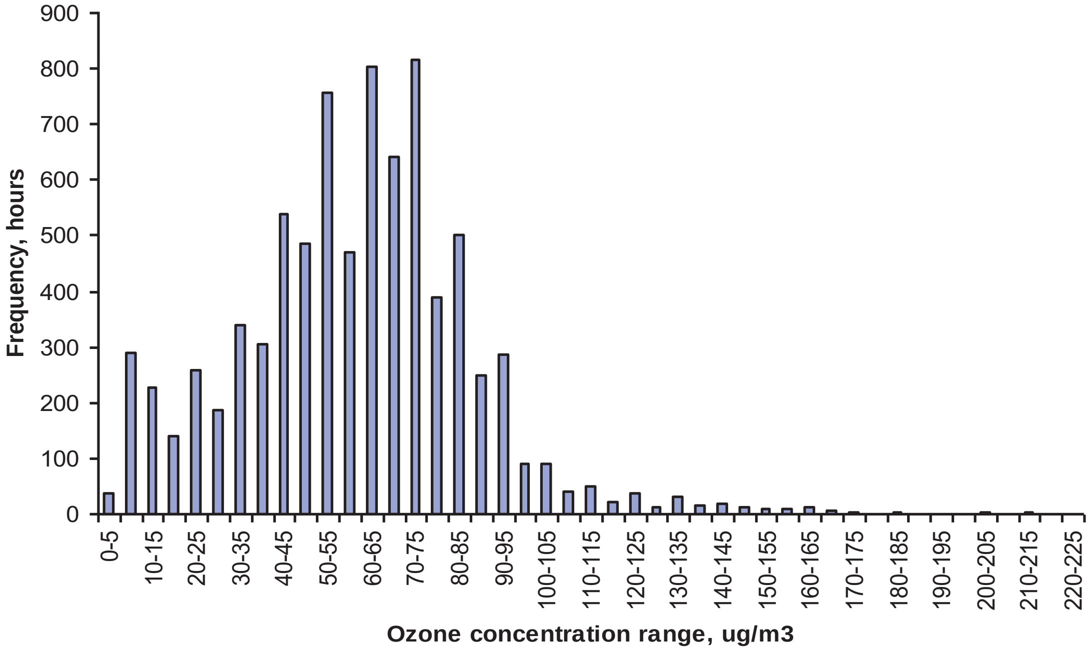


Figure 3.1 Frequency distribution of the hourly mean ozone concentrations observed at Harwell, Oxfordshire, during 2006 in µg m^{-3}.

113. The presence of pollution episodes is clearly seen as a 'tail' extending to high concentrations and there is also evidence of a 'tail' extending to low concentrations. This behaviour implies that there is a background source of ozone, so that even in the cleanest of situations the air masses arriving at the Harwell site almost always contain ozone. Further details of ozone frequency distributions and their trends are given in Chapter 2.

114. Generally speaking, observations in the cleanest of situations, when the influence of local man-made sources is minimal, are employed to define hemispheric background concentrations. Hence those observations at Mace Head, Ireland, that have been made in air masses that have recently been transported many thousands of kilometres across the North Atlantic Ocean, would be counted as such.

3.1.1 Ozone trends since the pre-industrial era

115. The change in tropospheric ozone since the pre-industrial era is difficult to evaluate on the basis of observations alone because ozone is highly reactive and atmospheric abundances cannot be retrieved from ice cores. Recent evaluations of surface observations in the 19th and early 20th centuries in Europe indicate much lower ozone abundances than today (Marenco *et al.,* 1994; Staehelin *et al.,* 1994, 1998; Volz and Kley, 1988). Volz and Kley (1988), for example, report ozone abundances for the Montsouris Observatory in Paris in the range 10-32 µg m^{-3} for the period between 1876 and 1910. It is not straightforward to scale these few measurements to establish global or even northern hemisphere abundances but it is likely that ozone abundances in the northern hemisphere have more than doubled since the pre-industrial era.

116. Chemical transport models predict that current man-made emissions of NO_X, VOCs and CO, as well as the increase in global methane burdens, should have increased tropospheric ozone by a similar amount as the observations, primarily in the northern hemisphere. IPCC (2001) provides a summary of 11 CTM studies of the growth in tropospheric ozone since the pre-industrial era. Comparisons of these model results with the reconstructed 19th century observations at continental sites indicates a systematic model overestimation of about 10 $\mu g\ m^{-3}$ (Wang and Jacob, 1998). There are also issues regarding seasonal cycles that are difficult to reconcile between the models and the pre-industrial observations. Correcting these systematic overestimations would require either a large missing sink for ozone or a downwards revision of the natural NO_X sources from lightning in the 19th century. Either of these is considered unlikely. There are also problems in reconciling model estimates of pre-industrial CO concentrations with observations derived from ice cores, suggesting considerable problems with the emission inventories for the pre-industrial era, particularly of biomass burning. Changes in tropospheric ozone since pre-industrial times have been modelled within the Atmospheric Composition Change: European Network of Excellence (ACCENT) PhotoComp Intercomparison, where increases in tropospheric ozone columns and radiative forcing changes are presented from ten models (Gauss *et al.,* 2006). Further simulations are described in the Royal Society (2008) study where global annual mean surface ozone levels increase from 34 ± 5 ppb to 56 ± 8 ppb in five chemistry-transport models.

3.1.2 Growth in the global ozone background

117. Reliable tropospheric ozone monitoring began at the surface and with ozone sondes during the International Geophysical Year in 1957. However, it is difficult to put a consistent picture together of the growth in background ozone since the 1960s. With photochemistry producing local ozone lifetimes as short as a few days in the boundary layer, local measurement of tropospheric ozone does not reflect the abundance over the same continent and a surface measurement is not representative of the bulk troposphere above (IPCC, 2001). Thus it is not contradictory for ozone trends in different atmospheric regions to be different because there may well be different trends in regional pollutants driven particularly by changes in NO_X emissions.

118. Surface ozone data collected at Arosa, Switzerland, during the 1950s are characterised by annual ozone abundances of about 36 $\mu g\ m^{-3}$ (Staehelin *et al.,* 1994). Measurements at the same location between 1989-1991 indicate an approximate doubling over a period of three decades.

119. Ozone sondes offer the best record of ozone throughout the troposphere. Weekly continuous data since 1970 are available from only nine stations in the latitude range from 36ºN to 59ºN (Logan, 1999). Most stations show an increase from 1970 to 1980 but no clear trend from 1980 to 1996. Of the 14 stations with records since 1980, only two, one in Japan and one in Europe, had statistically significant increases in the mid-troposphere between 1980 and 1995. In contrast, the four Canadian stations all showed significant decreases.

120. The ozone sonde record at Hohenpeissenberg, an alpine location in southern Germany, extends from 1966 to the present day. The pattern of long-term change in the mid-troposphere layer shows large increases from the mid-1960s to the mid-1980s, with smaller increases and even declines thereafter. The small declines seen in the 2000s at Hohenpeissenberg have not been seen at the high altitude Zugspitze site (also in the Alps), although they have been seen at the nearby alpine site of Wank (Oltmans *et al.,* 2006).

121. Surface ozone measurements from 17 background stations up to the mid-1990s also show no clear and consistent trend even in northern latitudes (Oltmans *et al.,* 1998). The largest negative trend in surface ozone was -0.7 ± 0.2% per year at the South Pole (1975 to 1997), while the largest positive trend was +1.5 ± 0.5% per year at the mountain-top Zugspitze site in southern Germany from 1978 to 1995.

122. Two stations in the eastern North Atlantic, Izana, Canaries and Mace Head, Ireland, show increasing ozone abundances over their period of record since 1987 (Oltmans *et al.,* 2006). While increases are seen in most months, the statistically significant changes are during the winter and spring months. The mid-Atlantic station in Bermuda also shows evidence of an increase during the winter and spring months and fits the pattern of the other two North Atlantic sites (Oltmans *et al.,* 2006). In contrast, Lelieveld *et al.* (2004) present results from ship cruises from 1977 to 2002 that show no statistically significant trends in the 40ºN to 50ºN latitude range.

123. Vingarazan (2004) details 21 near-surface or lower troposphere background locations that have reported increasing ozone trends over the period from 1967 to 2001. Mace Head, Ireland was included in this collection of sites. In contrast, eight surface or near-surface sites reported downwards trends over a similar period. Downwards trends have been reported for some Canadian arctic sites that have reversed during the 1991-2001 period (Vingarazan, 2004).

124. Background ozone has increased by up to 10 µg m^{-3} per decade over the last 20 to 30 years, according to measurements at sea level and on mountain tops that are less influenced by European sources (Raes and Hjorth, 2006). This has been attributed to the world-wide increase in anthropogenic activities, including growth in ozone precursor emissions from industry, road, air and ship transport, households and agriculture. In addition to an upwards trend, background ozone shows considerable year-to-year variability, partly due to precursor emissions such as forest fires, but also due to meteorological variability which can alter the efficiency of long-range transport from particular sources (Raes and Hjorth, 2006).

125. Trend analyses for surface ozone in Europe have been restricted to northern and western Europe, where the time series are long enough for meaningful studies (Monks *et al.,* 2003; Solberg *et al.,* 2004). Apart from the background sites on the western fringes of Europe, trends in surface ozone are not uniform across central Europe (Jonson *et al.,* 2006).

126. The differences in the reported trends between the ozone sondes, surface observations, mountain-top sites and other ozone records are difficult to resolve. There may be inherent difficulties with ozone sondes for long-term trend detection (Jonson *et al.,* 2006). There may well be different local site influences at mountain-top sites compared with surface sites that mean that surface sites are not fully representative of the lower and mid-troposphere (IPCC, 2001). However, surface sites are more relevant to policy formulation and for the assessment of environmental effects.

3.1.3 Observations of the trend in background ozone at Mace Head, Ireland

127. A number of analyses have been published of the long-term ozone monitoring data at the background site at Mace Head, Ireland (Carslaw, 2005; Derwent *et al.,* 2007a; Simmonds *et al.,* 1997, 2004). In all these analyses, care has been taken to separate the ozone data by air mass origins, whether maritime from across the North Atlantic Ocean or regionally-influenced from continental Europe. The basic unsorted monthly mean ozone data from 1987-2007 have shown no statistically significant trends. Attention is directed in the paragraphs below only to the unpolluted maritime data since these alone have shown evidence of trends over time.

128. During most years, around two-thirds of the time, the air masses arriving at the Mace Head, Ireland background station have travelled across the North Atlantic Ocean directly to the station and have had no recent influence from man-made pollutant emissions from Europe. These air masses are considered to be representative of northern hemisphere background conditions. Simmonds *et al.* (1997) used the simultaneous observations of man-made halocarbons, CO and methane and back-track air mass trajectories to sort the hourly ozone observations into two categories: background and regionally-influenced. Over the 1987-1995 period, Simmonds *et al.* (1997) reported an upwards trend of +0.4 $\mu g\ m^{-3}\ yr^{-1}$ in the annual mean hemispheric background ozone abundance.

129. Simmonds *et al.* (2004) using similar sorting techniques, reported trends in background ozone abundances over an extended period from 1987-2003. Annual mean trends were +1.0 $\mu g\ m^{-3}\ yr^{-1}$, with winter trends +1.2 $\mu g\ m^{-3}\ yr^{-1}$ and summer trends +0.8 $\mu g\ m^{-3}\ yr^{-1}$. It has become apparent subsequently that these trends have been influenced by unprecedented biomass burning during 1997/98 as revealed by simultaneous observations of the biomass burning gases: CO_2, CO, methane (CH_4), chloromethane (CH_3Cl) and bromomethane (CH_3Br) (Simmonds *et al.,* 2005). The 1997/98 biomass burning came towards the end of the 1987-2003 record and has apparently exaggerated the observed upwards trend in the background ozone data.

130. Analysis of the 20-year background record over the period 1987-2007 has recently been performed by Derwent *et al.* (2007a) in which an additional air mass origin sorting technique has been applied based on the Numerical Atmospheric Dispersion Modelling Environment (NAME) Lagrangian dispersion model to the data post-1995. Because of the reductions in European emissions, the original sorting method has been increasingly unreliable. Based on a 6-year overlap period from 1995-2001, the 1987-1994 data have been rescaled and the 1995-2006 data have been sorted using the NAME dispersion model. The combined monthly mean 1987-2006 data are presented in Figure 3.2 and annual mean data in Figure 3.3. Considering the time series overall, there is a highly statistically significant increase (at a 99% significance level) in ozone of 0.7 ± 0.3 µg m^{-3} yr^{-1} (95% confidence intervals). However, it is apparent from Figures 3.2 and 3.3 that this increase has not been gradual: there is evidence of two different periods from 1987 to 1997 and from 1998 to 2006. For each of these periods there is no statistically significant change in ozone concentration at a 95% significance level. The time series shows a shift in the mean from 71 to 81µg m^{-3} (i.e. 5 ppb) between these two periods.

131. The perturbation in the ozone record over the period 1997-1998 has been ascribed to global-scale biomass burning events by Simmonds *et al.* (2004). Figure 3.2 shows a rise to the all-time maximum background concentration of 104 µg m^{-3} in March 1999. While such events may explain an anomalous increase, they cannot explain the maintenance of this shift over a number of years.

132. As will be discussed in Chapter 4, methane plays a significant role in the formation of ozone in the background troposphere. Figure 3.4 shows the record of background methane concentrations at Mace Head over the period 1995-2008. Methane increases over this period, with the increase accelerating during the biomass burning events. A step change in background methane levels of 10 µg m^{-3} is apparent in Figure 3.4 during 1997-1998.

133. The persistence of the step change in ozone after 1999 suggests a perturbation to a long-lived mode of the chemistry system. The only mode with a sufficiently long timescale is the methane relaxation timescale (Prather, 1994). However using a rough scaling from section 4.2, where a 400 µg m^{-3} decrease in methane alone causes a 4 µg m^{-3} decrease in ozone, the step change in methane of around 10 µg m^{-3} in 1999 (Figure 3.4) would cause a step change in ozone of only 0.1 µg m^{-3}. Therefore the step change in methane is likely to be a symptom of the same process that affects the ozone, rather than a direct cause, consistent with the step change resulting from biomass burning.

134. The analysis of filtered background ozone concentrations at Mace Head clearly reveals changes that require further investigation and analysis. Given the magnitude and type of change shown in Figure 3.3, and its potential significance for ozone in the UK, it is important to establish the extent to which related changes are manifest at other ozone monitoring sites in the UK and Europe. Furthermore, more research is required to understand the underlying chemical and physical processes that lead to such changes.

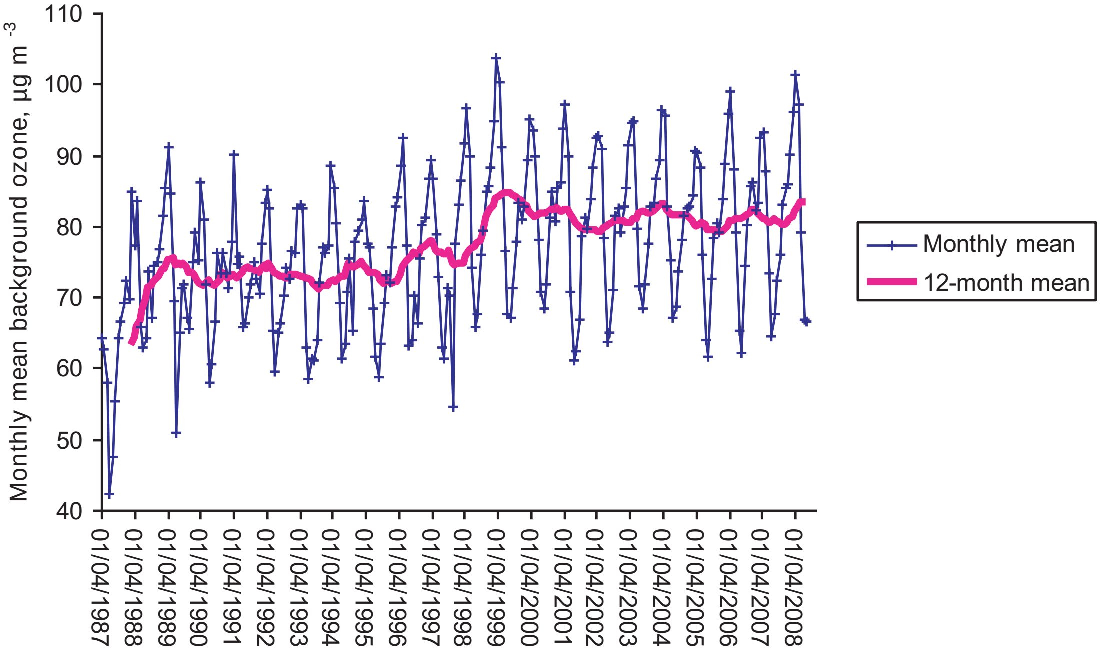


Figure 3.2 Monthly mean (blue line with + signs) and 12-month rolling mean (pink line) background ozone concentrations at Mace Head, Ireland, from 1987 to 2008.

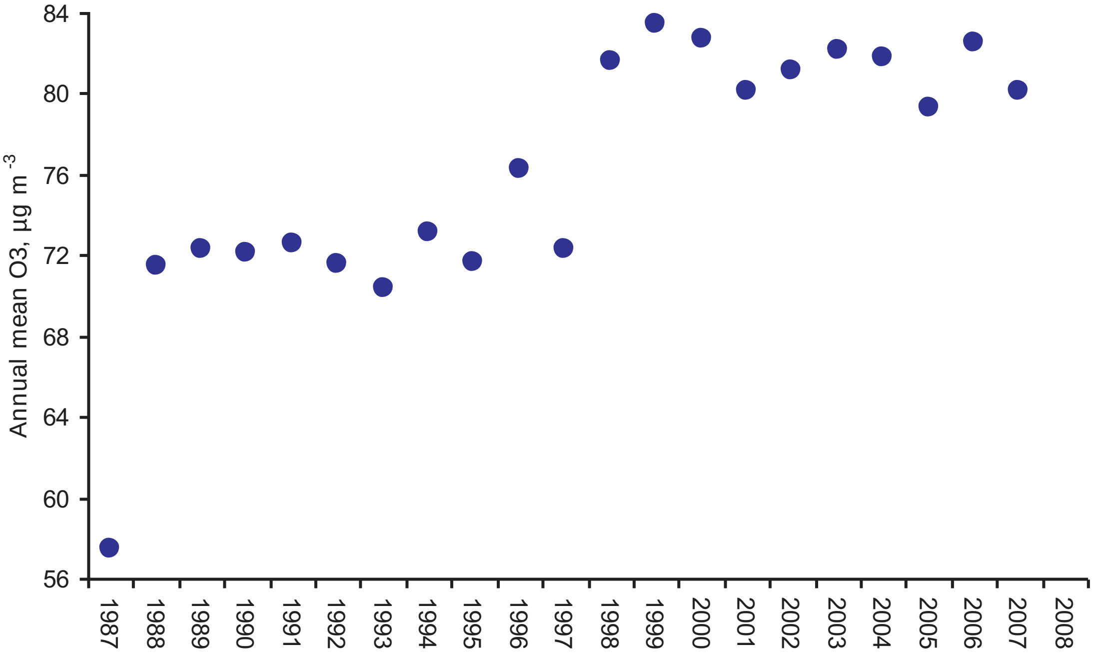


Figure 3.3 Annual mean background ozone concentrations for filtered air masses at Mace Head, Ireland, as determined by Derwent *et al.* (2007a).

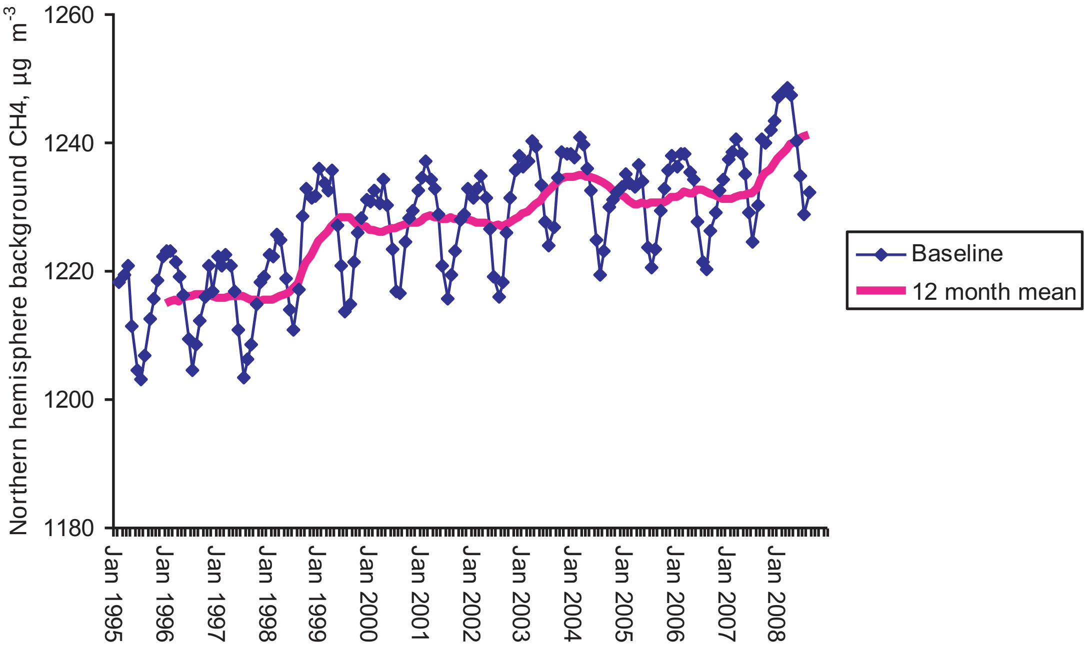


Figure 3.4 Monthly mean (blue line with diamonds) and 12-month rolling mean (pink line) background methane concentrations at Mace Head, Ireland, from 1995-2008.

135. Along the west coast of North America, three sets of observations have shown similar strong springtime increases in ozone as observed at Mace Head, Ireland (Jaffe and Ray, 2007). Measurements from the marine boundary layer surface sites at Lassen National Park in northern California and from aircraft have indicated a positive trend in ozone of +0.8 to +1.4 µg m^{-3} yr^{-1} depending on season between 1984 and 2002 (Jaffe *et al.,* 2003). Parrish *et al.* (2004) identify the cause of this increase as increasing emissions of ozone precursors at northern temperate latitudes.Oltmans *et al.* (2008) have reanalysed the observations on the Pacific coast of North America over the period 1981-2006 and conclude there has not been any significant impact of changing background ozone.

3.1.4 Relevance of the Mace Head background observations to the UK and Europe

136. The Mace Head observations define the trace gas concentrations in air masses that have recently been advected across the North Atlantic Ocean and on into Europe, the United Kingdom and Ireland included. Because of the large expanse of the North Atlantic Ocean, the Mace Head background site can be taken as representative of the Atlantic seaboard of Europe over a significant latitude range.

137. Back-track trajectories confirm that on most days, including most ozone episode days, the air masses arriving at UK monitoring sites and a significant number across north-west Europe, can be back-tracked eventually to the North Atlantic region. This may take up to 10 days under anticyclonic conditions. On this basis, the Mace Head background observations provide a reliable guide to the trace gas concentrations upon which European regional-scale pollution events are superimposed. Mace Head observations are, for example, used to modify the initial and boundary conditions for the Unified EMEP model (Simpson *et al.,* 2003).

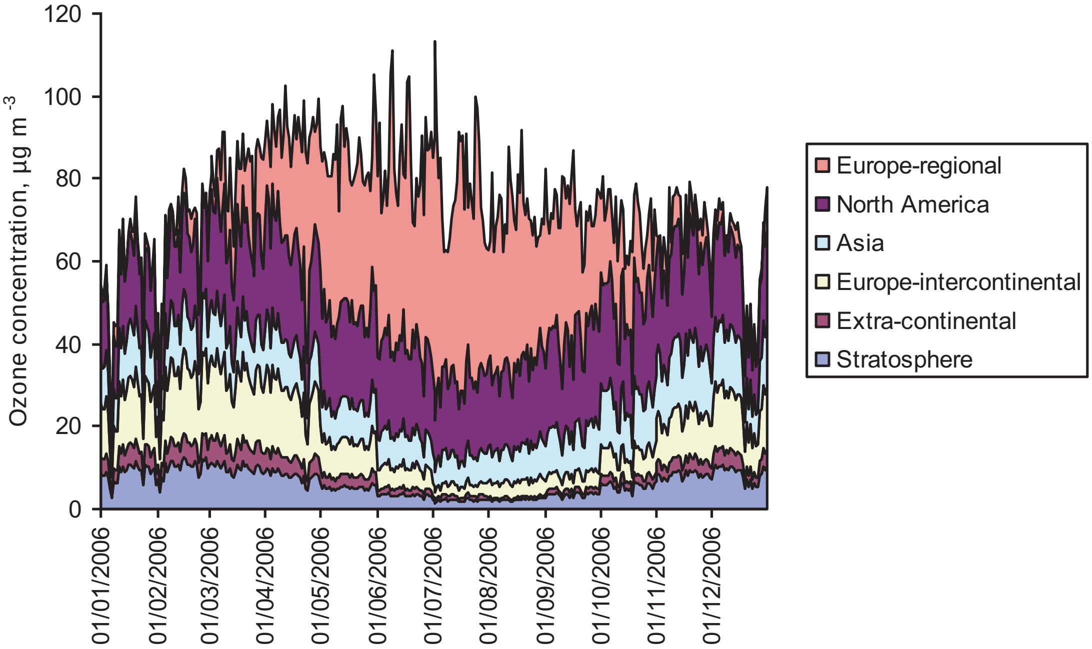


Figure 3.5 Source attribution of the ozone found at a rural location in southern England during each afternoon of 2006 using the UK PTM and STOCHEM models. Europe-regional refers to the ozone advected directly; North America to that formed over that continent and over the western Pacific; Europe-intercontinental to that advected around latitude circles and back into Europe.

138. Using the UK Photochemical Trajectory Model (PTM) and the global model STOCHEM, it has been possible to provide an attribution for the ozone modelled for Harwell, a rural location in Oxfordshire, during each afternoon of 2006, see Figure 3.5. The plot shows the daily contribution to ozone from regional-scale ozone formation within the model domain and from background sources, including exchange with the stratosphere and transport across the North Atlantic Ocean from sources in North America, Asia and, ultimately, from Europe having travelled around a latitude circle. Regional-scale ozone formation makes its largest contribution during the summer months, whereas background sources contribute most during springtime. On an annual average basis, regional-scale ozone formation accounts for about 22 µg m^{-3} and background sources for about 52 µg m^{-3} of the 74 µg m^{-3} annual mean daily maximum 1-hour mean ozone concentration estimated in the model.

139. Figure 3.5 and the annual average source attribution figures in the paragraph above are highly site specific. Generally speaking, it is anticipated that regional-scale ozone formation increases and background sources decrease relative to the Harwell case when moving eastwards and southwards into Europe. This is because the increasing frequency of photochemical episodes increases the former and the increasing travel time and hence dry deposition losses of background ozone decreases the latter.

140. Because of the dominant contribution from background sources to the source attribution of ozone at Harwell illustrated in Figure 3.5, it is apparent why the trend in background ozone levels is so important for the UK and for north-west Europe. The trend up to the year 2000 has apparently been stronger during the spring months when background sources are dominant. Furthermore, regional-scale ozone precursor emission controls have strongly reduced regional-scale ozone formation, particularly during the summer months, when the regional term dominates.

3.1.5 Modelling the global ozone background trend at Mace Head, Ireland

141. During the last few years, global CTM studies of tropospheric ozone have advanced significantly because of the availability of improved, consistent gridded emission inventories for the tropospheric ozone precursors that have resulted from the activities of the UNFCCC and of IIASA. These emission inventories have been used to complete multi-model ensemble simulations of present-day and near-future tropospheric ozone burdens (Stevenson *et al.,* 2006) and of CO (Shindell *et al.,* 2006) under the aegis of the EU ACCENT programme; the ACCENT output is discussed in further detail in Chapter 4. A detailed comparison has also been completed of two CTMs that participated in the EU ACCENT intercomparison, STOCHEM and TM3. Generally, both TM3 and STOCHEM represented the surface ozone concentrations observed at six background stations well (Dentener *et al.,* 2005). This study has been used to compile an analysis of the evolution of the methane, CO and tropospheric ozone burden from 1990-2030 at Mace Head, Ireland from 1990-2030 using STOCHEM (Derwent *et al.,* 2006).

142. The STOCHEM/TM3 study showed similarities between the model and observed abundances of methane, CO and ozone over the period from 1990 to 2002 for Mace Head. Seasonal cycles were in phase between the model and observations and model biases were minimal. Model trends in methane overestimated those observed as did those in CO. Model and observed CO abundances diverged particularly during 1998/99 when the observed record had been influenced by unprecedented levels of biomass burning which had not been included in the global emission inventories. Model seasonal cycles for ozone faithfully reproduced those observed in the background record at Mace Head. Again, the model failed to show the ozone anomaly observed during the winter of 1998/99 due to biomass burning.

143. Over the 1990-2002 period, the STOCHEM/TM3 model indicated an upwards trend in ozone at Mace Head of $+0.32\ \mu g\ m^{-3}\ yr^{-1}$. The STOCHEM study only considered the trends driven by man-made tropospheric ozone precursors and made no allowances for any trends in natural sources such as wetlands, tundra, natural fires, lightning, soil and vegetation emissions. The observed trend over the period from 1987-1995 was reported as $+0.38\ \mu g\ m^{-3}\ yr^{-1}$ by Simmonds *et al.* (1997) during the period where there was no apparent influence from global biomass burning events. It is therefore likely that the STOCHEM model may well have been able to account for much of the observed rise in background ozone levels during the 1987-1995 period.

144. Whilst it is generally accepted that increasing global man-made emissions of ozone precursors are an important, likely cause of the growth in background ozone levels (IPCC, 2001; Parrish *et al.,* 2004), other explanations are not ruled out. Lelieveld and Dentener (2000) point to the possibility that the observed inter-annual variability in lower tropospheric ozone is influenced by changes in stratospheric ozone. Ordóñez *et al.* (2007) demonstrate an impact of northern mid-latitude lowermost stratospheric ozone changes on background ozone in the lower troposphere at the Jungfraujoch and Zugspitze mountain-top sites in the Alps. Using the SLIMCAT model they are able to explain the time trend in lower tropospheric ozone anomalies from 1990 through to 2004 based on the upwards trend and year-to-year variability in lowermost stratospheric ozone. Oltmans *et al.* (2006) drew attention to the disparity in the annual growths in ozone between Mace Head, Ireland (+8.2 % per decade) and Zugspitze (+12.6 % per decade) and this stratospheric ozone signal found by Ordóñez *et al.* (2007) may well be an explanation.

3.1.6 Forecasts of future background ozone levels in the British Isles

145. Looking into the future, the STOCHEM study pointed to future ozone trends for Mace Head, Ireland, of +0.18-0.24 µg m^{-3} yr^{-1} in the IIASA CLE scenario over the period to 2030 and -0.28 µg m^{-3} yr^{-1} in the IIASA MFR scenario (Derwent *et al.,* 2006). Further details are given of these emission scenarios in Annex 1 and of the multi-model ensemble calculations for the whole of Europe in Chapter 4. For the southern United Kingdom, STOCHEM model experiments give an indication of the ozone trends for these global emission scenarios (Figure 3.4). This plot shows the observed annual mean ozone abundances from the seven Defra rural ozone monitoring network sites in southern England for 1990-2003, together with the model trends out to 2030. 60-month rolling mean ozone abundances increase from 66.4 µg m^{-3} in 2000 to 72.6 µg m^{-3} in 2030 in the IIASA CLE scenario corresponding to an annual increase of +0.3 % per year. In contrast, in the IIASA MFR scenario, rolling mean ozone abundances fall from 66.4 µg m^{-3} in 2000 to 62.8 µg m^{-3} in 2030, corresponding to a decline of -0.2% per year. Observed background ozone abundances over the 2000-2006 period have remained level and shown no overall trend, in the middle of the range of the STOCHEM model predictions (Derwent *et al.,* 2007a).

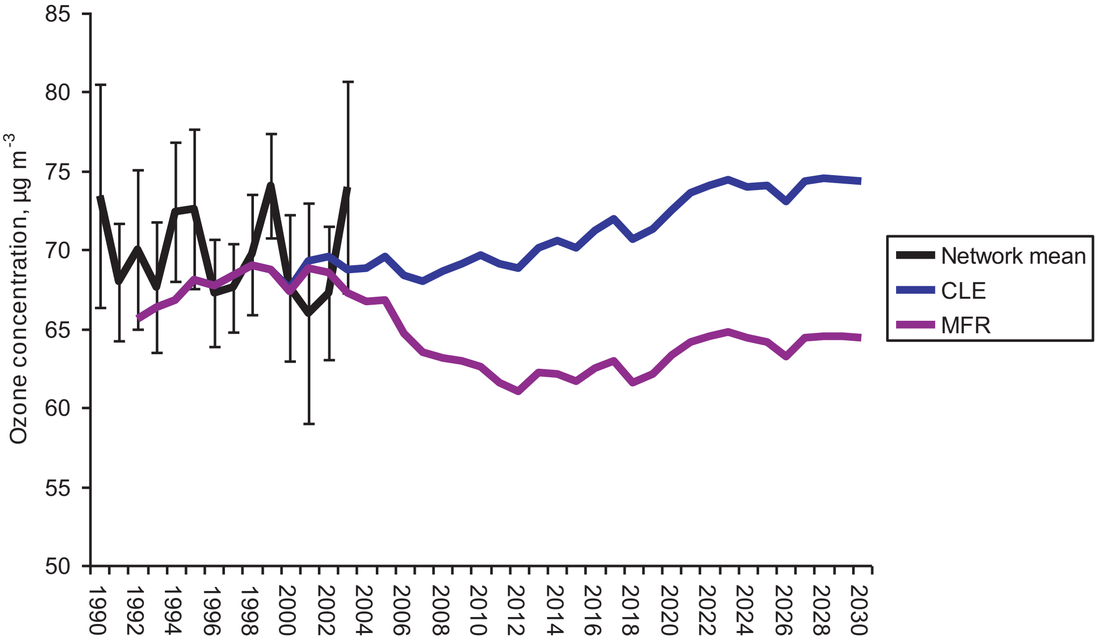


Figure 3.6 Observed annual mean rural ozone network concentrations and 60-month rolling mean STOCHEM model results for southern England in the IIASA CLE and MFR scenarios for 1990 through to 2030.

3.2 Recommendations

146. The Air Quality Expert Group makes the following recommendations to Defra:

- Analysis of ozone monitoring data is required to assess the impact of the increases observed at Mace Head on ozone concentrations in the UK.
- The Task Force on Hemispheric Transport of Air Pollution should consider the likely origin of the background ozone trends seen at some sites from the 1970s onwards.
- Research is needed on biomass burning and its influence on background ozone concentrations.
- The issue of changes in background ozone should be kept under review.
- Support is needed for work on hemispheric emission projections for tropospheric ozone precursors, similar to that done by IIASA for 2050.

Chapter 4

Short-term impact of climate change on ozone concentrations in Europe

Question C: *What is the likely impact of climate change on future ozone levels in Europe over the next two decades? What is the significance of such impacts compared to other influences, such as inter-annual variability or (global and regional) emission trends?*

Short answer to question C

147. The net impact of climate change on mean surface ozone levels over Europe on the 2030 time horizon is not known with any confidence but is likely to be small compared with the most important influence. This is the change in the anthropogenic emissions in Europe and throughout the whole northern hemisphere of the important precursor gases to ozone formation: nitrogen oxides (NO_X), methane (CH_4) and non-methane volatile organic compounds (VOCs), in particular, and carbon monoxide (CO). Climate change may have relatively greater influence on future peak episodic ozone in particular geographic areas through a number of different mechanisms such as changes in precursor emissions, ozone loss by deposition and meteorology. Inter-annual variability in annual mean surface ozone at a given location is large compared with the likely magnitude of net ozone change by 2030, so multi-year data series are necessary for unravelling the competing influences on ozone concentration at different locations.

Detailed answer to question C

148. Assuming trends in anthropogenic emissions of precursor gases around the world follow the projections of presently-planned controls ("current legislation"), then an ensemble of global models simulate an increase in annual mean surface ozone over Europe as a whole of 1.8 ppb between the years 2000 and 2030. For a more optimistic scenario in which all possible technical control options are implemented worldwide, the models simulate a decrease in average surface ozone over Europe of -2.8 ppb over this period. In contrast, for a more pessimistic high-growth scenario, average surface ozone over Europe is simulated to increase by 3.9 ppb. Precursor emission projections are continually re-evaluated, and actual emissions may vary from those projected for a given scenario, so the above simulations indicate the range in possible future annual mean surface ozone change over Europe over this time period.

149. The models show that the benefit to mean European ozone levels of current legislation control measures on European emissions of precursor gases is more than offset by increasing hemispheric and global ozone levels, driven by increasing precursor emissions elsewhere, and their subsequent long-range transport into Europe. However, projected ozone changes vary spatially across the region and with season, with the UK having an "ozone climate" that is often somewhat different to that of Europe as a whole. There is a general tendency for absolute ozone levels to be greater in southern and central Europe and for beneficial change to be more marked in these regions than in

north-west Europe. Also, although simulations of the current legislation scenario project an increase in ozone over Europe on average, they project a decrease in summertime ozone episode extremes but an increase in winter ozone levels. This will increase the metrics of long-term exposure to ozone, such as SOMO35 and AOT40, but is not expected to have an important impact on ozone metrics that are sensitive to short-term high peaks.

150. The net sign of the additional impact of climate change on ozone across particular geographic regions such as Europe, let alone the magnitude of the impact, is highly uncertain. This is because, although many different processes have been identified through which climate change can influence future ozone levels (e.g. effects on anthropogenic and biogenic precursor emissions, atmospheric chemistry, synoptic meteorology, deposition and stratospheric-tropospheric exchange), only a limited number of coupled climate-chemistry models have been run and many known climate impacts have yet to be included. The most substantive impacts quantified so far on global tropospheric ozone are increased gas-phase chemical destruction of ozone driven by increased atmospheric humidity, and increased downward transport of ozone from the stratosphere. Some model simulations suggest a pattern of net negative impact of climate on surface ozone over the oceans, but a net positive impact over polluted land surfaces, the latter likely driven, at least in part, by the net dominance of increased ozone production with water vapour concentration in high-NO_x environments. Overall, however, the size of modelled impacts of climate change on mean European surface ozone to 2030 to date are small compared with the modelled changes in ozone arising from anthropogenic precursor emission changes, and insignificant in comparison with the uncertainties in these emissions projections and with inter-model variability. This does not exclude the possibility that climate change may have proportionally larger influence on regional peak summertime surface ozone through, for example, drought-related depression of ozone dry deposition, increased incidence of wild fires, or extended air mass residence time in the boundary layer. Also, the climate change signal on surface ozone is likely to increase significantly beyond the 2030 time horizon specified for this report.

151. Changes in natural sources of ozone precursors in Europe to 2030 are anticipated to be of less influence on future mean ozone levels in Europe than the changes in the man-made precursor emissions of NO_x, VOC, CH_4 and CO over this period. Biogenic VOC emissions may increase in the future with increasing temperatures under climate change, although other factors will certainly also be influential, such as future changes to the vegetation mix caused by human activities or by environmental change itself. VOC emissions are found to contribute to ozone production in the UK and near-European continent, but may be less important to ozone generation in the NO_x-limited atmosphere of southern Europe. The relationship between biogenic emissions and major episodes of poor air quality in the context of both a changing climate and other precursor emission changes remains an area of major uncertainty.

152. Although there is variation in model simulations of future ozone, these inter-model differences are generally smaller (to the 2030 time horizon) than the differences in average simulations of future ozone for different

emission scenarios; so the main source of uncertainty in predicting future ozone is in projecting the emissions, rather than in the modelling. Uncertainty increases substantially when trying to model effects of climate change also, because of deficiency in what is currently incorporated within global- and regional-scale models.

153. Large-scale climate variability phenomena, such as the North Atlantic Oscillation, impose inter-annual variability on annual surface ozone concentrations at a given European location which is likely to be of comparable magnitude to the net trend over the next two to three decades caused by the other influences described above.

Supporting evidence for question C

4.1 Overview

154. **Note on units:** In this chapter, abundances of model-simulated ozone are expressed in parts per billion volumetric mixing ratio (ppb, 1 part in 10^9) for the reasons explained in Chapter 1. For ozone close to the surface, at ambient temperature and pressure (293 K and 101.3 kPa, the conversion is 1 ppb ≡ 2 $\mu g\ m^{-3}$.

155. The Air Quality Expert Group's (AQEG's) Third Report *Air quality and climate change: A UK perspective* (AQEG, 2007b) has previously summarised considerable background information on this question. The answer to question G in Chapter 8 of this report also provides complementary information, as does a concurrent report from the Royal Society, *Ozone in the 21st century* (Royal Society, 2008).

156. The concentrations of ozone over Europe are dependent on the global background of ozone (due to photochemical formation of ozone throughout the troposphere and downward transport from the stratosphere), on the local and regional emissions of ozone precursors within Europe, and on meteorology (via its influence on long-range transport of ozone and on regional air pollution episodes). Chapter 1 provides further information on the factors influencing ozone and the temporal and spatial scales on which they operate.

157. The most important precursors of ozone are methane (CH_4) and carbon monoxide (CO), which are long-lived gases, and nitrogen oxides (NO_X) and non-methane volatile organic compounds (VOC_S), which are relatively short-lived gases. Model calculations indicate that approximately half of the increase of tropospheric ozone from pre-industrial times to the present day is due to changes in the chemistry of the atmosphere induced by the increase in NO_X and approximately half to the combined increase of CH_4, CO and VOC emissions (~25% for CH_4, and ~25% for CO and VOC together) (Wang and Jacob, 1998).

158. Models, described below, indicate that the most important influence on changes in surface ozone over the period to 2030 will continue to be the global and regional mix in anthropogenic emissions of these short- and long-lived ozone precursor gases, which in turn is consequent on the particular trajectory of economic growth, and of air quality and climate change legislation implementation, that is followed. Changes in natural sources of

ozone precursors within Europe, predominantly biogenic VOCs from natural vegetation, are likely to be less influential on the 2030 timescale. Biogenic VOC emissions may increase in the future with increasing temperatures under climate change, although other factors will certainly also be influential, such as the future anthropogenically-driven or climate change-driven vegetation mix. VOC emissions are observed to contribute to episodic ozone production in the UK and near-European continent, but may be less important to ozone generation in the NO_X-limited atmosphere of southern Europe.

4.2 Impacts of trends in precursor emissions

159. A recent model intercomparison organised under the auspices of ACCENT has compared the results from 26 differently formulated chemistry models (Dentener *et al.,* 2006; Stevenson *et al.,* 2006). The models investigated the effect on global ozone levels in the year 2030 compared with the year 2000 of three different scenarios for future worldwide emissions available at the time of the research: “central”, “optimistic” and “pessimistic” (Table 4-1). A subset of ten of the models also investigated the effect of climate change on atmospheric chemistry for the “central” emissions scenario, using a simulated climate for 2030 as described in Stevenson *et al.* (2006). Further detail on ozone precursor emissions projections is given in Annex 1.

Table 4-1 ACCENT CTM (Chemical Transport Model) intercomparison experiments (adapted from Dentener *et al.,* 2006). Notes: GCM: General Circulation Model, SSTs: Sea Surface Temperatures; IIASA: International Institute for Applied Systems Analysis; IPCC: Intergovernmental Panel on Climate Change; SRES: Special Report on Emissions Scenarios.

Run	Descriptor	Meteorology	Emissions
S1	Y2000 (Background)	CTM 2000 GCM SSTs 1990s	2000
S2	CLE (Central)	CTM 2000 GCM SSTs 1990s	IIASA current legislation, to 2030
S3	MFR (Optimistic)	CTM 2000 GCM SSTs 1990s	IIASA maximum technically feasible reduction, to 2030
S4	A2 (Pessimistic)	CTM 2000 GCM SSTs 1990s	SRES A2, most pessimistic IPCC scenario, to 2030
S5	CLEc (Central + climate change)	GCM SSTs 2030s only	IIASA current legislation, to 2030

160. For the current legislation scenario (“central”), annual mean surface ozone averaged over Europe as a whole is simulated to increase by 1.8 ppb between 2000 and 2030 (+0.6 ppb/decade) (Dentener *et al.,* 2006). For the maximum technically feasible reduction scenario (“optimistic”), in which all possible technical control options are implemented worldwide, average surface ozone over Europe is simulated to decrease by –2.8 ppb (–0.9 ppb per decade). In contrast, under the more pessimistic high-growth A2 scenario of the Intergovernmental Panel on Climate Change (IPCC) Special Report on Emissions Scenarios (SRES), average surface ozone over Europe is simulated to increase by 3.9 ppb (+1.3 ppb per decade).

161. Europe-wide averages obscure the fact that the magnitude of the modelled changes in future ozone vary significantly geographically across Europe and with season. The spatial variation in the changes in annual mean and seasonal mean surface ozone over Europe for the year 2030 compared with the year 2000 are shown in the left-hand side of Figures 4.1-4.3 for the three different projections of global emissions. The extent of inter-model variation is illustrated by the corresponding maps of the model ensemble standard deviations of simulated ozone changes.

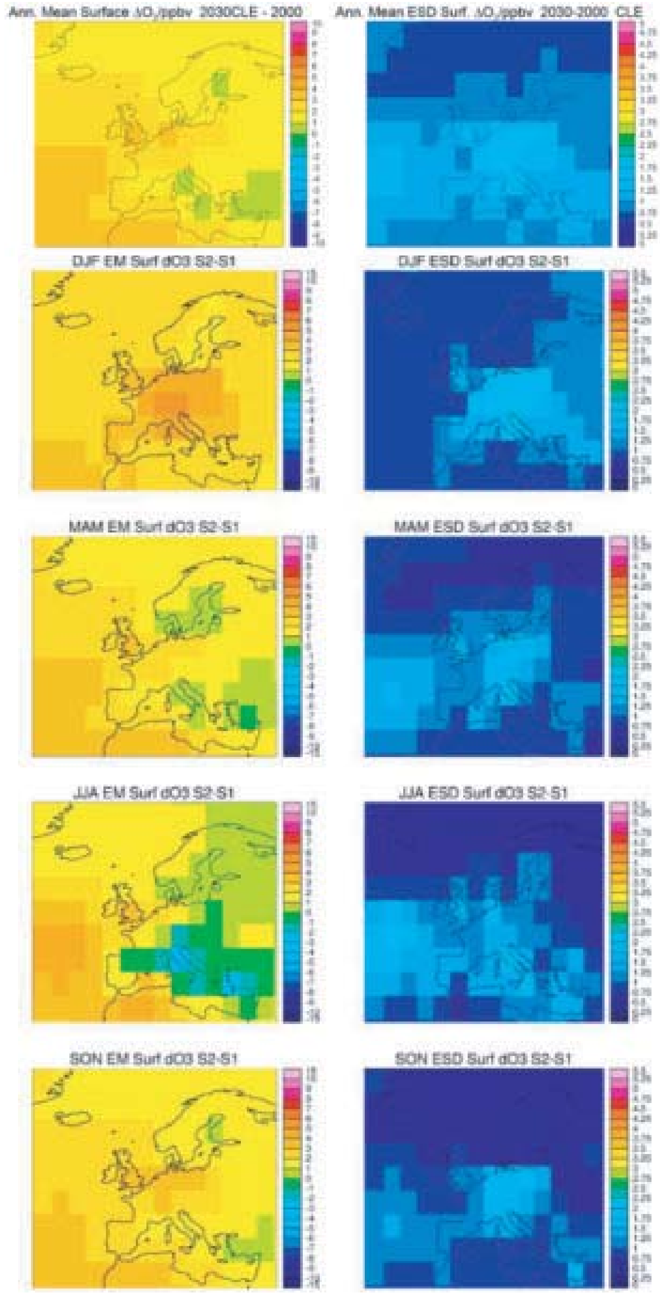


Figure 4.1 Left column: 26-model ensemble-mean <u>change</u> in annual mean surface ozone (top row) and 3-month seasonal mean surface ozone (subsequent rows) for year 2030 with "central" emissions scenario CLE (climate unchanged) (model run S2) compared with base year 2000 (model run S1). "DJF" refers to mean for the 3 months December, January, February, "MAM" refers to March April May, etc. Right column: Standard deviation of the corresponding 26-model ensemble simulations. Units are ppb (1 ppb $O_3 \equiv 2$ µg m^{-3} O_3). The models have been interpolated to a common resolution (5° x 5° horizontal); lowest model level depth is ~100 m. Source: Stevenson (pers. comm., 2008) from data of Dentener *et al.* (2006).

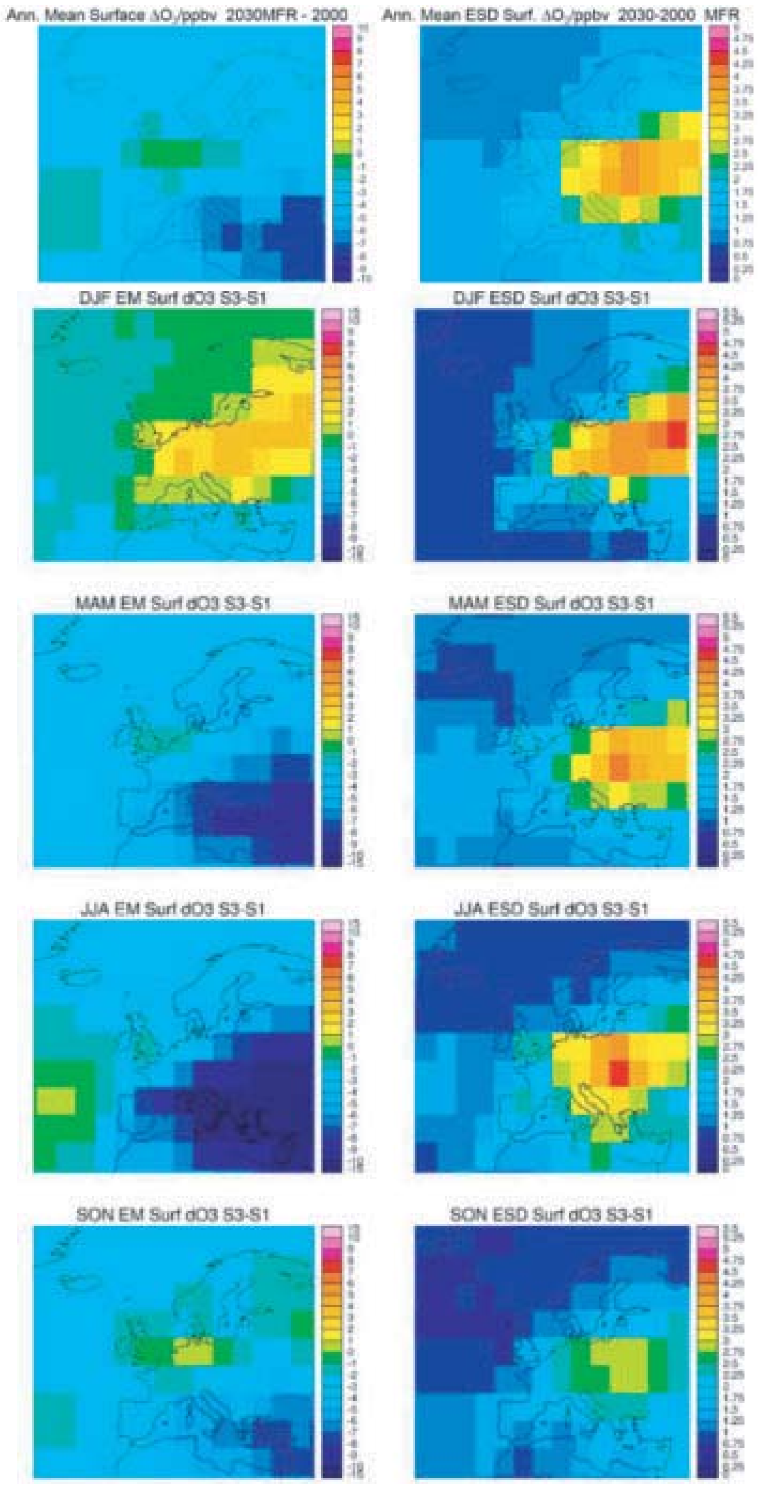


Figure 4.2 Left column: 26-model ensemble-mean change in annual mean surface ozone (top row) and 3-month seasonal mean surface ozone (subsequent rows) for year 2030 with "optimistic" emissions scenario MFR (climate unchanged) (model run S3) compared with base year 2000 (model run S1). "DJF" refers to mean for the 3 months December, January, February, etc. Right column: Standard deviation of the corresponding 26-model ensemble simulations. Units are ppb (1 ppb O_3 ≡ 2 μg m^{-3} O_3). The models have been interpolated to a common resolution (5° x 5° horizontal); lowest model level depth is ~100 m. Source: Stevenson (pers. comm., 2008) from data of Dentener *et al.* (2006).

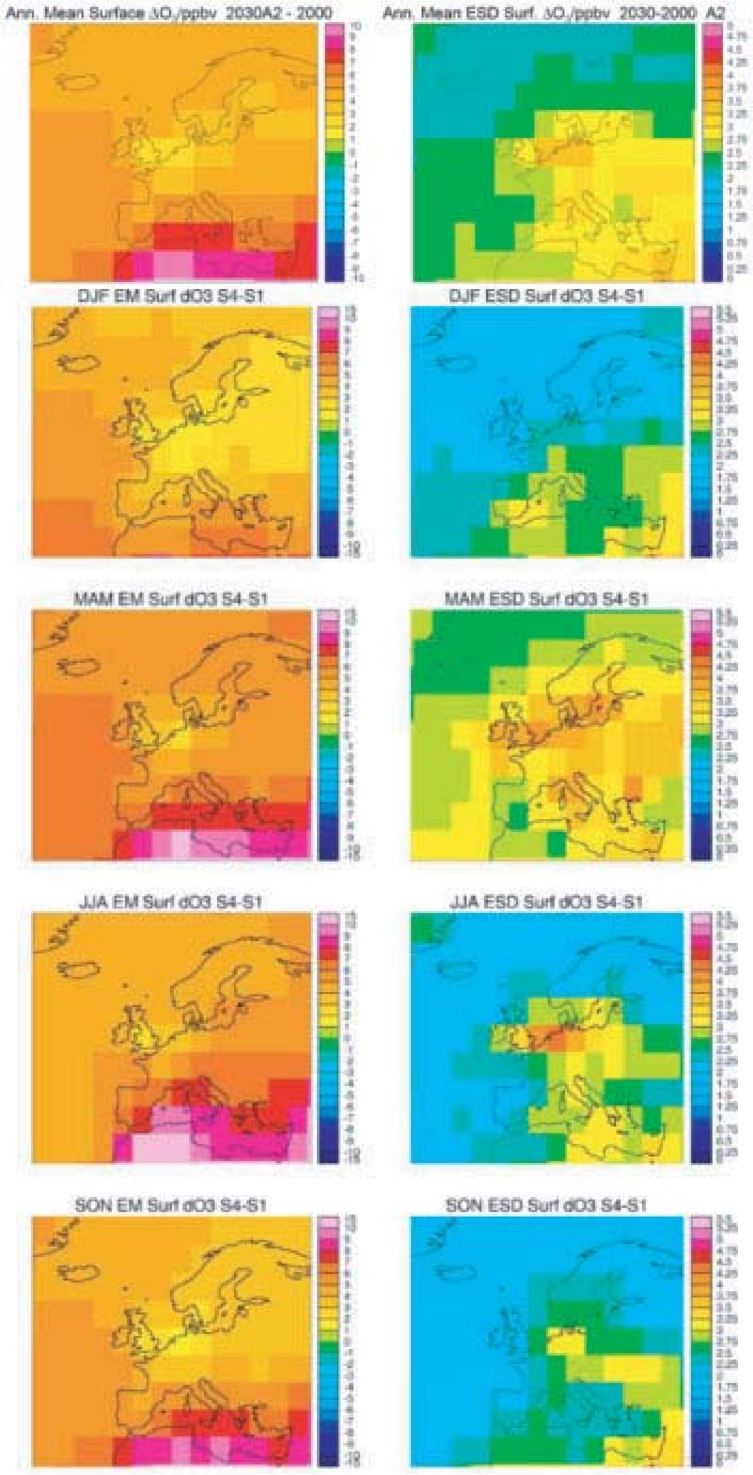


Figure 4.3 Left column: 26-model ensemble-mean change in annual mean surface ozone (top row) and 3-month seasonal mean surface ozone (subsequent rows) for year 2030 with "pessimistic" emissions scenario SRES A2 (climate unchanged) (model run S4) compared with base year 2000 (model run S1). "DJF" refers to mean for the 3 months December, January, February, etc. Right column: Standard deviation of the corresponding 26-model ensemble simulations. Units are ppb (1 ppb $O_3 \equiv 2\ \mu g\ m^{-3}\ O_3$). The models have been interpolated to a common resolution (5° x 5° horizontal); lowest model level depth is ~100 m. Source: Stevenson (pers. comm., 2008) from data of Dentener *et al.* (2006).

162. There is a strong gradient of greater annual average ozone from north to south across Europe reflecting gradients in ozone photochemistry, the transport patterns of ozone and its precursors into Europe, and the decreased removal of ozone by dry deposition over the Mediterranean Sea as compared with over land. The interplay between the factors determining future ozone is complex. For example, ozone levels over southern and eastern Europe are simulated to increase less (or actually to decrease) compared with the simulated increases in ozone levels over northern Europe for the "current legislation" (CLE) scenario (particularly in summer, Figure 4.1), but to increase more than ozone levels over northern Europe for the pessimistic SRES A2 emissions scenario (Figure 4.3). Also, although the optimistic "maximum technically feasible reduction" (MFR) scenario is simulated to lead to substantial declines in summertime surface ozone across most of Europe, this scenario gives rise only to marginal improvement in ozone over the southern UK and near continent (Figure 4.2). These latter simulations illustrate the consequence of the NO_X-saturated chemistry in this region (Collins *et al.,* 2007). In winter, in northern Europe, NO_X emissions generally decrease ozone production by removing HO_X radicals through the reaction $NO_2 + OH \rightarrow HNO_3$ (nitrogen dioxide + hydroxyl radical → nitric acid). For the CLE and MFR scenarios, the local NO_X emission reductions increase wintertime ozone. For the SRES A2 scenario, an expected local decrease in European winter ozone production is swamped by the import of increased ozone from outside Europe (Figure 4.3). The transport of ozone is most efficient in winter due to its increased lifetime.

163. In areas that better represent background conditions, and where inter-model agreement is generally better (such as over the Atlantic Ocean off the coast of the European continent away from the shipping lanes), the simulated changes in annual average ozone between 2000 and 2030 are up to +4, -3 and +6 ppb for the central, optimistic and pessimistic emissions scenarios, respectively.

164. Current modelling activity indicates that the benefit to average ozone over Europe from current legislation reductions on European emissions of precursor gases is more than offset by increasing hemispheric and global ozone levels, caused by increasing precursor emissions elsewhere, and their subsequent long-range transport into Europe (Derwent *et al.,* 2006). Again, it is important to note that there is considerable geographical and seasonal variability within this overall statement. A decrease in summertime average surface ozone of -3 (±1.5) ppb (as model ensemble average ± standard deviation) is calculated for 2030 for the CLE scenario over some southern areas of Europe, whereas an increase up to 4 (±1.5) ppb is calculated over parts of north-western Europe (panel "JJA" in Figure 4.1). On the other hand, the CLE scenario is expected to lead to a decrease in extreme summertime ozone episodes everywhere, but to an increase in wintertime ozone levels (panel "DJF" in Figure 4.1).

165. The offsetting effect of hemispheric ozone on average European ozone levels in the CLE scenario is illustrated in model estimates for changes to human health indices for exposure to ozone by 2030. The ensemble-mean data from the 26-model intercomparison is for the SOMO35 metric for the central Europe region (7°-17°E, 48°-54°N) to increase by 341 ppb.days by 2030 from the year 2000 modelled estimate of 2795 ppb.days; and for SOMO35 for the central Mediterranean region (5°-30°E, 35°-45°N) to increase by 234 ppb.days by 2030 from the year 2000 estimate of 5559 ppb.days (Ellingsen *et al.,* 2008).

(No objective is currently defined for this metric.) In contrast, the ensemble-mean simulation for the effect of the current legislation emissions scenario on the EU60 health metric (the number of days in the year with maximum 8-hour average exceeding 60 ppb), which has greater emphasis on ozone extremes, is for a decrease of 4 days by 2030 from 26 days in year 2000 in the central Europe region, and for a decrease of 2 days from 75 days in the central Mediterranean regions. (The currently defined threshold for this metric is 25 days per year as an average over 3 years.) Under the MFR scenario, the model-ensemble average change in the SOMO35 metric is a decrease of 935 ppb.days in the central Europe region and a decrease of 2318 ppb.days in the central Mediterranean region, and the model-ensemble average change in the EU60 metric is a decrease of 23 days in the central Europe region and a decrease of 55 days in the central Mediterranean region (Ellingsen *et al.,* 2008).

166. The magnitude of all these simulated changes should be set in the context of the spread of model results. The uncertainty in the calculations of future European ozone obtained by comparing results from the ensemble of different models is relatively large, of the order of 30% or 1-2 ppb in the simulated ozone changes for 2030 (Dentener *et al.,* 2006; Stevenson *et al.,* 2006). Inter-model uncertainty for individual grid domains is much larger (right-hand panels of Figures 4.1-4.3) than the inter-model uncertainty for European-scale averaged values, as expected. The inter-model uncertainty is also not spatially uniform, being generally substantially greater over eastern and southern Europe than elsewhere.

167. The individual model estimates for the values of the health metrics vary widely but the variation is substantially less for the SOMO35 metric than for the EU60 metric, suggesting that the former is probably the more robust indicator of changes in ozone exposure when considering results from individual models (Ellingsen *et al.,* 2008).

168. On a European-averaged spatial scale, inter-model variations are smaller than the differences in average simulations for future ozone for different emission scenarios, so the main sources of uncertainty in simulating future ozone are the future trends in global and regional precursor emissions, rather than the models. However, it should also be recognised that the majority of simulations so far are derived from global models, and results may be different if deploying higher resolution regional models.

169. Subsequent to the ACCENT model intercomparison described above, the International Institute for Applied Systems Analysis (IIASA) provided updated estimates for ozone precursor emissions for the year 2000 and into the future under the “current legislation” (CLE) scenario (Rafaj and Amann, 2007). The IIASA projections are only for anthropogenic emissions. The new IIASA emissions estimates, plus updated shipping emissions estimates with greater spatial disaggregation, have been incorporated into new model simulations of future ozone (Royal Society, 2008). The new 2000 global emissions totals for NO_X and CO are now ~11% and ~15% higher, respectively, than the global totals for 2000 used in the published ACCENT intercomparison. On the other hand, the new scenario suggests that the initial projections of future precursor emissions under a CLE scenario may have been too high and that a commitment to implementation of emissions reduction technology has

occurred more rapidly worldwide than was previously envisaged. Global emissions projections of NO_X and VOC in the updated CLE scenario now lie somewhere between the old CLE and MFR scenarios, although still closer to the former in 2030 because the impact of the new CLE scenario on these precursor emissions is relatively small before 2030 but greater thereafter. (See also note below about assumed global CH_4 concentrations in the scenarios.)

170. The preliminary model simulations indicate that, as expected, the lower future precursor emissions in the updated CLE scenario lead to lower surface ozone increase by 2050 than was obtained by 2030 using the old CLE scenario. Figure 4.4 compares model simulations of European ozone using the updated emissions with the ACCENT simulations (Royal Society, 2008). By 2050, simulations using the updated CLE scenario show European-averaged annual-average ozone to be lower than in 2000 (white solid and black solid lines, respectively, in Figure 4.4), although this is not uniform throughout the year – spring and summertime ozone is lower in 2050, but ozone in winter is higher. Model results are not available for the updated CLE scenario for 2030 but, given the relative values of the emissions estimates, simulated European surface ozone in 2030 would have values between the solid and dashed white lines in Figure 4.4.

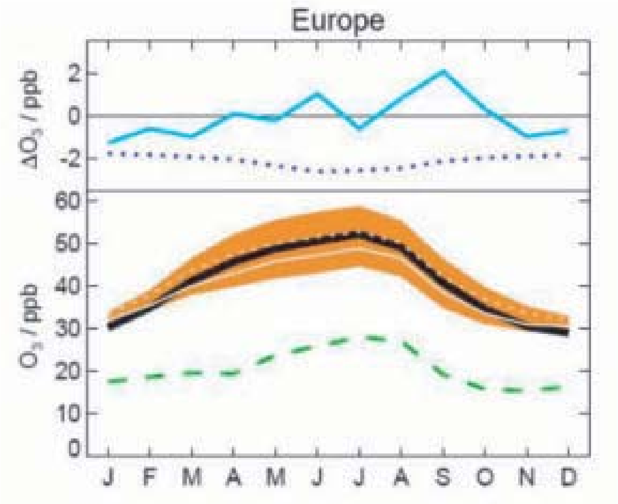


Figure 4.4 Lower panel: Seasonal cycle in future surface ozone averaged over Europe from various model simulations. The dashed green line is from simulation of a pre-industrial emissions scenario. The solid black line is for year 2000 using the updated IIASA year 2000 emissions. The solid white line is for year 2050 using the updated IIASA CLE emissions scenario. The shaded orange region shows the range of the ensemble-mean ACCENT-simulated European surface ozone for year 2030 using the "old" IIASA scenarios as already discussed in the text and illustrated in detail in Figures 4.1-4.3 (the white dashed line is the ACCENT simulation for 2030 using the "old" IIASA CLE scenario). Upper panel: Change in European surface ozone relative to the updated 2050 IIASA CLE scenario for two cases; inclusion of 2000-2050 climate change (solid cyan), and for methane fixed at 2000 levels (dotted blue). For the non-ACCENT data, between three and five different global models were used. Source: Royal Society (2008).

171. Model-simulated future ozone is also very sensitive to assumed future global CH_4 concentration, which is projected to be higher than it is presently. Although global CH_4 abundances have levelled off in recent years (at ~1760 ppb), it appears this may have been due to transient counteracting factors and that anthropogenic CH_4 emissions are again on the increase (Bousquet *et al.,* 2006; Rigby *et al.,* 2008).

172. This sensitivity of ozone to global CH_4 is clearly illustrated by the dotted blue line in the upper panel of Figure 4.4 which shows the substantial (~ 2 ppb) decrease in European-averaged surface ozone that might be anticipated in 2050 for the updated CLE scenario but with a year 2000 CH_4 abundance of 1760 ppb rather than the year 2050 projected CH_4 abundance of 2363 ppb used in these simulations (Royal Society, 2008). For comparison, the ACCENT intercomparison used an estimate of 2088 ppb for CH_4 abundance in its simulations of ozone in 2030.

173. The key messages to be taken from the revised IIASA projections is that they illustrate the uncertainty surrounding predictions of future global and regional precursor emissions (including CH_4) and that this is the major uncertainty in predicting future surface ozone. Also, that surface ozone in Europe is impacted by emissions in the rest of the hemisphere. The optimistic (MFR) and pessimistic (SRES A2) emissions scenarios used in the ACCENT modelling remain a guide to the range in changes in European surface ozone that may occur in the future depending on which trajectory of precursor emissions is ultimately followed. If the CLE scenario is taken up worldwide as per the latest IIASA projections then ozone in Europe in the longer term, i.e. beyond 2030, is likely to decrease. On the other hand, if some countries worldwide do not follow the CLE course, then background ozone in the longer term, including over Europe, is likely to increase.

4.3 Impacts of climate change

174. Climate change will additionally influence future ozone levels through its impacts on many different natural processes. Identified processes include those related to:

a. Emission fluxes of ozone precursors (e.g. VOC from vegetation, NO_X from soil and lightning, CH_4 from wetlands and NO_X, CO and VOC from wild fires);

b. Atmospheric chemistry (e.g. via changes in temperature and atmospheric water vapour content);

c. Atmospheric dynamics (e.g. boundary layer ventilation, convective mixing, prevalence of anticyclonic blocking highs, precipitation, and stratosphere-troposphere exchange);

d. Loss of ozone by dry deposition to vegetation. This depends on soil moisture content and CO_2 concentrations. Under dry soil conditions the stomata of vegetation are almost completely closed because the plants are conserving water, so loss of ozone by dry deposition decreases and ozone levels increase. Similarly, if CO_2 levels rise the stomata can open less for the same level of photosynthesis. For example, Sanderson *et al.* (2007) calculated an increase in surface ozone over Europe of 2-6 ppb solely due to the effect of doubling CO_2 concentrations on plant stomata.

Climate change may also influence future anthropogenic emissions of ozone precursors indirectly through mitigation and adaptation responses, such as reduced energy demand for space heating in winter but greater energy demand for air conditioning in summer.

175. Although these (and other) processes and feedbacks of climate change acting on future ozone levels have been identified, only a small number of coupled climate chemistry models have been run and many known climate feedbacks have yet to be included.

176. Stevenson *et al.* (2005) performed a detailed investigation of climate change and variability on future tropospheric ozone using the IPCC IS92a emissions scenario ("business as usual") and comparing results for the period 1990-2030 for a fixed climate and for a projected climate from the Hadley Centre HadCM3 Model with a global mean surface warming of ~1 K. They specifically investigated the impact of changes in temperature (on reaction rates and changes in isoprene emissions), humidity (on the $O(^1D) + H_2O$ reaction), convection (affecting mixing and lightning NO_X emissions), precipitation (wet removal processes) and large-scale circulation (e.g. stratosphere-troposphere exchange). In this study the largest influence on the tropospheric ozone burden was the increase in humidity, reducing ozone lifetime and, together with enhanced oxidants, leading to a decrease in methane lifetime and influence on ozone production. The influence of increased stratospheric-tropospheric exchange of ozone was less important.

177. Other model studies, for example Collins *et al.* (2003), Sudo *et al.* (2003) and Zeng and Pyle (2003), have found a greater impact on tropospheric ozone from increased mid-latitude stratosphere-troposphere exchange of ozone. A recent interpretation of European free-troposphere ozone trends has suggested that European ozone may have been more strongly influenced by downward transport from the lower stratosphere of air more concentrated in ozone than was previously recognised (Ordóñez *et al.,* 2007). Climate change and reductions in anthropogenic emissions of ozone-depleting substances are also projected to contribute to an increase in stratospheric ozone which, combined with the projected increase in transport to the surface, will lead to an increase in background ozone independent of the impact of precursor emissions.

178. Ten of the 26 global models in the ACCENT intercomparison were used to produce year 2030 global ozone fields for the CLE scenario and a simulated 2030 climate (Table 4-1). The ensemble-mean difference in European seasonal surface ozone calculated with and without climate change is shown in Figure 4.5. Climate change is simulated to reduce the 2030 annual mean surface ozone averaged over Europe by 0.4 ppb, i.e. average surface ozone over Europe increases by 1.4 ppb for this emissions scenario rather than by the 1.8 ppb calculated for 2030 using a year 2000 climate (Dentener *et al.,* 2006). However, the magnitude of this projected impact of climate change on this time horizon is not significant in comparison with the magnitude of the spread in model simulations shown in the right-hand panels of Figure 4.5.

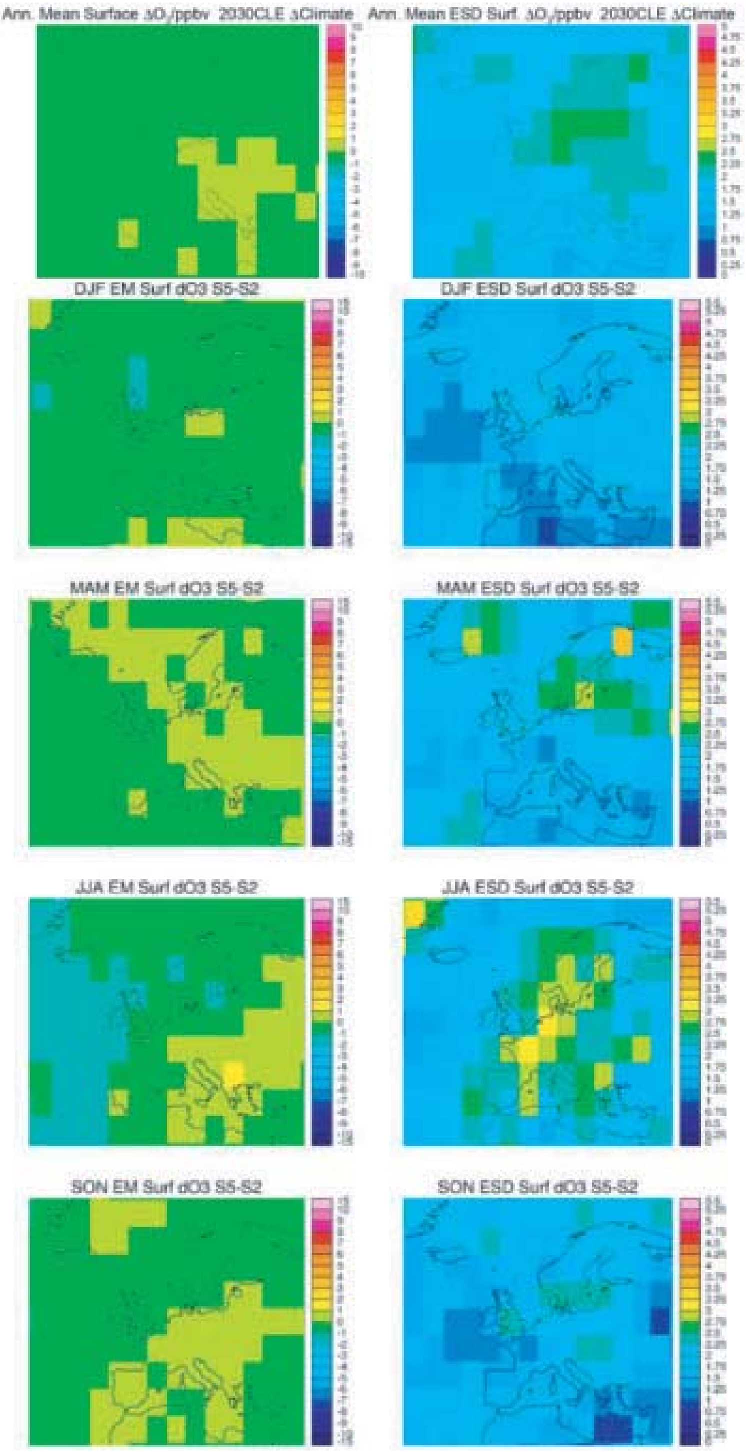


Figure 4.5 Left column: Ten-model ensemble-mean change in annual mean surface ozone (top row) and 3-month seasonal mean surface ozone (subsequent rows) for year 2030 for "central" emissions scenario CLE using a simulated 2030 climate in 2030 (model run S5) as compared with using 2000 climate in 2030 (model run S2), i.e. this figure illustrates the additional perturbation caused by modelled impacts of climate change between 2000 and 2030 to the simulated ozone changes for this period already illustrated in Figure 4.1. "DJF" refers to mean for the 3 months December, January, February, etc. Right column: Standard deviation of the corresponding 10-model ensemble simulations. Units are ppb (1 ppb $O_3 \equiv 2$ µg m^{-3} O_3). The models have been interpolated to a common resolution (5° x 5° horizontal); lowest model level depth is ~100 m. Source: Stevenson (pers. comm., 2008) from data of Dentener *et al.* (2006).

179. There is some evidence from this model intercomparison, and from other global model simulations including climate change effects (e.g. Royal Society, 2008), for a pattern of negative feedback of climate on surface ozone over the oceans, but positive feedback over polluted land surfaces (Figure 4.5). The negative climate feedback for lower altitude ozone in the models appears to be driven by the increased specific water vapour content of a warmer atmosphere increasing the rate of chemical ozone loss through the sequence of reactions:

$$O_3 + h\nu\ (\lambda < 310\ \text{nm}) \rightarrow O_2\ (a^1\Delta g) + O(^1D)$$

$$O(^1D) + H_2O \rightarrow 2OH$$

This loss process dominates particularly in low-NO_X environments, such as over the oceans, and offsets the increased rate of photochemical ozone production via increased temperature, and the increased influx of ozone via stratospheric-tropospheric exchange, although these latter processes are also important. The impact of climate change on surface ozone over polluted continents appears to have a positive net effect. In these areas increases in surface ozone may arise because of a decrease in formation of the NO_X reservoir compound peroxyacetyl nitrate (PAN) with temperature as shown, for example, for the US by Murazaki and Hess, 2006), or because of an increase in ozone production with increasing water vapour in a high-NO_X environment.

180. Many climate feedbacks, however, have generally not been included in these models. One important potential climate effect, for example, is the impact of temperature on emissions of VOC from vegetation. High concentrations of ozone were observed in northern Europe during the summer heat-wave of 2003 (Solberg *et al.,* 2005). Detailed analysis of measured species concentrations and chemical and meteorological modelling has indicated that the peaks in ozone experienced at the surface were due to regional-scale accumulation of pollutants entrained into the boundary layer each day (including, for example, from extensive forest fires caused by the drought and heat on the Iberian Peninsula (Solberg *et al.,* 2005)), coupled to strong additional stimulation of ozone production from the oxidation of biogenic VOC (Lee *et al.,* 2006) (see also section 2.9).

181. Two issues regarding future ozone arise from the experience of 2003. First, the extent of occurrence of meteorological conditions in the future which cause extended residence time of pollutants in the atmospheric boundary layer (regardless of changes in pollutant emissions), and second, the future response of biogenic VOC emissions to climate change and their impact on future ozone generation.

182. Meteorological conditions contributing to ozone episodes are summertime blocking high pressure systems which cause import of air westwards into the UK from the continent, together with low wind speed (i.e. stagnation) and high temperature. The evidence is currently equivocal regarding the future frequency of summertime blocking high pressure systems, but it is possible that such conditions will, if anything, slightly decrease in frequency (Barnes *et al.,* unpublished, cited in Royal Society, 2008), although the result should be treated with caution because of the difficulties inherent in the modelling and the large natural variability in blocking activity.

An analysis of future daily meteorological parameters simulated for southern UK by the Hadley Centre HadCM3 model driven by the IPCC IS92a (business-as-usual) scenario showed an increase in the number of days having both low wind speed (at least 1 hour with wind-speed <2 m s^{-1}) and high temperature (at least 1 hour with temperature >25 °C). The analysis suggested a tripling in frequency of such days by the 2030s compared with the 1990s (Department of Health, 2001). However, an update to this report (Department of Health, 2008) concluded that, although summer meteorological episodes in the UK will increase in frequency and intensity, episodic peak ozone concentrations should continue to fall over the period 2030 due to predicted falls in European emissions of ozone precursors.

183. There is considerably more confidence that periods with temperatures of the magnitude encountered in 2003 will rise both in frequency and duration across Europe as a consequence of climate change (Meehl and Tebaldi, 2004; Schar *et al.,* 2004; Stott *et al.,* 2004) than for future changes in the frequency of blocking high pressure systems. However, such a strong response of biogenic emissions to high temperature as observed in northern Europe in 2003 may not be observed in future decades or across all regions, for example, in the Mediterranean, where the plants are better adapted for water and temperature stress. Many other environment-related factors, in addition to temperature, influence biogenic VOC emissions and it is not clear what the net effect will be on biogenic emissions in particular regions as the environment changes. Example factors influencing biogenic VOC emission include:

- Increasing CO_2 levels. Experiments in chambers (Possell *et al.,* 2004, 2005) and in "free-air enrichment experiments" (Centritto *et al.,* 2004) have shown that isoprene emission rates from some species decrease with increasing CO_2 concentration. The degree of suppression caused by increasing CO_2 can be large, but there is disagreement on the net effect on total biogenic emissions from all species regionally and globally. Some model studies predict that the CO_2 suppression effect more or less offsets the increase in emissions predicted with increasing temperature in the future both globally (Lathiere *et al.,* unpublished, cited in Royal Society, 2008) and for Europe (Arneth *et al.,* 2008), whilst other studies predict substantial future increases in global isoprene (Guenther *et al.,* 2006).

- Changes to net primary productivity (NPP) and to NPP-CO_2 feedbacks through direct impact on plant health by ozone (Sitch *et al.,* 2007), or through indirect impact on stomatal conductance of changes in soil moisture (Sanderson *et al.,* 2007).

- Changes in individual species' tolerance as environmental conditions change.

- Changes in the species mix as environmental conditions change, i.e. successional changes in vegetation land-type.

- Response of species emissions to changes in insect herbivory as environmental conditions change.

184. Additionally, it is likely that anthropogenically-driven changes in land-use species mix (i.e. agricultural changes, whether directly policy driven, e.g. for biofuel crops, or otherwise) will have greater influence on the total biogenic VOC emission flux going forward than the net effect of climate change-driven influences on the plant emission processes *per se.*

185. Future changes in biogenic VOC emissions will occur alongside changes in anthropogenic precursor emissions (VOC and NO_X, in particular). Therefore, in addition to uncertainty on future biogenic emission rates, the extent to which any future increase in emissions of biogenic VOC increases future ozone will depend on the extent to which the future air-shed into which these emissions occur is VOC rather than NO_X sensitive, which will likely vary with region.

186. In summary, the net sign of the additional impact of climate change on surface ozone at the European and sub-European spatial scale, let alone the magnitude of the impact, is not known with confidence. Current levels of understanding suggest that the climate change feedback on annual average surface ozone over polluted land masses will be positive. However, the additional effects of climate change on precursor emissions, atmospheric chemistry, downward transport of stratospheric ozone, synoptic meteorology, etc., are currently anticipated to have a smaller influence on annual mean surface ozone levels on the 2030 time horizon than the effects of human-led changes in regional and global emissions of precursor gases already discussed. This does not exclude the possibility that climate change may have proportionally larger influence on regional peak summertime surface ozone through, for example, enhanced biogenic VOC emissions, drought-related depression of ozone dry deposition, increased incidence of wild fires, or extended air mass residence time in the boundary layer. Also, for time horizons longer than 2030, benefits to continental surface ozone levels accrued from precursor emissions controls may be reversed by climate change impacts and by consequences of land-use change.

4.4 Inter-annual variability

187. Inter-annual variability in European ozone is driven mainly by variations in the frequency of zonal air flows (westerlies) versus blocking high pressures (as mentioned above) which, in winters, is determined by the North Atlantic Oscillation. In summer this variation only occurs over northern Europe; summers with predominantly zonal flow have lower ozone and fewer episodes than summers with predominantly blocked flow. There is evidence that variations in the thermohaline circulation, leading to changes in the Atlantic (Ocean) Multidecadal Oscillation (AMO), may have been an important driver of multi-decadal variations in the summertime climate of western Europe (Sutton and Hodson, 2005). Such large-scale climatic/meteorological phenomena are major contributors to the considerable variability in year-on-year levels of ozone at different locations, as apparent in the historic time series of ozone measurements from UK network monitoring sites shown in Chapter 2, sections 2.6-2.8, and Chapter 3.

188. The magnitude of inter-annual variability is likely to be large compared with the net change in mean ozone over the next 2 to 3 decades caused by the other influences described above. This adds to the uncertainty in ascribing the various influences on ozone and emphasises the need for multi-year observational data and simulations.

4.5 Recommendations

189. Recommendations relevant to addressing question C include:

- Continued development, validation and intercomparison of models that provide bidirectional coupling between climate and atmospheric chemistry, and at higher spatial resolution. This will include continued refinement of global and regional precursor emission projections.

- Narrowing the uncertainties associated with emissions inventories for the UK and the rest of Europe for isoprene, in particular, and other relevant biogenic volatile organic compounds (BVOCs). This will include increased accuracy and precision in estimates of BVOC emissions by plant species, in current land cover data for plant species distributions and in process-based algorithms describing environmental influences on BVOC emissions (e.g. seasonal, diurnal, photosynthetically active radiation (PAR) intensity and temperature).

- Development of potential future scenarios of changes in land cover in Europe, particularly in respect of potential changes in agricultural and natural species distributions (driven, for example, by biofuel crop policies).

Chapter 5

Likely future trends in urban ozone concentrations

Question D: *What are the likely future trends in urban ozone concentrations over the next two decades and what is driving them?*

Short answer to question D

190. Urban ozone concentrations are expected to rise over the next two decades and to tend towards the concentrations found in the rural areas that surround them. These increases in urban ozone concentrations are largely driven by vehicle emission controls that have brought about a reduction in nitrogen oxides (NO_X) emissions in urban areas. Road traffic NO_X emissions have previously depressed urban ozone levels and although this scavenging effect is being diminished by pollution controls, many urban areas in the UK are still expected to have lower ozone concentrations in 2020 than those in the surrounding rural areas. Urban ozone concentrations will also respond to the changes occurring to ozone in the surrounding rural areas, largely driven by changes on the hemispheric/global scale. Depending on the strength and sign of these trends, these could also cause increases in urban ozone, which will be in addition to the NO_X-scavenging effect.

Detailed answer to question D

191. To answer this question on future urban ozone levels in the United Kingdom, we need to understand what have been the main lessons learnt from the large expansion that has taken place in urban ozone monitoring during the last decade. As the response to question A and supporting evidence in Chapter 2 indicated, almost all of the 49 urban centre, urban background, roadside and kerbside Automatic Urban and Rural Network (AURN) sites have shown upwards trends in the annual mean of the daily maximum 8-hour mean ozone concentrations over the period since monitoring began until 2003. Broadly speaking, these upwards trends are greatest for the most polluted sites and lowest for the least polluted sites. This behaviour has been explained by the progressive diminution of the NO_X-driven scavenging of ozone following the implementation of three-way exhaust gas catalysts on petrol-engined motor vehicles mandated by the European Commission.

192. What happens to future urban ozone levels at a given location can be considered to depend on the nitrogen NO_X emission density (strictly NO_X concentrations) in the immediate surrounding area. Once NO_X emission densities fall much below 10 tonnes per 1 km x 1 km grid square per year, then NO_X scavenging ceases to act as the dominant sink for ozone, compared with dry deposition, and the urban ozone concentrations approach those of the surrounding rural area. Under these conditions, it becomes increasingly unlikely that further NO_X emission controls will bring about further ozone increases. For grid squares above this threshold, further NO_X emission reductions would still bring about further urban ozone increases. The further above this threshold

a grid square is then the greater the potential for ozone increases. Whether urban ozone levels continue to increase in the grid squares with emission densities above the threshold over the next decade will depend on the extent of future road traffic NO_X emission reductions. Experience in London has however shown that it is also important to understand the fraction of the NO_X that is emitted as NO_2 as primary NO_2 emissions are a source of oxidant, and hence ozone, in urban areas.

193. Modelling using the process-based ADMS-Urban model and Ozone Source-Receptor Model (OSRM) provides further confirmation of the decrease in the local NO_X-scavenging effect and increases in ozone concentrations (i.e., a decrease in the urban ozone decrement). For example, calculations using the ADMS-Urban model gave annual mean ozone concentrations in 2001 which varied from 40 µg m^{-3} on the outskirts of London to less than 20 µg m^{-3} in central London. In 2020, using projected changes in London emissions and trends in oxides of nitrogen but with no change in meteorology, annual mean concentrations were predicted to have risen to more than 52 µg m^{-3} on the outskirts of London. Furthermore, the two models gave comparable percentage decreases for a number of ozone metrics as a function of the modelled NO_X concentration and these decrements were also comparable to those derived from the empirically-based approach used in the UK Pollution Climate Model (PCM).

194. Urban ozone monitoring network data from the London Air Quality Network (LAQN) during the intense photochemical ozone episode in August 2003 showed no evidence of elevated ozone concentrations within the London conurbation compared with rural levels. Intense urban-scale ozone production does not appear to occur in London as it does at lower latitudes, for instance, in Atlanta, Houston and Los Angeles in the USA. It appears that urban ozone levels are always lower than the levels in the surrounding rural areas and that this deficit is driven by depletion of ozone by NO_X emissions within the urban areas. This urban-rural relationship is expected to continue in the future.

195. Urban ozone concentrations will therefore also respond to the changes occurring to ozone in the surrounding rural areas. Model results obtained using the OSRM showed that, depending on the strength of the ozone trends on the hemispheric/global scale, these changes could also cause increases of a similar magnitude in urban ozone to the changes in the NO_X-scavenging effect.

196. Analysis of the $NO–NO_2–O_3–O_X$ relationships for a number of years and urban locations has revealed the presence of anomalously elevated regional background oxidant contributions for 1999 (see Figures 2.1 and 2.21 and 2.26 in Chapter 2). These elevated background oxidant levels appear to be associated with the 20-year maximum monthly mean background ozone concentration of 51.8 ppb (~103.6 µg m^{-3}) reported for Mace Head, Ireland, during March 1999 (see Figure 3.2). Indeed, this ozone anomaly is present in the records of 55 sites (located largely in rural locations) in the EMEP ozone monitoring network during the winter and early spring of 1998-1999. The origin of this anomaly appears to have been tropical and boreal biomass burning elsewhere in the northern hemisphere. Hemispheric-scale events have the capacity therefore to influence regional and hence urban ozone concentrations and are likely to contribute to year-on-year variability in the future.

197. On this basis, it is concluded that the main drivers for future urban ozone are likely to be:

- Local-scale NO_X emissions and the extent to which the diminution of NO_X scavenging of ozone due to the control of road traffic NO_X emissions continues in the future;
- Regional ozone levels, and the balance between any rise in hemispheric background ozone and any decline due to the control of regional-scale ozone precursor emissions; and
- The occurrence of hemispheric-scale events such as biomass burning.

198. Any changes to UK ozone arising from other aspects of climate change (temperature, humidity, etc.) are most likely to affect urban ozone through changes to regional ozone concentrations. Given the absence of local photochemical production of ozone in London during the 2003 episodes, it is unlikely that photochemical production of ozone in urban areas will become more significant in future when the frequency of such episodes is expected to increase.

Supporting evidence for question D

5.1 Overview

199. As shown in Chapter 2, urban ozone levels are lower than those in the rural areas surrounding them because of higher NO_X emissions, mostly from low-level vehicular traffic sources, which act as local ozone sinks. It is then straightforward to understand how measures to control vehicular NO_X emissions have led to a diminution in these local ozone sinks and hence to rising urban ozone levels. This difference between the values of the ozone concentration or metric at the urban location and the corresponding quantity at a surrounding rural site (taken to be representative of the regional ozone field) defines an urban ozone decrement.

200. The issue is then the extent to which rising ozone levels will continue into the future. In this supporting evidence, information is presented on diurnal cycles in ozone, ozone sinks and on projections of future low-level NO_X emissions. An analysis has been undertaken to determine the significance of the local NO_X-scavenging effect in 2020 and model calculations are also presented for the Greater London area for current and future years using the ADMS-Urban model.

201. As Chapters 2, 3 and 4 have indicated, in addition to a reduction in the NO_X-scavenging effect, there are two other main drivers determining UK ozone concentrations and their frequency distribution:

a. Regional controls on NO_X and VOC emissions at the European level, reducing peak ozone concentrations (i.e., reduced photochemical ozone production).

b. An increasing background concentration arising from global changes in atmospheric composition and hemispheric circulation.

202. The Ozone Source-Receptor Model (OSRM) has been used to investigate the significance of these other drivers on future UK urban ozone concentrations compared to the NO_X-scavenging effect.

203. A comparison of the urban ozone decrements derived from UK process-based models (OSRM and ADMS-Urban) and empirical approaches is then made. Further details are provided in a technical annex to this chapter (Annex 3). The supporting information concludes with a summary of recent European activities to model urban ozone.

5.2 Photochemical production of ozone in urban areas

204. There has always been a concern as to whether there may be photochemical ozone formation hot spots in UK urban areas. Urban ozone monitoring network data from the London Air Quality Network (LAQN) during the intense photochemical ozone episode in August 2003 showed no evidence of elevated ozone concentrations within the London conurbation compared with rural levels, as illustrated in Figure 5.1. This also appeared to be the case on a monthly, highest daily and highest hourly basis during August 2003, as shown earlier in Figure 2.35. Strictly, the comparison should be based on a rural measurement made some hours beforehand to allow for the time to travel to the urban site but such comparisons would also need to take account of the diurnal cycles observed in ozone. While urban-scale ozone production cannot be completely excluded in London (and hence the UK), it is unlikely to be as intense as it is at lower latitudes, for instance, in Atlanta, Houston and Los Angeles in the USA. On this basis, it appears that urban ozone levels are always lower than the levels in the surrounding rural areas and that this deficit is driven by depletion of ozone by NO_X emissions within the urban areas. The bulk of the ozone monitored in urban areas therefore had its origins in the surrounding rural areas and was the result of regional scale ozone formation. This urban-rural relationship is expected to continue in the future and regional-scale ozone levels are expected to continue to control future urban levels.

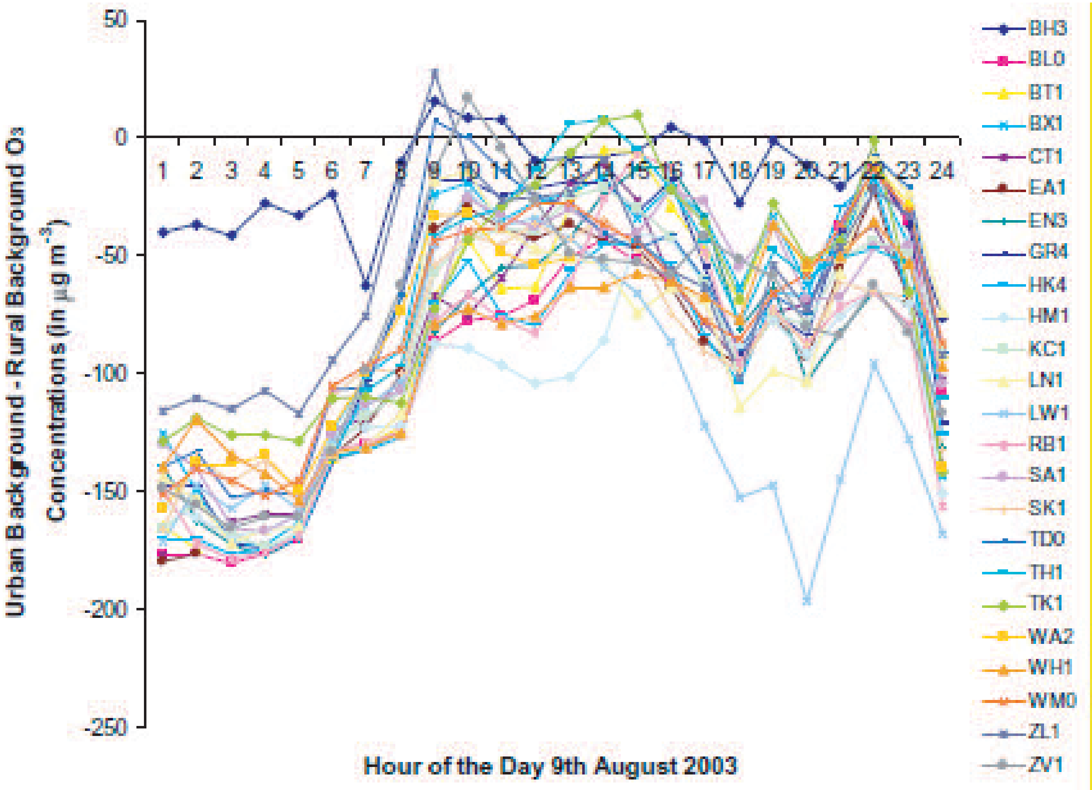


Figure 5.1 The difference in the hourly ozone concentrations measured at 24 sites in the London Air Quality Network and the rural background concentration (in µg m^{-3}) for the 9th August 2003.

5.3 The NO$_X$-scavenging driver

5.3.1 Ozone diurnal cycles and sinks

205. At almost all UK ozone monitoring sites, both rural and urban, ozone exhibits a characteristic diurnal cycle. Figure 5.2 illustrates these diurnal cycles for a pair of sites, a rural site at Harwell, Oxfordshire, and an urban background site, London Bloomsbury. Average diurnal cycles are presented for two years, 1995 and 2006, two strongly photochemically-active years, widely spaced in time. The average diurnal cycles for both sites and years are clearly evident in Figure 5.2 and show highest average concentrations in the mid-afternoon and lowest levels in the early morning.

206. Garland and Derwent (1979) have shown that the average diurnal cycle at the rural monitoring site is controlled by the diurnal cycle in the atmospheric boundary layer depth and the strength of turbulent mixing processes. The atmospheric boundary layer is generally well mixed during the mid-afternoon and surface ozone monitoring data are representative of those levels present in a considerable depth of the atmosphere. At night, boundary layer depths are generally considerably smaller and ozone levels close to the surface become depleted by surface deposition and local NO$_X$ sources because they are not efficiently replenished by turbulent transport and exchange. The average diurnal curves at the rural site have been shifted to higher concentration during the early morning and evening, reflecting the diminution in local NO$_X$ sources due to vehicle emission controls. Nocturnal depletion is still evident in 2006 because surface deposition under the shallow night-time stable

layer is the dominant night-time sink for ozone. Diminution of NO_x scavenging between 1995 and 2006 has led to a reduction in the urban ozone deficit in London and urban levels have clearly risen towards the rural levels immediately outside of London.

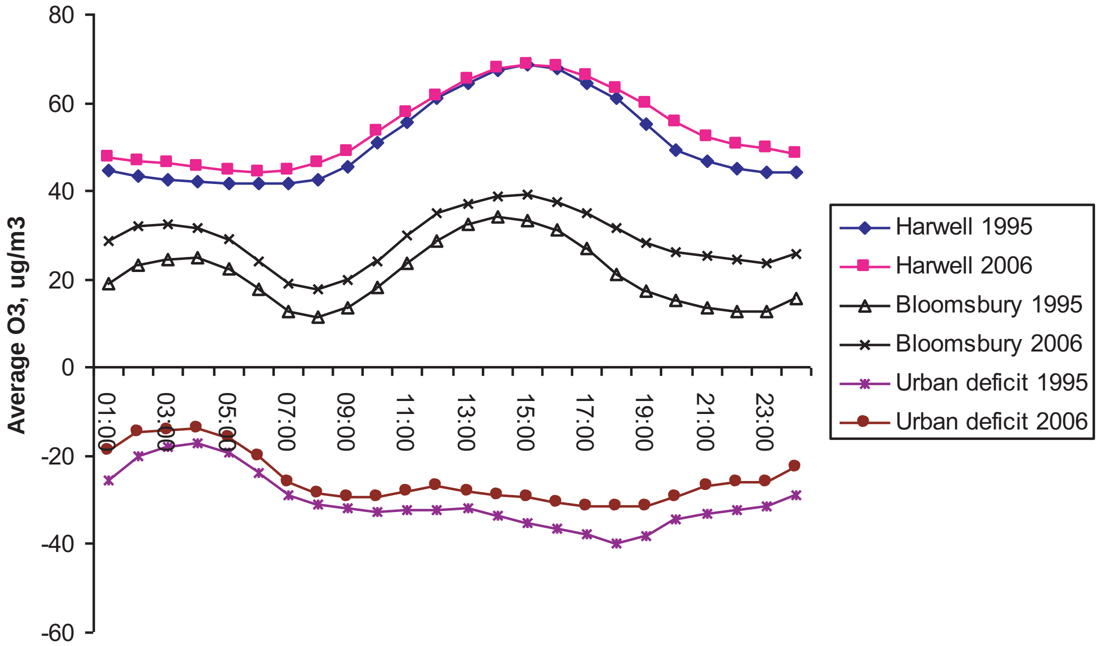


Figure 5.2 Average diurnal cycles in ozone observed at a rural and a London urban background site in 1995 and 2006, together with the London urban deficit in both years.

207. The average diurnal cycle at the London urban background site (Figure 5.2) exhibits a similar mid-afternoon maximum to that observed at the rural site. The apparent shift in the time of the mid-afternoon peak is not significant. However, London urban background levels are clearly depressed relative to rural levels and there is a London urban decrement of about 30-40 µg m^{-3} during the mid-afternoon, due largely to London-wide and local NO_x emissions. Over the period from 1995 to 2006, NO_x emission controls have reduced the strength of NO_x scavenging, reducing the London urban ozone decrement by about 10 µg m^{-3}, that is by about one quarter.

208. It is evident from Figure 5.2 that ozone concentrations at the London urban background site have been rising from 1995 to 2006 due to a diminution in NO_x scavenging because of NO_x emission reductions. London urban background ozone concentrations have been rising relative to rural levels and urban ozone decrements have been falling. Similar behaviour has been observed in most urban areas. Further details of urban ozone trends were given in the response to question A (see Chapter 2).

5.3.2 Changes in the NO_x-scavenging effect

209. Over the period from 1995 to 2006, it would appear that the NO_x sink for ozone at the London urban background site has been reduced by about one quarter due to NO_x emission controls. As further NO_x controls are enacted and promulgated, the average diurnal curve will rise further until it meets the average diurnal curve for the rural Harwell site and the London urban ozone

decrement is eroded away. At that point, NO_X depletion will have become of negligible importance in comparison with surface depletion and the difference between rural and urban areas, at least as far as ozone sinks are concerned, will have disappeared.

210. Turbulent transport to a vehicular NO_X-driven ozone sink or to a vegetated surface become of roughly similar magnitudes for a NO_X emission density of about 10 tonne km^{-2} yr^{-1}, making reasonable assumptions of deposition velocities (5 mm s^{-1}), aerodynamic resistances (0.1 cm s^{-1}) and nocturnal atmospheric boundary layer depths (100 metres). Support for this threshold can be gained from the UK urban ozone decrements calculated using process and empirical models which are described later in this chapter. Hence, this value can be used as an indicator of the relative importance of NO_X-driven and surface deposition sinks for ozone. Strictly, it is the NO–NO_2–NO_X concentrations that determine the threshold and this will vary throughout the year and from site to site.

211. For the purposes of this analysis, the threshold of 10 tonne km^{-2} yr^{-1} has been used to identify those urban areas in which local vehicular traffic sources will dominate over surface deposition as the major ozone sink. In these locations, urban ozone concentrations are expected to be depressed relative to rural levels and further NO_X emission controls will lead to increasing urban ozone concentrations. For urban areas with local NO_X emission densities below this level, NO_X depletion is expected to be relatively unimportant and the impact of further NO_X emission controls is likely to be small.

212. Figure 5.3 shows the spatial distribution of the low-level NO_X emissions across the United Kingdom in 2004 (left-hand panel) and 2020 (right-hand panel), highlighting the areas with emission densities in excess of 10 tonne km^{-2} yr^{-1}. The analysis used the latest emission estimates available at the time from the National Atmospheric Emissions Inventory (NAEI) for these years. From these figures, it is apparent the area with emissions in excess of 10 tonne km^{-2} yr^{-1} shrinks between 2004 and 2020 in response to further NO_X emission reductions. On this basis, it is likely that the upward trends in annual mean ozone concentrations in the urban areas with NO_X emissions below this threshold will cease relative to rural areas immediately surrounding them. Furthermore, there are a number of rural areas containing important road links that will also shift below this critical emission density, carrying the implication that ozone concentrations will cease rising in these grid squares relative to the rural areas. When this happens, the trends in rural and urban areas will become the same and the future prospects for both will be determined by those of the rural areas. The influence of the diminution of NO_X scavenging will drop out, leaving European pollution controls and the growth in northern hemisphere ozone background as the main controlling influences on ozone in the 'white' areas of the right-hand panel of Figure 5.3.

213. Table 5-1 presents details of the urban locations where ozone levels are depleted relative to the rural areas immediately surrounding them in 2004 and 2020, using the NO_X emission threshold of 10 tonne km^{-2} yr^{-1}. The prospects are that significant areas of the UK, and significant populations, will still have depleted ozone levels in 2020, despite the NO_X emissions reductions from vehicular traffic that are projected to occur between 2004 and 2020.

Urban ozone levels will continue to rise in these areas up to 2020 and beyond. This conclusion is confirmed by the calculations of process-based models in subsequent sections of this chapter.

214. The further above the 10 tonne km^{-2} yr^{-1} threshold a grid square is then the greater the potential for ozone increases. Whether urban ozone levels continue to increase over the next decade in the grid squares with emission densities above the threshold, will depend on further road traffic NO_X emission reductions. This in turns depends crucially on the extent to which diesel NO_X exhaust emission controls are introduced through EU legislation and penetrate through the respective vehicle fleets. Experience in London has shown how important it will be to understand the fraction of the NO_X that is emitted as NO_2 with these new technology diesel vehicles (AQEG, 2007a), as primary NO_2 emissions are a source of oxidant and hence ozone in urban areas (Clapp and Jenkin, 2001; Jenkin, 2004).

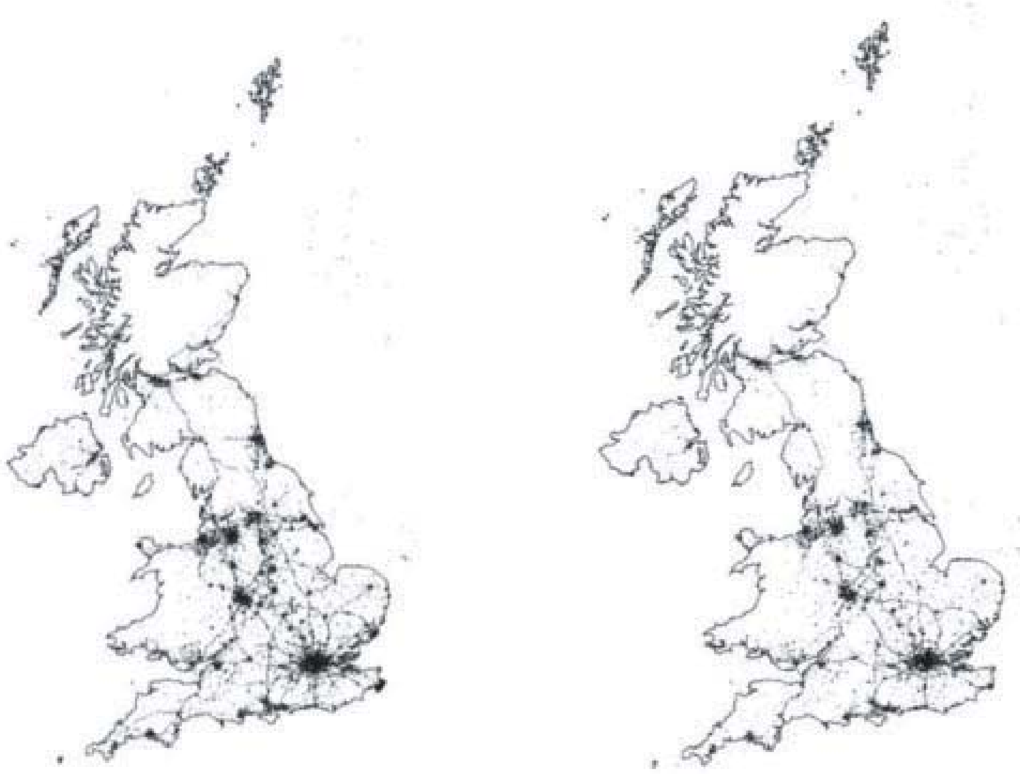


Figure 5.3 Maps showing areas (shown in black) where the NO_X emission density exceeded the threshold of 10 tonne km^{-2} yr^{-1} across the UK in 2004 (left-hand panel) and is projected to exceed the threshold in 2020 (right-hand panel).

Table 5-1 Populated areas of the United Kingdom and likely occurrence of depleted ozone levels relative to the rural areas immediately surrounding them in 2004 and 2020.
The percentage of the area with depleted ozone levels and the percentage of the population resident in these depleted areas have been estimated using the 10 tonne km^{-2} yr^{-1} threshold emission density, see text. Data for the regions in the lower part of the table exclude any urban areas in the regions listed separately in the table.

Urban area/ region	Total area, km^2	Percentage of area with depleted ozone, (km^2)		Total population	Percentage of population in area with depleted ozone	
		2004	2020		2004	2020
Greater London	1632	92.9%	82.7%	7784707	98.8%	94.6%
West Midlands	594	96.3%	86.7%	2083891	98.9%	92.5%
Greater Manchester	557	96.2%	85.8%	1846479	98.9%	91.8%
West Yorkshire	363	90.6%	77.7%	1150737	96.0%	86.1%
Tyneside	217	97.2%	87.6%	714326	99.1%	93.1%
Liverpool	185	93.5%	84.9%	697197	98.3%	92.5%
Sheffield	165	93.9%	83.0%	521984	97.5%	89.8%
Nottingham	169	94.1%	82.8%	558935	98.5%	90.4%
Bristol	142	91.5%	78.9%	488798	98.2%	85.7%
Brighton/Worthing /Littlehampton	98	83.7%	66.3%	387431	91.9%	81.7%
Leicester	102	94.1%	77.5%	374314	97.5%	84.7%
Portsmouth	93	83.9%	75.3%	355516	92.5%	84.6%
Teesside	113	84.1%	68.1%	301290	98.0%	82.6%
The Potteries	91	92.3%	85.7%	266188	96.2%	92.2%
Bournemouth	113	85.0%	66.4%	338103	91.6%	75.9%
Reading/Wokingham	97	86.6%	74.2%	305786	94.4%	83.4%
Coventry/Bedworth	76	94.7%	84.2%	277475	98.8%	91.5%
Kingston upon Hull	80	88.8%	75.0%	260201	94.3%	81.0%
Southampton	78	89.7%	82.1%	265011	94.0%	89.6%
Birkenhead	88	88.6%	75.0%	265019	98.5%	88.3%
Southend	64	85.9%	70.3%	217874	95.3%	84.7%
Blackpool	63	93.7%	77.8%	212909	99.1%	90.8%
Preston	58	93.1%	81.0%	180687	96.7%	88.3%
Glasgow	366	86.3%	73.2%	1083323	95.2%	85.4%
Edinburgh	118	85.6%	65.3%	429071	94.7%	84.0%
Cardiff	74	93.2%	81.1%	264259	97.6%	89.1%
Swansea	85	80.0%	60.0%	190228	92.4%	69.7%
Belfast Metropolitan	196	75.5%	58.2%	515484	85.8%	68.7%
Eastern	19137	11.2%	7.3%	4909876	60.6%	47.9%
South West	23546	6.4%	4.0%	4039462	49.0%	38.5%
South East	18664	14.0%	9.8%	6160629	60.4%	48.8%
East Midlands	15492	10.5%	6.6%	3261327	56.9%	42.1%
North West & Merseyside	13743	12.0%	8.2%	3470622	68.6%	52.5%
Yorkshire & Humberside	14792	9.4%	6.6%	3003872	58.6%	44.9%
West Midlands	12192	10.9%	7.1%	2624016	60.9%	45.9%
North East	8288	8.0%	4.9%	1443912	65.2%	51.1%
Central Scotland	9352	8.7%	5.8%	1883014	58.7%	45.2%
North East Scotland	18625	2.1%	1.1%	976022	55.7%	46.4%
Highland	39170	0.1%	0.1%	341329	19.8%	14.3%
Scottish Borders	11182	1.1%	0.8%	250529	38.2%	29.9%
South Wales	12236	5.0%	3.5%	1698082	48.1%	39.8%
North Wales	8372	3.1%	1.9%	702506	39.6%	30.0%
Northern Ireland	13969	2.1%	0.9%	1149153	24.4%	12.2%
Total	244837	8.6%	6.1%	58251571	72.4%	61.7%

5.3.3 Modelling of the Greater London area

215. Williams *et al.* (2006) have used the ADMS-Urban model to calculate ozone concentrations for the Greater London area for the-then current year (2001) and for future (2010 and 2020) years. The model runs used traffic flow and emissions data from the 2001 London Atmospheric Emissions Inventory,[1] together with meteorological data from Heathrow and background data from rural monitoring sites around London. The modelling of future years allowed for changes to traffic flows, pollutant emission rates and background concentrations.

216. Background NO_X concentrations for 2010 and 2020 were derived by multiplying the 2001 data by factors of 0.68 and 0.53 respectively, based on trends in national emission projections. To obtain future concentrations of NO_2, a best-fit curve was derived to relate NO_X and NO_2 concentrations in 2001 and this was applied to the projected 2010 and 2020 NO_X concentrations. Future 'background' ozone concentrations were calculated by assuming conservation of total oxidant ($O_X = O_3 + NO_2$), which led to some increases in background ozone but no allowance was made for changes in global or hemispheric ozone concentrations.

217. The model uses the Generalised Reaction Set (GRS) chemical mechanism. Comparison with measurements has shown good performance for annual means but some underestimate (typically 10%) in peak concentrations. Table 5-2 shows the annual average ozone concentration predicted at each of the AURN monitoring sites for 2001, 2010 and 2020. In 2001, annual mean ozone concentrations were calculated to vary from 40 μg m^{-3} in the outskirts of London to less than 20 μg m^{-3} in central London. In future years, ozone concentrations in London were predicted to increase and will be greater than 52 μg m^{-3} by 2020 in the outskirts of London. Concentrations will be greater than approximately 20 μg m^{-3} along major roads in central London.

Table 5-2 Measured and modelled annual average ozone concentrations (μg m^{-3}) for AURN Sites in London for 2001, 2010 and 2020. The model results are from ADMS-Urban calculations.

Site	2001 Observed	2001 Model	2010 Model	2020 Model
Marylebone Road	14	11	12	15
Bloomsbury	23	21	26	28
Hackney	29	27	32	35
London Southwark	29	26	30	32
North Kensington	34	28	32	34
Wandsworth	27	25	30	34
Eltham	37	36	41	44
Bexley	38	34	39	42
Brent	37	37	42	45
Hillingdon	26	24	32	36
Teddington	44	40	45	47
Average	31	28	33	36

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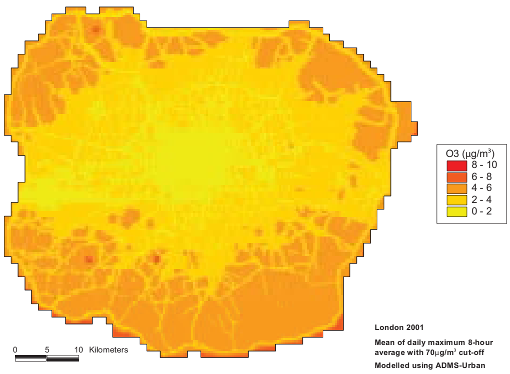
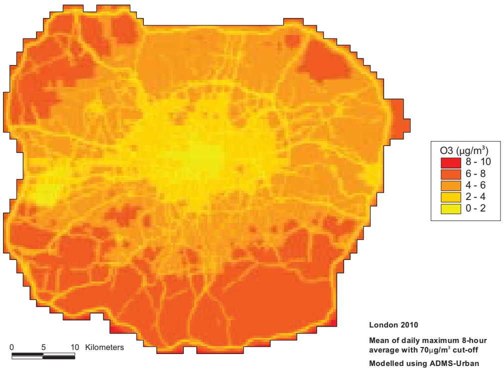
[1] The latest available emission inventory at the time of the work.

218. As an example, Figure 5.4 shows the annual mean of the daily maximum 8-hour rolling average concentrations with a cut-off of 70 µg m^{-3} for 2001, as a current year, 2010 and 2020. The increase in this metric for these base case runs is clear and is driven primarily by the reduction in local NO_X scavenging, the increase in background ozone resulting from the reduction in primary NO_2 and conservation of oxidant being small.

(a) 2001



(b) 2010



(c) 2020

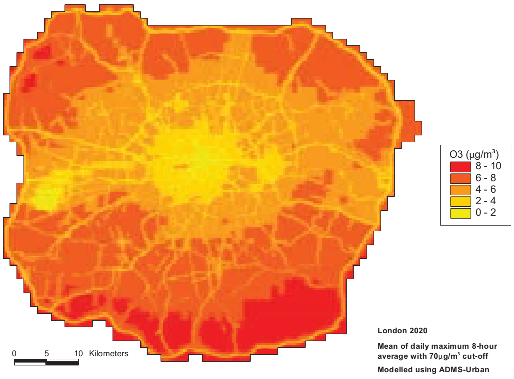


Figure 5.4 Maps of the annual mean of the daily maximum 8-hour average ozone concentrations with a cut-off of 70 µg m^{-3} (in µg m^{-3}) calculated for London for 2001 and 2010 and 2020 base cases, using ADMS-Urban.

219. In the supporting evidence to question A, Figure 2.33 presented the variation in three ozone metrics, as derived by the empirical mapping methods used in the Pollution Climate Model (PCM), along a transect from west to east across London for the years 1995, 2003 and 2005. Figure 5.5 illustrates the ozone metrics derived along the same transect (see Figure 2.34 for a map of the transect) for 2003 using the ADMS-Urban model. Figure 5.5 also includes the observed ozone metrics and those derived for 2003 using the empirical approaches. The figure again provides an illustration of the urban decrement and indicates a broad level of agreement between the two approaches. The ozone metrics are lowest (and thus the urban decrement is highest) in regions of highest NO_X emission density in central London.

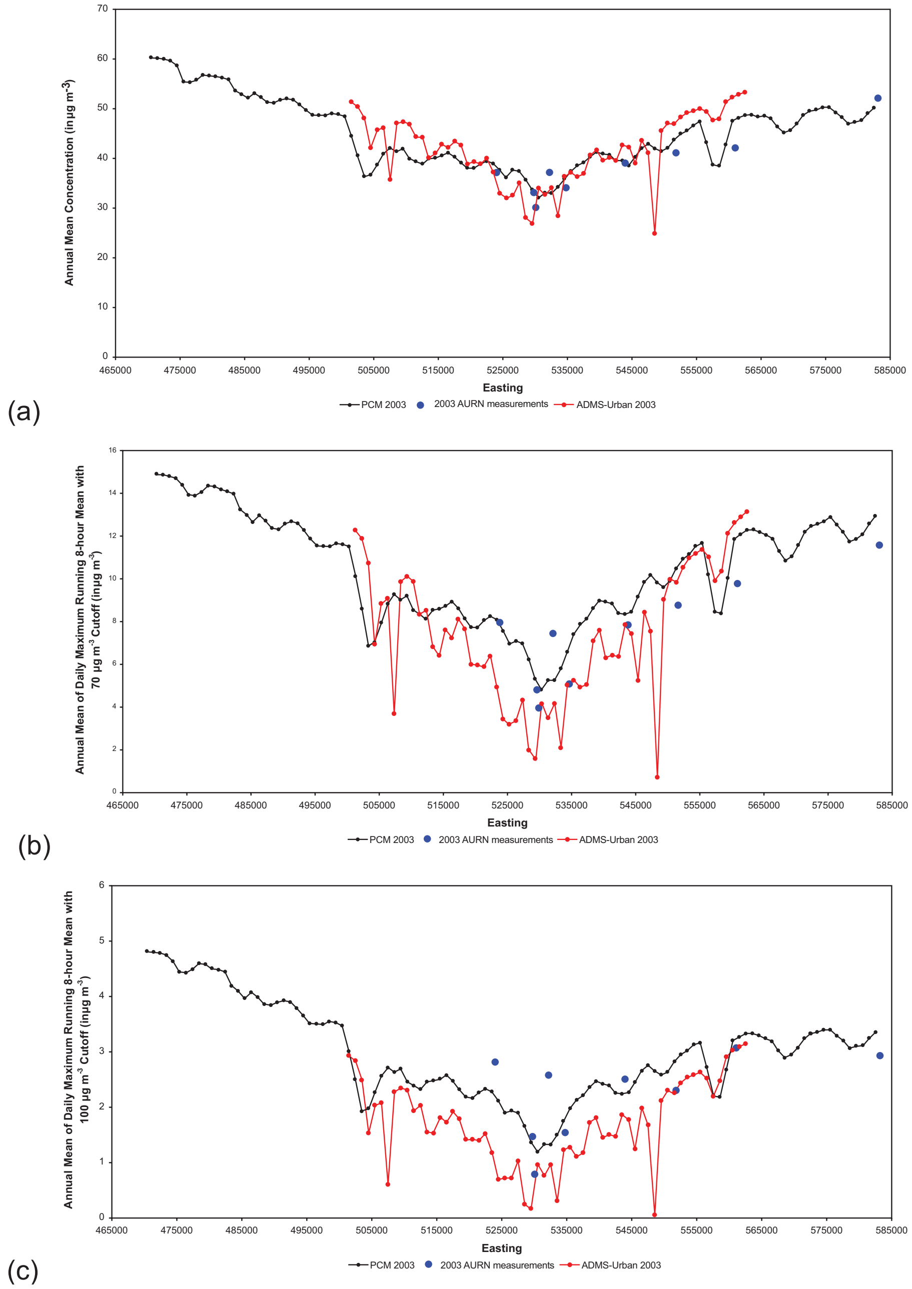


Figure 5.5 Transects of modelled (ADMS-Urban (red) and Pollution Climate Model (black)) and observed ozone metrics (blue) across London for 2003: (a) Annual mean ozone concentration; (b) Annual mean of the daily maximum of the running 8-hour mean ozone concentration with a 70 µg m^{-3} cut-off; and (c) Annual mean of the daily maximum of the running 8-hour mean ozone concentration with a 100 µg m^{-3} cut-off.

5.4 The regional and global determinants of ozone

220. In addition to a reduction in the NO_X-scavenging effect, two other drivers determine ozone concentrations in the UK: (a) regional controls on NO_X and VOC emissions at the European level, reducing peak ozone concentrations; and (b) an increasing background concentration arising from global changes in atmospheric composition and hemispheric circulation. The Ozone Source-Receptor Model (OSRM) has been used to investigate the significance of these other drivers on future UK urban ozone concentrations compared to the NO_X-scavenging effect (Hayman *et al.,* 2006, 2008a, 2008b).

221. Two sets of OSRM model runs were undertaken for 41 receptor sites – representing the locations of 20 rural, 10 London and 11 other urban background ozone monitoring sites – for the years 1990, 1995, 2000, 2003, 2005, 2010, 2015 and 2020. Year-specific emission inventories based on the 2004 NAEI and EMEP inventories were used in these calculations and the same meteorology – that of 2003 – was used to avoid complications arising from year-to-year variations in meteorology. Two sets of model runs were performed:

    a. The first set were initialised using daily concentration fields of ozone and key trace species derived from the STOCHEM model and modified for ozone to take account of the trend in background concentrations derived for a business-as-usual scenario with climate change (Hayman *et al.,* 2006a).

    b. The second set of runs, for the same years, used the same initial concentrations as used for the 2003 model run to illustrate the effect of changing atmospheric composition.

    All other model parameters were set to those used in the ozone modelling runs undertaken for the Review of the Air Quality Strategy (Hayman *et al.,* 2006a).

222. The hourly ozone concentrations were processed to derive the ozone metrics of interest. A surface conversion algorithm using site-specific hourly parameters has been developed to improve the performance of the model for urban ozone (Hayman *et al.,* 2008a). The difference between the 'unconverted' and 'converted' outputs is a measure of the ozone decrement at the location and reflects *inter alia* the effects of the local NO_X emissions.

223. The results for London Bloomsbury, London Bexley and Rochester Stoke, three closely related sites in urban, suburban and rural locations respectively, have been chosen to illustrate the model results. Figure 5.6 shows the values of two metrics

    a. The annual mean of the maximum daily running 8-hour average ozone concentration (upper panel – metric A); and

    b. The annual mean of the difference between the maximum daily running 8-hour average ozone concentration and a 100 µg m^{-3} (or 50 ppb) cut-off (lower panel – metric B).

Both metrics were calculated for the two sets of model runs. The runs with changing atmospheric composition are shown in the figure as solid lines and the runs with fixed composition use dotted lines. It is clear that the changes in atmospheric composition assumed are calculated to have a significant effect on the values of the metrics.

224. The two metrics shown in Figure 5.6 were selected because of their relevance to human health and also because of their differing sensitivity to peak ozone concentrations. As discussed in the supporting evidence to question A (Chapter 2), emission controls on NO_X and VOCs across Europe have largely reduced peak ozone concentrations and this can be seen in the downward trend at Rochester Stoke for metric B (lower panel of Figure 5.6) from 1990 to 2005. Metric A is more sensitive to changes in background ozone concentrations. The effect of the assumed changes in atmospheric composition was to increase future ozone concentrations and hence the values of metric A by ~3-5 $\mu g\ m^{-3}$ in 2020 compared to the constant composition case (see upper panel of Figure 5.6. Conversely, years prior to 2003 show lower values of metric A (a reduction of ~3-5 $\mu g\ m^{-3}$ in 1990), for variable composition. 2003, the year used for the constant initial composition case, is in the middle of the time period considered.

225. Figure 5.6 also shows the decreasing values of the two ozone metrics in moving from Rochester Stoke to the suburban London Bexley to the urban London Bloomsbury sites, consistent with the increase in NO_X emission densities and hence local NO_X-scavenging effect. To illustrate the change in the NO_X-scavenging effect more clearly, the modelled percentage ozone decrements, defined in paragraph 222, are shown in Figure 5.7 for the London Bexley and Bloomsbury sites for the two metrics.

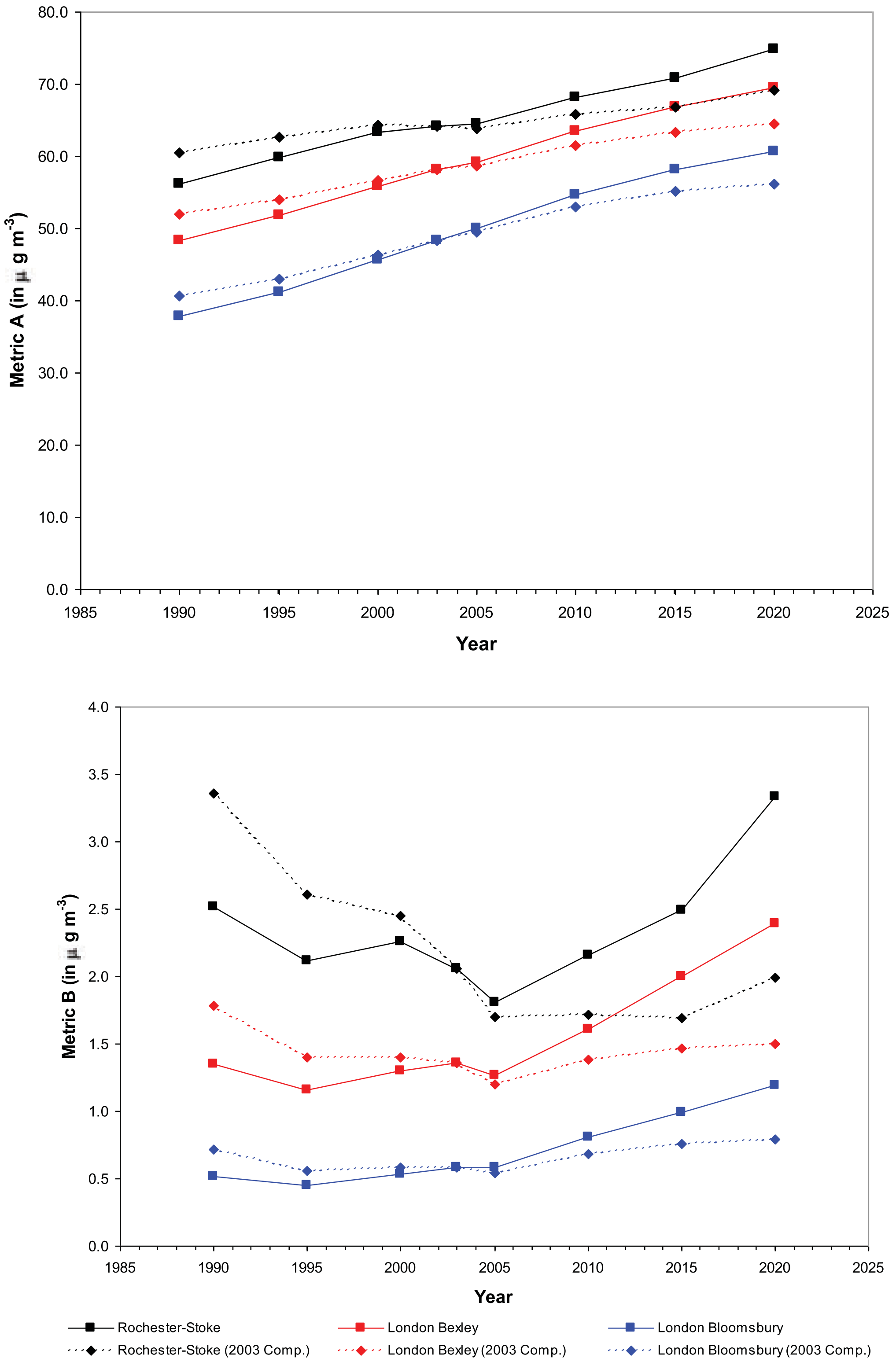


Figure 5.6 Annual mean of the maximum daily running 8-hour average ozone concentration (metric A, upper panel) and annual mean of the difference between the maximum daily running 8-hour average ozone concentration and a 100 µg m^{-3} (50 ppb) cut-off (metric B, lower panel) as calculated using the OSRM for changing (squares/solid lines) and constant (diamonds/dashed lines) atmospheric composition for selected years between 1990-2020 using 2003 meteorology for the three sites: Rochester Stoke (black), London Bexley (red) and London Bloomsbury (blue).

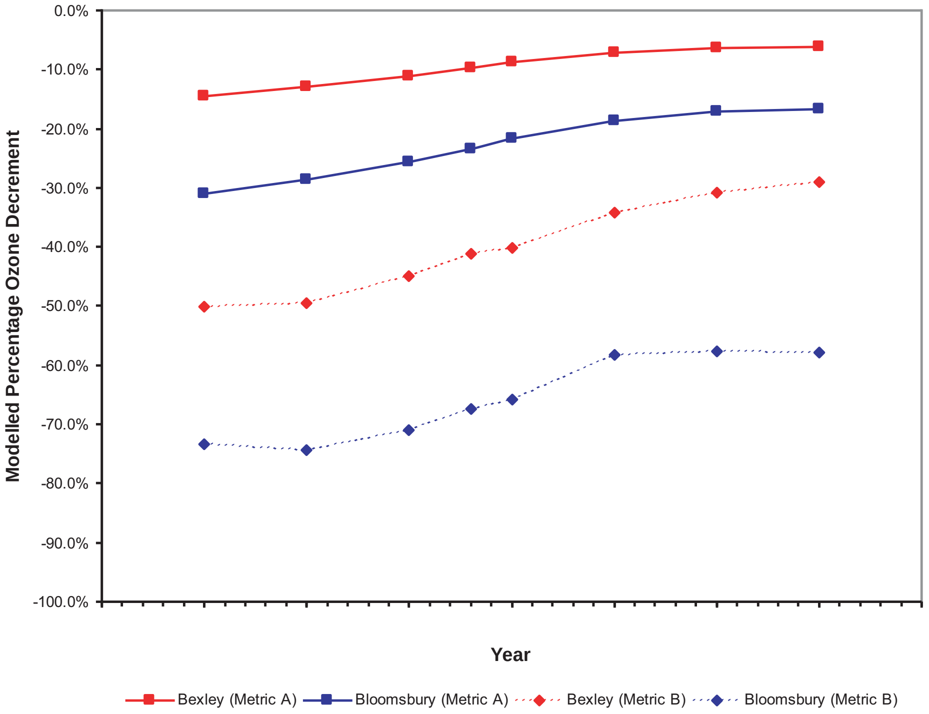


Figure 5.7 Modelled percentage decrement in the annual mean of the maximum daily running 8-hour average ozone concentration (squares/solid lines) and annual mean of the difference between the maximum daily running 8-hour average ozone concentration and a 100 μg m^{-3} (50 ppb) cut-off (diamonds/ dotted lines) as calculated using the OSRM for changing atmospheric composition for selected years between 1990-2020 using 2003 meteorology for the two sites: London Bexley (red) and London Bloomsbury (blue).

226. As expected, the decrements for both metrics, as measured by the differences between the three sites, decrease between 1990 and 2020, reflecting the reduction in NO_X emissions and hence the NO_X-scavenging effect. There is a difference however between the two metrics. As indicated in Figure 5.7, the urban decrements for metric B as a fraction of the rural value, appear larger than those for metric A. At London Bloomsbury, local NO_X scavenging causes a ~30% reduction in metric A in 1990 (reducing to ~17% in 2020) but a ~75% reduction in metric B in 1990 (reducing to ~60% in 2020). The corresponding values at London Bexley in 1990 are reductions of 15% (7% in 2020) and 50% (30% in 2020), respectively. The larger changes in the decrement for the metric B is a result of the sensitivity of this metric to relatively small changes in ozone concentrations close to the cut-off concentration (this high sensitivity was discussed by Sofiev and Tuovinen (2001) for the case of the AOT40 metric). The fractional decrements for the metric with the 70 μg m^{-3} (or 35 ppb) cut-off lie between the other two.

227. Figure 5.6 indicates that the changes in atmospheric composition assumed could cause an increase in metrics A and B of ~5 and between 0.5-1.5 μg m^{-3}, respectively, between 2003 and 2020. Over the same period, the reduction in the NO_X-scavenging effect causes an increase in the ozone metrics of ~6-7 (metric A) and ~0.5 (metric B) μg m^{-3}. Thus, it can be seen that, depending on their strength, ozone trends assumed on the hemispheric/global scale could, by 2020, cause increases in urban ozone of a similar magnitude to the changes

expected from the NO_X-scavenging effect. As discussed in Chapter 4, the trend in the ozone background over this period is however very uncertain, both in strength and the sign of the trend.

5.5 Modelling UK urban ozone decrements

228. The OSRM calculations presented in section 5.4 support the view that ozone can be represented by a regional component and an urban ozone decrement (see Clapp and Jenkin, 2001, and Jenkin, 2004). In this report, the urban ozone decrement is taken to be the difference between the values of the ozone concentration or metric at the urban location and the corresponding quantity at a surrounding rural site (taken to be representative of the regional component).

229. Urban decrements in ozone concentrations have been explored for a range of metrics using two UK process-based models (the Ozone Source-Receptor Model (OSRM) (Hayman *et al.,* 2008a, 2008b) and ADMS-Urban models (Williams *et al.,* 2006)) and an empirical approach based on monitoring data (Pollution Climate Model (PCM) (Kent *et al.,* 2006)). The relationships between the urban decrements and local NO_X concentrations have been examined and the results of the analysis indicate that the empirical approach to estimating urban ozone decrements currently used in mapping studies (as presented in the supporting evidence for Chapter 2) is in reasonably good agreement with process-based models. This section provides a short summary of the modelling and uses annual mean ozone to illustrate the results obtained. An accompanying technical annex to the report provides further information and also results for a wider range of ozone metrics (Annex 3).

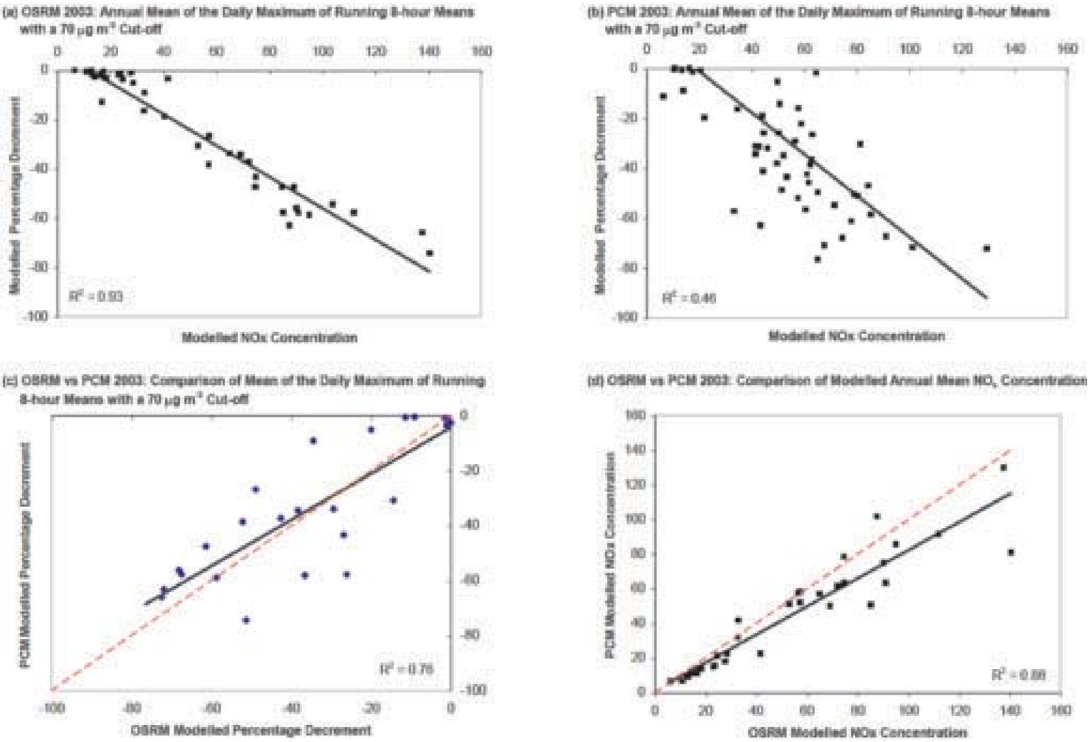


Figure 5.8 Ozone decrements calculated for UK ozone monitoring sites in 2003 using the Ozone Source-Receptor and Pollution Climate Models, as a function of the modelled NO_X concentration (μg m^{-3}, as NO_2).

230. The upper panels of Figure 5.8 show the dependence of the modelled percentage decrement in the metric, the annual mean of the daily maximum of the running 8-hour mean ozone concentrations with a 70 µg m^{-3} cut-off, on the modelled local NO_X annual mean concentration, as calculated using OSRM (panel a) and PCM (panel b). This linear dependence is also seen for other ozone metrics (see Annex 3). The lower panels of Figure 5.8 show scatter plots of the PCM results against the corresponding OSRM results for this ozone metric (panel c) and for NO_X concentrations (panel d).

231. The results presented in Figure 5.8 support the assumption in the empirical modelling approach that the urban decrement in ozone concentration varies approximately linearly with annual mean NO_X concentration (the regression lines are shown here for illustrative purposes only). There is, however, considerable scatter in these plots, which is to be expected since the regional ozone field will incorporate uncertainties associated with the interpolation procedure and very local effects may affect the measured ozone concentrations. The positive outliers are due to measured urban ozone metrics exceeding the values measured at rural sites in the same region.

232. Urban ozone decrements have also been calculated for monitoring site locations in London for the same ozone metrics using ADMS-Urban. These results again compare well with those derived by OSRM and PCM, as indicated in Annex 3. The OSRM model seems to generally predict a lesser decrement for the ozone metrics than ADMS-Urban. The PCM-derived values are based on ambient monitoring data and also incorporate additional uncertainties associated with spatial interpolation of data from rural monitoring sites and therefore show greater scatter than the process-based model estimates. These results indicate that process-based models are now available which can provide a good description of how the decrement in ozone varies spatially across UK urban areas.

5.6 The City-Delta study and integrated assessment modelling

233. The City-Delta study is the most relevant recent study on urban ozone in Europe. The City-Delta study was an open model intercomparison exercise, sponsored by a number of international bodies, as a contribution to the modelling activities in the Clean Air for Europe (CAFE) programme[2]. The aim of the study was to assess the changes in urban air quality predicted by different atmospheric chemical transport dispersion models for ozone (and particulate matter) in response to changes in urban emissions. Eight European cities were selected for the study: Berlin, Copenhagen, Katowice, London, Marseille, Milan, Paris and Prague. Scientific drivers (distinct differences in climatic conditions, the vicinity of the sea, differences in meteorological situations and emission densities, etc.) and practical considerations (the availability of suitable models, of emission inventories for gaseous and particulate pollutants, of sufficient meteorological information and monitoring data, etc.) were important in the choice of these cities. The results of the study have been published in a number of recent papers (for example, Cuvelier *et al.,* 2007; van Loon *et al.,* 2007; Thunis *et al.,* 2007; Vautard *et al.,* 2007).

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[2] http://aqm.jrc.it/citydelta/

234. In the paper by Vautard *et al.* (2007), six different models were used to simulate concentrations of ozone and particulate matter for 1999 over domains encompassing a large area around four major European cities: Berlin, Milan, Paris and Prague. Three of the models produced results at large-scale (typically 50 km) and small-scale spatial (5 km) resolution. For ozone, the study concluded that the models were able to capture *fairly well* the mean, daily maxima and variability of ozone concentrations, as well as the time and inter-city variability. However, there was a significant overestimation of ozone concentrations and hence metrics in city centres, especially for the large-scale models.

235. The City-Delta study used the SOMO35 metric to assess the impact of ozone on human health. It was ultimately intended that the City-Delta study would assist the development of a methodology for treating urban ozone in the integrated assessment modelling of the International Institute for Applied Systems Analysis (IIASA) (Amann *et al.,* 2006). As discussed in Chapter 7, the IIASA integrated assessment model uses source-receptor relationships derived from the EMEP Eulerian model at a 50 km x 50 km resolution to relate regional-scale changes in ozone to reductions in NO_X and VOC emissions. Although it was recognised that urban ozone levels should be lower through the NO_X-scavenging effect, the magnitude of this effect could not be quantified systematically for cities in different parts in Europe (see Figure 5.9) (Amann *et al.,* 2006). The contribution of the determining factors (such as meteorological conditions, emission densities, NO_X/VOC ratios, etc.) still has not been defined in a Europe-wide context. The implications of these issues for integrated assessment modelling and UK ozone are discussed further in Chapter 6.

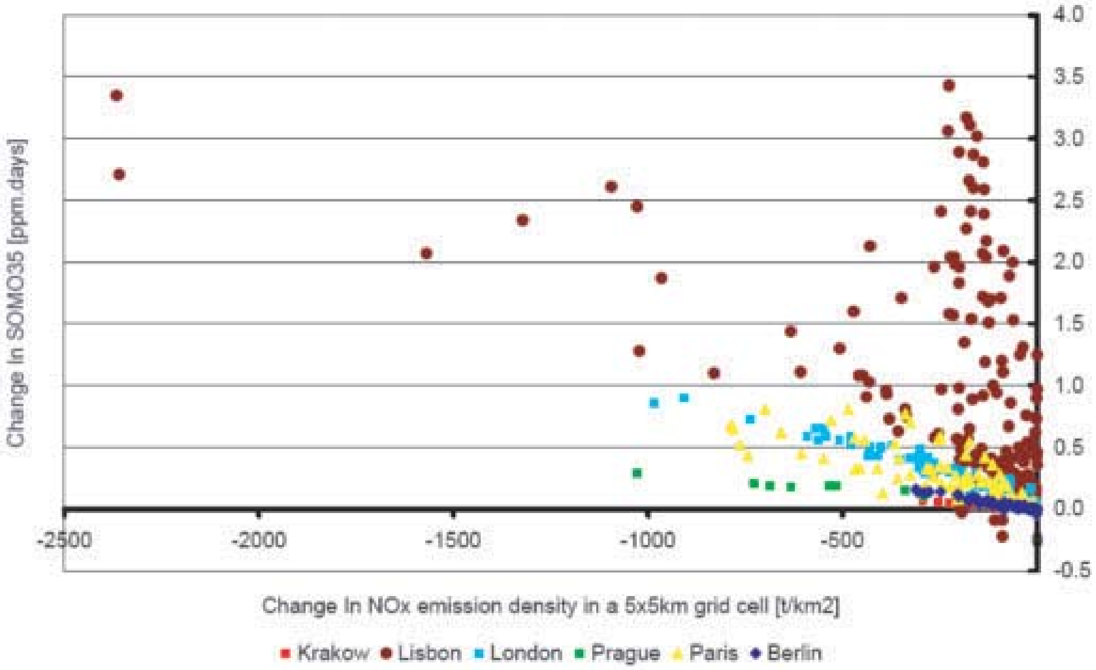

Figure 5.9 Change in the SOMO35 indicator in response to reductions of urban NO_X emissions as computed by the CAMx model for six European cities participating in the City-Delta project (taken from Amman *et al.,* 2006).

5.7 Urban ozone and climate change

236. The Air Quality Expert Group (AQEG) has considered the impact of climate change on air quality in a previous report (AQEG, 2007b). The response to question C addressed the impact of climate change on UK ozone concentrations. Section 5.4 has shown that changing atmospheric composition is a significant driver for UK ozone. Given that urban ozone in the UK can be represented in terms of a regional component and an urban decrement, any additional impact of climate change on the regional component will produce accompanying changes in ozone in the urban centre. The impact of climate change on ozone in the UK and the rest of Europe has been considered in detail in Chapter 4 of this report. On the 2030 time horizon, the impact of climate change on mean regional ozone is likely to be small compared with the effect of changes in precursor emissions although it may have greater influence on photochemical episodes that transiently impinge on urban areas.

237. As indicated in Chapter 4, the summer of 2003 in Europe is expected to become “typical” by the 2030s in terms of temperature. This may also be accompanied by an increase in stagnation conditions necessary for summertime photochemical episodes, although not necessarily an increase in summertime blocking high weather systems. Even under these conditions, it is unlikely that the intense urban ozone production typical of Los Angeles would occur in the UK. As indicated in section 5.2 and also in Chapter 2, there was no evidence for local photochemical production of ozone in London in 2003.

5.8 Recommendations

238. Significant progress has been made on modelling ozone in urban areas. We recommend that further improvements be made to improve the spatial resolution of ozone modelling tools so that the decrements in ozone in urban areas can be treated robustly in integrated assessment modelling.

239. While NO_x scavenging is the major driver on ozone concentrations in urban areas, changes in northern hemisphere background concentrations of ozone could be as important. We recommend that studies are undertaken to define the sign and strength of these changes for a number of realistic scenarios to 2020.

Chapter 6

Uncertainties in ozone models

Question E: *Ozone is currently modelled on a number of spatial and temporal scales. What are the main uncertainties associated with such work, and what research is required to reduce these uncertainties?*

Short answer to question E

240. Although a number of models address ozone on a range of temporal and spatial scales across the UK, there is no consistent and comprehensive understanding of model performance and the uncertainties that affect them. Research is required to understand and intercompare the influence of different spatial and temporal resolutions, chemical mechanisms and parameterisations upon predicted concentrations and their policy implications. This process would involve harmonising model performance evaluation and collecting information on uncertainties of the various model formulations. Research is also required to evaluate the relative importance of man-made and natural biogenic sources of ozone precursors.

Detailed answer to question E

241. A number of model studies have been performed and applied with the aim of understanding ozone formation in Europe and in the United Kingdom. A survey of some of the principal models used in these studies has been compiled and is summarised at the end of this chapter in Table 6-1. Some of these models have been used to support Defra in its policy development and air quality strategy formulation, in particular the Pollution Climate Model (PCM), the Ozone Source-Receptor Model (OSRM) and the UK Photochemical Trajectory Model (PTM). The EMEP model is the main tool for policy formulation within Europe through the aegis of the United Nations Economic Commission for Europe (UNECE) and the Commission of the European Communities. The use of models is an accepted way of incorporating understanding of the underlying science into environmental decision making. However, the complexity and spatial scale of processes leading to ozone production means that it is difficult to evaluate the accuracy of ozone models, even though decisions on how to reduce ozone concentrations have relied on the interpretation and forecasting of such models.

242. Ozone models have a long history of helping to explain observations such as the long-range transboundary transport of ozone, the diurnal cycle in ozone concentrations and in predicting outcomes where observations have a limited usefulness or availability. They have demonstrated that each different organic compound exhibits a different propensity to form ozone. They have shown how the observed downwards trends in urban concentrations of volatile organic compounds (VOCs) and nitrogen oxides (NO_X) due to vehicle emission controls can account for the observed downwards trends in episodic peak ozone levels. They have also been used to great effect in the underpinning of the integrated assessment models used to formulate air quality policies at the EU and UNECE levels.

243. Defra employs ozone models to predict the outcomes of particular control measures in support of the UK Air Quality Strategy and policy development. It also uses modelling to supplement observations in its reporting under the EU Daughter Directives. This activity is an alternative to obtaining a comprehensive set of observations for the UK which would not be feasible because of resource constraints. Ozone models are thus critical tools that help inform and set priorities within Defra. As a result, it is essential that these tools are fit-for-purpose, that the results are robust and produced in a timely manner, and that Defra's applications are consistent with the model's intended purpose and formulation.

244. The models detailed in the supporting evidence cover a wide range of complexity from simple empirical and interpolation-based mapping models to highly sophisticated Eulerian grid-based three-dimensional air-shed models. Models are always incomplete and efforts to make them more complex can be problematic. Increasing the complexity of a model by adding new features and capabilities may introduce more model parameters whose values are uncertain. More complex models may contain more parameters than can be estimated or calibrated with the available observations. Scientific advances will never make it possible to build a perfect ozone model. Models are necessarily simplifications and approximations of the real world and are inherently uncertain (National Academy of Sciences (NAS), 2007; Oreskes *et al*., 1994). The NAS report recognises the limitations of models but still recommends their use in environmental decision making. It recognises the difference between research and regulatory applications of models. It contains general warnings and guidance on how this should be done, especially that evaluation should continue throughout the life of a model. In particular, the report cites the history of ozone control policies to draw lessons about model use. The policy response to ozone issues in North America has been subject to step changes as the science and modelling has developed and this is something to be avoided. A more complete evaluation of uncertainties in modelling should have been undertaken before policy was initiated.

245. The ozone models reviewed in the supporting evidence, in common with all environmental models, contain a large number of simplifications in their formulation which for the purposes of this discussion may be grouped into four categories as follows:

- Simplifications to the modelling system arising from theoretical aspects of the system that are not fully understood or from model parameterisations.
- Boundary conditions and model input.
- Empirical aspects of the system that are difficult or impossible to measure.
- Temporal and spatial averaging within the modelling system.

246. The first area of model uncertainty concerns the theoretical aspects of ozone modelling that are not fully understood or processes which are simplified by model parameterisations. This includes both dynamic processes, for example the mixing of stratospheric ozone into the troposphere or mixing of free tropospheric air into the atmospheric boundary layer, and chemical processes, for example simplified models of natural biogenic emissions of isoprene and

other VOCs, and hence uncertainty about their contribution to ozone formation in the United Kingdom. Increased theoretical understanding also impacts on the best choice of model for a particular application. Broadly speaking, two major regimes of ozone formation have been identified: intense VOC-limited ozone formation in urban plumes and steady NO_X-limited ozone formation on the regional and transboundary scales. Whilst the extremes of these regimes have been well studied in field studies carried out elsewhere in Europe, for example in Milan and Berlin, and in Los Angeles and Texas in North America, it is not at all clear under which regime the ozone observed in a given episode in the UK has been formed. This lack of understanding precludes a definitive choice between the different model approaches, for example, between Eulerian grid and Lagrangian trajectory approaches. Eulerian models are well suited to describing the competition between vertical mixing and rapid chemistry that drives urban-scale ozone formation. In contrast, trajectory models, because they can handle more complex chemical mechanisms, are well suited to the detailed chemical description of ozone formation on the regional scale.

247. The second area of model uncertainty arises because of the uncertainties inherent in the large amount of input data that is used to set up and drive ozone models. This includes emissions, both man-made and biogenic, the boundary conditions for both the meteorological models and transport and chemical transformation models (if both models are utilised), model initialisation and data assimilation (if utilised). Both the data themselves, which may come from measurements or other larger area model outputs, and the implementation of these data in the ozone model have a degree of uncertainty associated with them. Further uncertainty may arise because the process descriptions in the models are themselves simplifications of real-world behaviour. Photolysis rate coefficients are a good example of this. These coefficients control the rate of photochemical ozone formation and should depend on cloud cover and take photochemical haze formation into account. Rarely are the large amounts of solar radiation data required for this purpose available and clear sky conditions are commonly assumed.

248. The third area of model uncertainty concerns those empirical aspects of ozone formation that are difficult or impossible to measure or observe and then to represent in ozone models. Many of these empirical uncertainties involve the measurement of the atmospheric concentrations of the many reactive free radical species, the emitted VOCs and their secondary organic reaction products that together control ozone formation over Europe. Even if all these measurements were available, it is not clear that the different ozone models would necessarily have the capacity to handle the detailed chemical mechanisms that would be needed to calculate the resulting ozone formation. This a particularly pressing issue for Eulerian grid models because they necessarily require highly compact and concise chemical mechanisms because of the limited computer resources available.

249. The fourth area of model uncertainty arises from the temporal and spatial averaging which is a feature of all models to a greater or lesser extent. The greater the temporal averaging in a model the less it is able to take account of aspects of the system and processes which vary with time, because the processes and the parameters that drive them are implicitly assumed to be constant over the averaging time considered, when in reality they may be

highly variable or sporadic. Scavenging by precipitation, resuspension and fires are classic examples of sporadic processes. Man-made emissions are most accurately quantified as annual and national totals. These totals have then to be broken down to an hourly basis and gridded at a spatial scale of 1 km x 1km or down to individual road links, across the country. In the real world, however, emissions from a single 1 km x 1 km grid square or a single road link are highly variable and unpredictable and this variability may introduce significant uncertainties in ozone model predictions and their evaluation against observations. Spatial averaging both in model input (e.g. emissions) and model formulation introduces further uncertainty since at scales similar to or smaller than the relevant input or model grid resolution the model will not be able to predict spatial variations in concentrations. This is particularly relevant in urban areas where there are sharper gradients in ozone concentrations, and requires due consideration when model calculations are compared with monitored data measured at a point.

250. Whilst it is relatively straightforward to characterise the main sources of uncertainty in ozone models in general terms, it is another matter to quantify their importance in a particular ozone modelling system, let alone in a particular model application. The effects of some sources of model uncertainty, such as those in the initial and boundary conditions, may decrease as the model calculation progresses, but the effects of others, such as those in meteorology or emissions, may grow. Some sources of uncertainty are crucial to the accuracy of an individual case but drop out when looking at the relative responses to emission controls and other policy measures. Such sources of uncertainty include photolysis rates and natural biogenic emissions. Uncertainties are best reduced by continual improvements to process descriptions, participation in model intercomparisons, development of real-time forecasting capabilities and continued comparisons against observations.

251. Comparisons against observations give an invaluable guide to overall model performance. Understandably, policy-makers expect modellers to establish the trustworthiness of their models. For ozone models, this almost always involves some form of comparison of model predictions against measured ozone concentrations. However, the ability of an ozone model to reproduce measured ozone concentrations from the past does not guarantee its adequacy for the future or for predicting the response to ozone control strategies. Agreement with observations is inherently partial. Models agree with some observations but not all. A model can certainly perform well against observations and the precision and accuracy of the fit can be quantified. The performance of models can be evaluated relative to observations, relative to other models or against our own theoretical expectations, but the performance of a model, especially for future projections of concentrations, cannot be ascertained precisely. Nevertheless, comparison against observations is a good first step in the evaluation of model performance.

252. An example of model uncertainty and the methods for assessing it can be seen in Figure 6.1, which shows a comparison between seven regional air quality models and observed ozone over the yearly mean diurnal cycle of ozone over Europe. Though the models capture the main features of the diurnal cycle many of them overestimate the observed values and there is a considerable spread in absolute values. The daytime ozone production rates also seem to vary widely. The authors ascribe the main factors leading to this difference to a

combination of several processes including vertical resolution, mixing, dry deposition, emission injection height and representation of ozone profile in the lower boundary layer.

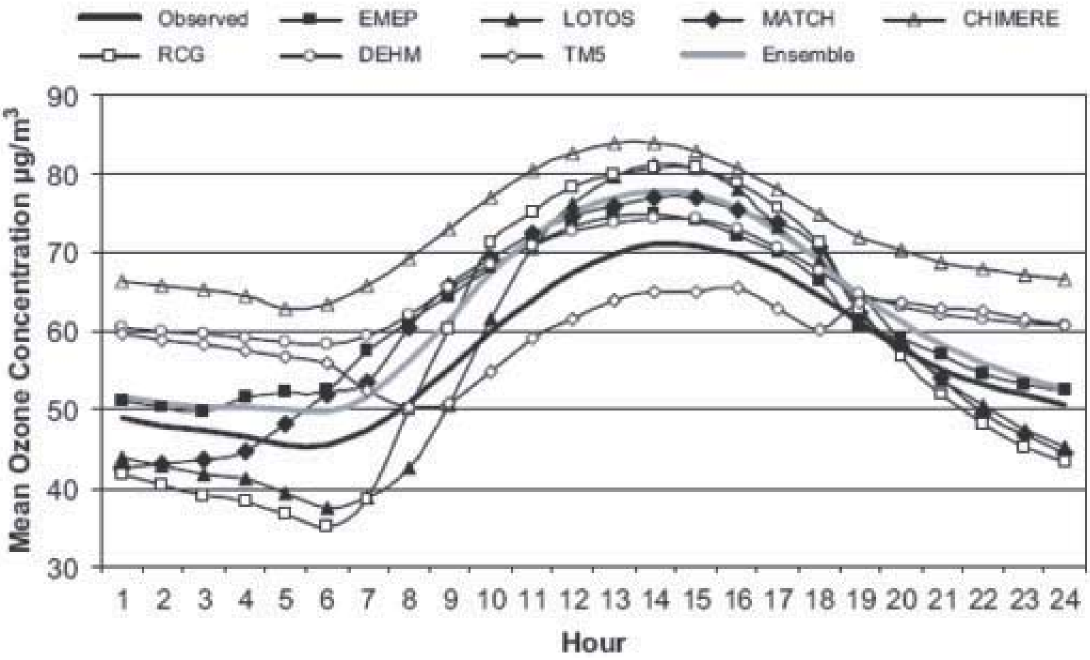

Figure 6.1 Yearly mean diurnal cycle of ozone from a number of regional air quality models compared to observations (van Loon *et al.*, 2007) (reprinted with permission from Elsevier).

253. The term 'validation' is widely used to describe evaluation of model performance against measured data. Oreskes et al. (1994) argue that comprehensive or complete validation and verification of models of natural environmental systems are not possible. This is because there may always be input parameters that are poorly known, fine-scale details of crucial importance which are inadequately understood, and assumptions and inferences which may not be valid under all circumstances. 'Complete validation' of models refers to their validation under all possible circumstances and scenarios. In practice, the term 'validation' is frequently used in a more restricted sense to describe the evaluation of model predictions against measured data, which might more appropriately be described as 'partial validation', as it applies only to a limited range of input variables, and hence outputs. The distinction between 'complete' and 'partial' model validation is an important one, as a model giving excellent predictions from within a limited range of input variables may perform poorly when used beyond that range of inputs. Such limitations need to be clearly recognised and the term 'evaluation' is to be preferred to 'validation', as it more accurately describes the process undertaken.

254. Assuming that the model code faithfully represents the model specification, there are three general questions to be answered when evaluating environmental or mathematical models, which, following Beck (2002) and Britter and Schatzmann (2007), can broadly be expressed as follows:

- Is the scientific formulation of the model broadly accepted and does it use state-of-the-art process descriptions?

- Does the model replicate observations adequately?

- Does the model reflect the needs and responsibilities of the model user and is it suitable for answering policy questions and fulfilling its designated tasks?

These questions can be applied effectively to ozone models and should lie at the heart of a review and evaluation of such models.

255. When relying on models to draw conclusions, the Air Quality Expert Group (AQEG) generally supports an approach which uses a range of available ozone models, including simple and advanced ones, in combination with measurements. The ozone model should be used to determine the relative response of ozone to estimated future emissions from a background determined from recent measurements. This assumes that current emissions and emission projections are reliable (US Environmental Protection Agency, 2007).

256. There is a constant need to review current modelling activities to ensure that the models used are 'fit for purpose'. They should reflect the current state of the art, assess the uncertainties inherent in such modelling studies, and be able to encompass changes both within the modelling art itself and the expected drivers for future policy, such as predicted changes in temperature and weather patterns. Table 6-1 summarises a survey of the principal ozone models of policy relevance to the UK and starts to gather together some of the basic information required for their review and evaluation.

257. In 2007, Defra commissioned a study with the primary objective of collating, evaluating and summarising information on tools for modelling ozone formation and assessing impacts on human health and ecosystems within the Defra policy context. The study undertook a wide-ranging review of European and US modelling tools and their application to the policy context. The study concluded that for ozone modelling, Defra requires a model which is reliable, well tested, as precise as possible, readily updatable and, as far as possible, future safe (Monks *et al.* 2007). There is a requirement for flexibility in modelling approach that allows a number of policy areas in air quality to be assessed (e.g. ozone and particulate matter (PM)). In order to achieve this ideal there are a number of issues and elements that require consideration.

258. It was recommended that future ozone models should use the Eulerian framework, but there is a need to ensure backward compatibility and continuity. Chemical schemes are an inherent part of models and these should be tested for ozone (e.g. the Master Chemical Mechanism (MCM) or CBM-IV). However, surrogate schemes which have a firm basis in explicit chemistry and which have been tested by comparison with experimental data can also be used as appropriate.

259. With respect to emissions, there is need to allow robust coupling between the speciation in the emission inventories and the chemical scheme allowing specific policy measures to be assessed more clearly and contain less simplification and tuning of mechanisms. Owing to the growing importance of biogenic/anthropogenic coupling, there is a requirement for the improved representation of biogenic species in models, for example, to be prepared for likely warmer summer periods in the future. Models used for ozone work should have transparent sources of emission estimates. There is a requirement

that models have recognised and realistic schemes for the spatial and temporal disaggregation of emission estimates. Some assessment is also required of how these might change in the future. Improved biogenic emission estimates, or land-use data in conjunction with biogenic emission factors, are required, coupled to a reassessment of the UK biogenic emission inventory.

260. A key element of ozone model performance is model evaluation and (inter)comparison. The performance of models should be tested by observation to ensure their continued performance levels, as well as regular comparison between UK ozone models choosing, perhaps, periods of peak and background ozone, to ensure that model performance is satisfactory. It is important that ozone policy models have a strong peer-reviewed evidence base and that UK ozone models are taking part in European-wide comparisons for policy purposes, to model observations from small groups of high quality stations in chosen countries, and to ensure the performance of its own models and of those used by the EU for regulatory purposes. Full details of how these recommendations were arrived at are available at: http://sciencesearch.defra.gov.uk/Document.aspx?Document=AQ0706_6733_FRP.pdf.

6.1 Recommendations

261. AQEG recommends that this ozone model review should be extended by:

- exchanging the experiences gained with different chemical mechanisms and model parameterisations;
- investigating further central policy issues such as the relative importance of man-made and natural biogenic sources of VOCs;
- assembling information on uncertainties; and
- setting out a protocol for a model intercomparison activity.

Table 6-1 Ozone models

Model	Type and resolution	Chemistry	Treatment of VOCs	Meteorology	Policy relevance
ADMS-Urban CERC	Gaussian type nested in trajectory model. Variable resolution down to 10 m	GRS* 6 reactions or CB4* 95 reactions, 36 species	GRS – 1 surrogate VOC CB4 7 man-made VOCs + isoprene	Standard met data from one measurement site or mesoscale model	Policy including for UK. Air quality forecasting for AirTEXT (London) – nested within Prevair.
CHIMERE INERIS, Paris	Eulerian grid 0.5° x 0.5° 50 km x 50 km emissions	MELCHIOR 80 species, 300 reactions	12 man-made VOCs + isoprene	Meteo France	Ozone and PM forecast model for Europe
EMEP4UK MSC-W	Eulerian grid 50 km x 50 km, 5 km x 5 km Emissions: EMEP 50 km x 50 km, NAEI 5 km x 5 km	EMEP 80 species, 140 reactions	9 man-made VOCs + isoprene (C_2H_6, nC_4H_{10}, C_2H_4, C_3H_6, C_8H_{10}, HCHO, MEK, CH_3OH, C_2H_5OH)	HIRLAM-PS WRF/NCEP	Policy formulation for UNECE CLRTAP. Input to RAINS, CAFE and NEC policy analyses
HARM/ELMO Universities of Nottingham and Lancaster	Lagrangian trajectory Emissions: NAEI 10 km x 10 km, EMEP 50 km x 50 km	STOCHEM + secondary organic aerosol formation	12 man-made VOCs + isoprene, terpenes (C_2H_6, C_3H_8, nC_4H_{10}, C_2H_4, C_3H_6, C_7H_8, C_8H_{10}, CH_3OH, acetone, MEK, HCHO, CH_3CHO)	HYSPLIT/NCEP	Defra mapping for SOA EU & UNECE assessments
MODELS-3/CMAQ Imperial College London	Eulerian grid Fully nested: 4 km x 4 km, 12 km x 12 km, 48 km x 48 km Emissions: NAEI 10 km x 10 km, EMEP 50 km x 50 km	CB4* 36 species, 93 reactions	7 man-made VOCs + isoprene ETH, PAR, OLE, TOL, XYL, FORM, ALD2	Met Office, UK BADC archive	Most heavily used research and policy model in USA.

Table 6-1 Ozone models (Cont.)

MODELS -3/CMAQ Universities of Manchester, Edinburgh and Hertfordshire Eon	Eulerian grid Fully nested: 5 km x 5 km, 45 km x 45 km Emissions: NAEI 10 km x 10 km, EMEP 50 km x 50 km	CB4* 36 species, 93 reactions RADM2* 57 species,158 reactions SAPRC-99* 72 species, 214 reactions	7 man-made VOCs + isoprene ETH, PAR, OLE, TOL, XYL, FORM, ALD2	MM5 – 5th generation three-dimensional meteorological fields	Most heavily used research and policy model in USA.
NAME Met Office	Lagrangian dispersion Source- or receptor-oriented Emissions: NAEI 10 km x 10 km, EMEP 50 km x 50 km	STOCHEM + reactive VOCs	13 man-made VOCs + isoprene (C_2H_6, C_3H_8, nC_4H_{10}, C_2H_4, C_3H_6, C_4H_6, C_7H_8, C_8H_{10}, CH_3OH, acetone, MEK, HCHO, CH_3CHO)	UK Met Office NAME archive 1995-2007 ECMWF 1957-onwards	Basic model (without ozone) used as Defra air quality forecast model. Emergency response (without chemistry)
OSRM AEA Technology & Environment	Lagrangian trajectory with surface post processing. Emissions: NAEI 10 km x 10 km, EMEP 50 km x 50 km	STOCHEM 70 species, 180 reactions	12 man-made VOCs + isoprene (C_2H_6, C_3H_8, nC_4H_{10}, C_2H_4, C_3H_6, C_7H_8, C_8H_{10}, CH_3OH, MEK, acetone, HCHO, CH_3CHO)	Met Office, UK NAME archive	UK policy applications Current Defra ozone tool Used for scenarios in the Defra Air Quality Strategy.
POLLUTION CLIMATE MODEL AEA Technology & Environment	Empirical- and interpolation-based mapping models	Parameterised treatment of oxidant partitioning	No VOC chemistry	n/a	Reporting for ozone Daughter Directives Interpretation of observations
UK PTM rdscientific, Imperial College London and Universities of Leeds and Birmingham.	Lagrangian trajectory Emissions: NAEI 10 km x 10 km, EMEP 50 km x 50 km	Master Chemical Mechanism* 4,414 species, 12,871 reactions CB4* 36 species, 93 reactions	175 man-made VOCs from NAEI + isoprene, α-pinene, β-pinene	Met Office, UK NAME archive HYSPLIT/NCEP BADC/UK MO	UK policy applications for O_3 and PM modelling

Notes: a. * denotes that this chemical mechanism has been compared against smog chamber data.

Chapter 7

Impact of European emissions reductions on ozone in the UK

Question F: *Integrated assessment modelling to support the European Commission's Thematic Strategy for Air Quality suggests that regional ozone levels in the UK are likely to remain relatively steady regardless of foreseeable emission reductions across Europe. Does the Group agree with this analysis and what is the explanation for this lack of response to reductions in precursor emissions?*

Short answer to question F

262. The Air Quality Expert Group agrees that under the specific emission scenarios considered for the European Commission's Thematic Strategy, regional ozone levels in the UK (based on the SOMO35 metric) would be likely to remain steady in the foreseeable future. However, this does not indicate that regional ozone levels in the UK are insensitive to precursor emissions in European countries and the surrounding seas, especially for episodes of high concentrations.

Detailed answer to question F

263. In order to understand why regional ozone levels in the UK were predicted to remain relatively steady within the integrated modelling carried out for the European Commission's Clean Air for Europe (CAFE) Thematic Strategy, it is firstly necessary to understand how potential future precursor emission reductions were handled. Some potential future precursor emission reductions were taken on board and incorporated into the 'CAFE background' emissions scenario. The remaining potential emission controls were made available to the optimisation routines within the integrated assessment model, to be taken up when and if required to achieve the identified targets. Owing to the manner in which the different contributions to environmental damage were weighted by the choice of gap closure targets,[3] optimisations that reduced acid rain and eutrophication, together with particulate matter (PM) levels, were generally favoured, rather than optimisations that reduced ground-level ozone formation.

264. The potential emission controls that came out strongly in the CAFE analysis turned out to be measures that focussed particularly on emissions of nitrogen oxides (NO_X) and reducing concentrations of PM. Apart from their effects on ozone, reductions in NO_X emissions are favoured because they contribute to nitrogen deposition (and hence eutrophication and acidification of ecosystems) as well as nitrate aerosol as a component of PM. Because of the associated improvements in lost days of life expectancy from human exposure to PM, reductions in PM rank highly in weighing costs and benefits of emission reductions. However, reducing NO_X emissions leads to an increase in urban ozone levels, partially offsetting improvements in regional ozone levels secured

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[3] Intended to close the gap between the projected "current legislation" (CLE) scenario and the scenario corresponding to the "maximum feasible reduction" (MFR) by 60%, that is, to achieve 60% of the maximum possible improvement implied by the MFR scenario.

elsewhere. Reductions in VOC emissions were relatively modest. The CAFE scenarios did not therefore show evidence of any potential for dramatic improvement in UK ozone.

265. The next issue to understand is the impact of the additional CAFE measures on regional ozone, through the selection of a target based on SOMO35 as the index of human health effects, and associated estimation of AOT40 for crops and vegetation damage. Both SOMO35 and AOT40 are strongly influenced by changes in northern hemisphere ozone background concentrations and so the influence of changes in regional ozone formation are heavily damped. This damping is greatest for countries on the Atlantic Ocean seaboard of Europe, such as the United Kingdom, Ireland, Norway and Portugal and smallest for countries in central Europe, such as Germany, where the influence of the northern hemisphere ozone background is least and the contribution from regional ozone formation greatest. In urban areas, any reductions in the NO_X emissions from road traffic would lead to higher SOMO35 levels, counteracting any benefits from reduced regional-scale ozone formation. As has been observed elsewhere in this report, ozone levels appear to behave differently, depending on the metric used.

266. The integrated assessment modelling carried out for the CAFE strategy reveals little about the response of regional ozone to foreseeable VOC emission reductions, because many of the projected control measures, such as the EU Directives addressing solvent emissions and petrol vapour recovery for example, were already implemented in the CAFE background. Furthermore, the VOC control measures available to the optimisation routines were relatively costly and had few side benefits for primary PM emissions and hence would not be seen as attractive in any optimisation that was, understandably, weighted towards reducing urban PM health effects rather than regional ozone. EMEP simulations show that VOC reductions show greater reductions in SOMO35 throughout Europe, including the UK, than do equivalent percentage reductions in NO_X. However, the reductions in VOCs included in the International Institute for Applied Systems Analysis (IIASA) Regional Air Pollution Information and Simulation (RAINS) analysis were significantly smaller than those in NO_X.

267. This combination of reasons explains why regional ozone levels over the United Kingdom have appeared to remain relatively steady under the European Commission's CAFE strategy, regardless of the implementation of a number of additional measures in the foreseeable future. The results are related to the sensitivity of the UK to the northern hemisphere background, coupled with the increases in ozone that are projected in urban regions on reducing NO_X. European emissions have the greatest effect in the UK on high episodic concentrations of ozone experienced under anticyclonic conditions. As a result of this combination of circumstances, measures which lead to significant reductions in ozone in central Europe are less effective in the UK when assessed against a low threshold metric such as SOMO35. A key issue in the CAFE assessment is that in a combined strategy designed to address several pollutants, the measures which win out are those that reduce PM, simply because of the greater health effects/benefits. Measures purely directed at ozone are less attractive in cost-benefit terms. Strategies for reducing ozone in the UK are addressed in Chapter 8 but a full cost-benefit analysis has not been performed.

Supporting evidence for question F

7.1 Overview

268. Chapter 7 addresses the results of the integrated assessment modelling carried out to support the European Commission's Thematic Strategy for Air Quality, Clean Air for Europe (CAFE) (Commission of the European Communities, 2005). This modelling work was undertaken using the Regional Air Pollution Information and Simulation (RAINS) integrated assessment model based at the International Institute for Applied Systems Analysis (IIASA) in Austria, which in turn used source-receptor relationships from the Unified EMEP model operated at the Meteorological Synthesising Centre-West (MSC-W) in Norway. The supporting evidence assembled here deals with the salient features of the Unified EMEP model. An overview is then provided of the RAINS model together with the CAFE strategy analyses. Some remarks are then made about the various policy-relevant ozone metrics. Finally, the supporting evidence contains some comments on the relative effects of VOC vs NO_X controls on ozone formation.

7.2 Salient features of the Unified EMEP model

269. The European-scale integrated assessment modelling carried out to support the United Nations Economic Commission for Europe (UNECE) international Convention on Long-range Transport of Air Pollution (CLRTAP) has always been based on an understanding of the transboundary fluxes and origins of the pollutants in question. At the time of the formulation of the multi-pollutant, multi-effect Gothenburg Protocol, these transboundary fluxes were quantified using the EMEP Lagrangian model (Iversen, 1990). In the intervening period since the Gothenburg Protocol, EMEP has developed the Unified Eulerian model (EMEP, 2003). This new model has the same basic representations of the emissions, chemistry and deposition processes as the original Lagrangian model, but differs in that they are assembled into a three-dimensional modelling framework on a 50 km x 50 km grid (see also Chapter 6). The performance of the EMEP Unified model has been reviewed by national experts as part of the EMEP Task Force on Measurement and Modelling (TFMM) at a workshop in Oslo during 2004 (TFMM, 2004). It was this version of the EMEP model that would be ultimately employed in the determination of the source-receptor relationships for the integrated assessment modelling to be carried out within the CAFE thematic strategy. Model performance against observations for acidifying substances and ground-level ozone appeared adequate for policy purposes on the regional scale. Preliminary evaluation of the source-receptor relationships determined with the EMEP Unified model against those determined with other European models also appeared satisfactory. However, problems were found in the modelling of some PM components. Further work was recommended to improve model performance for these PM components and to address urban ozone and PM.

270. During the CAFE process, further changes to the Unified Eulerian model were made by EMEP. They initially used two main metrics for ozone, AOT40 and AOT60. The former was used to assess ozone impacts on crops and vegetation, whereas the latter was used for human health impacts. Later, following advice from the World Health Organization (WHO), it was decided that SOMO35 provided a better metric for the assessment of human health effects, and so

EMEP, and hence IIASA (for the integrated assessment model), switched to SOMO35. A description of ozone metrics is given in the introduction to this report and is also discussed in section 7.4 below.

271. In 2006, for source-receptor calculations, EMEP introduced new assumptions concerning ozone background concentrations, including an increase of 6 µg m^{-3} in the annual mean over the mean 1990 level, when calculating 2010 ozone concentrations and an increase of 9 µg m^{-3} in the annual mean for year 2020 simulations. A comparison of maps of SOMO35 produced by the EMEP model with and without this increase illustrates how the higher hemispheric background leads to higher values of SOMO35 across the whole of the UK, indicating the sensitivity to changes in the global background (EMEP, 2006).

272. With this trend in northern hemispheric background included, modelled trends in SOMO35 averaged over the UK show a slight reduction between 1990 and 2000 with a subsequent increase to slightly higher levels between 2000 and 2010. This apparent slight increase can be explained by the influence of the increasing northern hemispheric background acting to offset the reductions in ozone caused by regional-scale pollution controls. This can be contrasted with the situation in Germany, where average SOMO35 levels are much higher but show a more marked decrease between 1990 and 2010. Since the relative contribution from hemispheric background ozone is much smaller in central Europe, the hemispheric background increase is not enough to offset the decrease due to regional pollution controls.

273. The RAINS analysis for the UK is sensitive to the assumed increase in background ozone. Chapter 3 shows that, while concentrations of background ozone, as measured at Mace Head, have increased significantly since 1987, the concentration has been steady since 2000, so that the increases anticipated in the EMEP analysis, and their effect on UK ozone, may not be realistic.

7.3 IIASA RAINS integrated assessment model

274. Integrated assessment modelling, in particular the RAINS model of IIASA, has played an influential role in both the UNECE development of the Gothenburg Protocol (1999), and the Clean Air for Europe (CAFE) programme of the European Union. The RAINS model (Amann *et al.*, 2004) is based on the atmospheric modelling of EMEP to simulate pollutant concentrations and deposition across Europe on a grid of spatial resolution 50 km x 50 km, together with the response of these concentrations and depositions to the scaling of national emissions from each country to represent the effect of emission reductions. The RAINS model links these data to emission projections and uses a database on potential emission reductions and their costs in each country to deduce the least cost solution to achieving targets set for improved environmental protection across Europe. Such optimisation procedures were used to derive proposed emission ceilings for NO_X, VOCs, sulphur dioxide (SO_2), ammonia (NH_3) and primary $PM_{2.5}$ in a series of scenarios for the CAFE programme in 2005 towards drafting of the European Commission's Thematic Strategy for Air Quality.

275. At the inception of the CAFE strategy analyses, the RAINS model was subject to peer review and the documentation prepared for that review fully details the methodologies and input data employed (Amann *et al*., 2004). Subsequently, as with the Unified EMEP model, further changes have been made to the RAINS model and these have had a major impact on the results of the scenario analyses (Amann *et al*., 2005a, 2005b, 2005c).

276. Parallelling EMEP and WHO, there has been a switch from AOT60 to SOMO35 in characterising human health impacts (see section 7.4 below). IIASA used a linear relationship between SOMO35 in each grid square and changes in NO_X and VOC emissions in each country based on the source-receptor relationships[4] calculated using the EMEP Unified model (EMEP, 2006). IIASA justified this linear relationship by assuming that in 2020 NO_X emission reductions will be sufficient to get over the non-linear 'hump' situation whereby reductions in NO_X can lead to enhanced ozone. As discussed in this report, and especially in Chapter 5, these may not be good assumptions for parts of the UK, and also the Netherlands and Belgium. Nevertheless, with these assumptions, the change in SOMO35 in any EMEP grid square is represented as a weighted sum of emission changes in NO_X and VOCs in each country in the RAINS model.

277. Recognising that ozone levels are lower within urban areas, IIASA distinguishes between urban and rural populations in deriving a population-weighted average. However in a regional model, it was not possible to model realistically these sub-grid scale phenomena in the SOMO35 values derived by EMEP, and no simple relationship was suggested by urban-scale modelling in the City-Delta project. Because of the resolution used (50 km x 50 km) in the RAINS model, any implied reduction in SOMO35 in an urban grid square is set to zero for the population in that square. Conversely, for rural populations any implied increase in SOMO35 due to reduction of NO_X emissions is also set to zero. Hence in the RAINS model, this reduces any response of SOMO35 to emission changes and does not truly reflect the urban ozone decrement (see Chapter 5, section 5.5), or allow for situations where NO_X concentrations may still be high in other areas of Europe.

278. The CAFE Thematic Strategy aimed to close the gap between the projected "current legislation" (CLE) scenario and the scenario corresponding to the "maximum technically feasible" reduction (MFR) by 60%; that is to achieve 60% of the maximum possible improvement implied by the MFR scenario. However, whereas the RAINS-optimised scenario for the CAFE Scenario Analysis Report implied a 22% reduction in SOMO35 for the EU25 by 2020 relative to the year 2000, for the UK it indicated a 17% increase. The UK was the only country to show such a projected increase, albeit from a lower start point than many other areas of Europe. One effect of this increase is a projected increase in premature deaths in the UK attributable to ozone exposure between 2000 and 2020 (Amann *et al*., 2005c). The reduction in SOMO35 in the UK from the additional effort beyond the CLE for the Thematic Strategy (involving emissions reductions from 817 (CLE) to 646 ktonnes of NO_X, and from 878 (CLE) to 766 ktonnes of VOCs, for the UK) is modest (~3%) compared with the average improvement in the EU25 (~7.3%). Ireland also remains very much the same (see Table 7-1).

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[4] EMEP's source-receptor relationships assign the proportion of emissions by country of origin that give rise to the concentrations of a given pollutant seen in each individual country, i.e. the origin of the pollution in each country in the model domain.

Table 7-1 Estimates of premature deaths attributable to exposure to ozone (cases per year). These calculations are based on regional-scale ozone calculations (50 x 50 km) and for the meteorological conditions of 1997. A cut-off value of 35 ppb has been applied.

	2000	**2020**	
		Background current legislation scenario	**The Thematic Strategy scenario**
Austria	422	316	287
Belgium	381	345	337
Cyprus	33	32	31
Czech Rep.	535	390	348
Denmark	179	161	153
Estonia	21	22	21
Finland	58	60	56
France	2663	2171	1973
Germany	4258	3316	3057
Greece	627	568	542
Hungary	748	573	511
Ireland	74	79	76
Italy	4507	3556	3328
Latvia	65	65	61
Lithuania	66	64	60
Luxembourg	31	26	24
Malta	22	20	19
Netherlands	416	369	356
Poland	1399	1112	1005
Portugal	450	437	412
Slovakia	239	177	157
Slovenia	112	82	75
Spain	2002	1687	1518
Sweden	197	189	178
UK	1423	1705	1665
EU-25	**20927**	**17522**	**16246**

7.4 Ozone metrics

279. In response to advice from the WHO and the UNECE Task Force on Health (WHO, 2008), changes were made to the ozone metrics employed in the integrated assessment modelling for the assessment of human health effects within the CAFE programme. This advice recommended moving away from formulations based on the AOT concept and moving towards daily metrics such as the annual mean of the maximum 8-hour mean ozone concentrations. On this basis, the integrated assessment models have replaced AOT60 with SOMO35, a metric that includes a specific 70 µg m^{-3} threshold (see also Chapter 1 for an explanation of ozone metrics).

280. Observations of the behaviour of SOMO35 show a definite inter-annual variability at UK monitoring sites. Thus, in general, higher levels are indicated in more photochemically-active years such as 2006 compared with 2005, suggesting that SOMO35 is responsive to meteorology and regional-scale

ozone formation, and is consequently sensitive to the occurrence of more severe episodes such as those occurring in 2003 and 2006 (see Chapter 2).

281. Measurements at rural sites clearly indicate a mixture of contributions, with the peak episodes of large exceedence superimposed on a large number of days with relatively modest exceedences of the 70 µg m^{-3} threshold, which even occur in winter. In urban areas there are clear indications of local-scale removal of ozone leading to near or complete elimination of many of the days of lesser exceedence and large reductions relative to the peak ozone level observed during episodes, consistent with the behaviour of the urban decrement.

7.5 VOC vs NO_X controls

282. Table 7-2 (EMEP, 2004) indicates the response of SOMO35 in each country to 15% reductions in emissions of NO_X and VOC in each other country and source region. It includes shipping, and provides an insight into the differing effects of NO_X vs VOC controls. The table indicates that the response of SOMO35 to reducing UK emissions of NO_X is negative, indicating a suggested worsening of health effects. The extent of the negative change outweighs the benefits to the UK of NO_X emissions reductions in some other countries (such as France) with a net negative effect from the EU25 as a whole. This is in contrast to central European countries such as Germany, where reductions in NO_X both in Germany and other EU25 countries give a clear improvement. It is also evident that changes in shipping emissions of NO_X in the Atlantic and North Sea have an impact, with NO_X reductions in the Atlantic reducing UK ozone because of reduced ozone production, and NO_X reductions in the North Sea increasing ozone because of reduced ozone scavenging. Note that while land-based emissions are being reduced, shipping emissions continue to increase steadily. In this context, the effect of increases in shipping NO_X emissions in the Atlantic are significant from an ozone exposure perspective.

283. Table 7-2 shows that reducing VOC emissions by 15% results in either no change or a reduction in SOMO35, that is an improvement in air quality. As with NO_X, it is also evident that the effects of emissions reductions in the EU25 has greater benefits in central European countries, such as Germany, than it does for countries at the western edge of Europe such as the UK. The table shows that, on an equivalent percentage reduction basis, VOC emissions reductions are more beneficial than NO_X emissions reductions. However, it is important to note that the additional emission reductions in the CAFE Thematic Strategy scenario beyond the CLE scenario were relatively modest for VOCs compared with NO_X (equivalent to ~8% of 2010 emissions of VOCs from the EU25 as opposed to 15% of NO_X emissions in 2010).

Table 7-2 Source-receptor tables indicating effects of 15% reductions in NO_X or VOCs in different countries/sea areas on SOMO35 and on AOT40 taken from EMEP (2004). Positive values indicate improvements in ozone health effects, i.e. reductions in SOMO35.

Country in which emissions are reduced by 15%	**Effect of cutting NO_X on SOMO35 in**		**Effect of cutting VOCs on SOMO35 in**	
	UK	**Germany**	**UK**	**Germany**
UK	-36	3	49	22
Germany	1	47	5	48
France	6	28	7	74
EU25	-20	111	74	128
Atlantic	19	11	0	0
North Sea	-7	3	0	0

	Effect of cutting NO_X on AOT40 in		**Effect of cutting VOCs on AOT40 in**	
	UK	**Germany**	**UK**	**Germany**
UK	-140	48	351	177
Germany	21	651	42	341
France	71	311	55	116
EU25	29	1371	537	926
Atlantic	101	77	0	0
North Sea	-38	101	0	0

Notes:

a. SOMO35 data are provided in ppb.days from EMEP (2004).

b. AOT40 data apply to forests and are provided in ppb.hours from EMEP (2004).

284. For comparison, corresponding figures are given for the response of AOT40, showing the effect of a higher threshold and less sensitivity to the influence of the hemispheric background concentrations. For AOT40, although the effect of reducing UK NO_X emissions leads to increasing ozone in the UK, the EMEP model results imply that equivalent reductions in other countries outweigh this, although they still have much more benefit for central European countries.

285. It is concluded that the EMEP model indicates a number of negative and positive responses of ozone levels in the UK to changes in NO_X emissions from the UK itself, from other countries, and from shipping, and that the global background also influences SOMO35. The net effect of projected changes depends on how much these different influences cancel out.

Chapter 8

Control options for reduction of exposure to ozone in the UK

Question G: *What are likely to be the most effective control options to reduce UK population exposure to ozone (in terms of precursors to be targeted) and on what scale should they operate? The Group may include discussion of the types of controls they consider to be feasible, but do not need to consider the policy implications of such measures.*

Short answer to question G

286. The ozone precursor compounds of relevance are methane, non-methane volatile organic compounds (VOCs), oxides of nitrogen (NO_x) and carbon monoxide (CO). While UK action can be beneficial, effective control of ozone concentrations in the UK will require emission reductions to be implemented throughout Europe and increasingly the entire northern hemisphere. Local actions, especially those of a short-term nature to address episodes of high ozone concentrations, have generally had, or been simulated to have, limited benefits.

- **Control of VOC emissions will almost always lead to an improvement in ozone air quality and a reduction in population exposure. Additional benefits result from concerted international action and from focussing the emission control on those source sectors making the largest contributions to ozone formation.**
- **Methane mitigation is seen as a cost-effective strategy on the global scale, bringing multiple benefits for air quality, public health, agriculture and the climate system.**
- **Less attention has been paid to global CO emissions but their reduction also has the potential to reduce ozone exposure.**
- **The picture is more complicated for control of NO_x emissions; large emission reductions are generally needed for urban areas to overcome the initial ozone disbenefit. Control of the rising emissions of NO_x from shipping would also be beneficial to annual and summertime mean ozone in western Europe.**

Detailed answer to question G

287. Improvements in ozone air quality across Europe are expected as a result of the Gothenburg Protocol to the United Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (CLRTAP), the European Union's National Emission Ceilings Directive (NECD) and its draft Air Quality Directive implementing the Clean Air for Europe (CAFE) Thematic Strategy. As question F stated, the emission reductions proposed in the CAFE strategy will not however improve ozone air quality over the UK. The aim of this answer is to identify additional measures, over and above those agreed in current base case scenarios for 2010-2020, which would be effective in reducing UK population exposure to ozone.

288. A number of additional control measures were considered as part of the latest UK Air Quality Strategy (published in July 2007), although not all those originally identified were subsequently taken forward. It should be noted that only one of the original measures specifically addressed ozone. All the other measures were designed to reduce concentrations of oxides of nitrogen (NO_X) (and/or particulate matter), and although the reductions of nitrogen oxide emissions will have benefits in the broader context, such as reduced nitrogen deposition or oxidant formation on the regional scale, it will potentially have an adverse impact on ozone concentrations mainly because of a reduction in the NO_X-scavenging effect, especially in urban areas (see Chapters 2 and 5).

289. The measure designed to reduce ozone exposure sought to control volatile organic compound (VOC) emissions, largely through implementation of Stage II petrol vapour recovery and abatement of emissions from onshore/offshore oil tanker loading operations (the reductions were equivalent to ~9% reduction in UK annual VOC emissions). There was a reduction in ozone concentrations, and hence metrics, compared to the corresponding base case for all metrics considered but the improvements were very modest (< 0.2%). Slightly larger improvements were seen if the same level of VOC emission reduction occurred across the board from all UK anthropogenic source sectors, a result of the spatial distribution of the VOC emissions and the reactivity of the VOC emitted from different source sectors.

290. The CAFE Thematic Strategy also considered a maximum technically feasible reduction (MFR) scenario, suggesting scope for additional control of UK (and European) VOC and NO_X emissions. Using the same assumptions and input datasets as those used in the review of the Air Quality Strategy (Hayman *et al.,* 2006a), a limited set of further model runs was carried out to investigate the effect of larger reductions in these emissions. The emission reductions used were not associated with specific measures but were selected simply to illustrate their effect on ozone concentrations. Reductions in VOC emissions in both the UK and the rest of Europe (from a 2020 base) led to corresponding reductions in ozone concentrations in rural and urban areas of the UK. Reductions in NO_X emissions in both the UK and the rest of Europe would generally reduce ozone concentrations in rural areas of the UK, but would initially lead to increases in ozone concentrations in urban areas. This trend would be reversed in urban areas for reductions in NO_X emissions in excess of ~30% (~60% in London) from the 2020 base. Although these reductions reverse the ozone concentration increases, they would not necessarily be sufficient to bring the values of the ozone metrics to levels below those modelled for 2003. The most effective way of doing this would be to reduce both NO_X and VOC emissions in the UK and the rest of Europe by at least 60%. Although not modelled, further benefits would be expected to accrue from concerted action taken on the global/hemisphere scale, as discussed in the following paragraph.

291. Reductions of NO_X and VOC emissions in Europe will not only reduce further high (peak) ozone levels over Europe but will also reduce Europe's contribution to hemispheric ozone. However, effective control of hemispheric ozone, and its contribution to background ozone, requires the reduction of emissions of NO_X, VOC, methane and carbon monoxide throughout the entire northern hemisphere:

- Reductions in global emissions of methane, in particular, could be effective at reducing levels of background ozone. There would be additional climate benefits since both ozone and methane are greenhouse gases. A 20% reduction in global methane concentrations was simulated to have the same impact on background surface ozone in Europe as a 20% reduction in both European NO_x and VOC emissions, although the latter has the more immediate impact. Although global methane concentrations have levelled off in recent years, there is no clear explanation of this change and it may be transient. Projected trends in anthropogenically-influenced sources of methane show a continuing increase, driven by increasing population and industrialisation. Regardless of future trends, however, methane mitigation could be a cost-effective strategy globally, bringing multiple benefits for air quality, public health, agriculture and the climate.

- Whilst NO_x control will generally lead to reductions in ozone, especially in summer, over northern Europe (including the UK), reduction of NO_x alone (that is, without reduction in VOC) may lead to increased ozone, particularly in winter and/or in areas close to busy shipping waters. Various studies have highlighted the importance of NO_x emissions from shipping. The exact spatial pattern of shipping's impact on ozone depends on the particular "photochemical climate". Whilst reduction in shipping emissions of NO_x was simulated to yield net benefit overall on surface ozone over Europe, in some localities (English Channel, southern North Sea and Baltic), NO_x emissions from shipping currently reduce local ozone by NO_x scavenging such that reducing these emissions may lead to an increase in ozone in these areas.

- Less attention has been paid to the assessment of carbon monoxide emissions and their potential to influence background ozone concentrations and radiative forcing. Model calculations have suggested that emissions of carbon monoxide in the northern hemisphere are currently substantially underestimated. However, since carbon monoxide has a lifetime of several months it is clear that, as for methane, the trajectory of future hemispheric carbon monoxide emissions has the potential to influence hemispheric ozone air quality significantly.

292. Within the EU, transport was the largest source of ozone precursor emissions in 2007. Because of the marked reductions in VOC emissions from the transport sector by 2020, a different range of source categories was found to be responsible for summertime photochemical ozone formation; these were stationary sources in sectors associated with the chemical, oil and gas industries and the manufacturing industries that use solvents. Subsequent work also investigated the ozone benefits resulting from the replacement of highly reactive aromatic species by low reactivity species and concluded that a much more focussed approach on the most important solvent sub-sectors identified by their Photochemical Ozone Creation Potential (POCP) would appear to be more effective than unselective reduction of VOC emissions.

293. As VOC emissions from man-made sources decrease, biogenic sources will become more significant. The extent of any changes in future ozone concentrations will be increasingly influenced by such biogenic emissions. In addition, if the incidence of high temperature episodes increases, then the very

strong temperature dependence of biogenic emissions may lead to increases over current levels (although other factors related to environmental change will also certainly affect future emissions, as discussed in greater detail in Chapter 4). Human influence in determining which tree species are planted could be significant, as emissions vary considerably between species.

294. The modelling results described in the preceding paragraphs are generally based on reductions in annual emissions of ozone precursor species. Article 7 of the 3rd Air Quality Daughter Directive on ozone addresses the issue of short-term action plans when hourly ozone concentrations exceed the alert threshold of 240 $\mu g\ m^{-3}$. There have been few exceedences of this threshold in the UK in recent years. A study of an episode that occurred in the UK in July 1999 concluded that it was difficult to identify any realistic and beneficial short-term actions in the UK. Although this conclusion was based on a single episode at a single site, it concurred with the outcome of experiments in Germany and France that concerted large-scale interventions were needed for decisive reductions of peak ozone concentrations. More recent calculations have again shown that VOC emission control reduces ozone concentrations throughout the year, but control of NO_X emissions would have a more complex effect with day-to-day differences.

Supporting evidence for question G

8.1 Overview

295. Controls on NO_X and VOC emissions introduced throughout Europe since the late 1980s have been effective in reducing photochemical ozone production and peak ozone concentrations in both the UK and across Europe (see responses to questions A and D; Jonson *et al.*, 2006; NEPAP, 2005) but there are still widespread exceedences of ozone air quality objectives across Europe (see for example EEA, 2007). Further improvements in ozone air quality across Europe were expected as a result of the Gothenburg Protocol to the UNECE Convention on Long-Range Transboundary Air Pollution (CLRTAP), the European Union's National Emission Ceilings Directive (NECD) and its Air Quality Directive implementing the Clean Air for Europe (CAFE) Thematic Strategy. As Chapter 7 indicates, the emission reductions proposed in the CAFE strategy do not, however, improve ozone air quality in the UK for the metrics considered. The aim of this response is to identify additional specific measures (where possible) over and above those agreed in current base case scenarios for 2010-2020, which would be effective in reducing UK population exposure to ozone.

296. A reduction in the UK population exposure to ozone implies a focus on suburban and urban areas, although exposures are still higher in the surrounding rural areas. As indicated in Chapter 2 and Chapter 5, the reduction in NO_X emissions in urban areas has led to increased ozone concentrations and hence exposure.[5] The existence, or otherwise, of a threshold for effects on human health is critical as it will determine (a) the

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[5] The reduction in NO_X emissions has been driven largely to reduce the impact of nitrogen dioxide (NO_2) on human health but this has had an adverse effect on ozone air quality (albeit from a generally low base). While urban ozone air quality would improve if NO_X emissions were allowed to increase, this is not seen as a desirable outcome. The integrated assessment modelling undertaken for the CAFE strategy specifically excluded this outcome.

ozone metric to use for the assessment and (b) the extent to which the increases in urban areas drive the policy response. The ozone metrics with and without the cut-offs are likely to respond differently both spatially and to the level of emission reductions that may be required. The increasing background concentration arising from global changes clearly indicates that future policy action on ozone air quality is inextricably linked to that on climate change and greenhouse gas emissions.

297. The UK's particular geographic position on the north-west, and generally upwind, coastal fringe of Europe is a further important consideration. It means that, with the exception of south-east England, the UK's "chemical climatology" can frequently be somewhat different from that of central and southern Europe. This has a major influence in determining the level of emissions reductions needed and whether these need to be implemented at the national, European or global scales.

298. Ground-level ozone not only affects human health but also crops and other vegetation. These impacts will shift the focus away from urban areas. Further, different ozone metrics are used to assess the impacts. The response of these metrics may again differ from those used to assess the impact on human health, both spatially and in terms of emission reductions required to achieve relevant ozone air quality standards and objectives.

299. In this supporting evidence, we start by considering additional specific measures (where possible) which would reduce ozone exposure over extended periods. The ozone modelling undertaken on the national scale and for London for the UK Air Quality Strategy published in 2007 is reviewed. There is scope for additional reductions of NO_X and VOC emissions beyond those considered in the Air Quality Strategy and model calculations are presented to show the effect of further UK and UK/European emission reductions of these species in 2020. Other modelling studies emphasised the significant changes expected in the contribution and hence importance of different VOC source sectors to ozone formation. With the increasing recognition of the influence of global and hemispheric changes in ozone on national and local concentrations, modelling studies investigating possible regional or hemispheric controls on ozone precursor emissions (i.e. methane, oxides of nitrogen, carbon monoxide) are summarised. As VOC emissions from man-made sources are reduced, biogenic VOCs will become more significant and a section is included on this topic. The supporting information concludes with a consideration of exposure during ozone pollution episodes. Model results are presented that describe the sensitivity to reductions of NO_X or VOC emissions and the limited effectiveness of possible short-term actions to reduce ozone concentrations during such episodes.

300. The studies described in this supporting information present results of emission reduction calculations. It should be noted that many of the emission reductions used were not necessarily associated with specific measures but were chosen to indicate the response or sensitivity to a significant emission reduction. Additional information is given in Annex 4.

8.2 Ozone exposure over extended periods

8.2.1 Ozone policy options at the national scale

8.2.1.1 The UK Air Quality Strategy

301. As part of the Review of the UK Air Quality Strategy,[6] modelling was undertaken to assess the impact on ground-level ozone for future base case scenarios (2010, 2015 and 2020) and a number of additional measures developed for the review (Hayman *et al.*, 2006a; Williams *et al.,* 2006). Only one of the original measures (Measure M) was specifically designed to address ozone through control of VOC emissions. Most of the other measures were designed to reduce concentrations of NO_X (and/or particulate matter) and as such would potentially have an adverse impact of ozone (see Table 8-1 for a list of the measures and the emission reductions associated with the measures).

302. Hayman *et al.* (2006a) used the Ozone Source-Receptor model (OSRM) to assess the impact on ozone at the national scale. UK emission projections for 2010, 2015 and 2020 were taken from the UK National Atmospheric Emissions Inventory (NAEI) programme (see Table 8-1) and the non-UK emissions were based on the International Institute for Applied Systems Analysis (IIASA) scenario – "current legislation including climate change". Allowance was made for a change to atmospheric composition arising from climate change. Using the monthly trends provided by Derwent *et al.* (2006), this would cause a change in the initial daily ozone concentrations from 2003 values ranging from -1.7 to +3.2 µg m^{-3} by 2010 and from -3.1 to +5.9 µg m^{-3} by 2020. The calculations were undertaken to a 10 km x 10 km grid covering the UK and the model runs used meteorology for 2003, a photochemically-active year (although some runs were undertaken with 2000 or 2002 meteorology to assess the sensitivity to year-to-year variability in meteorology).

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[6] The UK Government and the Devolved Administrations published the latest Air Quality Strategy for England, Scotland, Wales and Northern Ireland on 17th July 2007 (Defra, 2007). This followed a consultation on the Review of the Air Quality Strategy in 2006.

Table 8-1 Projected UK NO_X and VOC emissions for the base case and the measures in the Review of the UK Air Quality Strategy modelled using the OSRM for 2010, 2015 and 2020.

Measure(s)	Description	Total UK NO_X emissions (ktonnes per annum)		
		2010	**2015**	**2020**
Base	Base case projections	1118.5	992.1	869.1
A	Introduction of EURO V/VI vehicle emission standards (low reduction scenario)	1115.9	958.3	803.7
B	Introduction of EURO V/VI (high reduction scenario)	1109.6	914.1	727.8
B*	An earlier version of the introduction of EURO V/VI (high reduction scenario)	1109.6	11.6	712.7
C	Early uptake of EURO V/VI	1107.8	946.5	799.4
E	Introduction of low emission vehicles	1117.0	986.6	857.9
J	Domestic combustion	1115.6	984.3	856.2
K & L	Control on power stations, iron and steel industry, and oil refineries	924.1	796.0	785.0
L	Control on small combustion plant (20-50 MW) (= base case in 2010 as no NO_X emission reduction until 2013)	1118.5	976.4	852.9
M	Petrol vapour recovery from petrol stations and abatement of VOC emissions from onshore and offshore loading of crude oil (= base case emissions as only VOC emission reduction)	1118.5	992.1	869.1
O	A combination of Measures C and E	1106.4	941.8	790.1
P	A combination of Measures C and L	1107.8	930.8	783.2
Q	A combination of Measures C, E and L	1106.4	926.1	773.9
M & Q	A combination of Measures C, E, L (=Q) and M	1106.4	926.1	773.9
Measure(s)	**Description**	**Total UK VOC emissions (ktonnes per annum)**		
		2010	**2015**	**2020**
Base	Base case projections	1026.2	1034.7	1061.3
M	Petrol vapour recovery from petrol stations and abatement of VOC emissions from onshore and offshore loading of crude oil	952.2	958.4	983.0
M & Q	Combination of selected NO_X and VOC control measures	952.2	958.4	983.0

Notes: The above VOC emission totals include a contribution from natural sources of 178 ktonnes per annum. This sectoral source was not used in the OSRM modelling and the VOC emissions were generated using a biogenic emission potential inventory.

303. Seven ozone metrics were derived from the calculated hourly ozone concentration, of which four were related to impacts on human health. The results for these four metrics were presented as population-weighted means for the whole of the UK and for specific regions, as shown in Tables A4-1 to A4-4 (Annex 4). The full set of results for all the metrics calculated can be found in Hayman *et al*. (2006a). The population-weighted mean values were combined with risk functions in subsequent cost-benefit analyses to determine the population affected. The population exposure results were presented in the main consultation document for the review of the Air Quality Strategy (Defra, 2006).

304. As shown in Tables A4-1 to A4-4 (Annex 4), there was a progressive increase in ozone concentrations for the base case runs and hence the values of all the ozone metrics (i.e. a decline in ozone air quality) from 2003 through to 2020. As most of the measures that were modelled were focussed on control of NO_X (and particulate matter) emissions, the model results generally showed further increases in ozone concentrations and in the population-weighted metrics, and hence in population exposure. This was mainly because of a reduction in the NO_X-scavenging effect in urban areas (see Chapter 2 and Chapter 5)

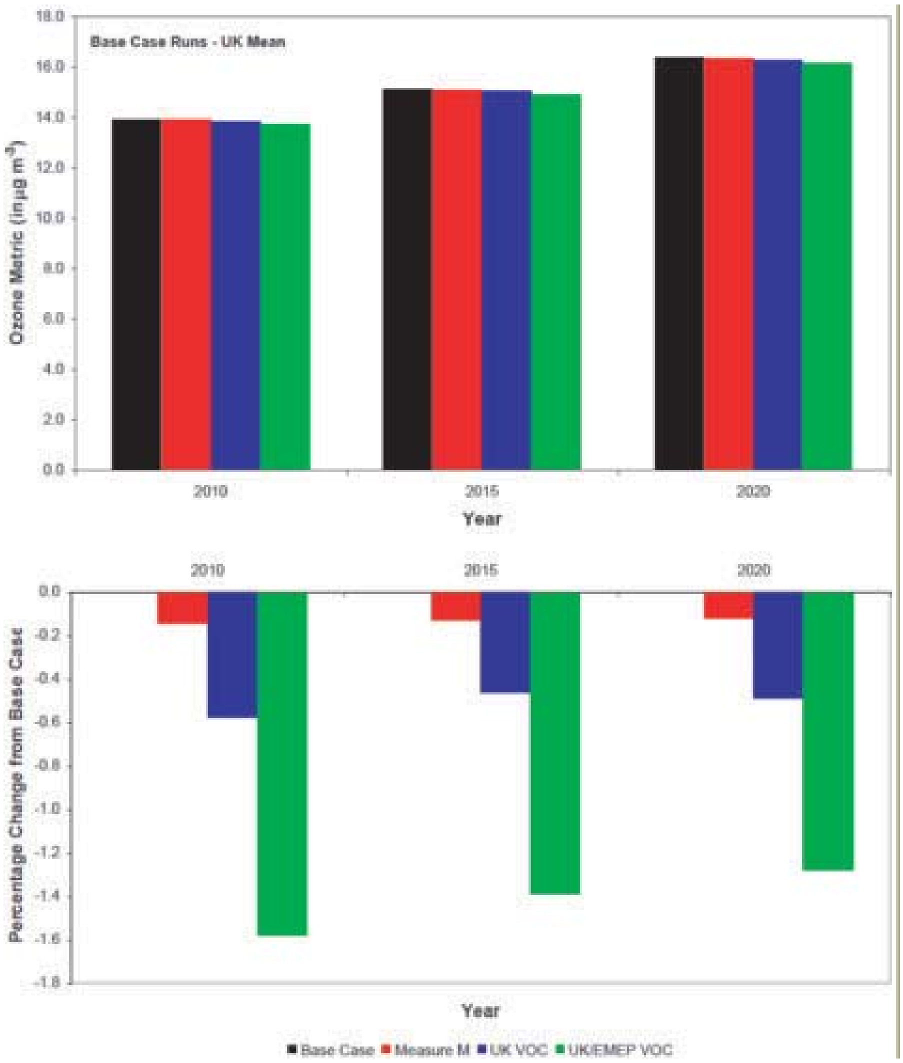

Figure 8.1 The response (upper panel) and percentage change from the base case (lower panels) of the metric – annual mean of the daily maximum of the 24 daily running 8-hour mean ozone concentrations using a 70 µg m^{-3} (i.e. 35 ppb) cut-off – calculated by the OSRM for the base case and for three VOC emission reduction scenarios: (a) Measure M in the Review of the UK Air Quality Strategy; (b) equivalent 9% across-the-board reductions of UK VOC emissions; and (c) 9% across-the-board reductions of UK and non-UK VOC emissions.

305. The exception was Measure M, which addressed reduction of UK VOC emissions (equivalent to ~9% of all UK emissions), largely through implementation of Stage II petrol vapour recovery and abatement of emissions from onshore/offshore oil tanker loading operations. There was a reduction in ozone concentrations (i.e. an improvement in ozone air quality) over the corresponding base case for all metrics considered but the improvements were very modest (<0.2% reduction in the value of the metric) and insufficient to offset the underlying increases in ozone concentrations from 2003 to 2020. This is illustrated in Figure 8.1 for the metric, the annual mean of the difference between the maximum daily running 8-hour average ozone concentration and a 70 µg m^{-3} (or 35 ppb) cut-off. As also shown in this figure, slightly larger improvements were however seen if the same level of emission reduction occurred from (i) all UK anthropogenic source sectors (0.5%-0.6% reductions in the value of the metrics) and (ii) all UK and European anthropogenic source sectors (1.3%-1.6% reductions). In part, this reflected differences in (a) the spatial pattern of the emissions and (b) the VOC speciation and hence reactivity profiles of the source sectors.

306. For the Review of the Air Quality Strategy, limited modelling of ozone concentrations was also undertaken using the ADMS-Urban model for the Greater London area (Williams *et al.*, 2006). As described in the response to question D (Chapter 5), the model runs used traffic flow and emissions data from the 2001 London Atmospheric Emissions Inventory,[7] together with meteorological data from Heathrow and background data from rural monitoring sites around London. The modelling of ozone in future years took into account changes to traffic flows, pollutant emission rates and background concentrations. A single additional measure – Measure Q – was investigated. This measure represented a combination of other measures: the early uptake of EURO V/VI (Measure C, see Table 8-1), the introduction of low emission vehicles (Measure E) and the control on small combustion plant (20-50 MW) (Measure L). As presented in Table 8-2, the ADMS-Urban calculations also showed an increase in ozone for all four metrics used to assess human health impacts. These results are not directly comparable with those of the OSRM because of differences in the emission inventories, meteorology and assumptions of atmospheric composition. The results do however give an indication of the range of potential future ozone concentrations and metrics.

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[7] The latest available emission inventory at the time of the work.

Table 8-2 Population-weighted means calculated for Greater London for the Review of the Air Quality Strategy by ADMS-Urban for the four ozone metrics used to assess human health impacts for 2001, 2010 and 2020 base cases and for Measure Q in 2020.

	2001 Base	2010 Base	2020 Base	2020 Measure Q
Annual mean of daily maximum running 8-hour ozone concentrations	47.3	55.7	58.5	59.6
Annual mean of daily maximum running 8-hour ozone concentrations with a cut-off of 70 µg m^{-3}	2.42	3.96	4.67	4.88
Annual mean of daily maximum running 8-hour ozone concentrations with a cut-off of 100 µg m^{-3}	0.28	0.56	0.69	0.72
Number of days on which a running 8-hour ozone concentration exceeds 100 µg m^{-3}	7.6	13.1	15.6	16.0

307. In summary, the modelling of future ozone concentrations for the Review of the Air Quality Strategy indicated that, without additional measures, ozone air quality would deteriorate for both average and peak levels. Concentrations will still exceed the Air Quality Strategy's objective in 2020. Average levels were likely to rise in urban and rural areas.

8.2.1.2 Beyond the Review of the Air Quality Strategy

308. The CAFE Thematic Strategy considered larger reductions in both VOC and NO_X emissions in both the UK and Europe. UK NO_X and VOC emissions in the "maximum technically feasible reduction" (MFR) scenario for 2020 were 518 and 663 ktonnes per annum respectively, whereas the lowest levels modelled in the Review of the Air Quality Strategy were 713 and 805[8] ktonnes per annum respectively. The reduction in VOC emissions considered in Measure M of the Review of the UK Air Quality Strategy was about a third of that of the MFR scenario, suggesting scope for additional reduction.

309. A limited set of site-specific OSRM runs was subsequently undertaken to investigate the level of emission reduction needed to achieve ozone air quality objectives and target values. 41 receptor locations, corresponding to UK ozone monitoring sites, were modelled using the same assumptions and input datasets as the OSRM runs undertaken for the Review of the Air Quality Strategy. The site-specific calculations considered additional across-the-board emission reductions (20% to 80%) in 2020 NO_X and/or VOC emissions (see Table 8-3). Two series of runs were undertaken (i) UK action alone and (ii) combined UK and European action. These emission reductions were not associated with specific measures but were chosen simply to scope the scale of the reductions needed.

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[8] This does not include the contribution of 178 ktonnes per annum from biogenic sources which is included in the UK totals given in Table 8-1.

Table 8-3 The implied UK NO_X and VOC emissions used in the post-2020 OSRM model runs.

Year	NO_X emissions (ktonnes per annum)	VOC emissions (without natural) (ktonnes per annum)	VOC emissions (with natural) (ktonnes per annum)
2003 current year	1569.7	1088.6	1266.6
2020 base case	869.1	883.3	1061.3
with 20% emission reduction	695.3	706.6	884.6
with 30% emission reduction	608.4	618.3	796.3
with 40% emission reduction	521.5	530.0	708.0
with 50% emission reduction	434.6	441.6	619.6
with 60% emission reduction	347.7	353.3	531.3
with 70% emission reduction	260.7	265.0	443.0
with 80% emission reduction	173.8	176.7	354.7

Note: The NAEI assumed a contribution to VOC emissions from natural sources of 178 ktonnes per annum. This sectoral source was not used in the OSRM modelling and the VOC emissions were generated using a biogenic emission potential inventory. The emission reduction was only applied to the anthropogenic VOC source sectors.

310. As shown in Chapter 5, urban ozone concentrations can be represented by a regional component and an urban ozone decrement. Hence, results for rural sites are considered initially. Figure 8.2 presents the response at the Harwell and Strath Vaich sites of a number of ozone metrics: (a), (b) and (c) annual mean of the difference between maximum daily running 8-hour mean ozone concentrations with no cut-off and cut-offs of 70 and 100 µg m^{-3} and (d) AOT40 – crops.

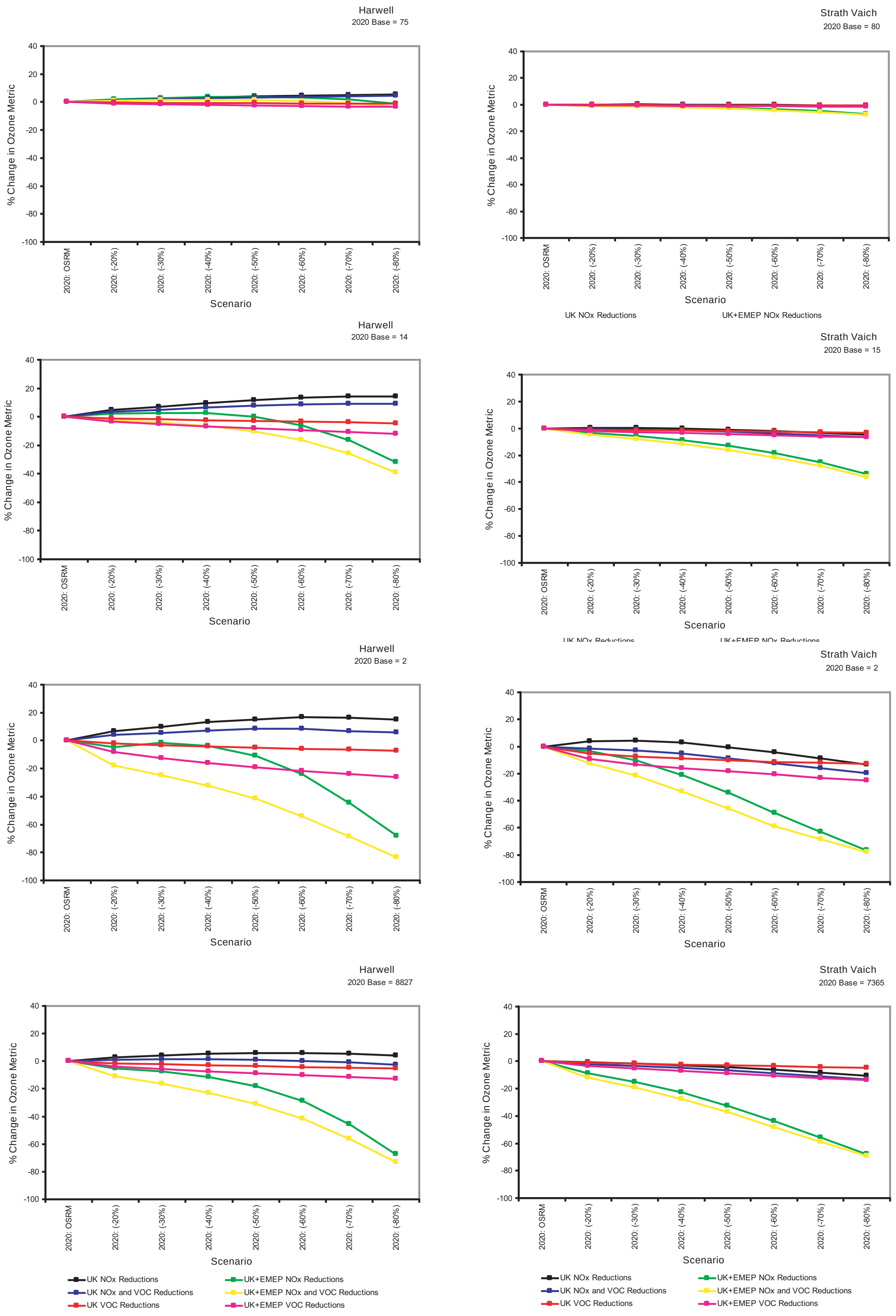

Figure 8.2 The percentage change in the ozone metrics: (a), (b) and (c) annual mean of the difference between maximum daily running 8-hour mean ozone concentrations with no cut-off and cut-offs of 70 and 100 µg m^{-3} and (d) AOT40 – crops, for additional post-2020 across-the-board reductions of NO$_X$ and/or VOC emissions at the Harwell (left-hand panels) and Strath Vaich (right-hand panels) sites, as calculated using OSRM.

311. Although the model results indicated that the effectiveness of the emission reduction was dependent on site and the ozone metric chosen, a number of general comments can be made:

- Metrics sensitive to higher ozone concentrations in the concentration distribution, such as the annual mean of the difference between maximum daily running 8-hour mean ozone concentrations with a cut-off or AOT40 – crops, show a larger response to emission reduction than metrics sensitive to the entire concentration distribution (e.g. annual mean of the maximum daily running 8-hour mean ozone concentrations);
- There are a range of responses to the reduction of UK NO_X emissions which depend on the site and metric. For some sites, there is an initial increase in the metric and substantial reductions in NO_X emissions are needed before there is a decrease in the metric;
- VOC emission reductions alone always improve ozone air quality;
- Combined reductions of VOC and NO_X emissions are more effective than reducing NO_X emissions alone;
- UK and European action is always more effective than UK action alone.

312. The influence of the higher NO_X emissions in urban areas is illustrated in Figure 8.3 for the sites: London Bloomsbury, London Eltham and Rochester Stoke, in urban, suburban and rural locations, respectively. As expected, the rural site at Rochester Stoke has higher ozone concentrations (and hence a higher value of the metric) than the suburban site at London Eltham, which is also higher than the urban site at London Bloomsbury. The ozone metrics at the urban sites do however tend towards the surrounding rural value as the local NO_X-scavenging effect decreases (see Chapter 5).

313. Overall, the results suggested that, as in the CAFE work, health-related ozone metrics would increase in the base case scenario between now and 2020. From the 2020 base and as above, reductions in VOC emissions in both the UK and in the rest of Europe led to a proportionate improvement in ozone in rural and urban areas of the UK, with this combined action being more effective than UK action alone. Reductions in NO_X emissions alone in both the UK and the rest of Europe will reduce ozone levels in rural areas of the UK, but will lead to increases in urban areas until this trend is reversed in urban areas for reductions in excess of ~30% (~60% in London) from a 2020 base. Although these reductions reverse the increasing ozone trend, they are not necessarily sufficient to bring the ozone metric to levels below those modelled for 2003. The most efficient way of doing this is to reduce both NO_X and VOC emissions in both the UK and the rest of Europe. The results suggest that reductions of the order of 60% or more for both pollutants will achieve this in rural and urban areas of the UK, whereas reductions of ~60%-70% would be required to achieve this in London. Although not modelled, further benefits would be expected to accrue from concerted action taken on the global/hemispheric scale, as discussed later in section 8.2.3.

8.2.2 Ozone policy options at the European scale

314. The Gothenburg Protocol to the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP), the European Union's National Emission Ceilings Directive (NECD) and its Air Quality Directive implementing the Clean Air for Europe (CAFE) Thematic Strategy have been agreed to reduce the impacts of ozone on human health and ecosystems. The Gothenburg Protocol and NECD mandate country-specific emission limits to be achieved by 2010 for NO_X and VOCs. Emission limits are also defined for sulphur dioxide (SO_2) and ammonia (NH_3). The Gothenburg Protocol and NECD are subject to periodic reviews, which are currently in progress.

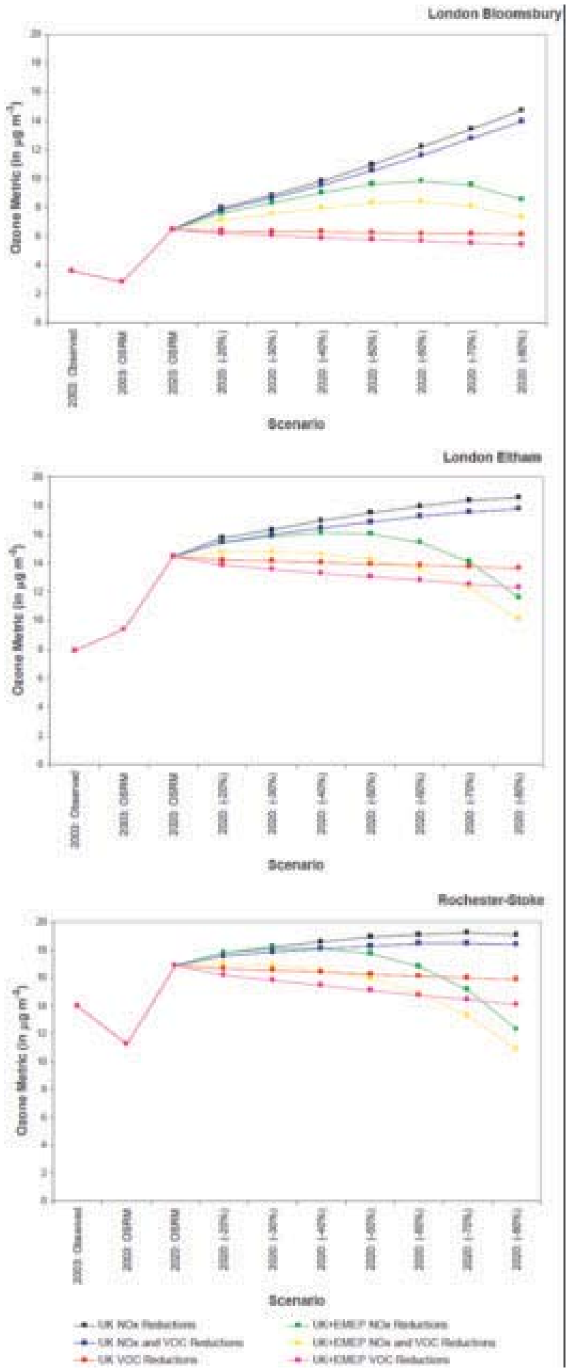

Figure 8.3 The effect of across-the-board reductions of 2020 NO_X and/or VOC emissions in (a) the UK and (b) UK and EMEP on the ozone metric – annual mean of the difference between maximum daily running 8-hour mean ozone concentrations with a 70 µg m^{-3} cut-off – as calculated by OSRM for the sites, London Bloomsbury, London Eltham and Rochester Stoke.

315. On ozone, the CAFE strategy aimed to cut mortality from ozone pollution by 10% by 2020 compared with the 2000 levels (as well as reducing the impacts on ecosystems). Overall, the CAFE strategy proposed reductions in the emissions of nitrogen oxides by 60% and in VOC emissions by 51% within the EU between 2000 and 2020. These European policy objectives have been underpinned by the integrated assessment modelling of IIASA, which was the subject of question F (Chapter 7) and is not considered further here.

316. The focus of this section is on the contribution to ozone formation made by different VOC source sectors and how the relative contributions and hence the importance of the sectors will change by 2020 because of planned emission controls. Subsequent studies have investigated the benefits to ozone resulting from the replacement of highly reactive aromatic species by low reactivity species.

8.2.2.1 Ozone formation from different VOC source sectors

317. Within the European Union, transport was the largest source of ozone precursor emissions (NO_X, VOCs, carbon monoxide (CO) and methane (CH_4)). Figure 8.4 shows the sector split of emissions of ozone precursors for member countries and groupings of the European Environment Agency in 2007. The emissions of the individual pollutants have been multiplied by a tropospheric ozone formation potential factor prior to aggregation. The factors used were VOCs: 1.00; NO_X: 1.22; CO: 0.11; and CH_4: 0.014 (de Leeuw, 2002). The resulting emissions are expressed in terms of VOC equivalents (ktonnes).

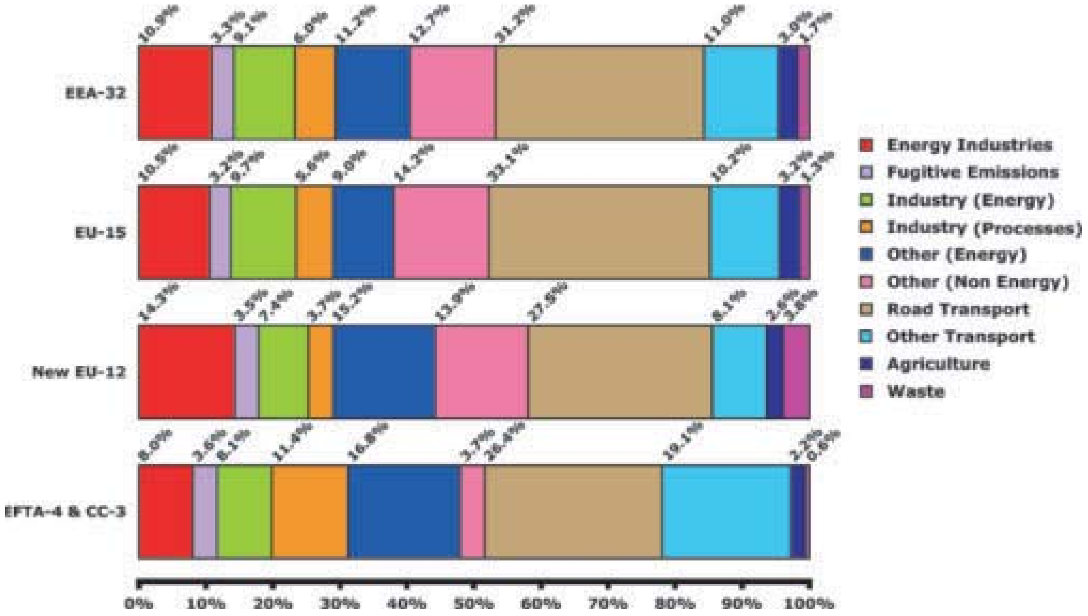

Figure 8.4 Sector split of weighted emissions of ozone precursors (NO_X, VOCs, carbon monoxide and methane) in 2007 for Member Countries of the European Environment Agency (EEA) and groupings used to derive the EEA's Core Indicator CSI002 on ozone precursor emissions (taken from http://themes.eea.europa.eu/IMS/CSI).

318. As part of the UK National Atmospheric Emissions Inventory (NAEI) programme, speciated VOC emission inventories (664 VOCs from 248 source sectors) have been derived for the years 2000, 2010 and 2020 (based on the 2002 NAEI inventory year).

Derwent and co-workers (see section 8.5 of Hayman *et al.,* 2006b) used these inventories in the UK Photochemical Trajectory Model (UK PTM). The model incorporated an extended version of the Master Chemical Mechanism (MCM), which considered 176 of the 664 VOCs, accounting for ~90% of the total mass emission in the 2000 inventory.

319. The model described photochemical ozone formation along the idealised photochemical episode trajectory giving the highest ozone concentration over the UK. The same speciation of the VOC emissions was assumed along the trajectory as that derived from the NAEI emission inventory. A series of sensitivity experiments was performed. At each point along the trajectory, the VOC emissions contributing to a specific source sector were increased by a nominal fraction (the choice of 7.3% was completely arbitrary and had no policy significance) and these additional emissions increased photochemical ozone production over the base case experiment. The additional ozone present at the end-point of the trajectory was taken as a measure of the propensity for regional-scale ozone formation from that source category and was used to derive the incremental reactivity of that source category.

320. The source categories that made the largest contribution to photochemical ozone formation in 2000 were associated with road transport (cars with and without catalysts). These categories were in first and second places by virtue of their large percentage contributions to total VOC emissions and their relatively high incremental reactivities. The source category with the largest incremental reactivity was chemical waste incineration but, because of its relatively low percentage contribution to total VOC emissions, it contributed little to ozone formation.

321. Assuming that there would be no change in the incremental reactivities between 2000 and 2010, peak ozone was reduced by 25.3 μg m^{-3} on the standard base case UK PTM model trajectory from Austria to the United Kingdom, when the 2000 VOC inventory was substituted with that for 2010 (i.e. reflecting the overall mass emission reduction that has occurred between 2000 and 2010). The reduction was somewhat smaller at 24.4 μg m^{-3} when the substitution was made with the 2020 inventory (corresponding to the trend in the total VOC emissions, see Table 8-1).

Table 8-4 The VOC emission source categories that gave the greatest contributions to the decline in episodic peak ozone concentrations between the years 2000 and 2010.

Source category	Contribution to the decrease in peak ozone concentrations between 2000 and 2010 (in µg m^{-3})
– Cars without catalysts	9.6
– Cars with catalysts	2.2
– LGVs without catalysts	0.8
– Rigid HGVs	0.6
All road transport	13.2
Onshore loading of crude oil	1.4
Offshore loading of crude oil	1.0
Surface cleaning with trichloroethene	0.6
Domestic combustion of coal	0.6
Offshore oil and gas industries	0.6
Chemical industry	0.6
Decorative paint trade	0.6
Gas leakage	0.6
House and garden machinery	0.6

322. Table 8-4 gives the VOC emission source categories that contributed most to the decrease in episodic peak ozone concentrations between the years 2000 and 2010 using these Defra) VOC emissions projections. By far the largest contribution to the decline in ozone of 13.2 µg m^{-3}, or more than about one half of the total, came from the reduction in VOC emissions in the road transport sector (cars with and without catalysts, LGVs and HGVs). The next largest combined contribution, amounting to about 15% of the ozone decline, came from VOC emission reductions in the stationary source sectors of the chemical, oil and gas industries.

323. In 2020, because of the marked reductions in VOC emissions in the transport sector, a different range of source categories will be responsible for the bulk of the ozone formation. Table 8-5 shows, in order of importance, the major ozone-producing sectors in 2020. The entries were dominated by stationary VOC sources associated with the chemical, oil and gas industries, and with the manufacturing industries that use solvents. The only sector listed associated with road transport was that of petrol stations and the emission of VOC from refuelling motor vehicles with unleaded petrol. Table 8-5 points to a major shift in policy if ozone levels are to be reduced beyond 2010 levels so that stationary VOC sources are targeted rather than motor vehicles.

Table 8-5 The VOC emission source categories that gave the greatest contributions to episodic peak ozone concentrations in 2020.

Source category	Contribution to peak ozone concentrations in 2020 (in μg m^{-3})
Chemical industry	3.2
Spirit manufacture (maturation)	2.2
Onshore loading of crude oil	1.8
Road transport (cars with catalysts)	1.4
Refineries (process fugitives)	1.4
Industrial adhesives	1.4
Petrol stations (vehicle refuelling with UP)	1.4
Offshore loading of crude oil	1.4
Industrial coatings (metal plastic)	1.2
Aerosols (cosmetics and toiletries)	1.2
Other solvent use	1.2
Car painting and refinishing	1.0

Notes: UP = unleaded petrol

324. Derwent *et al.* (2007a, 2007b) have also investigated the ozone benefits resulting from the replacement of highly reactive aromatic species by low reactivity species. Three simple illustrative strategies were formulated in which the emissions of the xylenes, trimethylbenzenes and all aromatic species were replaced or substituted by the emission of one of a potential range of 94 organic compounds. Of these, eight alcohols, six esters, six ketones, three ethers, a cycloalkane and a glycol ether were found to give greater ozone reductions on substitution for the aromatic compounds than would be obtained from the corresponding mass-based control policy. Derwent *et al.* (2007b) concluded that a much more focussed approach on the most important solvent sub-sectors identified by their Photochemical Ozone Creation Potential (POCP) would appear to be more effective, environmentally and financially.

325. It is also worth noting an earlier study by Derwent and Nelson (2003) for the Environment Agency. The UK PTM, with a previous version of the Master Chemical Mechanism, was used to derive an Integrated Downwind Ozone Potential (IDOP) for ~120 VOCs. The IDOP was defined as the excess ozone production, integrated over the downwind environment, for a 1 tonne hr^{-1} industrial emission source. This ozone production can also be expressed as an ozone-equivalent emission ceiling for each emitted VOC species (in tonne hr^{-1}) which, when exceeded, would lead to ozone concentrations in the downwind environment that were ~10 μg m^{-3} higher than in the absence of that industrial VOC source. The IDOP and POCP have similar rankings of VOC reactivity.

326. Houston, Texas, is noted for some of the highest ozone levels in the USA and measurement campaigns have linked these ozone excursions to emissions of ethene and propene from industrial sources. As part of the Texas 2000 Air Quality Study, Ryerson *et al.* (2003) drew upon the work of Derwent and Nelson (2003) and its conclusion that downwind ozone production was dependent upon only a limited set of all the possible organic compounds known to be emitted from petrochemical sources. Furthermore, the ozone productivities of ethene and propene defined in the Environment Agency study were able to account qualitatively for the formation of the ozone excursions in Houston.

8.2.3 Ozone policy options at the global scale

327. Allowance was made in the OSRM model runs for the Review of the Air Quality Strategy for a change to atmospheric composition arising from global changes and this had a significant impact on the calculated future ozone air quality. Sensitivity runs were undertaken for the emission projections for 2010 and 2020 with a current atmosphere (2003) and future atmospheric composition based on the business-as-usual scenario with climate change (see section 4.4.2 of Hayman *et al.*, 2006a). The changes in the initial daily ozone concentrations from the 2003 values ranged from -1.7 to +3.2 µg m^{-3} in 2010 and from -3.1 to +5.9 µg m^{-3} in 2020. Figure 8.5, shows the sensitivity of the different population- and area-weighted ozone and nitrogen dioxide metrics for the UK for the 2010 and 2020 base case runs, with and without changing atmospheric composition, relative to the 2003 current year (=1).

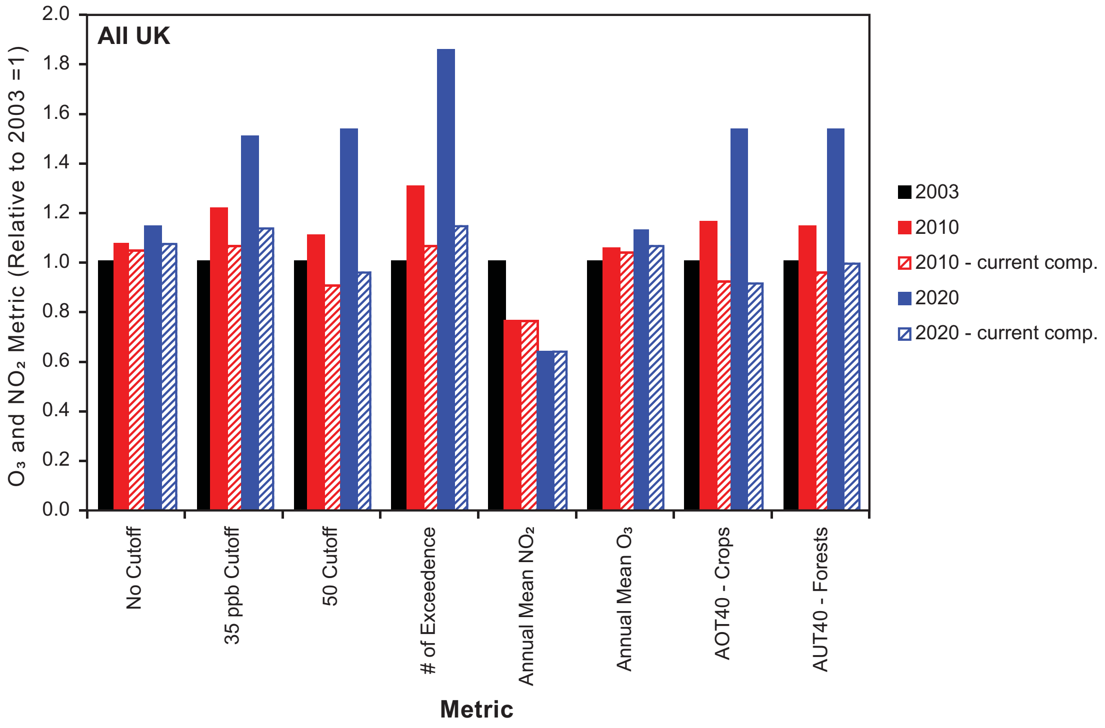

Figure 8.5 The sensitivity of different ozone and nitrogen dioxide metrics calculated by the OSRM for the 2010 (red) and 2020 (blue) base case runs (with changing (solid bars) and current (hashed bars) atmospheric composition) relative to 2003 (=1, solid black bars) for the UK.

328. There were significant and increasing differences for the ozone metrics between the two sets of runs. The greatest differences occur for the ozone metrics with an exceedence of a threshold, i.e. annual mean of the maximum daily running 8-hour average ozone concentration with a 70 or 100 µg m^{-3} cut-off, the number of days when the maximum of the 24 possible running 8-hour mean concentrations in each day exceeded 100 µg m^{-3} and the two AOT40 metrics. There was little effect on the annual mean NO_2 concentrations between the runs with and without a changing atmospheric composition.

329. The increasing background concentration arising from global changes clearly indicated that future policy action on ozone air quality was, and is, inextricably linked to that on climate change and greenhouse gas emissions.

8.2.3.1 Methane

330. Background (hemispheric) concentrations of tropospheric ozone are sensitive to methane emissions, yet methane mitigation is predominantly considered in respect of its impact on radiative forcing, rather than its impact on air quality. Methane is relatively long-lived in the atmosphere (~8-9 years) so controls on global methane will produce ozone reductions that are widespread globally, and which are sustained on the same timescale (West and Fiore, 2005). Model simulations have shown that, globally, tropospheric ozone responds approximately linearly to anthropogenic methane emissions changes. For example, reducing global anthropogenic methane emissions by 20% beginning in 2010 was simulated to decrease the average daily maximum 8-hourly surface ozone concentrations by approximately 2 µg m^{-3} on average globally by 2030. This strategy was estimated to be cost effective in terms of avoided global ozone-related mortality alone (West *et al.*, 2006).

331. Observations have now clearly revealed a slow-down in the rate of increase of global methane, with atmospheric concentrations currently levelling off at ~1750 ppb (Khalil *et al.,* 2007). The reasons for the trend towards apparent steady state (constant emission rates) were not entirely clear, but a strong possibility was that decreased emissions from agriculture, particularly from rice growing in China (Khalil and Shearer, 2006), and from fossil fuel processing following the break-up of the Soviet Union (Bousquet *et al*., 2006) had offset increases from other sectors. Other suggestions included an increased prevalence of tropical thunderstorms creating more NO_X that has enhanced methane destruction, and a decline in total methane emissions from tropical land plants because of deforestation (Houweling *et al*., 2006; Keppler *et al*., 2006). However, despite the current observations, recent analyses have suggested that anthropogenically-influenced sources of methane have increased again (Bousquet *et al*., 2006), driven by increasing population and industrialisation, so that global methane control remains a viable option for control of background ozone (and of an important contribution to radiative forcing).

8.2.3.2 NO_X and VOCs

332. In contrast to methane, controls on NO_X and VOC emissions, which are relatively short-lived in the atmosphere, will tend to impact more on local and regional ozone generation than on hemispheric background ozone, and consequently predominantly will act to reduce regional high-ozone episodes in regions where emissions of these precursors are relatively high. At the same time however, reductions of NO_X and VOC will also increase the lifetime of methane (by decreasing the average hydroxyl radical (OH) concentration), which will in turn cause an increase in average ozone in the long-term. This indirect influence on ozone was found to partially offset some of the direct benefit of decreased ozone production resulting from methane and NO_X emissions reductions (Dentener *et al*., 2005; West *et al*., 2007).

333. The UN Task Force on Hemispheric Transport of Air Pollution (HTAP) has compiled results from more than 20 global models that have simulated the impact on surface ozone over Europe (and other receptor regions) of a 20% reduction in NO_X, VOC and carbon monoxide emissions from each of a number of industrialised source regions for these pollutants (LRTAP, 2007). The geographical regions defined in the HTAP study are illustrated in Figure 8.6, and the mean model-simulated decreases in average surface ozone over the Europe source region for the months of March, April and May are shown in Figure 8.7.

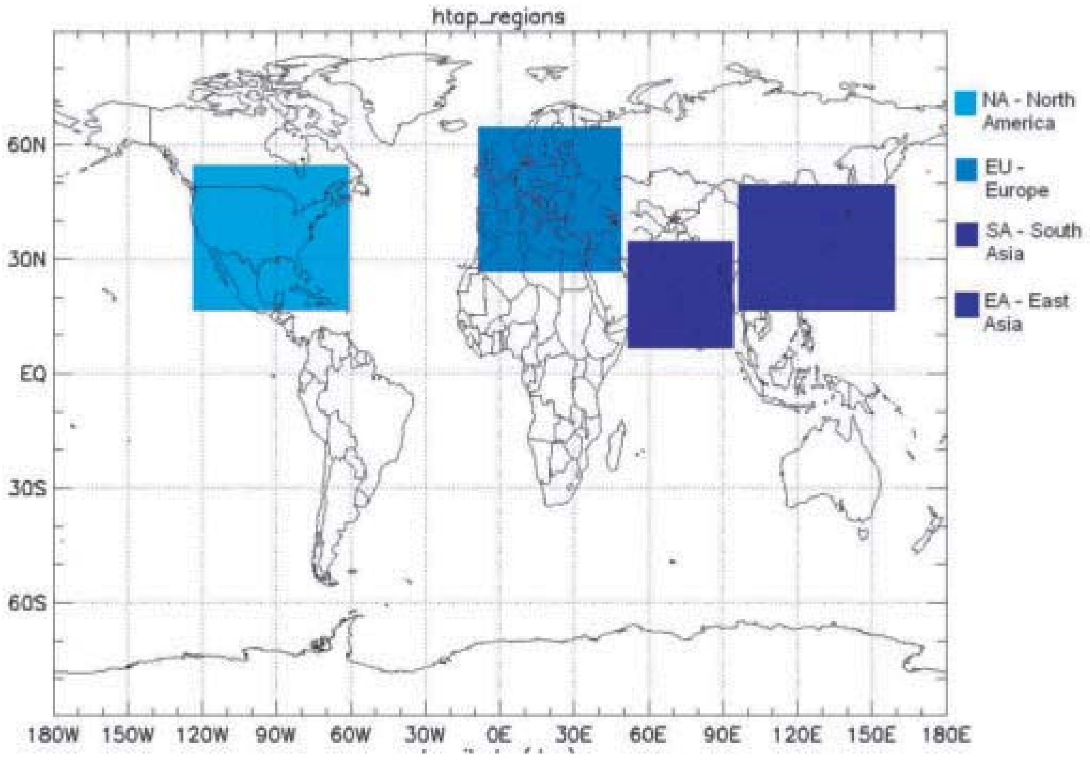

Figure 8.6 Source-receptor regions defined by the HTAP Task Force: (LRTAP, 2007).

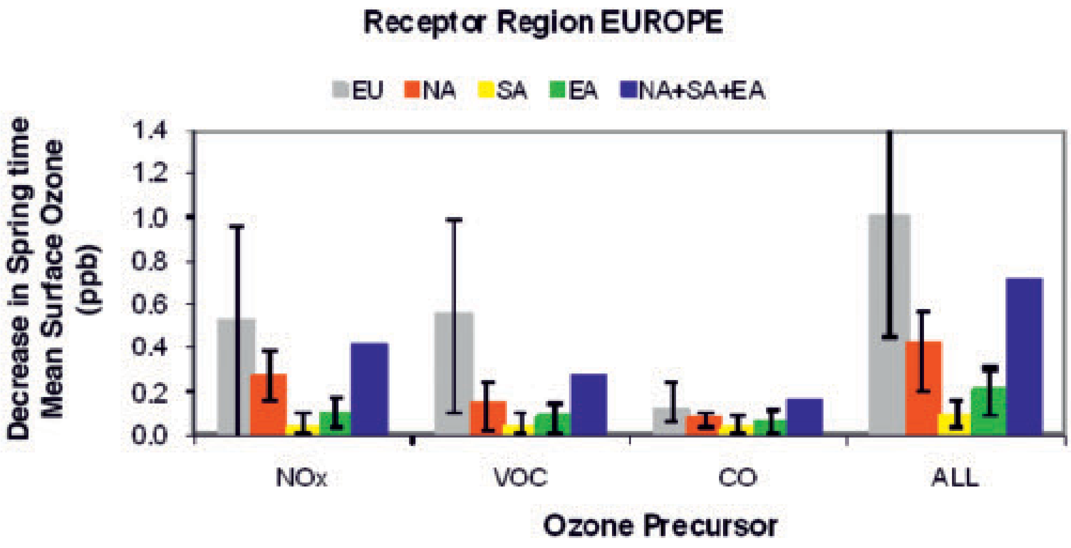

Figure 8.7 The decrease in springtime (March-April-May) mean surface ozone (ppb) in the Europe HTAP receptor region resulting from 20% lower emissions of ozone precursors NO_x, VOC and carbon monoxide in each of the four source regions (EU-Europe, i.e. application of "domestic" emissions reductions only; NA-North America; SA-South Asia; EA-East Asia). The ALL category is the result of applying the 20% reduction to all NO_x, VOC and carbon monoxide emissions simultaneously in the stated source region. The 5th bar in each group is the impact on European ozone of precursor reductions in the three non-European source regions together. The bars denote the Multi-Model Mean Response and the whiskers span the full range of the individual model responses.(LRTAP, 2007).

334. Figure 8.7 shows that, when considering these particular ozone precursor species, springtime surface ozone in Europe was most sensitive to reductions of precursors within Europe itself, as expected. The next greatest sensitivity was to reductions in emissions in North America, reflecting the prevailing west-east air mass transport at these latitudes. Nevertheless, even for these relatively short-lived precursors, the ensemble-mean sensitivity of European springtime ozone to 20% decreases in ozone precursors in the three non-Europe source regions together was always greater than half the ozone response to the equivalent decreases within Europe. (In the case of CO, the response of European ozone to carbon monoxide reductions in the three non-Europe source regions was as great as the response to carbon monoxide reduction in Europe alone.) The response of annual mean surface ozone in Europe to reductions of precursors from the three non-Europe source regions was even greater than the response of peak-season ozone.

335. A key observation is that European surface ozone was as sensitive to VOC emission reductions as to NO_x emission reductions, particularly with respect to domestic VOC emissions. This greater sensitivity to VOC emissions across Europe as a whole was in contrast to the NO_x-limited regime that exists in the other three HTAP receptor regions. It is also important to note that the model outputs were for net changes in simulated mean surface ozone across Europe as a whole, and these masked the spatial variations within Europe in the response of ozone to NO_x and VOC emissions reductions. For example, winter ozone was simulated to increase over northern Europe in the scenario of a 20% reduction in NO_x alone.

336. The magnitude of the reductions in European average springtime surface ozone shown in Figure 8.7 for 20% reductions in NO_X, VOC (and CO) emissions should be compared with the simulations showing that a 20% reduction in global methane concentration alone led to a reduction in background surface ozone, in the long term, of ~2 μg m^{-3}. Figure 8.7 shows that European reductions of 20% in both NO_X and VOC emissions together were necessary to achieve the same impact on average European ozone as a 20% reduction in global methane concentrations (but the timescales on which the different emissions reductions impact on ozone are different).

337. In the ACCENT intercomparison of 26 global models (see Chapter 4 and the response to question C for more detail and illustrations), the ensemble-mean simulation for a scenario in which all possible technical control options on ozone precursors were implemented worldwide (the MFR scenario) is a reduction of –5.6 μg m^{-3} in average surface ozone over Europe between the years 2000 and 2030 (Dentener *et al.,* 2006). This net reduction was predominantly driven by the significant reductions (of several μg m^{-3}) in the concentration of hemispheric background ozone in the air masses entering Europe (Derwent *et al*., 2006). The reduction in European ozone under the MFR scenario should be compared with the ensemble-mean simulated increase of 3.6 μg m^{-3} in average surface ozone across Europe by 2030 for implementation of the current legislation scenario on precursor emissions (Dentener *et al*., 2006). The MFR scenario illustrated the significant benefit to European ozone levels of implementation of stringent global-scale precursor emission control acting in addition to improvements brought about by precursor emission control at the regional scale.

8.2.3.3 NO_X emissions from shipping

338. One important, and comparatively under-regulated, source of ozone precursors (principally NO_X) is shipping (Derwent *et al*., 2005; Endresen *et al*., 2003; Eyring *et al*., 2005, 2007; Lawrence and Crutzen, 1999). World shipping has increased by 35% over the last 50 years and model simulations suggest that shipping currently contributes ~12 μg m^{-3} to annual mean and summertime surface ozone over the North Atlantic and western Europe (Derwent *et al*., 2005; Eyring *et al*., 2007). The exact impact of shipping on ozone at a more local scale in particularly congested sea-ways (e.g. English Channel, North Sea, Mediterranean, etc.) will depend on the particular local NO_X emission densities and photochemical climate. In some localities, NO_X emissions from shipping may currently limit local ozone generation, such that reducing emissions may lead to an increase in ozone in these areas. In addition, care must be taken in case models overestimate the impact of ship emissions on ozone photochemistry because of the shorter lifetime of NO_X within a ship exhaust plume compared with its lifetime within a larger model grid volume into which ship emissions are assumed to be diluted (von Glasow *et al*., 2003).

339. Emission scenarios predict that if no further control measures were implemented beyond existing International Maritime Organisation regulations, global NO_X emissions from shipping might increase with an annual growth rate of 1.7% between 2000 and 2030, becoming comparable in magnitude to global NO_X emissions from road traffic by 2050 (Eyring *et al*., 2005). On the other hand, this is a sector with large potential for NO_X reduction, and the aggressive

application of abatement measures that is suggested could reduce shipping NO_X emissions by 85% from year 2000 emissions by 2050, despite a growing fleet (Eyring *et al.*, 2005).

340. The impact of different shipping emission scenarios on future European surface ozone, derived from the ACCENT intercomparison of output from ten chemical transport models (CTMs) or general circulation models (GCMs), is shown in Figure 8.8 (Eyring *et al.*, 2007). The panels show the simulated changes in ozone in 2030 compared with 2000 assuming, from left to right, complete abatement of all shipping emissions in 2030, the same shipping emissions in 2030 as in 2000, and a flat rate increase of 2.2% per year in shipping emissions between 2000 and 2030. (Other emissions in the models follow the Intergovernmental Panel on Climate Change (IPCC) Special Report on Emissions Scenarios (SRES) A2 scenario.) Surface ozone increased in all three scenarios in 2030 (due to the increased emissions of precursors from other sources both regionally and globally under the SRES A2 scenario, see Chapter 4), but there was some additional sensitivity of ozone levels to shipping emissions. The sensitivity was greatest over the near-Atlantic areas, the British Isles and the North and Baltic Seas, and there were important contrasting trends. For example, ozone over the near-Atlantic in 2030 was simulated to increase by ~4 $\mu g\ m^{-3}$ less if shipping emissions are entirely removed (cf year 2000 emissions) and by ~4 $\mu g\ m^{-3}$ more if shipping emissions increased. In contrast, ozone over the English Channel, southern North Sea and Baltic areas was simulated to increase more in 2030 if shipping emissions were removed, and to increase less, or not at all, if shipping emissions increase. The latter reflected the sensitivity of ozone formation to the whole chemistry mix. Where NO_X was already relatively high, further NO_X (such as from shipping) limited ozone directly by scavenging (reaction of nitric oxide and ozone, $NO + O_3$) and/or indirectly by reducing oxidant capacity (reaction of nitrogen dioxide with hydroxyl radicals, $NO_2 + OH$). Collins *et al*. (2008) showed that the magnitude of the same shipping impact on European ozone varied by a factor of two depending on the assumptions for the land-based emissions.

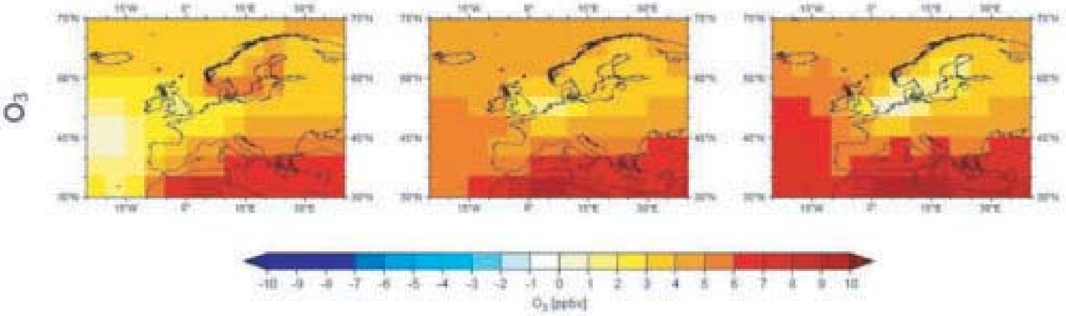

Figure 8.8 Changes in surface ozone between 2000 and 2030 over Europe under the IPCC SRES A2 scenario with different assumptions for ship emissions. Left: Changes in a world without ship emissions in 2030; Middle: Changes in a world with ship emissions remaining at 2000 levels; and Right: Changes under a constant growth scenario of a flat increase of 2.2% per year compared to the year 2000. Data are ensemble-mean values from ten global atmospheric chemical transport models (Eyring *et al.*, 2007).

8.2.3.4 Carbon monoxide

341. The influence of global and regional trends in carbon monoxide emissions on global and regional ozone has been less extensively studied. The ACCENT model intercomparison, using best estimates of present-day carbon monoxide emissions, showed large underestimates in simulated northern hemisphere extratropical carbon monoxide compared with both surface observations and satellite observations from the MOPITT instrument (Shindell *et al*., 2006). The interpretation was that year-round emissions, probably from fossil fuel burning in east Asia, and seasonal biomass burning emissions in south-central Africa, were greatly underestimated in current inventories, such as those from IIASA. Although carbon monoxide has a shorter atmospheric lifetime (of only a few months) than methane, it is sufficiently long-lived to have an impact on both hemispheric and regional-scale ozone concentrations, for example by scavenging the OH radical and increasing the lifetime of methane. Control of hemispheric carbon monoxide emissions will therefore also reduce background concentrations of ozone and its associated contribution to radiative forcing.

342. IIASA recently prepared a revised set of emission projections for methane, oxides of nitrogen and carbon monoxide to inform the report by the Royal Society, *Ozone in the 21st century.* The NO_X and carbon monoxide emissions were lower than those given in previous emission projections as the effects of recent air pollution control measures had been incorporated. These new emission projections could have a significant implication for future global ozone concentrations and emission control policies.

8.3 Natural vs man-made emission sources

343. The extent of any changes in future ozone concentrations will increasingly be influenced by biogenic VOC emissions. As man-made emissions decrease, biogenic sources will contribute an increasing proportion of ozone precursors. Biogenic VOCs describe a wide range of compounds, of which only a few are generally of interest and represented in chemical transport models. Isoprene is the compound of most importance for ozone modelling for example. Emissions of the various terpenes (e.g. α- and β-pinene) are also important, for both ozone and aerosol formation. The remaining VOC species (other VOC or OVOC) play some role, but little is known about the chemistry of many components or the quantitative emissions of individual species. Emissions may be large, however. Annex 1 provides further information on the compilation and comparison of different biogenic VOC emission inventories.

344. If the incidence of high temperature episodes increases as a result of climate change, then the very strong temperature dependence of biogenic emissions may lead to increases over current levels (although other factors related to environmental change will also certainly affect future emissions, see Chapter 4). Human influence in determining which tree species are planted could be significant in this context, as emissions vary considerably between species. Some of these changes were considered by Donovan *et al*. (2005) in their study to quantify the effects of trees on urban air quality in scenarios of high photochemical pollution.

345. Donovan *et al.* (2005) used a trajectory model to evaluate the effects of the emissions of biogenic volatile organic compounds from the urban forest in the West Midlands metropolitan area (as well as their effects on pollutant deposition of ozone, NO_2, carbon monoxide and nitric acid (HNO_3)). While all tree species investigated were found to be beneficial to air quality in terms of pollutant deposition, some tree species were found to have the potential for photochemical production of ozone. An urban tree air quality score (UTAQS) was developed to rank tree species in order of their potential to improve air quality. Of the 30 species considered, pine, larch and silver birch were calculated to have the greatest potential to improve urban air quality, while oaks, willows and poplars could worsen downwind air quality if planted in very large numbers. There have been several instances where local authorities have specified in planning consents that developers must take note and act upon the UTAQS in the implementation of approved planning applications [Hewitt, private communication].

346. The role of trees in urban air quality management (and other environmental issues) has also been recognised in the United States. The USEPA identifies multiple net benefits from planting trees and vegetation in urban areas despite the fact that trees and vegetation are themselves sources of VOCs[9]. For air quality, these were:

- increased removal of ozone (and other pollutants) from the air through deposition;
- lower urban temperatures through direct shading and evaporation of water from leaves. Lower urban temperatures would reduce the demand for air conditioning and hence the emissions from energy generation (also of benefit for climate change). In the US context, a further air quality benefit of reduced urban temperatures would be lower rates of photochemical ozone production within urban areas; and
- shading of, for example, car parks would reduce evaporative emissions of VOCs from parked cars.

The USEPA identified certain types of pine and maple trees as low-emitting species. High VOC-emitting trees included eucalyptus, sycamore, willow and certain oak varieties.

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[9] see http://www.epa.gov/heatisland/strategies/level3_vegairquality.html

8.4 Exposure during ozone pollution episodes

8.4.1 Action plans

347. The EU's 3rd Daughter Directive (2002/3/EC) makes provision for 'short-term action plans' specifically for extreme episodic ozone events, where there is a risk of exceedence of the alert threshold (240 µg $m^{-3} \approx$ 120 ppb) for three or more consecutive hours.

348. There have been a number of campaigns in the UK (for example, the Natural Environment Research Council (NERC) PUMA (Pollution of the Urban Midlands Atmosphere)[10] and TORCH[11] campaigns) to understand aspects of atmospheric chemistry in urban and rural areas. One of the TORCH campaigns was held during July and August 2003 when a major ozone episode occurred in southern Britain (see section 8.4.2). Here however, we consider the few UK case studies that were specifically undertaken to assess possible policy control measures which could form part of short-term action plans to address ozone pollution episodes in the UK.

349. As reported by Jenkin (see Section 5.3.2 in Hayman *et al.*, 2002), a version of the UK Photochemical Trajectory Model (PTM) was used to investigate the impact of a series of UK precursor control measures on the peak ozone concentration at the Barnsley Gawber site at 13:00 hr on 1 August 1999. This event was selected because an exceedence of 240 µg m^{-3} was recorded, within a more general and widespread UK ozone episode, which persisted from 30 July – 2 August 1999.

8.4.1.1 Modelling of a UK episode at Barnsley

350. Figure 8.9 shows the 96-hour trajectory arriving at Barnsley Gawber at 13:00 hours on 1st August 1999. The event was characterised by airflow from the European continent (as typically experienced during ozone episodes), with the trajectory passing over the UK for the final 18 hours prior to arrival. For much of this time, the trajectory path travelled over rural areas of Suffolk, Norfolk and Lincolnshire and therefore did not receive particularly significant precursor emissions. However, the final six hours travel involved passage over the general area of Workshop (Nottinghamshire) and Rotherham (South Yorkshire) and into the M1 corridor, prior to arrival at Barnsley Gawber, about 1 hour downwind of Rotherham.

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[10] http://badc.nerc.ac.uk/data/urgent/Projects-Doc/pumaco.html

[11] http://badc.nerc.ac.uk/data/polluted-tropo/projects/torch.html

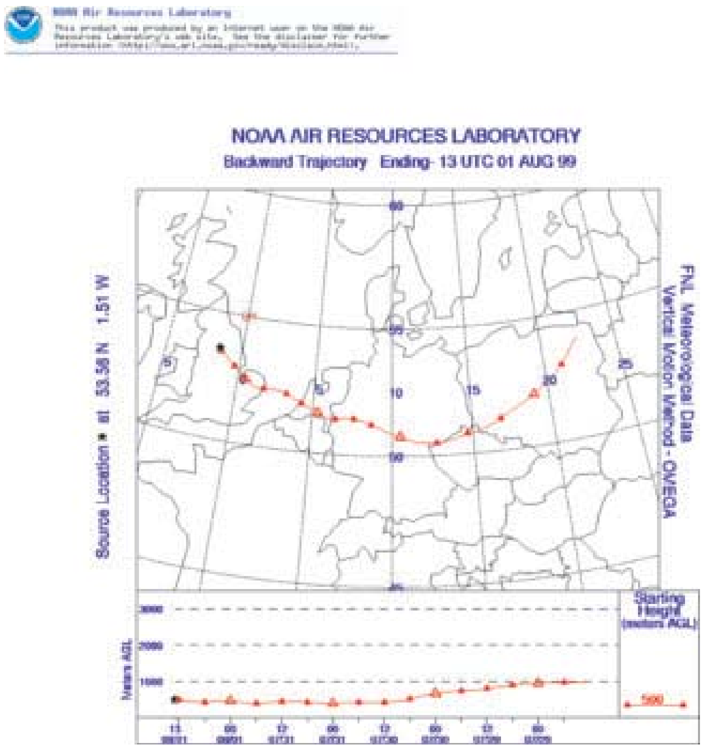

Figure 8.9 96-hour trajectories to Barnsley Gawber (13:00 hours on 1st August 1999), which correspond to reported hourly average ozone ≥ 120 ppb at this site. The trajectories were obtained from the US National Oceanic and Atmospheric Administration (NOAA) interactive on-line trajectory service (http://www.arl.noaa.gov/ready/hysplit4.html).

351. The PTM, containing the Common Representative Intermediates (CRI) mechanism, was used to simulate the ozone concentration along this trajectory for the base case scenario, employing mapped emissions based on the 1998 NAEI (this is described subsequently in Table 8-6. The simulated ozone concentration at the trajectory end-point, 236 μg m^{-3}, was slightly lower than the observed value of 248 μg m^{-3}. In order to provide a base case simulation in which the 240 μg m^{-3} threshold was exceeded, the VOC emissions throughout the model domain were slightly increased (by *ca*. 6%), to bring the simulated concentration at the end-point up to the observed value of 248 μg m^{-3}.

352. A set of 11 precursor control measures was investigated. Of these, only the final two were considered to be physically feasible: the remainder were to test the sensitivity of the simulated ozone level to purely illustrative measures, which cannot be achieved in isolation. Figure 8.10 shows the change in the simulated ozone levels at the trajectory end-point that result from these 11 control measures. It should be noted that none of the measures resulted in a large change. Even eliminating all UK anthropogenic emissions of VOC and NO_X (measure 1) only lowered the simulated concentration by 5.4 μg m^{-3}. Interestingly, a similar effect was achieved by doubling UK emissions (measure 2). This was because the Barnsley Gawber site was directly downwind of Rotherham under the conditions of 1st August 1999, and the ozone level was reduced as a result of increasing the NO_X emissions shortly before the air mass arrives at the end-point.

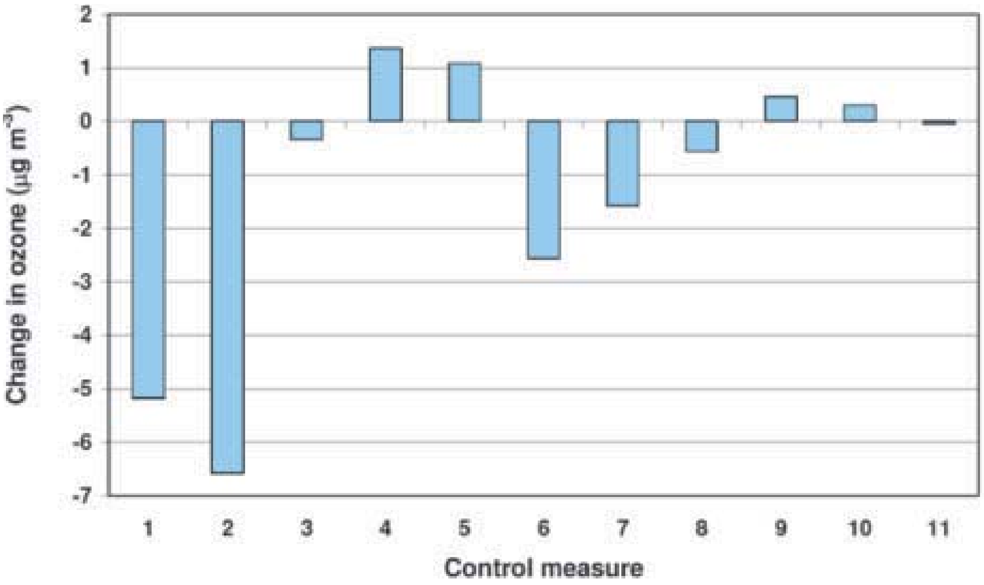

Figure 8.10 Illustration of the influence of 11 UK precursor controls on the simulated peak ozone level at Barnsley Gawber (1st August 1999 trajectory with assumed Friday arrival: base case ozone mixing ratio = 244.8 µg m^{-3}). The measures were: (1) No UK emissions of anthropogenic VOC or NO_X; (2) UK emissions of anthropogenic VOC and NO_X doubled; (3) No anthropogenic VOC emissions in the region of Rotherham/M1; (4) No NO_X emissions in the region of Rotherham/M1; (5) No anthropogenic VOC or NO_X emissions in region of Rotherham/M1; (6) No anthropogenic VOC emissions in Nottinghamshire or South Yorkshire; (7) No NO_X emissions in Nottinghamshire and South Yorkshire; (8) 22.3% anthropogenic VOC reduction in Nottinghamshire and South Yorkshire; (9) 23.8% NO_X reduction in Nottinghamshire and South Yorkshire; (10) No cars in the region of Rotherham/M1; and (11) No cars in Nottinghamshire and South Yorkshire.

353. The specific influence of control measures 3, 4 and 5 involving Rotherham and the M1 (i.e. the final two hours of the trajectory) confirmed that the air mass was strongly VOC-limited and also inhibited by elevated NO_X levels in a comparatively polluted plume. Thus removal of VOC emissions (measure 3) led to a reduction of ozone, removal of NO_X emissions (measure 4) led to an ozone increase, and removal of both (measure 5) led to a smaller increase. Expanding the scope of the illustrative measures to Nottinghamshire and South Yorkshire (i.e. the final six hours of the trajectory) led to ozone reductions when either VOC or NO_X emissions were eliminated (measures 6 and 7). However, partial elimination of NO_X over the widened area (measure 9) still resulted in increased ozone. Measures 10 and 11 represented actions that were potentially physically achievable. The elimination of car traffic in Rotherham and on the M1 resulted in a small simulated increase in the ozone concentration of 0.3 µg m^{-3}, whereas the elimination throughout Nottinghamshire and South Yorkshire led to a very small simulated decrease of 0.04 µg m^{-3}. The magnitude and sense of the influence of these controls were logical, within the context of the effects of the illustrative measures described above.

354. The results of these model simulations provided a series of peak ozone concentrations, from which the duration of the exceedence of the alert threshold was inferred. This was achieved on the basis of a relationship between these two quantities, derived from archived monitoring data. The results indicated that none of the measures would lead to changes in the exceedence duration of more than 27 minutes, and the potentially achievable actions 10 and 11 would result in changes of only +70 and -10 seconds, respectively.

355. The study concluded that it was difficult to identify any realistic and beneficial UK short-term actions for the type of extreme ozone events that had been recorded in the UK. The calculations also showed that some local measures (especially those affecting NO_X emissions) could increase ozone levels under certain circumstances.

8.4.1.2 UK vs European action

356. The UK PTM described above was also applied to assess the effectiveness of a series of emission reduction scenarios related to the National Emission Ceilings Directive (NECD), on simulated peak ozone concentrations over a wider geographical area in the southern UK during the same ozone episode (see Section 8.3.2 in Hayman *et al*., 2002). The emissions scenarios are summarised in Table 8-6, with the 1998 emissions used as the base case (referred to as the "1998 ref" scenario). The remaining scenarios refer to 2010 and represent either emission reductions within the UK only, or throughout Europe.

Table 8-6 Summary of NECD-related emissions scenarios used for investigation of the influence of precursor emission controls on episodic ozone levels.

Abbreviation	Description	UK VOC emissions (ktonnes per annum)	UK NO_X emissions (ktonnes per annum)
1998 (base case)	1998 UK emissions of CO, NO_X, SO_2 and VOC as defined by NAEI.[1] Other relevant country total scaled to latest EMEP values.[2]	1958	1753
2010 UK ref	2010 UK emissions of CO, NO_X, SO_2 and VOC as projected by NAEI.[3] Emissions outside UK unchanged from 1998 ref.	1252	1167 (NECD)
2010 UK 1200	UK VOC emissions reduced to meet NECD according to NAEI scenario.[3] Emissions outside UK unchanged from 1998 ref.	1200 (NECD)	1167 (NECD)
2010 UK 1150	UK VOC emissions further reduced according to NAEI scenario.[3] Emissions outside UK unchanged from 1998 ref.	1150	1167 (NECD)
2010 NECD	As 2010 UK 1200, with other EU country totals set at NECD values. Other relevant country totals set to EMEP 2010 projection.[2]	1200 (NECD)	1167 (NECD)

Notes: (1) 1998 emissions available from National Atmospheric Emissions Inventory (NAEI) (http://www.naei.co.uk); (2) EMEP website (http://www.emep.int/index.html) accessed August 2000. 'Latest' values generally corresponded to 1997. Projections for 2010 based on reduction plans in force at that time; (3) NAEI 2010 reference scenario supplied by J. Goodwin, AEA Technology; VOC reduction scenarios based on cost curve analysis supplied by N. Passant and J. Goodwin, AEA Technology.

357. In this case, the model was used to simulate the chemical development over a 96-hour period along the trajectories arriving at 16:00 hours (on 31st July 1999) at eight southern UK sites: Yarner Wood, Aston Hill, Somerton, Harwell, Lullington Heath, Wicken Fen, Rochester Stoke and Sibton. The 31st July 1999 was selected because the highest ozone concentrations were generally observed on that day. The calculation was carried out for each of the emissions scenarios summarised in Table 8-6. The results showed some variability from one site to another, particularly for those scenarios in which only UK controls were implemented. Figure 8.11 summarises the information in terms of the percentage reduction in simulated peak ozone at each site, relative to the 1998 base case. The sites were presented in order of location from west to east to emphasise the geographical dependence of the effectiveness of the control measures.

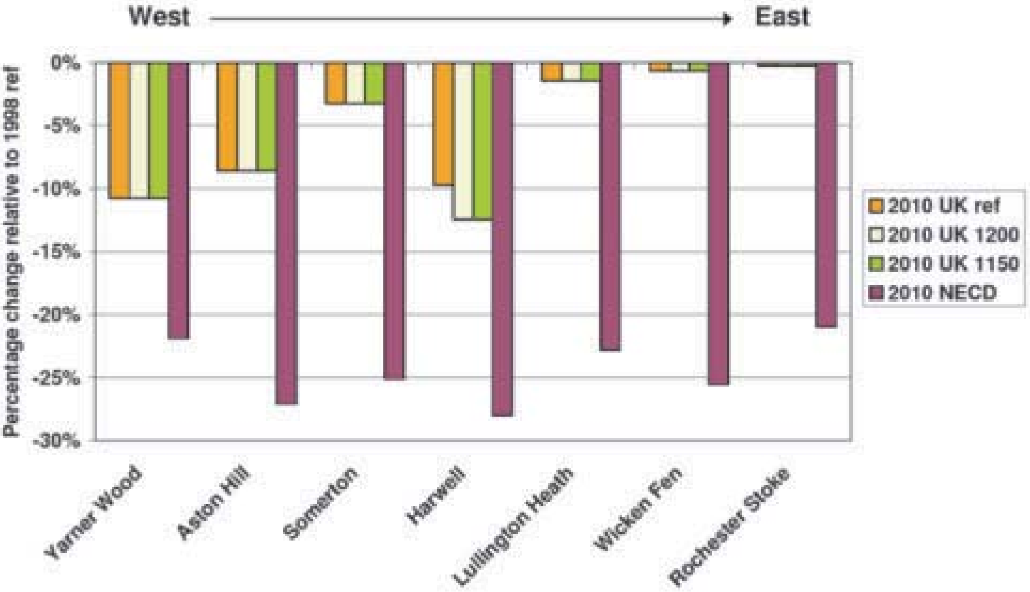

Figure 8.11 The percentage reduction in simulated peak ozone at each site, relative to the 1998 reference, for each of the scenarios in Table 8-6. Results represent the average of the results for each day of the week.

358. Relative to the 1998 reference, there was generally a clear decreasing trend in percentage ozone reduction from west to east as a result of purely UK measures, reflecting that the prevailing air flow was broadly from the east. In the limit, the Sibton site was totally unaffected by any UK reductions under the meteorological conditions of the present case study. The Harwell site was an exceptional case, in that the corresponding trajectory passed directly over London prior to arrival, and therefore received substantial UK emissions. As a result, the simulated influence of UK reductions was significantly greater than for sites further west (e.g. Aston Hill). Under the precise conditions of 31st July 1999, Harwell could be regarded as being representative of sites directly downwind of major UK population centres during an ozone episode.

359. Figure 8.11 demonstrated that the reduction of precursor emissions under the 2010 NECD scenario (involving controls throughout Europe) was accompanied by notable reductions (*ca*. 20%-25%) in the simulated peak ozone concentrations at all eight southern UK sites. This emphasised the transboundary nature of the ground-level ozone problem, and the importance of internationally co-ordinated strategies. The results indicated that the agreed precursor controls outside the UK had a greater influence on simulated peak ozone than those within the UK at all eight sites, and that controls outside the UK would be essential for any significant reduction in peak values at locations to the east/south-east of the country.

360. The conclusions from the above model runs mirrored the outcome of the Heilbronn ozone field experiment in Germany in June 1994 (e.g. Moussiopoulos *et al*., 1997). The aim of the study was to investigate whether peak ozone concentrations during summer smog periods could be reduced by the aid of short-term local-scale interventions. The measurements made during the experiment showed that local interventions led to lower concentrations of the emitted pollutants but only to minor changes in peak ozone concentrations. The main conclusion from the Heilbronn ozone experiment was that concerted large-scale interventions to the primary pollutant sources are needed for decisive reductions of peak ozone concentrations in Central Europe.

8.4.2 Ozone formation during the August 2003 episode

361. In conjunction with the NERC TORCH project, a modified version of the PTM was used to simulate regional-scale ozone formation during late July and August 2003 (Figure 8.12). The model is based on that of Derwent *et al.* (1996), with recent updates described by Johnson *et al*. (2006) and Utembe *et al.* (2005).

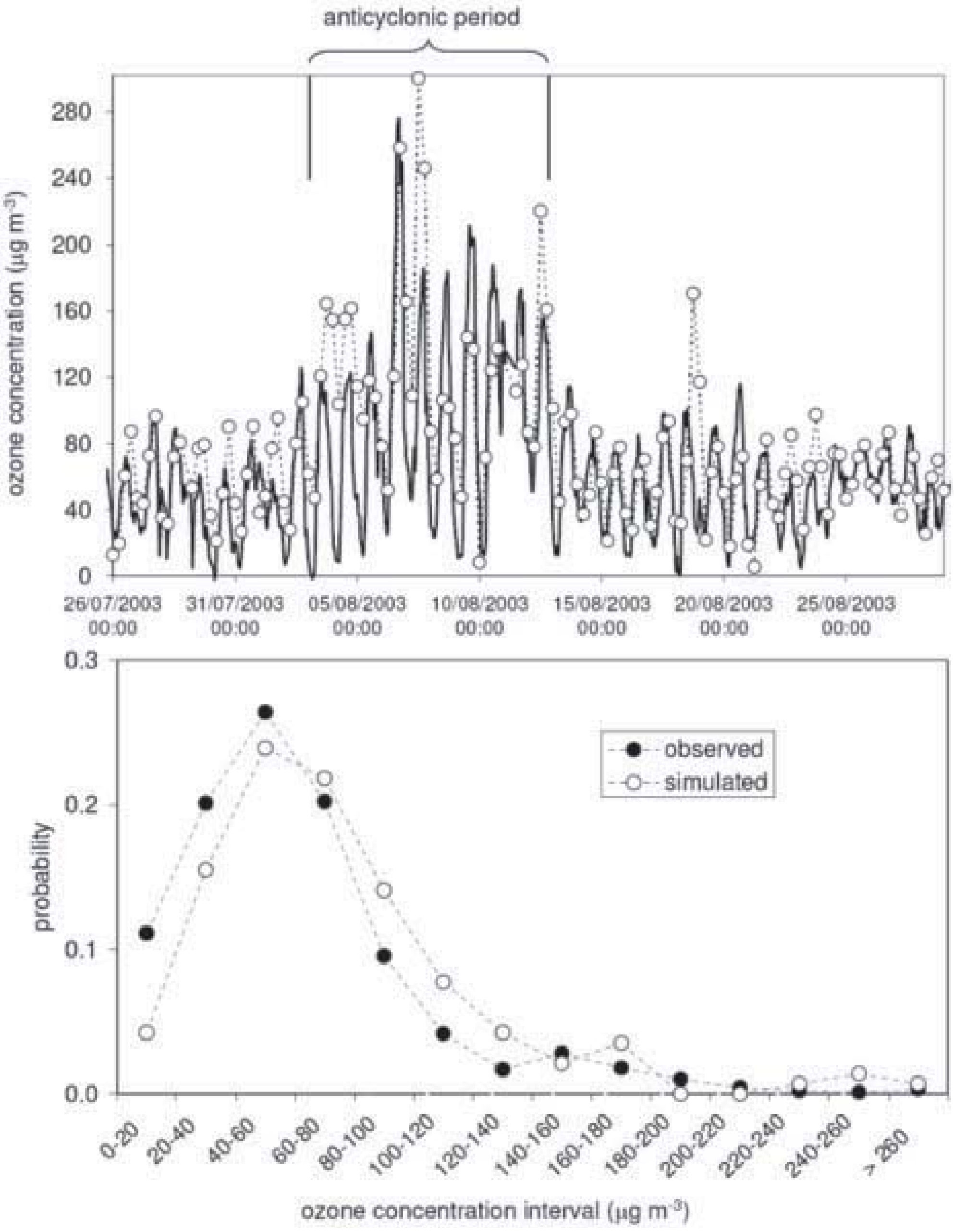

Figure 8.12 (Upper panel) Comparison of observed ozone concentrations (line) and those simulated in the base case (open points) for the period of the TORCH campaign. The broken line joining the simulated points is to guide the eye, and is not intended to infer intermediate simulated concentrations. (Lower panel) Comparison of the observed and simulated distributions of ozone concentrations for the campaign period. The ozone concentrations were measured by the University of York, as reported in Utembe *et al.* (2005).

8.4.2.1 Sensitivity to emission variations over the period 1990-2020

362. The 2003 campaign has also been simulated using historical emissions of NO_X, anthropogenic VOC, carbon monoxide and sulphur dioxide appropriate to the years 1990, 1995 and 2000 (based on NAEI and EMEP), and projected emissions for the years 2005, 2010, 2015 and 2020, based on relative UK figures reported in AQEG (2004, 2005). Figure 8.13 shows the trend in the simulated ozone distribution, which indicated that the same conditions in

earlier years would have been accompanied by broader ozone distributions with greater daytime maxima and increased overnight depletion. The decreasing trend in the top-end ozone concentrations was driven primarily by reductions in emissions of anthropogenic VOC since 1990. The simulated campaign maximum concentration was *ca*. 30% lower for 2005 conditions, compared with 1990 conditions, which agreed well with the observed decline at long-running rural sites, as presented in section 2.7. The maximum ozone concentration was projected to show some modest decline with future emissions trends, followed by a slight increase towards the end of the period. This was a consequence of the increasing trend in the emitted VOC/NO_X ratio (i.e. as VOC emissions level off and NO_X emissions continue to fall), and the fact that ozone formation generally tended to be VOC-limited for the conditions of this campaign (see below).

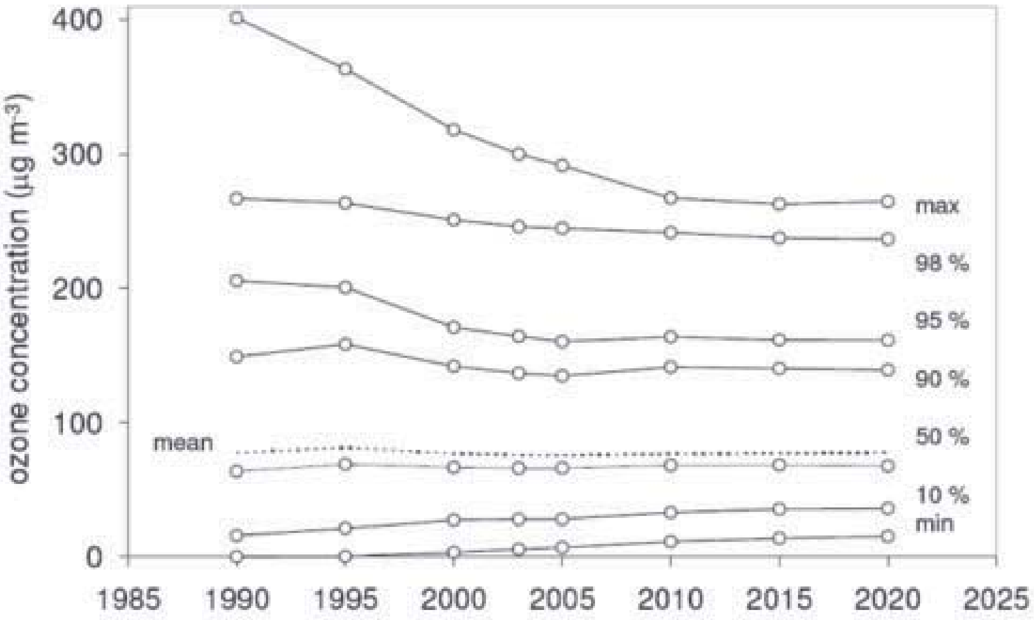

Figure 8.13 Simulated trend in ozone distribution statistics for the August 2003 episode at Writtle (Essex) as a function of emissions for the period 1990-2020.

363. The increasing (night-time) minima were a consequence of associated NO_X emissions reductions, resulting from decreased local removal of ozone by reaction with NO emitted into the shallow night-time boundary layer. The trend in the distribution thus showed similar features to those observed at rural sites (see section 2.7), although the observations relate to measurements throughout the year.

364. Figure 8.14 presents the same data in terms of the number of hours that ozone concentrations reached or exceeded 180 µg m^{-3}. Once again, the simulations with the earlier years' emissions showed a greater number of exceedences, and displayed a similar trend to that observed at long-running rural sites in the UK, particularly for the heat-wave years of 1990 and 1995 relative to 2003. This elevated ozone metric was projected to show no further decline with future emissions trends (consistent with the 2006 heat-wave year observations), but a slight increase towards the end of the period owing to the increasing trend in the emitted VOC/NO_X ratio, as indicated above.

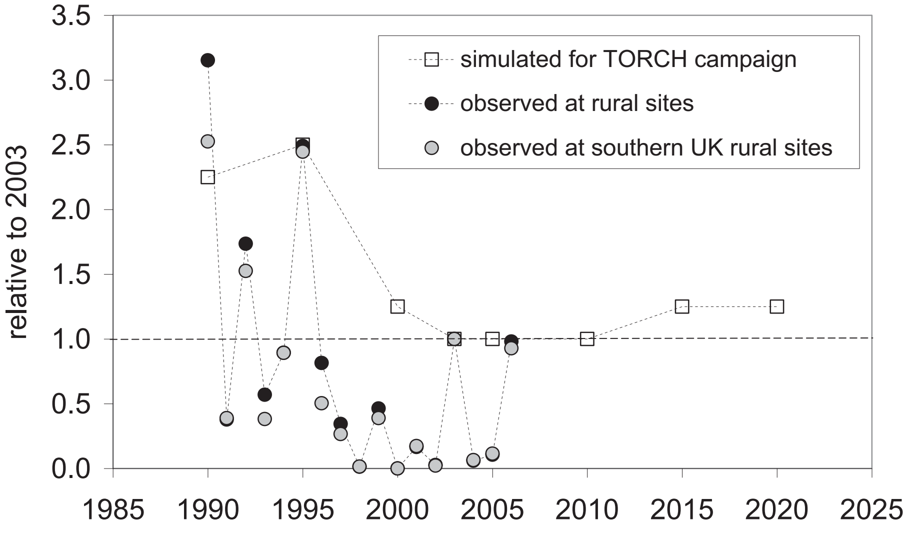

Figure 8.14 Simulated trend in the number of hours with ozone ≥ 180 µg m^{-3} for the August 2003 episode at Writtle (Essex) as a function of emissions for the period 1990-2020 (squares), compared with observed statistics at long-running UK rural sites (black circles) and at southern UK long-running rural sites (grey circles) for the period 1990-2006. Data are presented relative to the value simulated or observed in the reference 2003 case. The long-running rural sites which contribute to the data are identified in Figure 2.3 in section 2.7.
The southern UK sites are Lullington Heath, Sibton, Harwell, Yarner Wood and Aston Hill.

8.4.2.2 Effects of incremental NO_X and VOC emissions reductions

365. The effects of 10% incremental reductions in the emissions of NO_X and anthropogenic VOC for the years 1990, 2003 and 2020 were modelled to investigate whether ozone formation is limited by the availability of NO_X or VOC. Figure 8.15 shows the resultant simulated changes in ozone concentrations for the three years:

- The 1990 scenario demonstrated strong VOC limitation throughout the 2003 TORCH campaign period, with NO_X reductions almost always leading to an increase in ozone concentration, such that VOC emissions controls were clearly the favoured option for reducing ozone concentrations. The approximately compensating influences of VOC and NO_X reductions also qualitatively explained the small change in the simulated 180 µg m^{-3} exceedence in the early part of the time series (Figure 8.14).

- The results for the 2003 scenario demonstrated a shift towards NO_X limitation, with an increased number of days (relative to 1990) where NO_X control led to a reduction in ozone concentration. Although there were two days within the campaign period when 10% NO_X control led to slightly greater ozone reduction than 10% VOC control, it was clear that the campaign period was still dominated by VOC-limited conditions.

- The results for the 2020 scenario showed that 10% NO_X control led to comparable numbers of days when the ozone concentration was increased and decreased, and a further increase in the number of days (relative to 2003 and 1990) when NO_X control was more beneficial in reducing ozone concentrations than VOC control. Despite this, VOC reductions remained the favoured option when the whole campaign was considered.

366. The results therefore demonstrated that ozone formation for the conditions of the campaign at this location was predominantly VOC-limited for the complete time series, but showed a trend towards NO_X limitation. Although this was in general agreement with previous assessments for south-east England, this sensitivity study also showed that VOC and NO_X limitation were not intrinsic properties of a location, and that it was possible to get events when either condition prevailed.

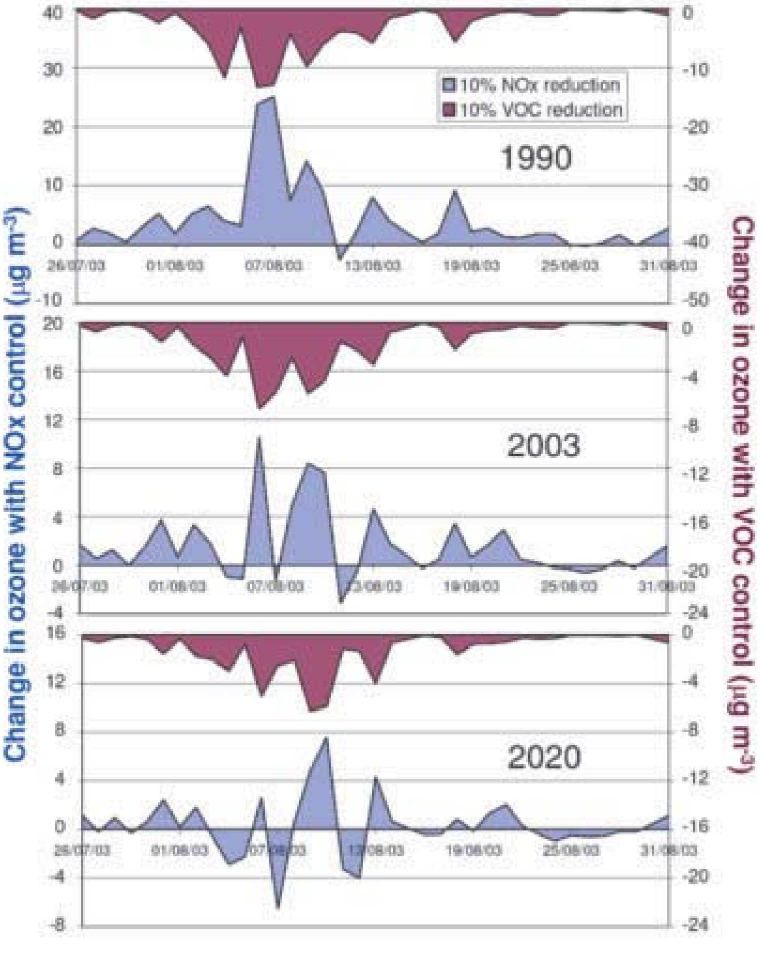

Figure 8.15 Simulated change in ozone concentration at the arrival point (Writtle, Essex) resulting from an incremental decrease of 10% in emissions of NO_X and anthropogenic VOC for the 1990 (upper panel), 2003 (middle panel) and 2020 (lower panel) emission scenario.

8.4.3 Further assessment of emissions reduction scenarios

367. Derwent (2008) has performed a similar study to that of the TORCH campaign by Utembe *et al.* (2005). Derwent (2008) used the UK Photochemical Trajectory Model (UK PTM) to calculate ozone concentrations at Harwell for each day of 2005. The model made use of three-dimensional 96-hour back trajectories provided by the Met Office NAME atmospheric dispersion model. Thirty trajectories were used for 15:00 hours each day because a single trajectory may not be a reliable guide to air mass origins. The chemical mechanism employed was Carbon Bond IV. Base case emissions were taken for 1999 using EMEP and NAEI inventories.

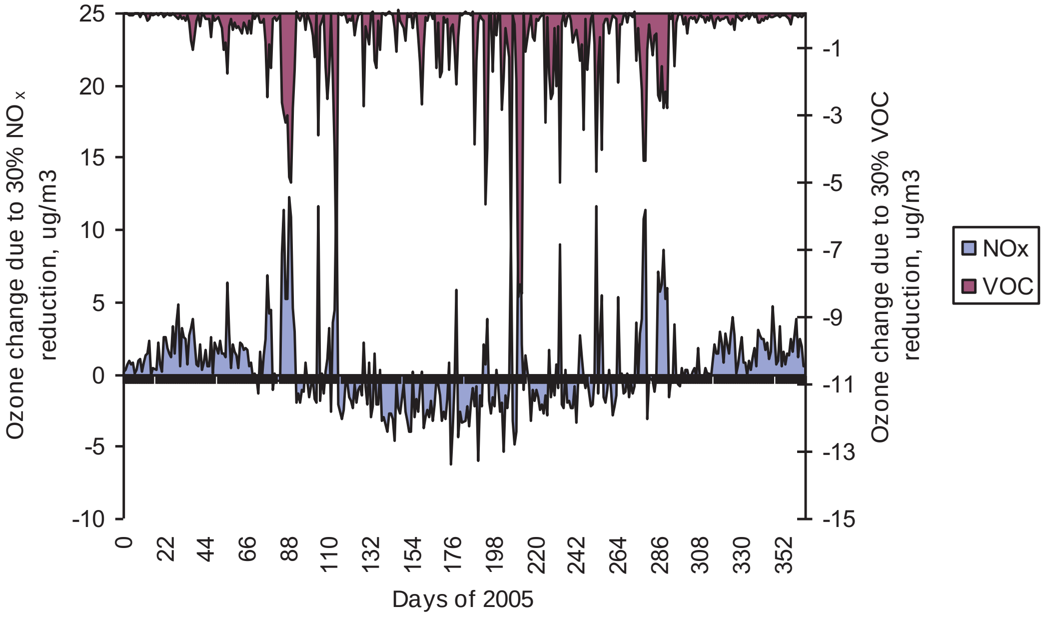

Figure 8.16 Changes in the maximum daily ozone concentration (in µg m^{-3}) calculated at Harwell for 2005 for 30% reductions in the emissions of (a) oxides of nitrogen and (b) volatile organic compounds.

368. NO_X and VOC emissions were then separately reduced by 30% across the board and the ozone changes were calculated for each day of the year, as shown in Figure 8.16. In response to NO_X reductions, daily maximum ozone levels sometimes increased and sometimes decreased. Ozone increases tended to occur during the winter and ozone decreases tended to occur during summer. On the other hand, daily maximum ozone levels only decreased in response to VOC reductions. Overall, the average of the daily ozone maximum values increased from 64.2 µg m^{-3} in the base case to 64.6 µg m^{-3} with 30% reduction in NO_X emissions but decreased to 63.4 µg m^{-3} with a 30% reduction in VOC emissions. This complemented the scenario calculations based on the results from the TORCH campaign in 2003 (see section 8.4.2).

369. Yu *et al.* (2007) have used the comprehensive US Community Multi-scale Air Quality (CMAQ) modelling system to investigate three UK air pollution episodes (occurring in September 1998, and June and December 2001), as part of a project for the Environment Agency. The overall aim of the study was to assess the use of advanced models as a tool to evaluate contributions of the Agency-regulated sources to regional ozone production (and also sulphur dioxide and NO_2 concentrations) under a range of episodic and typical situations. For ozone, the model was used to simulate the June 2001 episode and the effect

of a notional additional VOC source, located in the Thames Estuary area, was investigated. Even with the notional source emitting only a very reactive VOC (ethene) at a constant rate of 10 tonnes hr^{-1}, the overall contribution of this point source to ground-level ozone concentrations was found to be very small. This more sophisticated modelling approach based on an actual air pollution episode again confirmed the limited impact of local action on local ozone concentrations.

8.5 Recommendations

370. Future ozone concentrations will be influenced by changes on the global and hemispheric scales and policy control measures will increasingly need to be agreed and implemented internationally. The Air Quality Expert Group (AQEG) recommends that the UK continue to take an active part in European and international activities in this area (for example, the UN Task Force on Hemispheric Transport of Air Pollution). Further, future assessments should take account of the benefits (or disbenefits) not only to air quality but also to the climate system.

371. Modelling tools and systems are required that are capable of covering spatial scales from urban to global and of addressing air quality and climate change in an integrated manner. In this regard, many of the recommendations made in AQEG's earlier report, *Air quality and climate change: A UK perspective,* still hold.

372. As VOC emissions from man-made sources fall, those from natural sources become more significant. AQEG reiterates the recommendation made in Chapter 4 for further research to narrow the uncertainties associated with emissions inventories for the UK and the rest of Europe for isoprene, in particular, and other relevant biogenic volatile organic compounds. This will include increased accuracy and precision in estimates of VOC emissions by plant species, in current land cover data for plant species distributions and in process-based algorithms describing environmental influences on biogenic VOC emissions (e.g. seasonal, diurnal, PAR intensity and temperature).

Chapter 9

Progress on recommendations made in the Fourth Report of the Photochemical Oxidants Review Group in 1997

Question H: *Of the recommendations made in the Fourth Report of the Photochemical Oxidants Review Group in 1997, which remain to be implemented, to what extent do they remain valid and which have been superseded by scientific understanding?*

Short answer to question H

373. The Air Quality Expert Group (AQEG) has reviewed progress on the recommendations made by the Photochemical Oxidants Review Group (PORG) for the topics *ozone monitoring, NO_X and chemistry, hydrocarbons and synthesis and interpretation*. Recommendations related to impacts on *human health effects, vegetation* and materials have not been reviewed, because these topics are not discussed in the present AQEG report. The progress on 21 relevant recommendations is outlined in this chapter.

Detailed answer to question H

374. Each recommendation from the 1997 PORG report (PORG, 1997) is given in turn in bold type. The recommendations in the areas of human health effects, vegetation and materials are not addressed, because they are not directly discussed in this AQEG report.

9.1 Ozone monitoring

- **Maintain the rural network to provide the spatial distribution of ozone in the UK and trends with time. Increase the number of monitoring sites to eliminate large areas of uncertain ozone exposure, notably the East Midlands of England, north and west Wales, the Central Highlands of Scotland and inland rural areas of SE England.**

375. Figure 9.1 shows maps with the distribution of ozone sites within the Automatic Urban and Rural Network (AURN) in 1996 and 2007. Sites with co-located nitrogen dioxide (NO_2) measurements are indicated.

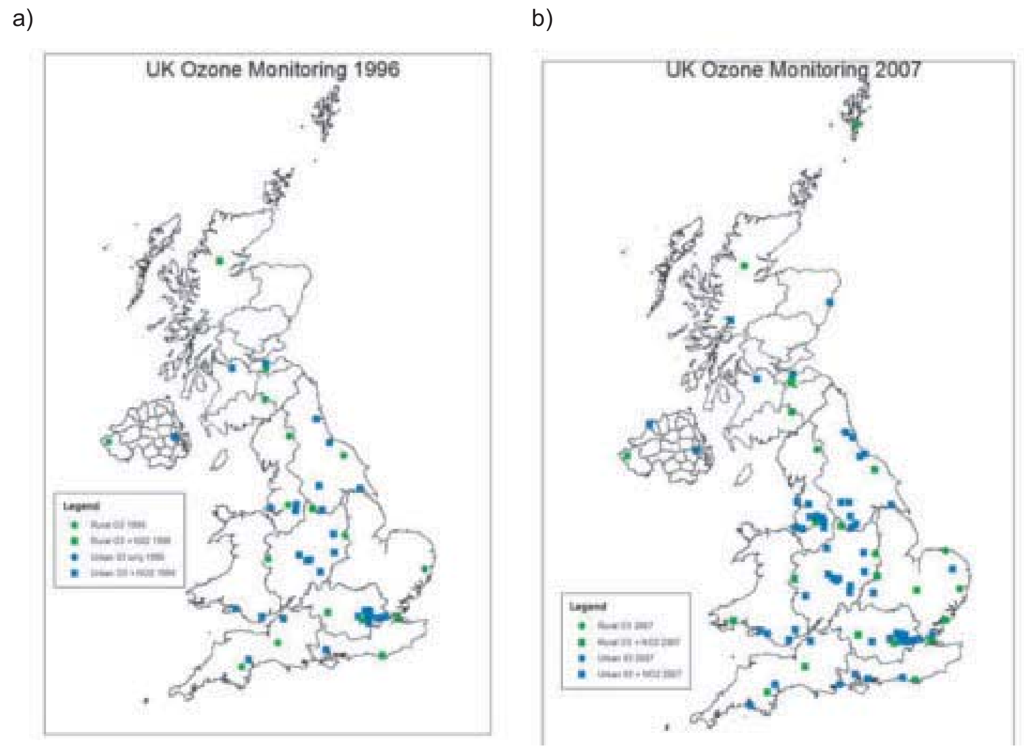

Figure 9.1 The distribution of ozone sites within the AURN in 1996 (panel a) and 2007 (panel b). Sites with co-located NO_2 measurements are indicated.

376. Rural site numbers increased from 17 (6 with NO_2) to 24 (16 with NO_2) and urban sites increased from 33 to 66; all except one have co-located NO_2 monitoring. These changes are largely due to the Third Daughter Directive. The Directive also tightens up the measurement requirements of ozone monitoring by making explicit reference to the European Committee for Standardisation (CEN) standard BS EN 14625:2005 *Ambient air quality – Standard method for the measurement of the concentration of ozone by ultraviolet photometry.* The standard specifies performance characteristics for instruments to be type approved (and certified), and requirements for calibration and ongoing quality assurance such that the measurement uncertainty of ozone measurements demonstrably meets the Directive's Data Quality Objective of 15% (at the 95% confidence level). The effect of this on UK network measurements is unlikely to be significant, as most of the requirements have been network practice for many years, but it will increase confidence in the data and in its comparability with other European data. The standard is currently being revised to take into account practical experience of its use.

377. For the specific areas identified by PORG:

- **East Midlands:** one additional rural site at Market Harborough.
- **North and West Wales:** one additional rural site at Narbeth in west Wales (Pembroke).
- **Central Highlands of Scotland:** suburban site in Fort William.
- **Inland rural areas of SE England:** one additional rural site at Wicken Fen (East Anglia) and an urban site in Reading.

Status report: Considerable progress has been made.

- **Encourage the development of low-cost ozone monitoring techniques, such as solid state sensors and diffusion tube methods.**

378. Work is in progress on low-cost ozone monitoring techniques based upon solid state sensors. Market forces for their deployment largely dictate product development. Little work has been done in the UK on diffusion tube methods, which have been developed significantly in other countries. They do, however, have the disadvantage of giving a long-term average and therefore no information on diurnal trends.

379. The development and validation of new techniques will be helped by the existence of the CEN standard method for ozone measurement cited above, together with the EU guidance document *Demonstration of equivalence of ambient air monitoring methods,* available through the EU website. This sets out procedures for laboratory and field tests that determine whether non-reference methods give results that are equivalent to the reference method for EU monitoring purposes.

Status report: Little progress has been made.

- **Develop further the modelling techniques to quantify the influence of local site factors (land use and shelter).**

380. This recommendation reflected the importance at the time of the preparation of the 4th PORG report of empirical interpolation-based maps of various ozone metrics. There was concern that local site factors limited the representativeness of some monitoring sites. The expansion of both rural and urban monitoring networks and the greater availability of process-based models means that this is now less of a priority area for further research.

Status report: No longer a priority.

- **Increase the number of stations with co-located rural monitoring of O_3, SO_2, NO, NO_2 and PM_{10} or $PM_{2.5}$.**

381. Table 9-1 shows the pollutants measured at the 2007 rural AURN sites.

Status report: Progress has been made in the enhancement of the rural network for co-located parameters.

Table 9-1 Pollutants measured at the 2007 rural AURN sites.

Sibton	O_3	NO_2						
Harwell	O_3	NO_2	PM_{10}	$PM_{2.5}$	SO_2	1,3-butadiene	Benzene	Toluene
Bottesford	O_3	NO_2						
Bush Estate	O_3	NO_2						
Eskdalemuir	O_3	NO_2						
Great Dun Fell	O_3	NO_2						
Aston Hill	O_3	NO_2						
Lullington Heath	O_3	NO_2	SO_2					
Strath Vaich	O_3	NO_2						
Lough Navar	O_3	PM_{10}						
Yarner Wood	O_3	NO_2						
High Muffles	O_3	NO_2						
Glazebury	O_3	NO_2						
Ladybower	O_3	NO_2	SO_2					
Rochester Stoke	O_3	NO_2	SO_2	PM_{10}	$PM_{2.5}$			
Somerton	O_3	NO_2						
Narberth	O_3	NO_2	SO_2	PM_{10}				
Wicken Fen	O_3	NO_2	SO_2					
Weybourne	O_3	NO_2						
St Osyth	O_3	NO_2						
Market Harborough	O_3	NO_2	CO					
Lerwick	O_3	NO_2	CO					
Auchencorth Moss	O_3	PM_{10}		$PM_{2.5}$	other pollutants			

- **Quantify the effects of urban areas on ozone climatology to improve estimates of materials and human exposure to the potentially damaging ozone concentrations.**

382. The issue of ozone concentration in urban areas, in the context of human health effects has been extensively addressed in this AQEG report.

Status report: Achieved.

- **Provide analysis and interpretation of UK ozone monitoring data for effects assessment and to monitor changes in ambient ozone as precursor emissions decline.**

383. Extensive analysis has been reported in this AQEG report and in the 2001 report on Transboundary Air Pollution (NEGTAP, 2001).

Status report: Achieved.

- **Develop validated models which describe the small-scale spatial variation of ozone within urban and rural areas and its diurnal variation.**

384. This issue has been discussed in the present report, especially in Chapter 8. In 2007, Defra commissioned a review of tools used for modelling ozone formation and assessing impacts on human health and ecosystems (see http://randd.defra.gov.uk/). The report concluded with a set of recommendations for further action by Defra which covered improvements to chemical schemes, emissions estimates, quality control processes and recommendations for the review and evaluation of the performance of chosen ozone models. Defra is in the process of taking forward the recommendations from this review.

Status report: Progress made, further work in progress.

9.2 NO_x and chemistry

- **Elucidate in detail the NO_y composition at UK sites ranging from the most polluted urban centres to the least polluted areas.**

385. The detailed composition of NO_y species (total reactive nitrogen oxides) has rarely been elucidated at UK sites. When and where a reasonably detailed speciation has been measured, this has been limited to campaigns at selected locations. For example, Harrison *et al.* (1999) measured concentrations of total NO_y as well as the major individual species comprising NO_y (i.e. nitric oxide (NO), NO_2, peroxyacetyl nitrate (PAN), nitric acid (HNO_3), nitrous acid (HONO) and nitrate aerosol), during three Natural Environment Research Council (NERC) campaigns at the rural coastal site at Weybourne, Norfolk, in 1993-1995, and were able to draw conclusions about missing species. Measurements of HONO, PAN and, more recently, alkyl nitrates have also been made for other campaigns (e.g. EU PRIME at Ascot in 1999 and NERC TORCH at Writtle in 2003), and concentrations of HONO have been measured at Marylebone Road in 1999 (Martinez-Villa, 2001) and in the Buncefield plume in December 2005 (Mather *et al.*, 2007).

386. Routine measurements of PAN up to the early 1990s were reported for Bush and Harwell in PORG (1997), with more recent data (1994-1998) for Bush being published by McFadyen and Cape (2005).

387. Models containing appropriately detailed chemical mechanisms provide a method of simulating NO_y composition for a range of ambient conditions. For example, preliminary box modelling studies using the Master Chemical Mechanism, (MCM), have examined the contributions from organic oxidised nitrogen components (e.g. alkyl nitrates, PAN and nitroarenes) in representative urban plumes (Jenkin et al., 2006).

Status report: Only limited information available. Plenty of scope for additional studies.

- **Determine the small-scale spatial variations in nitrogen dioxide concentrations within urban areas on a measurement scale of one hour or less.**

388. The density of sites reporting NO_2 concentrations at hourly resolution in urban areas (particularly London) has increased since PORG (1997). As reported in AQEG (2004), there are also some data characterising NO_2 concentrations with distance from roads, at a spatial resolution of a few metres. Measurements at high temporal resolution using portable devices have been carried out in London as part of the NERC DAPPLE project, but have not currently been reported.

Status report: Partially achieved. Plenty of scope for additional studies.

- **Elucidate fully the concentration and formation mechanism of HONO to quantify the role of this compound in the supply of the hydroxyl radical (OH) to the lower atmosphere.**

389. There has been continued national and international effort to elucidate mechanisms which form HONO. It is known that it can be produced from reactions of NO_2 with water on a variety of surfaces (producing HONO and HNO_3), and also from surface reactions with semi-volatile exhaust organics. Recent advances have demonstrated that the conversion of NO_2 to HONO on appropriate surfaces can be enhanced by light, as demonstrated for organic surfaces (George *et al.*, 2005; Stemmler *et al.*, 2006) and recently for titanium dioxide by Cox and co-workers (Gustafsson *et al.*, 2006). The motivation for this latest study was reactions on mineral dust, although the results may also have implications for the impact and effectiveness of titanium dioxide (TiO_2) coatings (e.g. for paving slabs and building materials) designed to improve air quality through photosensitised NO_X removal.

390. It has also been shown that o-nitrophenols, products of aromatic hydrocarbon degradation, can form HONO upon photolysis (Bejan et al., 2006). The photosensitised emission of HONO from snowpack has also been observed (e.g. Zhou et al., 2001), and has been interpreted in terms of photolysis of dissolved or surface-adsorbed nitrate ions (e.g. Jacobi and Hilker, 2007).

Status report: Substantial progress, but probably still much to be learnt.

- **Assess the extent to which chemical interference in the chemiluminescent method of measurement are influencing the reported concentrations of nitrogen dioxide at UK sites.**

391. A number of older studies have demonstrated that many oxidised nitrogen species can be converted into NO in heated molybdenum converters, and can therefore contribute to the NO_2 signal measured by chemiluminescence NO_X analysers (for example, Fehsenfeld *et al.*, 1987; Grosjean and Harrison, 1985; Winer *et al.*, 1974). Recently, Dunlea *et al.* (2007) investigated the level of interference by comparing measurements made by direct spectroscopic techniques (tunable infrared laser differential absorption spectroscopy, TILDAS, and differential optical absorption spectroscopy, DOAS), with those made by

chemiluminescence monitors in Mexico City. The interfering signals correlated with the extent of photochemical processing of the air mass, maximising at about 50% of the NO_2 mixing ratio (average 22%). The main contributors to the interference were identified as gas phase HNO_3, alkyl nitrates and multifunctional alkyl nitrates.

392. Some limited consideration was given in AQEG (2004) to assessing the potential magnitude of interferences from HONO and PAN, based on some of the NO_X speciation measurements referred to above. With the exception of HONO (which can be emitted and therefore interfere close to source), the potential interference of most oxidised nitrogen species increases with air mass age (i.e. as NO_X is progressively converted to other NO_y species).

Status report: Limited progress in the UK.

9.3 Hydrocarbons

- **To improve understanding of the UK VOC emission it is necessary to attempt closure between the hydrocarbon measurements and emissions inventories at a number of key sites and locations.**

393. Dollard *et al.* (2007) have carried out a comprehensive analysis of concentrations of up to 26 C_2 to C_8 hydrocarbons monitored for varying subsets of the period 1993-2004 at 11 urban background, one rural and one kerbside site within the UK hydrocarbon network. Although these hydrocarbons represent only a fraction of the 660 species in the National Atmospheric Emissions Inventory (NAEI) speciation (making complete closure effectively unattainable), the observed trends are compatible with the declines in anthropogenic volatile organic compound (VOC) emissions from relevant sources, as represented in the NAEI for the same period, and it is reasonable to infer that similar trends have occurred for other emitted, but unmeasured, VOCs which contribute to the same source sectors. Some limited validation of NAEI VOC source strengths and speciation has been achieved using comparisons of model output with ambient hydrocarbon data (for example, Utembe *et al.*, 2005).

394. Analysis and assessment of isoprene concentrations observed at UK sites has revealed evidence of both man-made and natural biogenic sources (see for example section 2.9 in Chapter 2). There may be additional isoprene sources not currently covered in the UK emission inventories arising from grassland and other vegetation and evaporative emissions from petrol-fuelled motor vehicles. AQEG has given preliminary consideration to the latter source and concluded that it is minor because of the low total diene content of UK motor spirit. There are currently no measurements of more complex biogenic hydrocarbons (e.g. monoterpenes) with which to assess the inventory source strength, which is estimated to be greater than that of isoprene (see for example, Table A1-2 in Annex 1).

Status report: Further work is required to evaluate source strengths of isoprene and other more complex biogenic hydrocarbons, and additional closure studies are required for VOC emissions in general.

- **Complete an assessment of the contribution made to ozone formation from all emission source categories treated in the NAEI.**

395. This has been considered in some detail in a number of studies using the NAEI speciation in conjunction with the Master Chemical Mechanism (MCM) most recently and comprehensively by Derwent *et al.* (2007b). Reactivity-based strategies for photochemical ozone control in Europe have also been identified (Derwent *et al.*, 2007c).

Status report: Completed for latest NAEI speciation of source categories.

- **Complete spatially disaggregated inventories of the most important individual hydrocarbon emissions, so that we can identify those areas in which photochemical oxidant production is under NO_X or hydrocarbon control and whether regionally differentiated strategies for hydrocarbon and NO_X reductions would be more cost effective.**

396. Spatially disaggregated VOC emissions maps have been compiled by the NAEI, and are routinely reported for total VOC emissions in the UK. They have also been developed for a series of individual VOC source sectors, and this information has been used in UK ozone modelling assessments. Such assessments have included consideration of whether ozone formation is controlled by VOC or NO_X, and how this varies both regionally, and temporally at a given location.

Status report: Completed for the most important NAEI source categories.

- **Investigate the feasibility of identifying those hydrocarbons which are unreactive and make no significant contribution to ozone formation, so that emissions of these species could be deregulated and so that they could be used as substitutes for more reactive hydrocarbons.**

397. Reactivity-based strategies for photochemical ozone control, based on species substitution, have been identified, using the NAEI speciation in conjunction with the MCM (Derwent *et al.*, 2007c).

Status report: Completed for the latest NAEI speciation.

- **Further research is required to quantify the emissions fluxes of reactive oxygenated VOCs from vegetation in the UK and contiguous areas of continental Europe, and to assess the contribution these compounds of biogenic origin make to ozone formation in the region.**

398. Although flux measurements of biogenic VOC have received continued attention, and new tools to quantify and represent their emissions have been developed, these activities have not been focussed specifically on reactive oxygenated species. More generally, the contribution of biogenic VOC to regional-scale ozone formation has been estimated in modelling studies, within the limitations of current representations of emissions source strength and speciation (see also progress with the first recommendation in this 'Hydrocarbons' section).

Status report: No progress in the specific area of biogenic emissions of oxygenated VOCs and their impact. Improved characterisation of biogenic hydrocarbon sources in general is also required.

9.4 Synthesis and interpretation

- **The current monitoring of urban and rural photochemical oxidants and volatile organic compounds provides an excellent resource for the UK research community. Mechanisms to analyse and interpret these very expensive measurements should be initiated.**

399. Since the PORG report, the monitoring of NO_2 and O_3 has grown significantly, whereas the monitoring of VOCs has decreased in favour of fewer strategic sites and the increased monitoring of specific pollutants such as benzene and 1,3-butadiene. Data from these networks have been extensively analysed through various mechanisms and used to develop an improved understanding of the factors controlling the concentrations of these species. Examples of the use of these data include AQEG (2004, 2007a), Clapp and Jenkin (2001), Coyle *et al*. (2002), NEGTAP (2001), and many other references cited in the current report. These analyses have been supplemented by research council-led field campaigns where a wider range of VOCs and other species have been measured, e.g. the 2003 NERC TORCH campaign.

400. The present AQEG report illustrates the extensive analysis and interpretation that has been achieved using these measurements.

Status report: Achieved.

- **Specifically, the government's air quality strategy for ozone recognises the importance of taking further steps on the European scale to reduce ozone exposure levels. PORG therefore recommends that, in three years time, the government commissions an assessment of:**

- **The precursor emissions control actions taken across Europe by the year 2000, and their impact on ambient NO_x and hydrocarbon concentrations;**

- **The resulting change in ozone exposure levels and their likely impact on human health and damage to vegetation; and**

- **The requirement for and timing of additional policy actions to control hydrocarbons and NO_X across Europe, and their likely impact over the period up to 2010.**

401. There has been an ongoing commitment since the publication of the PORG report to continuously assess the effects of controls taken at a European level to reduce ozone concentrations. NEGTAP (2001) summarises many of the points raised above and additionally considers other pollutants such as sulphur dioxide (SO_2). That report provides a synthesis of research carried out over the three-year timeframe since PORG (1997) and considers human health effects, effects on vegetation and the effects of European-wide initiatives to reduce ozone precursor emissions. The effects of complying with the EU National Emission Ceilings and Ozone Directives were considered in a wide-ranging report (AEAT, 2000). Exposure to ozone in the UK was assessed using outputs from UK and European modelling studies up to 2010 (Stedman *et al.*, 1999). Other assessments include the analysis of urban VOC trends and peak ozone concentrations in north-west Europe and the effect of controls over motor vehicle emissions of VOCs (Derwent *et al.*, 2003).

Status report: Achieved by several means from local to regional assessments, both in the UK and elsewhere in Europe.

Annex 1

Emissions of ozone precursors

A1.1 Emissions of ozone precursors

402. Chapter 6 of this report discussed the performance of ozone models. The accuracy of historic and future emissions is just as important in making reliable decisions as the accuracy of the models themselves and emission estimates for future scenarios are always going to be much more uncertain. The Air Quality Expert Group (AQEG) recommends that the use of a future emissions estimate should be a transparent process in which a decision-maker can clearly see what the assumptions behind an estimate are. The decision-maker can then decide which scenario is more appropriate or decide to use more than one emission scenario to obtain a range of results. This Annex has been provided to help this process.

403. Later in the Annex, examples where emission estimates are subject to great uncertainty will be given. These include differences between the Intergovernmental Panel on Climate Change (IPCC) and International Institute for Applied Systems Analysis (IIASA) emission projections, and in volatile organic compound (VOC) speciation, biogenic VOC emissions and natural emissions.

404. ozone precursor emissions at different spatial scales have been reported and interpreted throughout the report to support the answers to each question.

405. Emission projections for the precursor pollutants carbon monoxide (CO), methane (CH_4), non-methane VOCs and nitrogen oxides (NO_X) have been considered on the global or northern hemispheric scale in order to interpret trends in global background ozone levels in the UK in Chapter 3; the impacts of climate change in Chapter 4; and the impacts on future global background ozone in Chapter 8. The analysis has drawn on the “current legislation” (CLE) and the “maximum technically feasible reduction” (MFR) global emission scenarios reported by IIASA and the Special Report on Emissions Scenarios (SRES) scenarios reported by IPCC.

406. Historic and future trends in NO_X and VOC emissions have been considered on a European scale in Chapter 2 on the interpretation of trends in urban and rural network ozone concentration data; in Chapter 4 on climate change impacts on ozone; in Chapter 7 on integrated assessment modelling of the ozone response to European emission reductions; and in Chapter 8 on the effects of UK and European emission control measures.

407. Historic and future trends in NO_X and VOC emissions have been considered on a UK scale to interpret temporal and spatial trends in background and peak ozone levels in Chapter 2. Trends in urban UK NO_X emissions from traffic have been used to interpret the urban ozone decrement through the NO_X-scavenging effect in Chapter 5. This issue also required consideration of trends in UK VOC emissions as potential drivers in local photochemical events. Projected UK NO_X and VOC emissions have provided the platform for discussions and quantitative analysis on effective control options to reduce UK population exposure to ozone discussed in Chapter 8.

408. This section gives a brief overview of the sources of emissions data on a global, European and UK scale used in the report.

409. When comparing and evaluating emission projections provided from different sources it is important to understand the differences in scenarios and assumptions used. Each organisation reporting emission projections has considered more than one set of core assumptions and generated emissions for a range of future scenarios. Versions of emission projections are periodically updated and changed by the same organisation that first developed them as the basic input parameters are better refined, so it is important to note which version of emission projections are used in any analysis. This report has stated several times that models have shown how future emissions are the most sensitive parameters affecting future trends in ozone concentrations.

A1.2 Global emission projections

410. On a global scale, the report has made particular reference to the CO, CH_4 and NO_X projections reported by IIASA. Using a global version of the Regional Air Pollution Information and Simulation (RAINS) model, IIASA has generated two anthropogenic emission scenarios known as the "current legislation" (CLE) scenario and the "maximum technically feasible reduction" scenario (MFR). Details of each of these scenarios were given in Chapter 1 and they have been widely applied in published modelling studies. Recently, during 2007, IIASA updated its precursor emissions estimates for 2000 and for the CLE and MFR scenarios going forward, and preliminary modelling studies using these new IIASA emission estimates are referred to specifically in Chapter 4. (Note that none of the IIASA scenarios include estimates of emissions from aviation or shipping.)

411. The IPCC Special Report on Emissions Scenarios (SRES) (IPCC, 2000) also provides global emission projections for ozone precursor gases for a wide range of scenarios covering the main emission driving forces from demographic to technical and economic development. These are broadly grouped into four families following different "storylines", each assuming a distinctly different direction for future developments. One of these, the SRES A2 scenario, has been referred to in this report as a more pessimistic, high-growth scenario. The SRES A2 scenario describes a very heterogeneous world based on self-reliance, regional differences in economic and technological development, and continuous increase in global population.

A1.2.1 Global methane emissions

412. Figure A1.1 shows the IIASA global anthropogenic emission projections of CH_4 by source sector for the CLE and MFR scenarios (Cofala *et al*., 2006).

413. The CLE scenario predicts a growth in global methane emissions of around 36% between 2000 and 2030 due mainly to increases in emissions from enteric fermentation, losses from natural gas distribution and waste treatment. In the MFR scenario, methane emissions fall in the short term, but then increase to levels in 2030 about 6% lower than 2000 levels. The differences in the MFR and CLE scenario emissions in 2030 are mainly due to lower emissions from enteric fermentation and wastewater treatment in the MFR scenario.

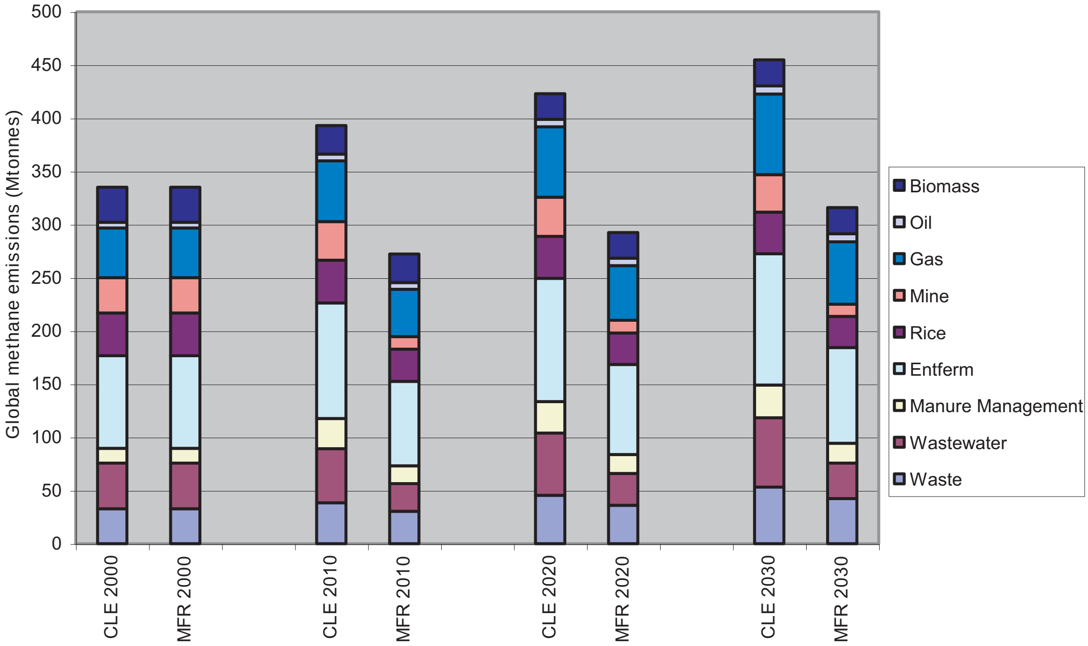

Figure A1.1 Global anthropogenic CH_4 emissions by source sector for IIASA CLE and MFR scenarios.

414. Figure A1.2 shows global anthropogenic emissions of CH_4 projected to 2030 by world region from the IIASA-RAINS model compared with projections from the SRES A2 scenario. From a similar 2000 base, the more pessimistic SRES A2 scenario predicts a faster growth (51%) in global methane emissions to 2030 than the CLE scenario.

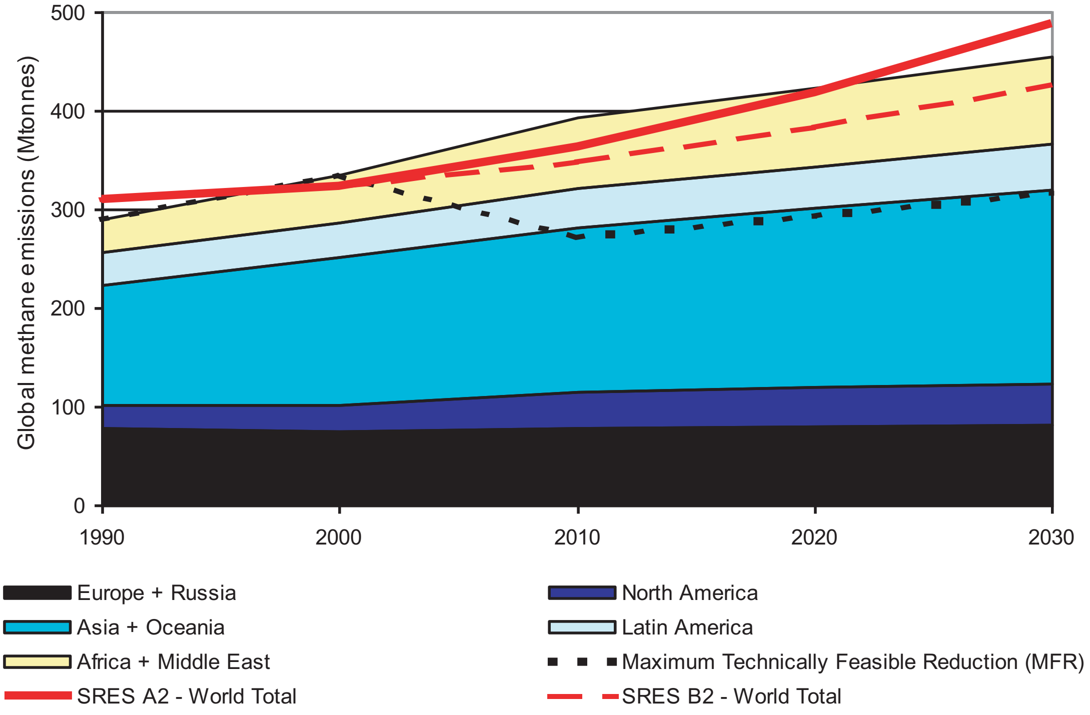

Figure A1.2 Global anthropogenic emissions of CH_4 showing a comparison of IIASA-RAINS CLE projections with IPCC SRES projections and RAINS MFR projections (world total)

A1.2.2 Global carbon monoxide emissions

415. Figure A1.3 shows the IIASA global anthropogenic emission projections of CO by source sector for the CLE and MFR scenarios (Cofala *et al.,* 2006).

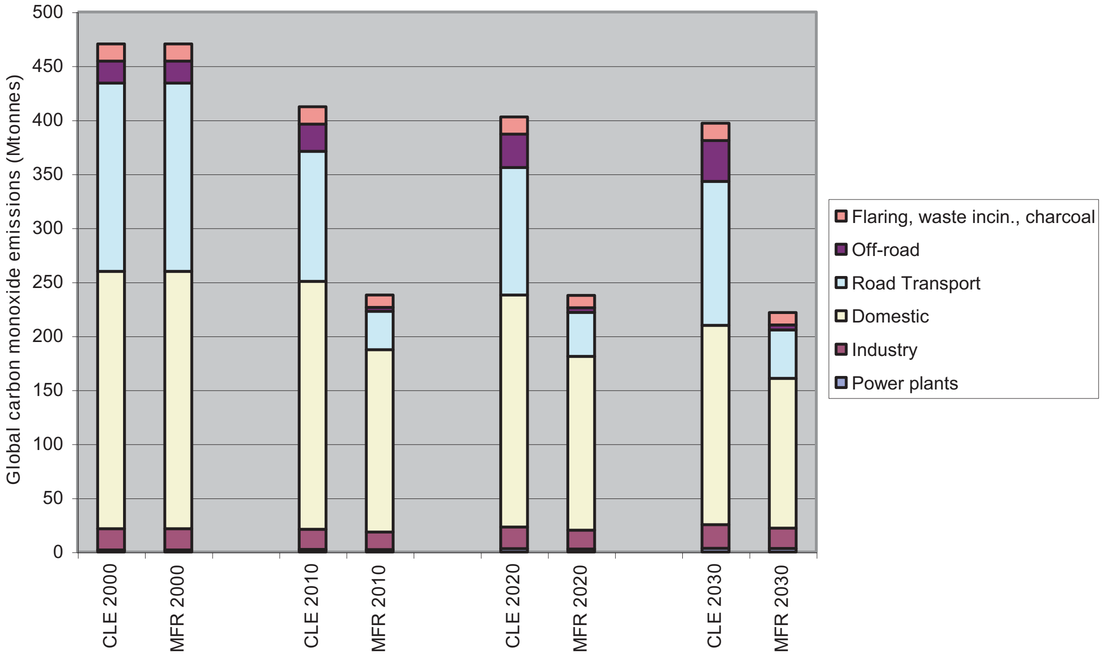

Figure A1.3 Global anthropogenic CO emissions by source sector for IIASA CLE and MFR scenarios.

416. The CLE scenario predicts a fall in global CO emissions of around 14% between 2000 and 2030 due mainly to a decrease in domestic combustion and road transport emissions, although transport emissions are on the rise again after initially falling substantially in the first decade to 2010. The MFR scenario predicts a more significant 53% reduction in global CO emissions due to bigger falls in the road transport and domestic combustion sector. In both scenarios, the decline in emissions occurs by 2010, with little further overall decline after this in either scenario.

417. Figure A1.4 shows global anthropogenic emissions of CO projected to 2030 by world region from the IIASA-RAINS model compared with projections from the SRES A2 scenario. From a rather lower 2000 base than the IIASA global emissions inventory, the more pessimistic SRES A2 scenario predicts a doubling in global CO emissions to 2030.

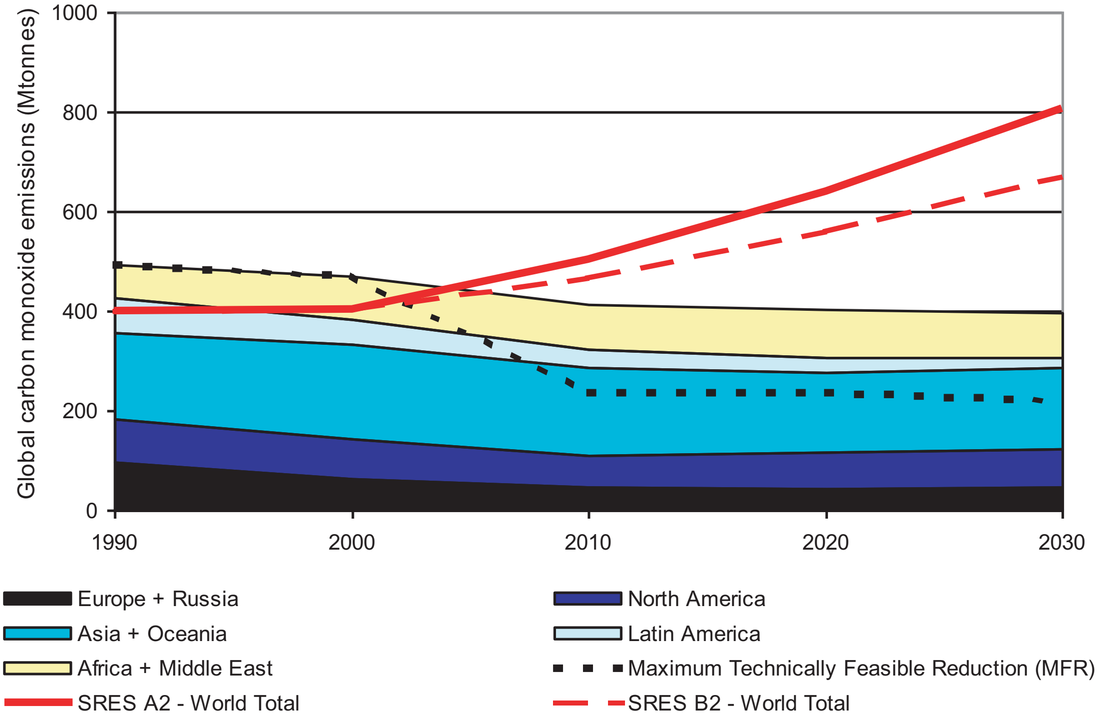

Figure A1.4 Global anthropogenic emissions of CO showing a comparison of IIASA-RAINS CLE projections with IPCC SRES projections and RAINS MFR projections (world total).

A1.2.3 Global nitrogen oxide emissions

418. Figure A1.5 shows the IIASA global anthropogenic emission projections of NO_X by source sector for the CLE and MFR scenarios (Cofala *et al.*, 2006).

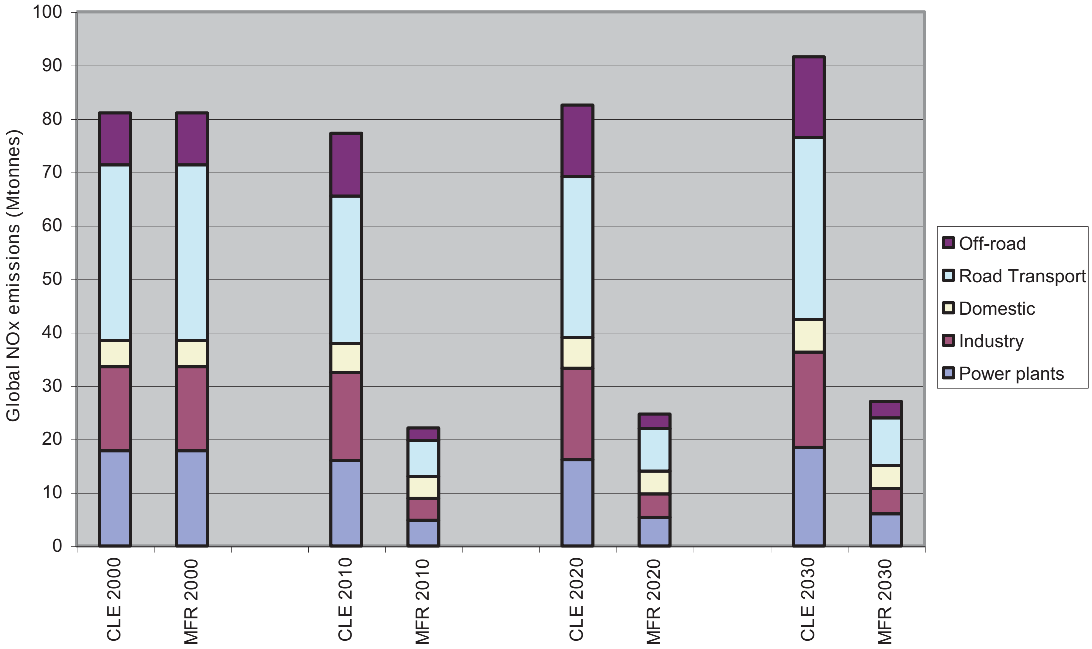

Figure A1.5 Global anthropogenic NO_X emissions by source sector for IIASA CLE and MFR scenarios.

419. The CLE scenario predicts a growth in global NO_X emissions of around 13% between 2000 and 2030. The dominant sector is road transport, but it is growth in emissions from off-road transport and machinery that is mainly responsible for the overall increase in global emissions over this time horizon, although after initially falling during the first decade, road transport emissions are increasing, surpassing 2000 levels by 2030. In the MFR scenario, NO_X emissions fall significantly, to levels in 2030 that are 66% lower than 2000 levels. The MFR scenario involves significant falls in emissions from all sectors, but the largest fall is from the road transport sector.

420. Figure A1.6 shows global anthropogenic emissions of NO_X projected to 2030 by world region from the IIASA-RAINS model compared with projections from the SRES A2 scenario. From a similar 2000 base, the more pessimistic SRES A2 scenario predicts an almost doubling (94% increase) in global NO_X emissions to 2030.

A1.2.4 Global non-methane volatile organic compound emissions

421. There are no assessments of global VOC emission projections from IIASA-RAINS. However, Dentener *et al*. (2005) suggested anthropogenic emissions of VOCs would closely follow the development of CO emissions given the importance of road transport to both pollutants. They suggested global emissions of VOCs of around 250 Mtonnes in 2000 (including biomass burning) that would increase by 17% to 2030 in a CLE scenario and would decrease by 10% to 2030 in a MFR scenario. The more pessimistic SRES A2 scenario predicts a 43% increase in global VOC emissions over this period (IPCC, 2001).

A1.2.5 Global emissions from international shipping and aviation

422. In modelling the impact of global emission controls on tropospheric ozone, Dentener *et al*. (2005) considered the changes in emissions from international sea traffic and aviation. International shipping makes an important contribution to global NO_X emissions and Dentener *et al*. (2005) assumed a 1.5% yr^{-1} growth in activity. Global aviation emissions of NO_X were predicted to rise from 2.6 Mtonnes in 2000 to 5.7 Mtonnes in 2030. This would be around 20% of global NO_X emissions from other anthropogenic sources of NO_X in the MFR scenario for 2030. Eyring *et al*. (2007) also studied the impact of shipping emissions on near-surface ozone concentrations. Two scenarios were modelled, one of which assumed a constant annual growth rate of 2.2% between 2000 and 2030, with emissions of NO_X growing unabated. Using an ensemble of atmospheric chemistry models, Eyring *et al*. (2007) showed that ozone would grow almost linearly with increases in international shipping NO_X emissions under the same background conditions. The maximum contributions from shipping to annual mean ozone concentrations was predicted to be 8 ppb over the North Atlantic in 2030. The study also pointed out the importance of ship plume dispersion in global models, with the lifetime of NO_X in ship plumes being significantly reduced compared to the background due to high in-plume oxidation.

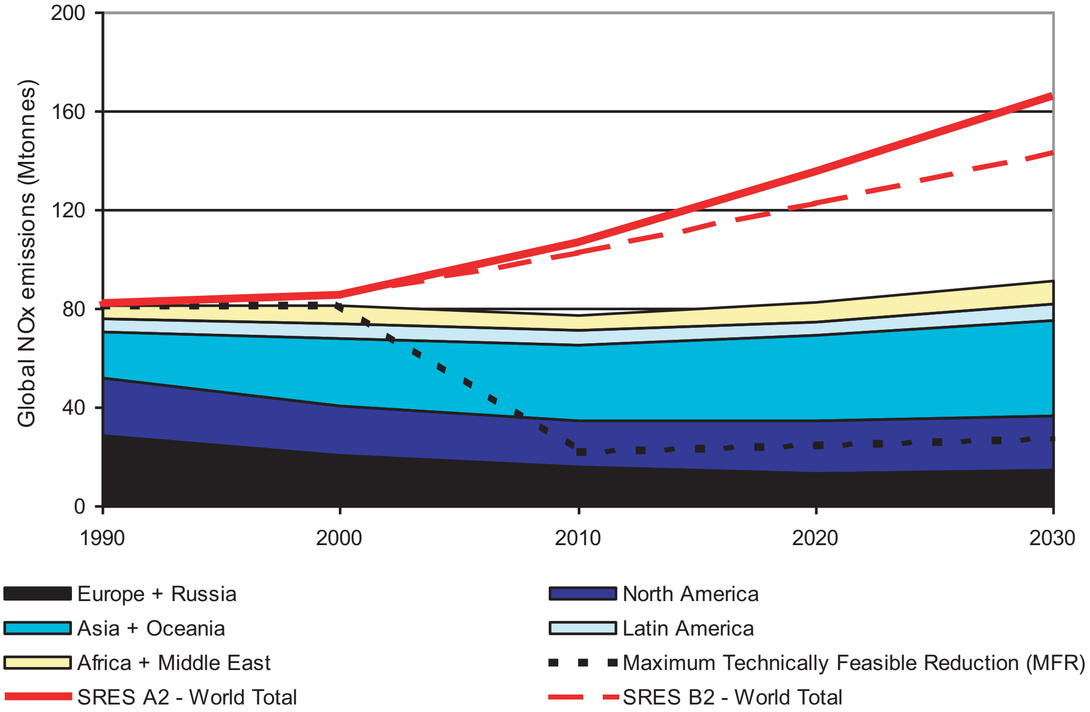

Figure A1.6 Global anthropogenic emissions of NO_X showing a comparison of IIASA-RAINS CLE projections with IPCC SRES projections and RAINS MFR projections (world total)

A1.3 European emission projections

423. Emission projections for the ozone precursors NO_X and VOCs, as well as ammonia (NH_3), sulphur dioxide (SO_2), particulate matter (PM) and carbon dioxide (CO_2), have been generated on the European scale by IIASA up to 2020. The projections were calculated using the GAINS-Europe model (Greenhouse Gas and Air Pollution Interactions and Synergies), an extension of the RAINS model, and are available on-line at http://www.iiasa.ac.at/web-apps/apd/RainsWeb/.

424. Several different versions of emission projections have been produced for individual European countries and regions following the CLE scenario. This report will focus on the NEC02 Version which contains recent scenarios used for the analysis of the revision of the National Emission Ceilings Directive (NECD). In particular, these scenarios include all information that has been collected during the bilateral consultations between IIASA and the teams of national experts during 2005/06. It contains the background scenarios for the revision of the NECD, as well as emission control scenarios that meet the objectives of the EU Thematic Strategy on Air Pollution. Data are shown here from the national background CLE scenario, referred to as NEC_NAT_CLE4REV which uses national background activity paths (as of December 2006) with emission controls reflecting current legislation on national and international emission, fuel quality, and product standards.

425. Regional-scale ozone forecasting models generally apply the IIASA European emission projections by sector and country to the EMEP 50 km x 50 km emission grids for the current years.

A1.3.1 European NO_X and VOC emissions by source sector

426. Figures A1.7 and A1.8 show projections of anthropogenic NO_X and VOC emissions from the EU27 countries (the 27 member countries of the EU after 1st January 2007) by source sector at SNAP1 level (Selected Nomenclature for Air Pollution – a system for defining the level of source sector detail in reporting emissions to EMEP/CORINAIR) for the version shown.

427. NO_X emissions are predicted to decline by 43% in 2020 relative to 2000 levels. This reduction is mostly due to reductions in emissions from road transport as petrol and diesel vehicles meeting the tighter European emission standards penetrate the European fleet. Road transport contributed 45% of EU27 NO_X emissions in 2000 and emissions from this sector are projected to fall by 67% by 2020. Small reductions are expected in emissions from power generation and from other transport and off-road machinery.

428. VOC emissions are also predicted to decline by 43% in 2020 relative to 2000 levels. This reduction is also mostly due to reductions in emissions from road transport as petrol and diesel vehicles meeting the tighter European emission standards penetrate the European fleet. However, there are also predicted to be significant reductions from solvent use, with implementation of the Solvent Emissions Directive, and from domestic and non-industrial combustion. Solvent use made the largest contribution to EU27 VOC emissions in 2000 and emissions from this sector are projected to fall by 30% by 2020. Road transport contributed 28% of EU27 VOC emissions in 2000 and emissions from this sector are projected to fall by 81% by 2020.

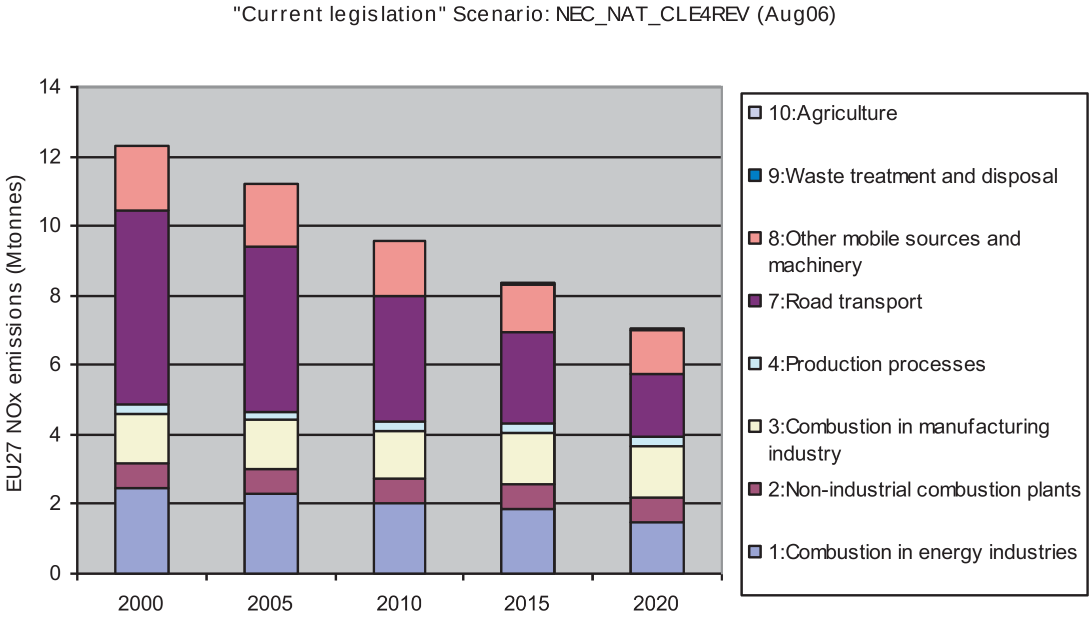

Figure A1.7 Anthropogenic NO_X emissions for EU27 countries by source sector for CLE scenario.

A1.3.2 Emissions from shipping in European waters

429. The EU27 emission projections presented above do not include emissions from shipping in European sea regions, but these have been estimated for sea traffic within the EMEP area, provided for EMEP models by the Centre for Integrated Assessment Modelling (CIAM, see Expert Emissions at http://webdab.emep.int/). EMEP international shipping emissions are divided into five regions: Baltic Sea, Black Sea, Mediterranean Sea, North Sea (including the English Channel) and the remaining North-East Atlantic Ocean within the EMEP region. For NO_X, the EMEP projections suggest the sum of international shipping emissions for these five sea regions will increase from 3.35 Mtonnes in 2000 to 3.99 Mtonnes in 2020, an increase of 19%. By 2020, the shipping emissions will be 57% of land-based anthropogenic emissions for the EU27 countries. For VOCs, the EMEP projections suggest the sum of international shipping emissions for these five sea regions will decrease from 0.114 Mtonnes in 2000 to 0.070 Mtonnes in 2020, a decrease of 38%, but shipping makes a very small contribution to VOC emissions relative to land-based anthropogenic emissions. By 2020, the shipping emissions will be 1% of land-based anthropogenic emissions for the EU27 countries.

430. A more detailed investigation into European shipping emissions has been carried out by Entec for the European Commission (Entec, 2005). This study provided estimates of European shipping emissions projected to 2020 broken down in various forms of country assignment. The different assignments distinguished between where international shipping emissions actually occur and which country is responsible on the basis of, for example, the ship’s flag, sale of fuel, country of departure and destination, among various other methods of assignment. For modelling ozone, it is the location of the emissions that is obviously the most relevant.

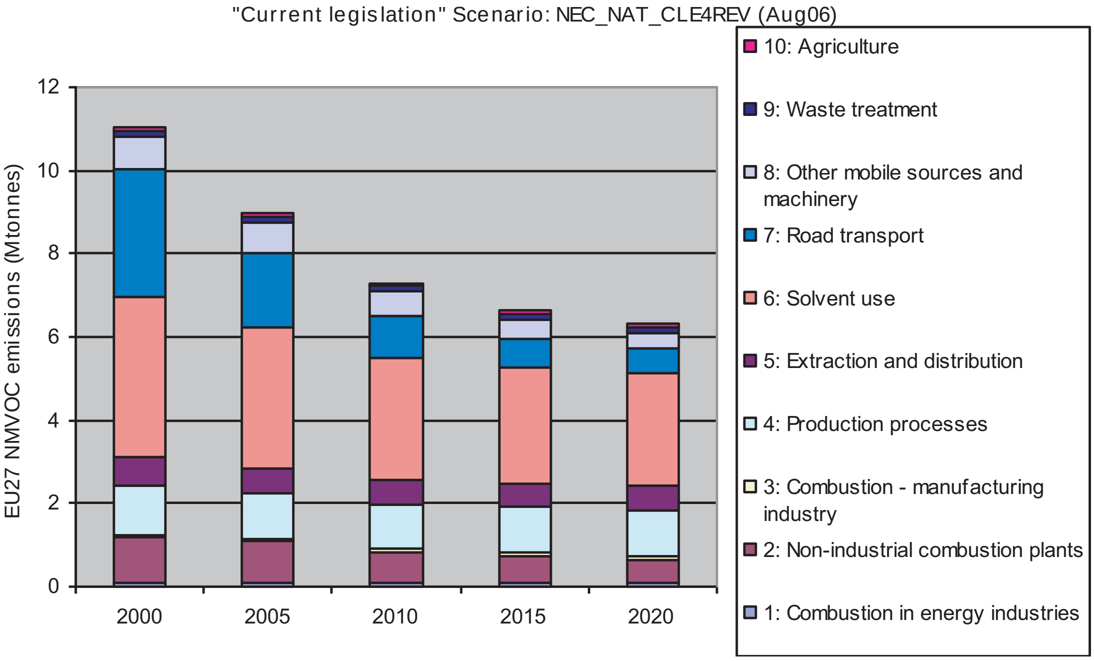

Figure A1.8 VOC emissions for EU27 countries by source sector for CLE scenario.

431. Method A in the Entec study referred to assignment according to location of emissions within a 12-mile zone area that equates to territorial waters, and within what is referred to in the UN Convention on the Law of the Sea (UNCLOS) as a 200-mile zone area, known as an Exclusive Economic Zone. The amount of emissions occurring in the 12-mile or 200-mile zones cannot be directly compared with the EMEP international shipping emissions that cover whole sea regions. However, the Entec study indicated NO_X emissions from shipping within the 200-mile zones for the EU27 countries plus Turkey and Croatia would increase from 2.81 Mtonnes in 2000 to 4.06 Mtonnes in 2020, an increase of 45%. This is greater than the relative increase in international shipping emissions for the five sea regions predicted by EMEP. Emissions of VOCs from shipping in the same area were predicted to increase from 0.099 Mtonnes in 2000 to 0.165 Mtonnes in 2020, an increase of 67%, in contrast to a decline in international shipping emissions of VOCs for the five sea regions predicted by EMEP. The Entec study indicates that these zonal shipping emissions of VOCs are very small compared with land-based anthropogenic emissions for these countries (2.5% of EU27 emissions in 2020), but the zonal shipping emissions of NO_X are very significant compared with land-based anthropogenic emissions for these countries, reaching 58% of EU27 emissions in 2020. This is similar to the situation predicted by EMEP for the five sea regions. Moreover, for the 12-mile zones, Entec (2005) concluded that the UK has the highest emissions assigned from ships.

A1.4 UK emission projections

432. The National Atmospheric Emissions Inventory (NAEI)[12] provides UK emission projections for the ozone precursors NO_X and VOCs and all the other NECD pollutants, SO_2, NH_3, and PM_{10}. The projections are primarily based on Department for Business, Enterprise and Regulatory Reform (BERR) (formerly the Department of Trade and Industry, DTI) energy forecasts (by fuel and sector), Department for Transport (DfT) traffic forecasts and various other economic drivers. The BERR energy forecasts limit emission projections to 2020, though DfT's road transport projections go further to 2025.

433. The NAEI emission projections are updated each time the BERR updates its energy projections or the DfT updates its traffic forecasts. The projections used in most of the modelling referred to in this report are based on BERR's UEP26 "Favourable to Coal" energy scenario and are from a 2004 base. For VOCs, the projections are based on the slightly older BERR UEP21 scenario, as VOC projections had not been run for the UEP26 scenario. However, the main differences between UEP21 and UEP26 projections are in the forecasts of coal and gas use for power station and domestic consumption, which make a relatively small contribution to VOC emissions, so the UEP26 projections for VOC emissions will not be significantly different to the UEP21 projections.

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[12] www.naei.org.uk/

A1.4.1 UK NO_X and VOC emissions by source sector

434. Figures A1.9 and A1.10 show UK projections of anthropogenic NO_X and VOCs.

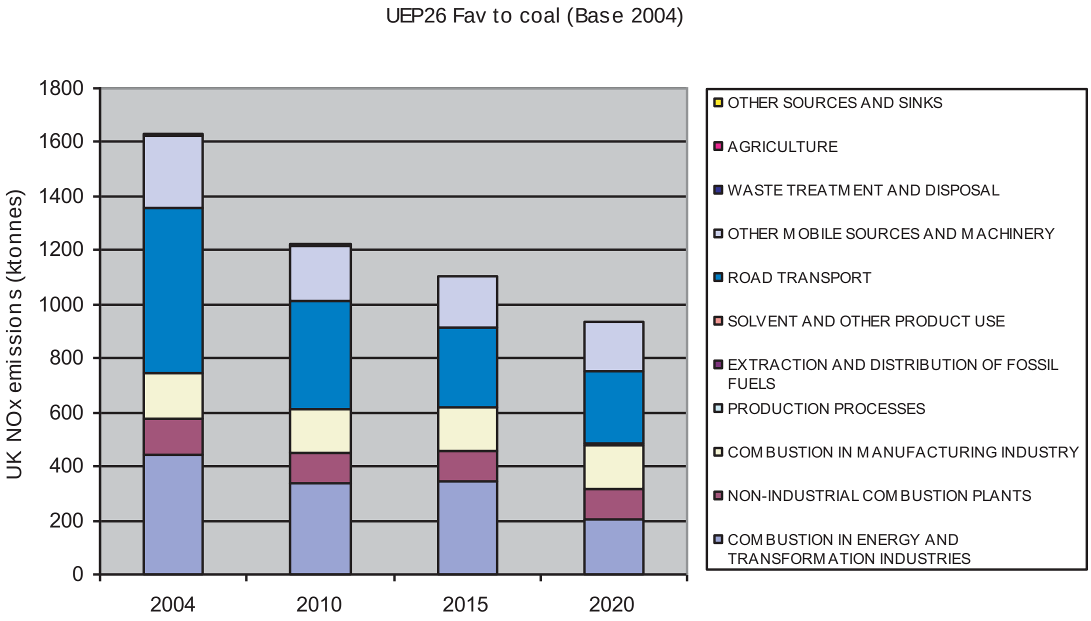

Figure A1.9 UK anthropogenic NO_X emissions (UEP26) from the NAEI (2004 base).

435. UK anthropogenic NO_X emissions are predicted to fall by 42% from a 2004 base by 2020. This is mainly due to the reduction in emissions from road transport which are predicted to fall by 56% over this time period due to the penetration of new vehicles meeting the tighter European emission standards in the UK fleet. The road transport emission projections presented here and used in all the air quality modelling work described in this report exclude the recently agreed EURO V/VI emission standards for light duty vehicles that will further reduce emissions of NO_X and VOCs from the road transport sector after 2010. After road transport, emissions from power generation are the next largest source of NO_X emissions and these too are expected to have decreased significantly (by 54%) by 2020. In urban areas, road transport emissions will further dominate NO_X emissions and a decrease in urban road transport emissions of 58% is expected between 2004 and 2020 (Murrells and Hobson, 2006), contributing to the reduction in the NO_X-scavenging effect on ozone in urban areas.

436. A smaller reduction in UK VOC emissions is predicted: 15% from a 2004 base by 2020. Solvent use is the largest source of VOC emissions in the UK, but is only forecast to decline by 9% over this time period. Reductions in road transport emissions are the major contributor to the overall reductions in VOC emissions. Emissions from this sector are expected to decrease by 63% over this time period.

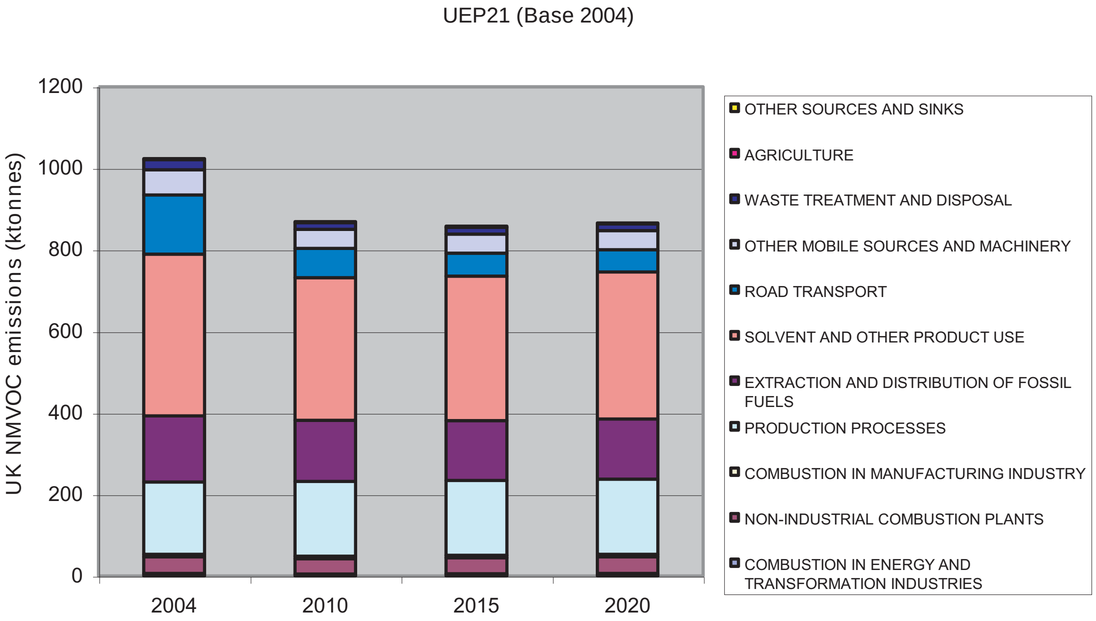

Figure A1.10 UK anthropogenic VOC emissions (UEP21) from the NAEI (2004 base).

A1.4.2 Speciated anthropogenic VOC emissions

437. The NAEI produces a speciated VOC inventory that is widely used in UK ozone models, including the Photochemical Trajectory Model (PTM) and the Ozone Source-Receptor Model (OSRM). A profile is constructed for around 250 different sources in the inventory involving altogether 664 individual or groups of VOCs. Each year, when a new VOC emissions inventory is compiled for the UK on the basis of total VOC emission factors and activity data, the speciation profile is applied to provide a new speciated VOC inventory for that year. An assumption is made that with the exception of benzene and 1,3-butadiene which are modelled explicitly, the speciation profile for a given source sector remains unchanged each year.

438. The speciation profile was last reviewed in 2002 by Passant (2002). The report provides the mass fraction of individual VOCs emitted by each individual source, together with their Photochemical Ozone Creation Potential (POCP) values.

439. The speciation profile for each sector comes from a number of sources. One of these profiles is from the US EPA and is used mainly for the combustion sources. More UK-specific sources are used for solvents and the chemicals industry. These are based on discussions with industry representatives, site measurements and product data. Profiles for road transport sources are taken from the European road transport emission factor model, COPERT III (EEA, 2000).

440. A considerable amount of uncertainty must be assigned to some of the sources. The profiles known with most accuracy are likely to be for the solvents sector, as this industry has knowledge of the chemical constituents of the solvents used. Some other sources associated with process industries are also known with reasonable accuracy because the industries are aware of the chemicals involved in the processes. For other sources, the profiles can be quite varied. For example, the profile for petrol vapour will vary to a certain degree with refinery source and also with time of year (e.g. winter blends of petrol are

more volatile than summer blends and contain more lighter butanes). The chemical composition of petroleum fuels can vary with year in order to comply with European fuel quality directives.

441. The UK's speciation profile for VOCs is currently the one most widely used across Europe. Others have been developed and used in some chemical transport models such as EMEP, including the recent speciation profile developed by Theloke and Friedrich (2007). This VOC profile covered 306 single species or classes of VOCs in 87 source sectors. It would be useful to compare this profile with that developed for the NAEI which is more UK specific.

A1.5 Emissions from natural sources and biomass burning

A1.5.1 Biogenic VOC emissions

442. Biogenic VOCs describe a wide range of compounds, of which only a few are generally of interest and represented in chemical transport models. Isoprene is the compound of most importance for ozone modelling for example. Emissions of the various terpenes (e.g. α- and β-pinene) are also important for both ozone and aerosol formation. The remaining VOC species (these are termed in inventories as 'other VOC', or OVOC, and are non-methane VOC species other than isoprene and monoterpenes emitted by vegetation, including oxygenated VOCs, but also non-oxygenated species) play some role, but little is known about the chemistry of many components or the quantitative emissions of individual species. Emissions may be large, however.

443. The emission flux of biogenic VOCs for all types of vegetation is most often described on an hourly basis by the algorithm (Guenther *et al.,* 1996):

$$\text{Flux } (\mu\text{g m}^{-2}\text{ yr}^{-1}) = \int \varepsilon \, D \, \gamma \, dt$$

where ε is the average emission potential (μg g^{-1} h^{-1}) for the particular species, D is the foliar biomass density (g dry weight foliage m^{-2}), and γ is a dimensionless environmental correction factor representing the effects of short-term (e.g. hourly) temperature and solar radiation changes on emissions. Guenther *et al.* (1991, 1993) had previously shown that, to a very good approximation, the short-term (hourly) variations in emissions of isoprene could be described by the product of a light-dependent factor, C_L, and a temperature-dependent factor, C_T. This is the basis of the Biogenic Emissions Inventory System (BEIS)[13] and the calculation of biogenic VOC emissions in most current models for ozone (including the EMEP Unified and the US Community Multiscale Air Quality (CMAQ) models). As such, annual emission estimates are not a routine input or output, making it difficult to compare the emission estimates used in different models.

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[13] see http://www.epa.gov/asmdnerl/biogen.html

A1.5.1.1 Global biogenic emissions

444. The Global Emissions Inventory Activity (GEIA) provides global emission inventories on a one degree grid for the entire world for a number of atmospheric species, including VOC emissions from biogenic sources. The VOC emissions from biogenic sources are available as monthly datasets for 1990[14]. This inventory dataset is effectively that of Guenther *et al*. (1995). The EDGAR database – Emission Database for Global Atmospheric Research – only considers VOC emissions from biomass burning[15].

445. For the 3rd IPCC Assessment, emissions of isoprene, monoterpenes, and other VOC were calculated using the GLOBEIS model of Guenther et al. (1999)[16]. The foliar densities, emission capacities and algorithms used to determine emissions activities for both 2000 and 2100 were the same as those described by Guenther *et al*. (1995). One difference between this work and that of Guenther *et al*. (1995) was the use of hourly temperatures for each hour of a month to determine the monthly average emission rate, as opposed to the use of monthly average temperatures to drive emission algorithms. This resulted in about a 20% increase in isoprene emissions in 2000 and 10% increase in emissions of other biogenic VOC. Global annual emissions of isoprene for 2000 were ~600 Mtonnes (compared to 506 Mtonnes in Guenther *et al*. (1995)) and emissions of monoterpenes for 2000 were 146 Mtonnes (compared to the estimate of 127 Mtonnes in Guenther *et al*. (1995)). These increase by 23% for 2100 relative to the 2000 scenario.

446. Guenther *et al.* (2006) developed MEGAN (Model of Emissions of Gases and Aerosols from Nature) as an improved approach to estimating emissions on the global and regional scales. MEGAN provides emission estimates at a spatial resolution of ~1 km x 1 km using a combination of modelled, ground and remote sensing information and data. Guenther *et al*. (2006) gave a range for the global annual emission rate of isoprene of 500 to 750 Mtonnes (as isoprene), depending on the assumptions made about the driving variables, such as temperature, solar radiation, leaf area index and plant functional type. The same changes in the driving variables could give rise to larger local differences in the isoprene emission rates (greater than a factor of 3). Using a 'standard set' of driving variables, the global annual isoprene emission was estimated as ~600 Mtonnes. Using temperature distributions from global climate models, isoprene emissions were predicted to be a factor of two higher by 2100.

447. Dentener *et al*. (2005) considered global contributions from natural sources in their study on the impact of emission controls on tropospheric ozone. They estimated 507 Mtonnes VOC emissions from vegetation (predominantly isoprene) in 2000, approximately double the global anthropogenic emissions in 2000 (including biomass).

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[14] http://www.geiacenter.org/

[15] http://www.mnp.nl/edgar/

[16] see http://www.globeis.com/

A1.5.1.2 European biogenic emissions

448. Simpson *et al*. (1995) used the BEIS emission algorithms, together with meteorological data from the EMEP MSC-W ozone model, to generate estimates of the emissions of isoprene from European forests and agricultural crops over several summer periods. European isoprene emissions were estimated to be about 4,000 ktonnes carbon (C) per annum, approximately 50-100% greater than previous estimates. Estimates were also made of the emissions of OVOCs from forests. These estimates were improved further and documented by Simpson *et al*. (1999) and were the basis of Table 2.1 in Chapter 11 of the CORINAIR Emission Inventory Guidebook (CORINAIR, 2002).

449. As part of the EU Pan-European Land Use and Land Cover Modelling (PELCOM) project, and building on the work of Simpson *et al*. (1999), Winiwarter *et al*. (2001) used the land cover dataset derived in the project to improve existing information on emissions of terpenes and isoprene from forests. As there was insufficient discrimination between deciduous/mixed/coniferous forest types in the land cover dataset, the total PELCOM forest coverage was used and combined with a detailed forest species distribution taken from an assessment by TNO (Nijenhuis, 1999). A forest atlas was produced to derive species percentages on a 1° x 2° grid over Europe. Table A1-1 gives the biogenic VOC emissions derived by Winiwarter *et al*. (2001) by country and is used to illustrate the contribution of nine representative species. As the study used the same emission factors and country environmental correction factors, the emission totals agreed well with the earlier estimates of Simpson *et al*. (1999). Table A1-1 also includes reported national estimates of VOCs from man-made sources in 1990 and the National Emission Ceilings for 2010 agreed in the Gothenburg Protocol. To put into context, the biogenic VOC emissions for the EU27 countries from Winiwarter *et al*. (2001) in Table A1-1 make up 65% of the anthropogenic emissions from this region in 2005 shown in section 2.6.

450. The recently completed EU NATAIR project (Improving and Applying Methods for the Calculation of Natural and Biogenic Emissions and Assessment of Impacts on Air Quality, http://natair.ier.uni-stuttgart.de/) has been developing a new high-resolution (10 km) inventory for all relevant pollutant emissions from natural and biogenic sources in Europe. The emission database will cover a number of current and future years (including 2000 and 2010) with hourly temporal resolution. Steinbrecher *et al*. (2008) derived estimates of European isoprene and monoterpene emissions for July 2003 for the domain used, ranging from 1124-1446 Mtonnes and from 338-1112 Mtonnes respectively. Small-scale deviations could be as high as +0.6 ktonnes km^{-2} for July 2003, reflecting the current uncertainty range for biogenic VOC estimates. Key sources of errors in the inventories were identified as insufficiently detailed land-use data and little or no information on the chemical speciation of the plant-specific emission potentials. Table A1-2 shows the annual biogenic VOC emissions derived for the UK for the years 1997, 2000, 2001 and 2003. The NATAIR estimate appears slightly higher than other recent UK emission estimates shown in Table A1-3 but well within the current uncertainties for these inventories.

Table A1-1 Comparison of the emissions of volatile organic compounds by tree species and country derived in the PELCOM project with, national estimates of non-methane VOCs from man-made sources in 1990 and the National Emission Ceilings as given in the Gothenburg Protocol for 2010. Units: ktonnes per year.

Emission Species Country	Coniferous				Deciduous					Total	Anthropogenic	
	Pine	Spruce, Fir	Larch	Other	Quercus ssp.	Quercus ilex	Beech	Birch, Poplar	Other		1990	2010
Albania	1	2	1	–	28	14	1	–	2	51	–	–
Austria	8	95	9	–	14	2	2	–	1	132	351	159
Belarus	117	57	–	–	45	–	–	35	5	260	533	309
Belgium	2	10	1	–	23	–	1	–	–	38	324	144
Bosnia and Herzegovina	2	20	5	1	28	4	5	–	4	68	–	–
Bulgaria	25	12	2	–	173	12	5	–	4	234	217	185
Croatia	3	13	2	1	42	9	6	–	4	80	105	90
Czech Republic	18	83	5	1	18	–	1	–	1	127	435	220
Denmark	1	10	–	1	3	–	–	–	–	15	178	85
Estonia	19	22	–	–	4	–	–	–	2	47	–	–
Finland	149	146	–	–	–	–	–	12	2	309	209	130
France	119	71	49	8	593	66	11	2	17	935	2957	1100
Germany	84	236	25	1	88	1	9	1	3	448	3195	995
Greece	24	13	9	1	136	61	2	–	2	248	373	261
Hungary	6	18	2	–	74	–	2	–	4	107	205	137
Ireland	2	7	–	–	1	–	–	–	–	11	197	55
Italy	25	57	19	1	210	70	10	–	13	405	2213	1159
Latvia	32	22	–	–	5	–	–	3	2	64	152	136
Lithuania	22	21	–	–	5	–	–	2	1	52	103	92
Luxembourg	–	1	–	–	3	–	–	–	–	5	20	9
Macedonia	3	1	–	–	25	11	1	–	1	43	–	–
Netherlands	4	2	1	–	5	–	–	–	–	12	502	191
Norway	30	60	–	–	2	–	–	3	1	95	310	195
Poland	164	61	11	–	42	1	3	2	5	288	831	800
Portugal	62	–	–	2	138	45	–	–	3	251	640	202
Republic of Moldova	–	1	–	0	24	–	–	–	1	27	157	100
Romania	1	118	17	0	167	–	12	–	9	323	616	523
Russia	732	1452	6	5	282	–	2	3033	23	5534	3566	–
Russia – PEMA	–	–	–	–	–	–	–	–	–	–	203	165
Serbia and Montenegro	6	16	4	1	105	9	9	–	7	158	–	–
Slovakia	7	40	6	1	24	–	3	–	2	83	149	140
Slovenia	3	17	2	–	12	4	2	–	1	41	42	40
Spain	337	–	11	9	406	302	5	1	12	1083	1094	669
Sweden	120	285	–	–	17	–	1	14	4	441	526	241
Switzerland	1	16	6	1	3	1	1	–	–	29	292	144
Turkey	115	19	25	11	248	11	4	–	9	443	–	–
Ukraine	98	90	12	–	243	–	5	24	11	484	1369	797
United Kingdom	15	36	7	1	6	–	1	1	1	67	2555	1200
Sum	2357	3130	237	46	3242	623	104	3133	157	13038		
Sum without Turkey	2242	3111	212	35	2994	612	100	3133	148	12595		
Sum Simpson (without Turkey)	2036	4356	137	159	2408	360	56	994	211	10716		

Table A1-2 Estimate of annual UK VOC emissions from natural sources (in ktonnes per annum) derived for 1997, 2000, 2001 and 2003 as part of the EU NATAIR project.

Source	1997	2000	2001	2003
Biomass burning (open fires)	–	0.01	0.01	0.01
Forest	152.21	139.33	138.33	152.14
Other land use	128.12	115.24	114.20	129.28
Total	**280.33**	**254.58**	**252.55**	**281.43**

A1.5.1.3 UK biogenic emissions

451. The current estimate of UK biogenic VOC emissions by the NAEI programme for the 2003 and 2004 emission inventory years is 91 ktonnes per annum[17] and this emission estimate is used for all years in the emission time series. This estimate is based on the work of Stewart *et al*. (2003), who combined land-use information with the plant and tree species likely to be found in each class, based on the Natural Environment Research Council (NERC) Countryside Survey. Stewart *et al*. (2003) also developed a database of the VOC emission potentials of around 1000 plant species, obtained from the literature, although in practice only around 50 species were of importance for the UK. The meteorological parameters required (temperature and photosynthetically-active radiation flux) were modelled for the year 1998 using output from the MM5 mesoscale meteorological model on a 12 km x 12 km grid with hourly resolution. The estimates had a high uncertainty (up to a factor of four). Dore *et al*. (2003) undertook a detailed comparison of the inventory of Stewart *et al*. (2003) with the inventory derived in the PELCOM project for the UK (Winiwarter *et al.,* 2001). The NAEI estimates are compared with these other estimates in Table A1-3.

452. Hayman (2006) used the Ozone Source-Receptor Model (OSRM) to estimate the UK VOC emissions from biogenic sources (trees and forests) for the years 1997 to 2003 using the emission potential inventory described by Dore *et al*. (2003). The calculated annual emissions of isoprene, terpenes and other volatile organic compounds (OVOCs) are also presented in Table A1-3.

453. The isoprene and total emissions derived from the OSRM appear to be in good agreement with the current NAEI estimates. The lower terpene emissions are offset by the OVOC emissions, which were not estimated by Stewart *et al*. (2003). The previous NAEI estimate of 178 ktonnes per annum was taken from the work of Simpson *et al*. (1999) and the emissions of isoprene, terpenes and OVOCs were estimated to be 58, 31 and 89 ktonnes per annum respectively. The OSRM isoprene and OVOC emission estimates are lower but the terpene emissions are higher. It should be noted that the OVOC emissions derived by Simpson *et al.* (1999) included emissions of 43.6 and 25.7 ktonnes per annum from pasture and crops respectively; the former and possibly the latter sources are not included in the OSRM inventory. These are not, however, large sources of isoprene and terpenes in the UK.

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[17] For 2002 and earlier emission inventory years the estimate was 178 ktonnes per annum.

454. The OSRM terpene and OVOC emissions are reasonably consistent with those given in Simpson *et al*. (1999) but the isoprene emission estimate is significantly lower. Part of the difference in the emission estimates is almost certainly due to assumptions concerning emission potentials and possibly land cover.

455. Models generally use emission potential inventories and implicitly take account of the seasonal differences in emissions. Annual estimates of emissions do provide a useful way of comparing different emission inventories. The NAEI 2003 and OSRM estimates of UK biogenic emissions are both around 9% of anthropogenic sources of VOCs in the UK in 2005. The rather lower PELCOM figure implies biogenic emissions of around 7% of anthropogenic sources of VOCs in the UK in 2005.

Table A1-3 Comparison of annual UK VOC emission estimates from biogenic sources (ktonnes per annum).

Inventory estimate	Isoprene	Terpenes	OVOC	Total emissions
EMEP/CORINAIR (2002)	53	39	27	119
NAEI 2002 Inventory(1)	58	31	89	178
NAEI 2003 Inventory(2)	8	83	–	91
Stewart *et al*. (2003)	8(3)	83(3)	–	–
PELCOM (Winiwarter *et al*., 2001)	–	–	–	67
OSRM – 1997	6.7	49.0	34.8	90.5
OSRM – 1998	5.7	46.0	33.0	84.7
OSRM – 1999	6.6	47.0	33.4	87.0
OSRM – 2000	6.3	46.0	32.8	85.1
OSRM – 2001	6.5	45.8	32.7	85.0
OSRM – 2002	6.3	48.4	34.5	89.3
OSRM – 2003	7.1	51.7	36.7	95.5

Notes (1) See Dore *et al*. (2004), biogenic VOC emissions taken from paper of Simpson *et al*. (1999); (2) Passant (2006), biogenic VOC emissions taken from work of Stewart *et al*. (2003); (3) Emissions reported for 1998 in ktonnes carbon per annum.

A1.5.2 Emissions from biomass burning

456. Dentener *et al*. (2005) also considered contributions from biomass burning in their study based on the EDGAR database and other sources. The amounts of biomass burning are not well established and fluctuate from year to year, but it does make a particularly large contribution to global CO emissions and, to a lesser but still important extent, to NO_X and VOC emissions. Dentener *et al*. (2005) made estimates of global CO, NO_X and VOC emissions from biomass burning in 2000, which represent 122%, 33% and 47% of global anthropogenic emissions of these pollutants from other sources, respectively. Emissions of NO_X and CO are dominated by biomass burning in much of Africa and South America.

A1.5.3 Emissions from other natural sources

457. Global emission estimates have been made for other natural sources such as soils, wetlands, oceans and lightning. The ACCENT website has links to a number of global emission inventory datasets (POET, EDGAR, RETRO) covering these sources[18].

458. Dentener *et al.* (2005) provided estimates of global emissions from various natural sources. These included:

- 38 Mtonnes NO_X emissions from soils and lightning – 47% of global anthropogenic emissions in 2000;
- 150 Mtonnes CO emissions from soils and oceans – 32% of global anthropogenic emissions in 2000; and
- 240 Mtonnes CH_4 emissions from wetlands and termites – 72% of global anthropogenic emissions in 2000.

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[18] http://www.aerojussieu.fr/projet/ACCENT/database.php

Annex 2

Additional question A supporting evidence

A2.1 Ozone trends at 18 rural/remote and 45 urban sites based on data up to 2005

Table A2-1 Trends in ozone concentrations (µg m^{-3} yr^{-1}) at rural and remote sites.[a]

Site	Annual mean concentration	Annual average of the daily maximum running 8-hour mean	99.9th percentile of the hourly mean concentrations	Annual average of the daily maximum of the running 8-hour mean with a 70 µg m^{-3} cut-off
Aston Hill	0.3	0.1	**-3.0**	0.0
Bottesford	**0.7**	**0.8**	-0.3	0.0
Bush Estate	**0.3**	0.1	**-2.0**	0.0
Eskdalemuir	-0.1	-0.1	**-2.3**	-0.2
Glazebury	0.4	0.4	-1.3	0.0
Great Dun Fell	0.3	0.2	-2.3	-0.1
Harwell	0.3	0.2	-2.2	-0.1
High Muffles	**0.4**	0.2	**-2.5**	0.0
Ladybower	0.2	-0.1	**-3.6**	-0.1
Lough Navar	-0.2	-0.2	**-2.6**	-0.1
Lullington Heath	0.1	0.1	**-2.6**	-0.2
Rochester Stoke	0.3	0.1	-2.9	-0.2
Sibton	0.2	0.1	-3.1	-0.1
Somerton	0.0	-0.1	-2.9	**-0.4**
Strath Vaich	0.3	**0.3**	-0.7	**0.1**
Wharleycroft	**2.0**	**2.1**	2.9	**0.8**
Wicken Fen	**1.5**	**2.0**	2.9	0.9
Yarner Wood	0.0	-0.1	**-4.5**	-0.2

[a] The statistical significance of trends has been calculated using the Mann-Kendall approach, with the slopes estimated using the method of Sen (Gilbert, 1987). Numbers in bold show sites where the trend is statistically significant at the 95% confidence level.

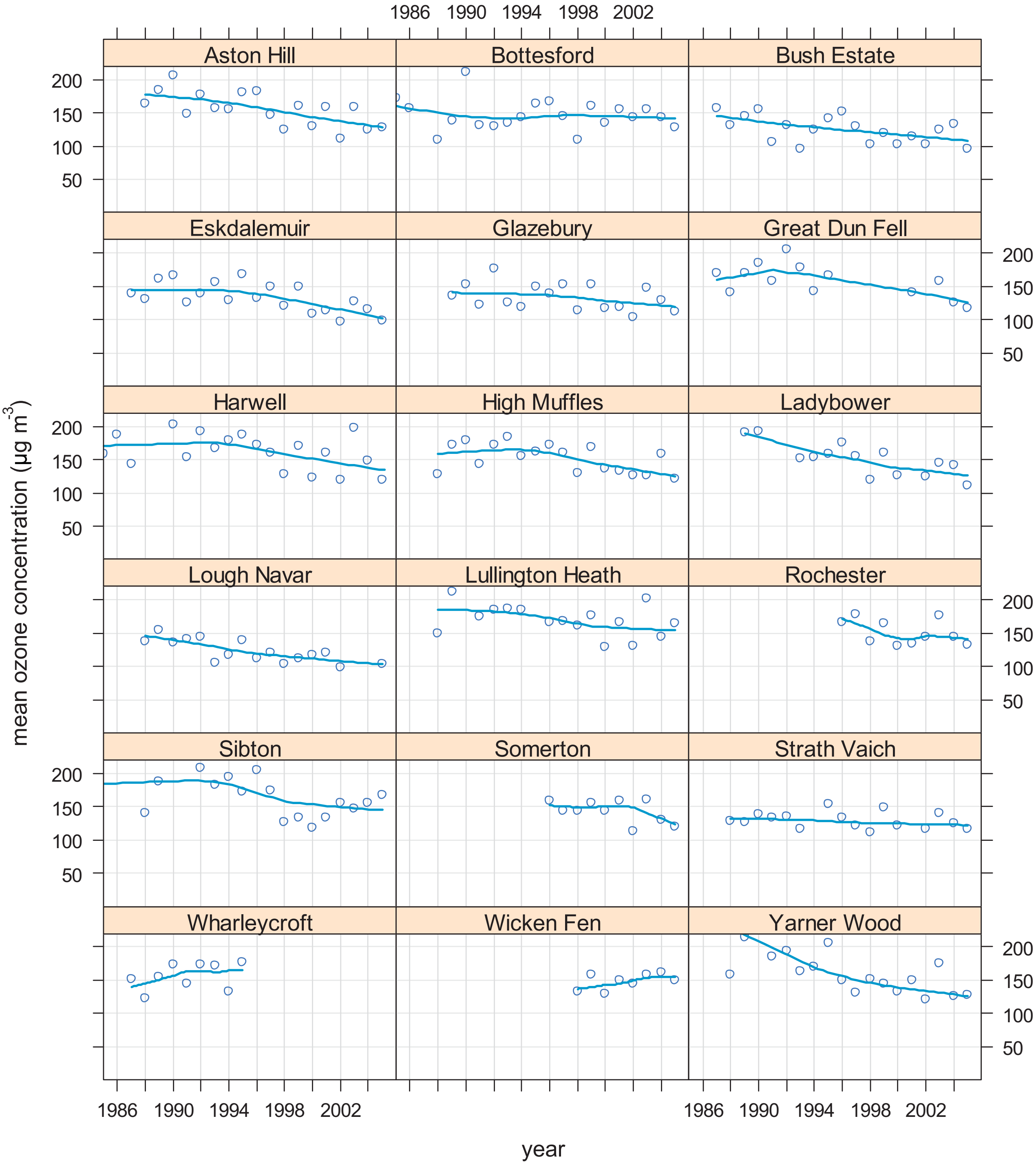


Figure A2.1 Trends in the 99.9th percentile of the hourly mean ozone concentration at rural/remote UK locations, based on data up to 2005.

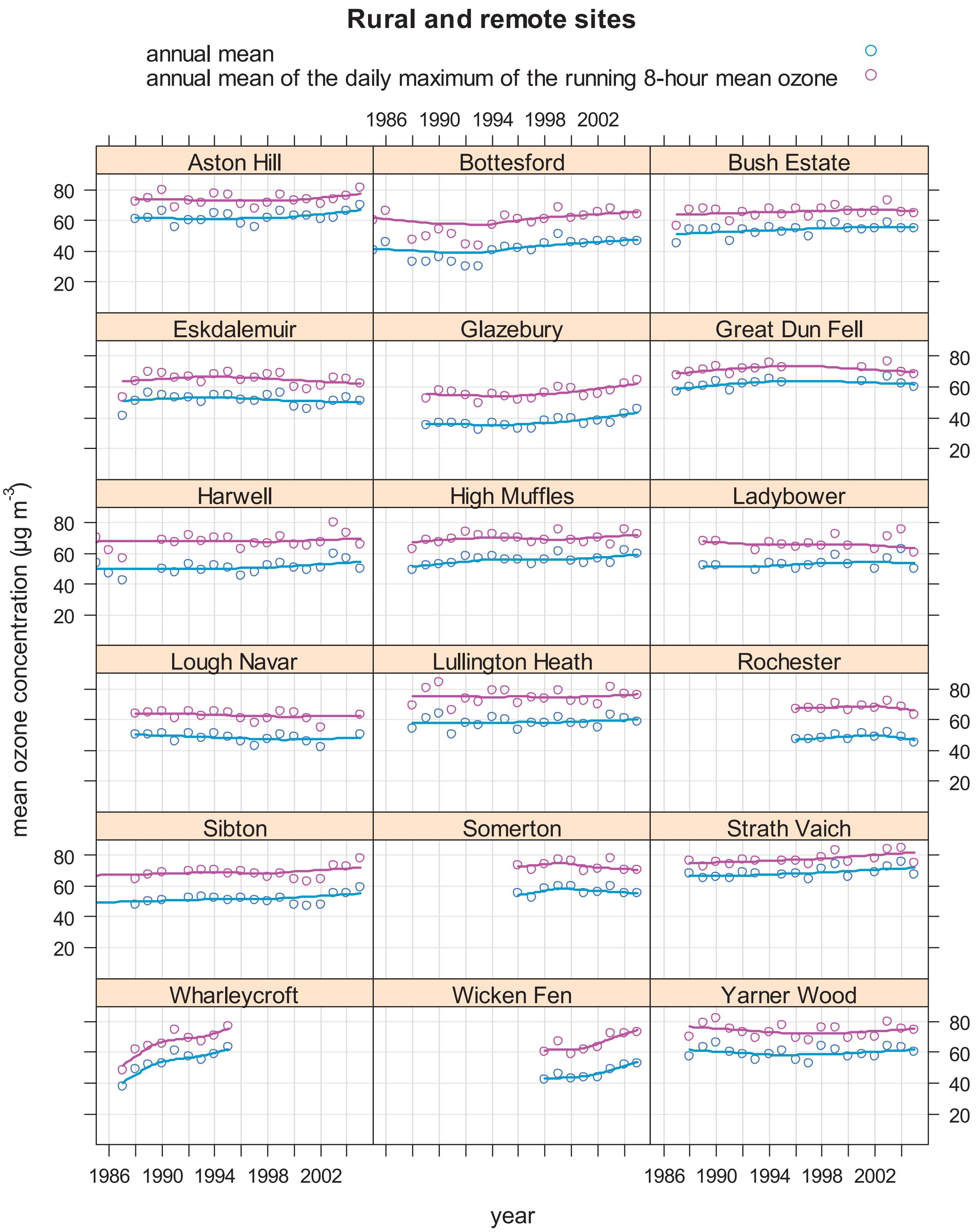


Figure A2.2 Trends in the annual mean and annual mean of the daily maximum running 8-hour mean ozone concentration at rural/remote UK locations, based on data up to 2005.

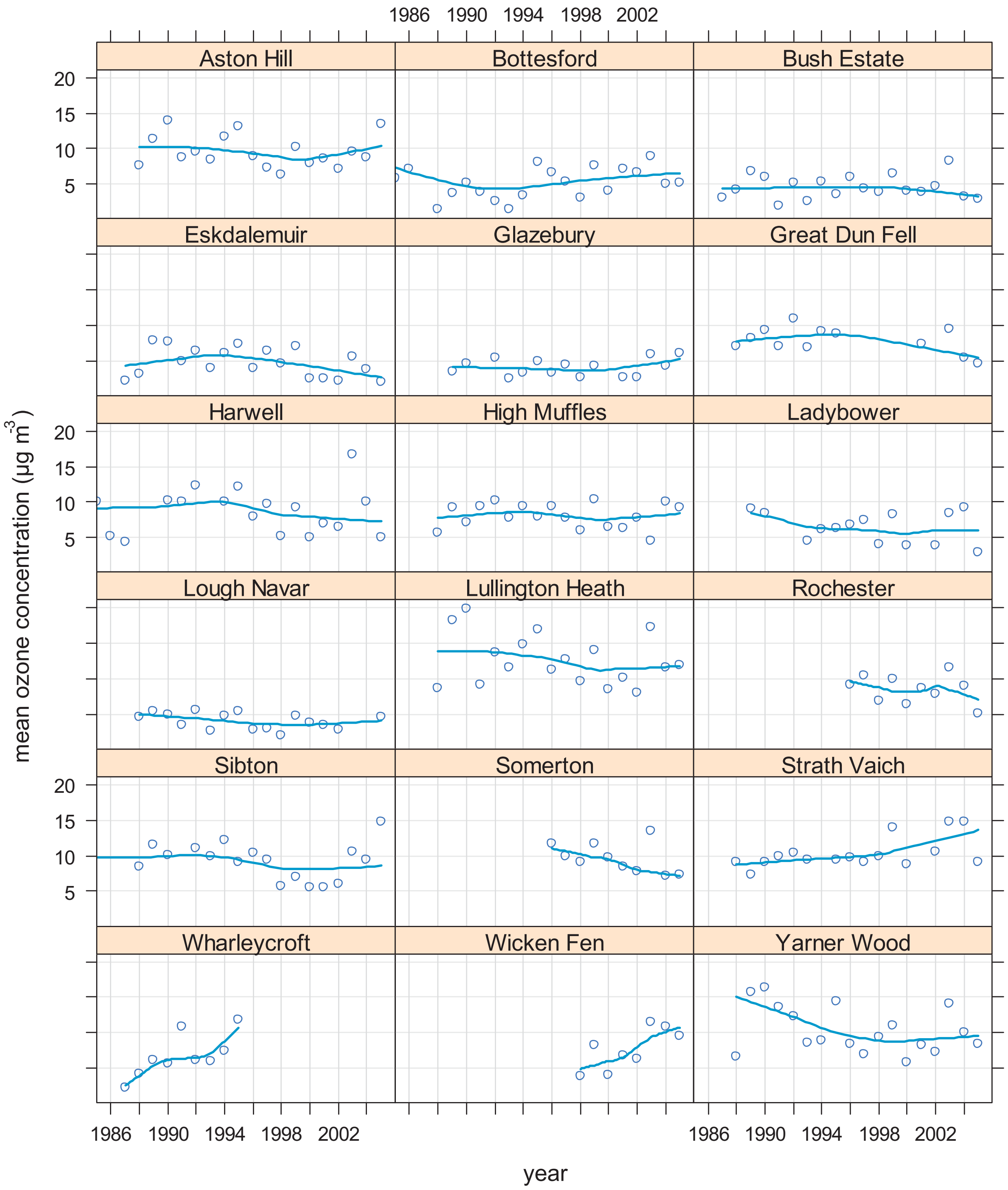


Figure A2.3 Trends in the annual average of the daily maximum of the running 8-hour mean concentration with a 70 µg m^{-3} cut-off at rural/remote UK locations, based on data up to 2005.

Table A2-2 Trends in ozone concentrations (μg m^{-3} yr^{-1}) at urban sites.[a]

Site	Annual mean concentration	Annual average of the daily maximum running 8-hour mean	99.9th percentile of the hourly mean concentrations	Annual average of the daily maximum of the running 8-hour mean with a 70 μg m^{-3} cut-off
Barnsley Gawber	-0.5	-0.5	-1.5	-0.1
Belfast Centre	**0.5**	**0.6**	-0.3	0.1
Birmingham Centre	**0.9**	**0.9**	0.0	0.1
Birmingham East	0.5	0.4	-1.8	-0.1
Bolton	**1.0**	1.2	-1.2	0.1
Bradford Centre	1.0	1.0	1.1	0.1
Bristol Centre	0.5	0.5	-2.0	0.0
Cardiff Centre	0.9	**1.0**	-2.1	0.1
Derry	0.0	0.4	-0.3	0.4
Edinburgh Centre	0.6	0.7	-1.0	0.0
Glasgow Centre	**0.6**	**1.0**	0.6	0.1
Hull Centre	0.8	0.7	-1.1	0.1
Leamington Spa	0.5	0.6	-1.4	0.0
Leeds Centre	**0.9**	**1.0**	-0.3	0.1
Leicester Centre	**0.4**	**0.6**	-1.3	0.0
Liverpool Centre	0.3	0.4	-2.6	0.0
London Bexley	**0.5**	**0.7**	-1.5	0.0
London Bloomsbury	**0.5**	**0.5**	-1.1	0.0
London Brent	0.7	0.8	0.1	0.2
London Bridge Place	**1.5**	**1.9**	3.7	0.1
London Eltham	**0.5**	0.6	0.9	0.2
London Hackney	1.0	1.2	2.8	0.2
London Haringey	**1.1**	**1.3**	2.5	0.4
London Hillingdon	**0.7**	1.0	0.9	0.1
London Lewisham	**1.7**	**2.6**	6.7	0.4
London N. Kensington	**0.8**	1.1	-0.3	0.2
London Southwark	0.3	0.3	-1.0	0.0
London Teddington	**1.0**	**1.0**	-1.1	0.4
London Wandsworth	0.6	0.7	0.8	0.0
Manchester Piccadilly	0.5	0.5	-2.0	-0.1
Manchester South	0.4	0.5	-1.4	0.0
Newcastle Centre	**1.0**	**1.1**	0.3	0.1
Norwich Centre	-0.6	-0.5	0.9	-0.1
Nottingham Centre	**0.8**	**0.9**	0.3	0.1
Plymouth Centre	-0.7	-0.6	-2.2	-0.1
Redcar	0.1	0.6	1.6	0.4
Rotherham Centre	0.0	-0.3	-2.0	-0.1
Sandwell West Bromwich	-0.3	0.2	-2.5	0.1
Sheffield Centre	0.5	0.5	-2.6	0.0
Southampton Centre	0.4	0.2	-2.2	-0.1
Stevenage	1.0	1.1	5.9	0.1
Stoke-on-Trent Centre	0.5	0.3	-0.6	-0.1
Swansea	0.4	0.5	-2.2	0.0
Thurrock	0.4	0.6	0.9	0.1
Wolverhampton Centre	0.6	**1.0**	-3.4	0.1

[a] The statistical significance of trends has been calculated using the Mann-Kendall approach, with the slopes estimated using the method of Sen (Gilbert, 1987). Numbers in bold show sites where the trend is statistically significant at the 95% confidence level.

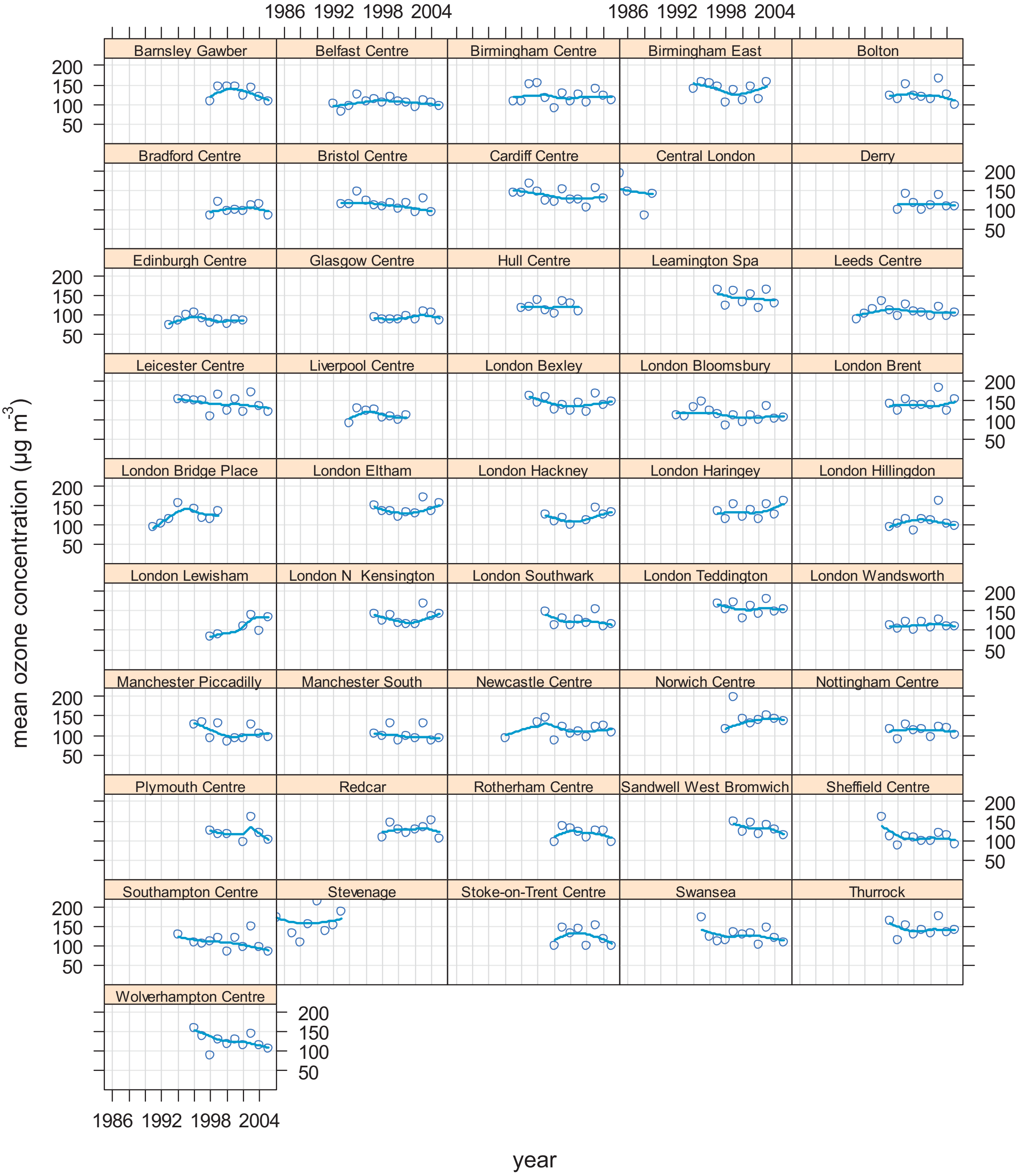


Figure A2.4 Trends in the 99.9th percentile of the hourly mean ozone concentration at urban UK locations, based on data up to 2005.

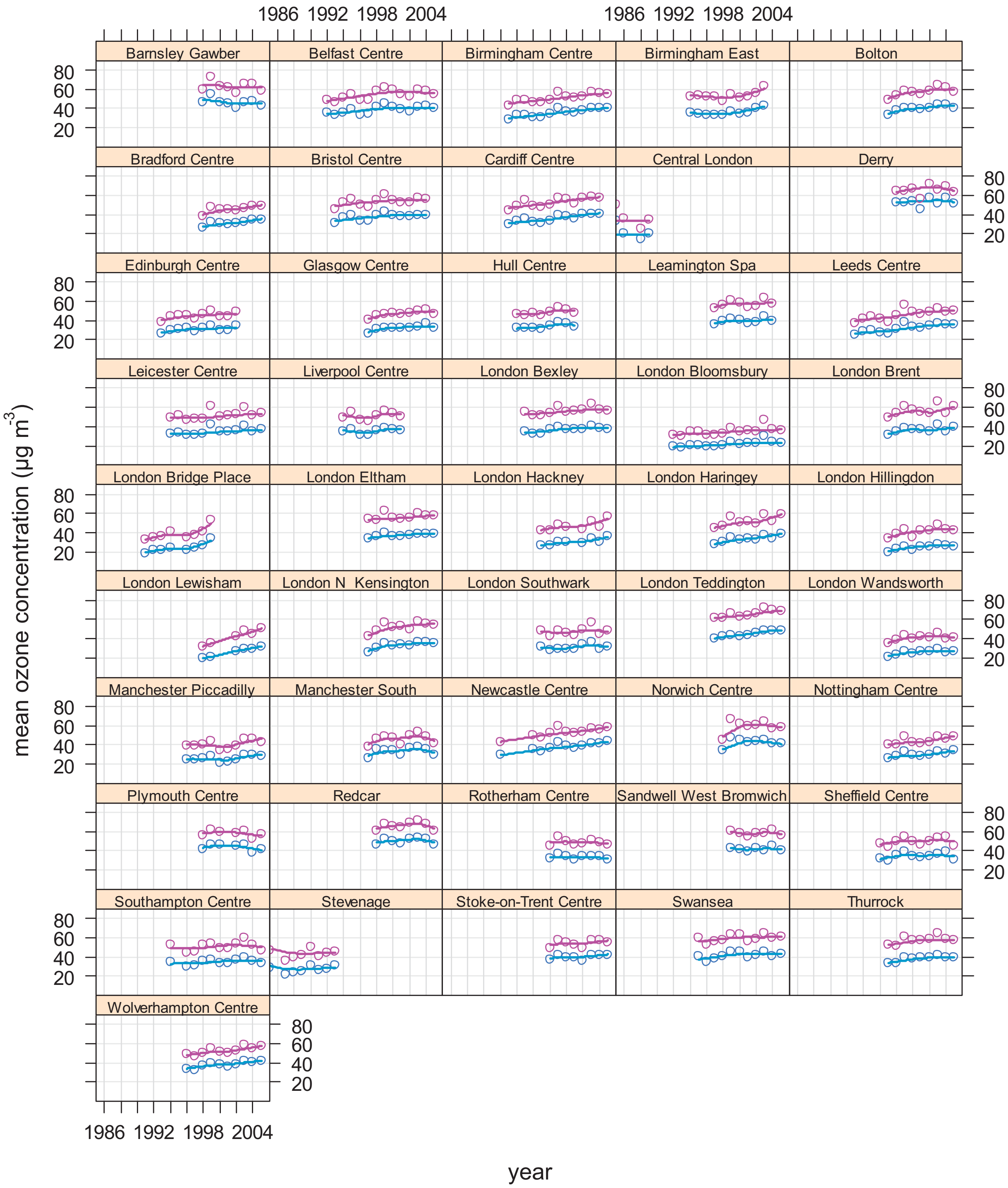


Figure A2.5 Trends in the annual mean of the daily maximum running 8-hour mean ozone concentration and the annual mean ozone concentration at urban UK locations, based on data up to 2005.

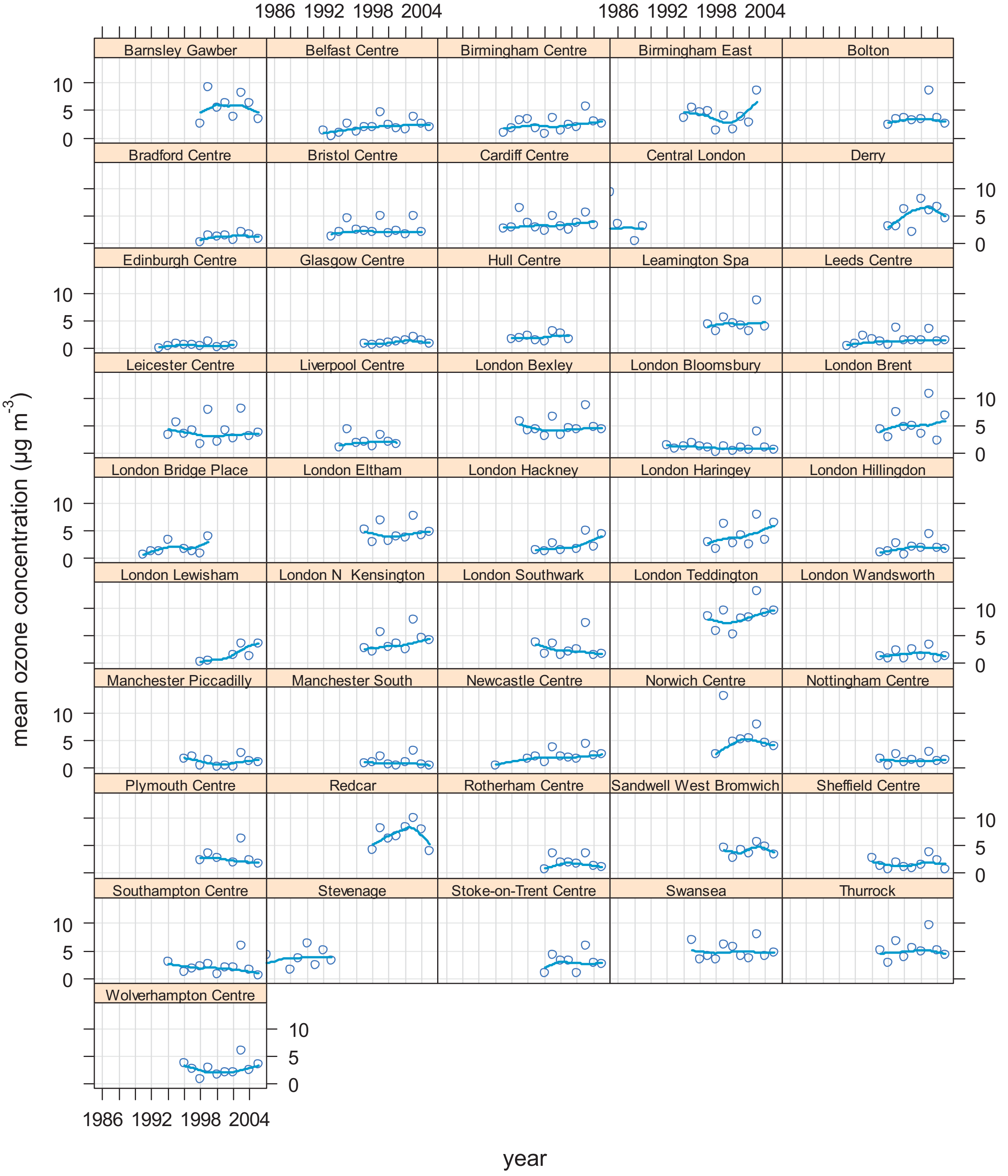


Figure A2.6 Trends in the annual average of the daily maximum of the running 8-hour mean concentration with a 70 μg m^{-3} cut-off at urban UK locations, based on data up to 2005.

Table A2-3 Trends and their statistical significance, in the annual average daily maximum 8-hour mean ozone concentrations for 46 EMEP rural sites with elevations below 500 m over the period from 1990-2002, together with the 1990 values of the ozone metric. Derived from data compiled by Solberg and Hjellebrekke (2004).

Site	Initial value $\mu g\ m^{-3}$	Slope $\mu g\ m^{-3}\ yr^{-1}$	Significance	Site	Initial value $\mu g\ m^{-3}$	Slope $\mu g\ m^{-3}\ yr^{-1}$	Significance
AT02	83.6	0.10		GB15	74.7	0.17	
AT30	87.7	0.12		GB31	78.0	-0.13	
AT45	75.4	-0.16		GB32	54.2	1.63	**
AT46	75.6	0.84	+	GB33	67.7	0.21	
AT47	77.8	-0.03		GB34	56.5	0.09	
BE32	60.9	1.74	***	GB36	68.5	-0.24	
BE35	74.4	0.83		GB38	81.2	-0.40	
DE01	66.1	1.47	**	GB39	68.2	-0.59	***
DE02	67.0	0.32		IE31	76.5	0.39	
DE07	46.1	0.97	+	IT04	78.3	-0.10	
DE09	60.4	0.93	*	LT15	59.6	1.23	
DE12	54.3	0.24		NL09	76.2	-0.36	
DE26	78.3	0.13		NL10	72.6	-0.61	+
DK31	61.9	0.20		NO01	67.1	0.00	
DK32	60.6	-0.05		NO39	56.4	0.96	**
DK41	61.3	-0.10		NO43	62.4	1.04	**
FI09	73.9	0.23		NO45	71.5	-0.33	
FI17	67.1	0.48		PT04	65.6	1.81	
FI22	71.9	-0.18		SE02	67.8	0.27	
GB02	68.2	-0.42		SE11	68.6	0.44	*
GB06	64.9	-0.33		SE12	69.4	0.82	+
GB13	80.8	-0.49		SE32	78.7	0.09	
GB14	66.5	-0.07		SE35	62.8	0.53	+

Notes: *** implies statistical significance at the 99.9%, ** at the 99%, * at the 95% and + at the 90% levels.

AT02: Illmitz, Austria
AT30: Pillersdorf, Austria
AT45: Dunkelsteinerwald, Austria
AT46: Ganserndorf, Austria
AT47: Stixneusiedl, Austria
BE32: Eupen, Belgium
BE35: Vezin, Belgium
DE01: Westerland, Germany
DE02: Langenbrugge, Germany
DE07: Neuglobosow, Germany
DE09: Zingst, Germany
DE12: Bassum, Germany
DE26: Ueckermunde, Germany
DK31: Ulborg, Denmark
DK32: Frederiksborg, Denmark
DK41: Lille Valby, Denmark
FI09: Uto, Finland
FI17: Virolahti, Finland
FI22: Oulanka, Finland
GB02: Eskdalemuir, United Kingdom
GB06: Lough Navar, United Kingdom
GB13: Yarner Wood, United Kingdom
GB14: High Muffles, United Kingdom
GB15: Strath Vaich, United Kingdom
GB31: Aston Hill, United Kingdom
GB32: Bottesford, United Kingdom
GB33: Bush, United Kingdom
GB34: Glazebury, United Kingdom
GB36: Harwell, United Kingdom
GB38: Lullington Heath, United Kingdom
GB39: Sibton, United Kingdom
IE31: Mace Head, Ireland
IT04: Isra, Italy
LT15: Preila, Lithuania
NL09: Kollumerwaard, Netherlands
NL10: Vreedepeel, Netherlands
NO01: Birkenes, Norway
NO39: Karvatn, Norway
NO43: Prestebakke, Norway
NO45: Jelly, Norway
PT04: Monte Vehlo, Portugal
SE02: Rorvik, Sweden
SE11: Vavihill, Sweden
SE12: Aspreveten, Sweden
SE32: Knorra-Kvill, Sweden
SE35: Vindeln, Sweden

Annex 3

Technical Annex to Chapter 5

459. This is a technical annex to the report of the Air Quality Expert Group (AQEG) *Ozone in the United Kingdom*. It provides more detailed information on the modelling of urban ozone decrements in the UK to support the response to question D, *What are the likely future trends in urban ozone concentrations over the next two decades and what is driving them?*

460. Various analyses presented in this report confirm the view that ozone can be represented by a regional component and an urban ozone decrement (see also Clapp and Jenkin, 2001, and Jenkin, 2004). In this report, the urban ozone decrement is taken to be the difference between the values of the ozone concentration or metric at an urban location and the corresponding quantity at a surrounding rural site (taken to be representative of the regional component).

461. Urban decrements in ozone concentrations are explored for a range of metrics using two UK process-based models (the Ozone Source-Receptor Model (OSRM) (Hayman *et al.,* 2008a, 2008b) and ADMS-Urban model (Williams *et al.,* 2006)) and an empirical approach based on monitoring data (Pollution Climate Model, PCM (Kent *et al.,* 2006)). The relationships between the urban decrements and local nitrogen oxides (NO_X) concentrations have been examined and the results of the analysis indicate that the empirical approach to estimating urban ozone decrements currently used in mapping studies (as presented in the supporting evidence to Chapter 2) is in reasonably good agreement with process based models.

A3.1 Modelling of urban ozone decrements

A3.1.1 A comparison of estimated urban decrements for a range of metrics at sites in the national monitoring network

462. OSRM runs were undertaken for 41 receptor sites – representing the locations of 20 rural, 10 London and 11 other urban background ozone monitoring sites – for each hour of 2003 using year-specific emission inventories and meteorology. The model runs were initialised using daily concentration fields derived from the STOCHEM model and modified for ozone to take account of the trend in background concentrations (see Section 4.4 of Hayman *et al.,* 2006a). All other model parameters were set to those used in the ozone modelling runs undertaken for the Review of the Air Quality Strategy (Hayman *et al.,* 2006a).

463. The hourly ozone concentrations were processed to derive the ozone metrics of interest. The output OSRM hourly concentrations are taken to represent mid-boundary layer concentrations and an algorithm can be used during the post-processing phase to take account of the vertical gradient in ozone concentrations arising from losses (i) to the surface by deposition and (ii) by scavenging in areas of high NO_X emissions. In this analysis, the urban ozone decrement is taken to be the difference between the 'unconverted' and 'converted' outputs. As there are still local NO_X emissions, albeit small, at the rural sites, this, together with any deposition, explains why there is a decrement at the rural sites.

464. Panel (a) of Figure A3.1 shows the dependence of the modelled percentage decrement in annual mean ozone in 2003 on the modelled local NO_X annual mean concentration within the surface conversion algorithm of the OSRM model. The decrement in annual mean ozone due to the local scavenging effects is almost linearly related to the modelled annual mean NO_X concentration, as would be expected. The annual mean NO_X concentration has been calculated by applying a dispersion kernel to the 1 km NO_X inventory. Of more interest, the model also predicts that the relationship between the annual ozone decrement and annual mean NO_X is approximately linear for all of the health-based metrics and the number of days with 8-hour mean ozone concentrations greater than 100 µg m^{-3} and 120 µg m^{-3} (as shown in panels (b)-(f) of Figure A3.1)

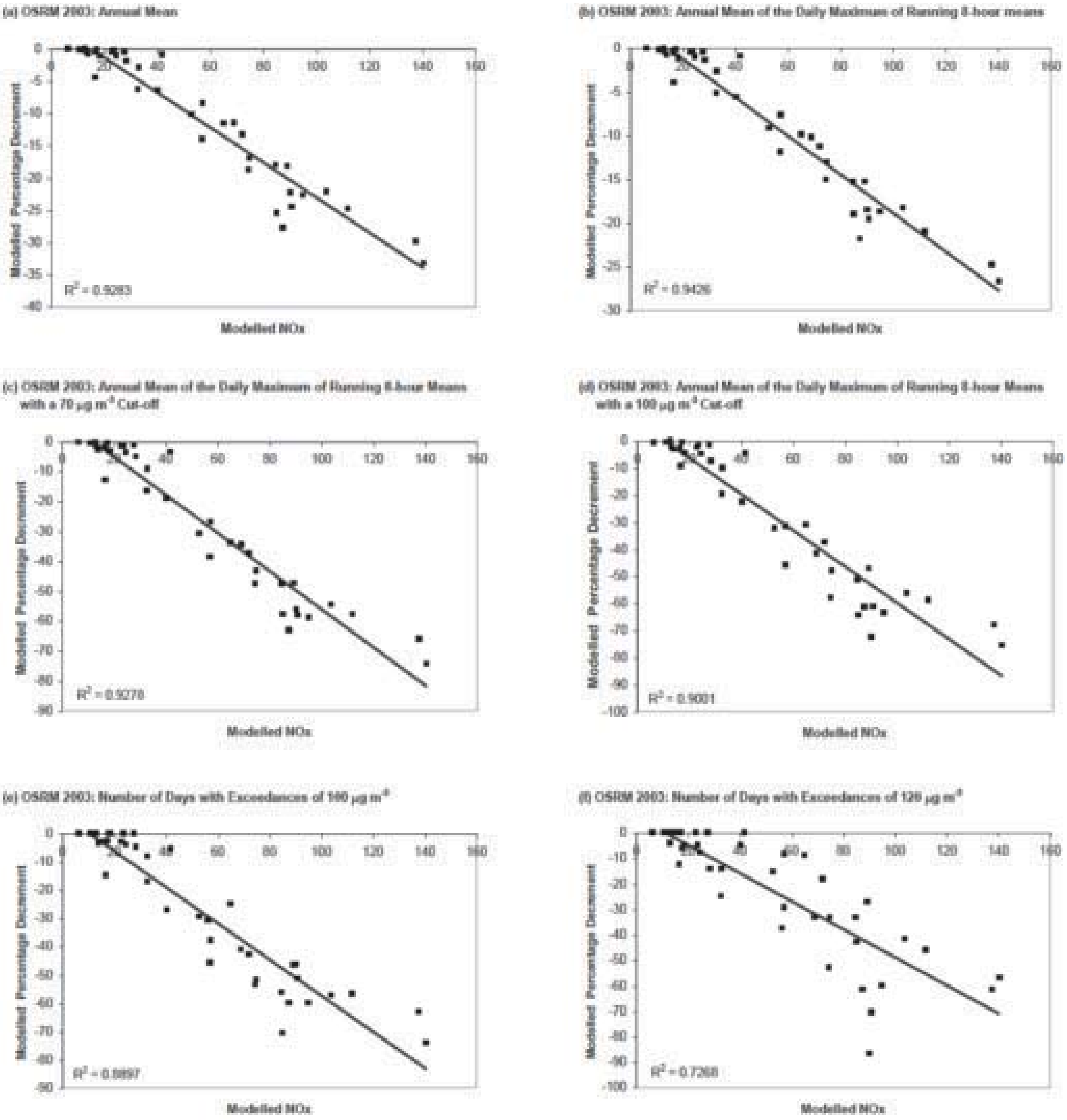
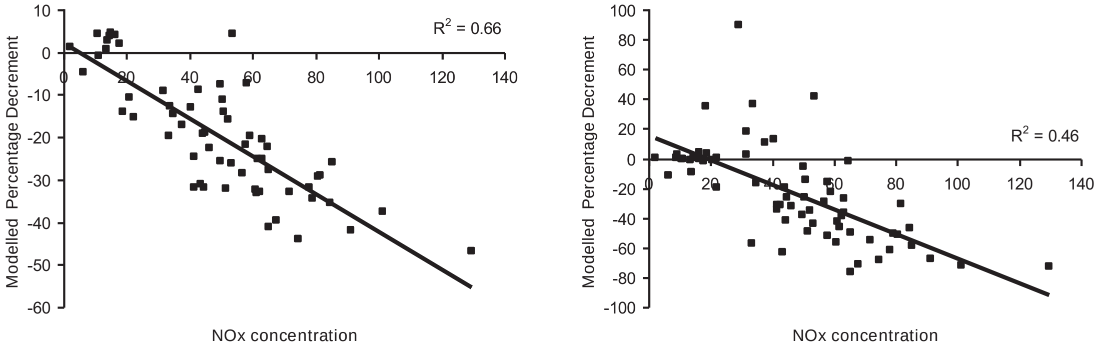


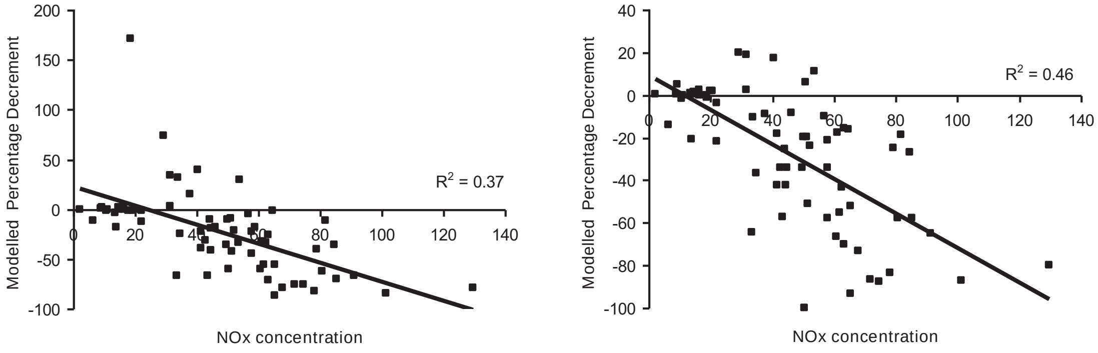
Figure A3.1 Ozone decrements calculated for 41 UK ozone monitoring sites in 2003 using the Ozone Source-Receptor Model as a function of the modelled NO_X concentration (µg m^{-3}, as NO_2).

465. Figure A3.2 shows the corresponding dependences of the urban ozone decrements derived using the empirical pollution climate modelling approach on the modelled annual mean NO_X concentrations. In this instance, the PCM decrement has been calculated by subtracting a regional value for the ozone metric estimated by interpolating rural monitoring sites from the measured value of the ozone metric at the monitoring site location. An altitude correction has been applied for the regional annual means, as described by Coyle *et al*. (2002) and Kent *et al*. (2006). The modelled annual mean NO_X concentrations have been derived using the PCM model (Stedman *et al.,* 2005). The contribution of point sources is modelled explicitly while area sources are modelled using a dispersion kernel approach (similar to that used in the OSRM). These are then combined with a regional rural NO_X background concentration.



(a) 2003: Annual mean concentration

(b) 2003: Annual average of the daily maximum of running 8-hour mean with a 70 µg m^{-3} cut-off



(c) 2003: Annual average of the daily maximum of running 8-hour mean with a 100 µg m^{-3} cut-off

(d) PCM 2003: Number of days with exceedences of 120 µg m^{-3}

Figure A3.2 Ozone decrements calculated for UK ozone monitoring sites in 2003 using the Pollution Climate Model, as a function of the modelled NO_X concentration (µg m^{-3}, as NO_2).

466. The results presented in Figure A3.1 and Figure A3.2 support the assumption in the empirical modelling approach that the urban decrement in ozone concentration varies approximately linearly with annual mean NO_X concentration (the regression lines are shown here for illustrative purposes only). There is however considerable scatter in these plots. This is to be expected since the regional ozone field will incorporate uncertainties associated with the interpolation procedure and very local effects may affect the measured ozone concentrations. The positive outliers are due to measured urban ozone metrics exceeding the values measured at rural sites in the same region. Extreme positive outliers are generally caused by the percentage difference between two small numbers.

467. Figure A3.3 shows a comparison of the modelled NO_X concentrations from the OSRM and PCM models and Figure A3.4 shows a comparison of the decrements for the ozone metrics derived by both the PCM and OSRM, with the 1:1 lines shown in red. To recap, the OSRM decrement has been calculated by estimating the impact of local NO_X emissions on an hourly basis on the modelled regional ozone concentration and the PCM decrement has been calculated by subtracting an interpolated rural field from the measured value for the metric.

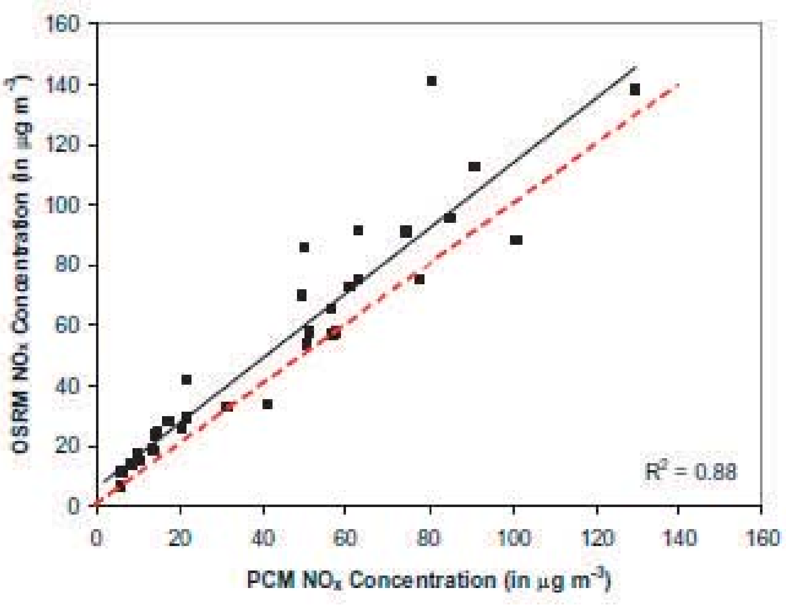


Figure A3.3 Comparison of the 2003 annual mean NO_X concentrations (µg m^{-3}, as NO_2) calculated using the PCM and OSRM. The dotted red line is the 1:1 line.

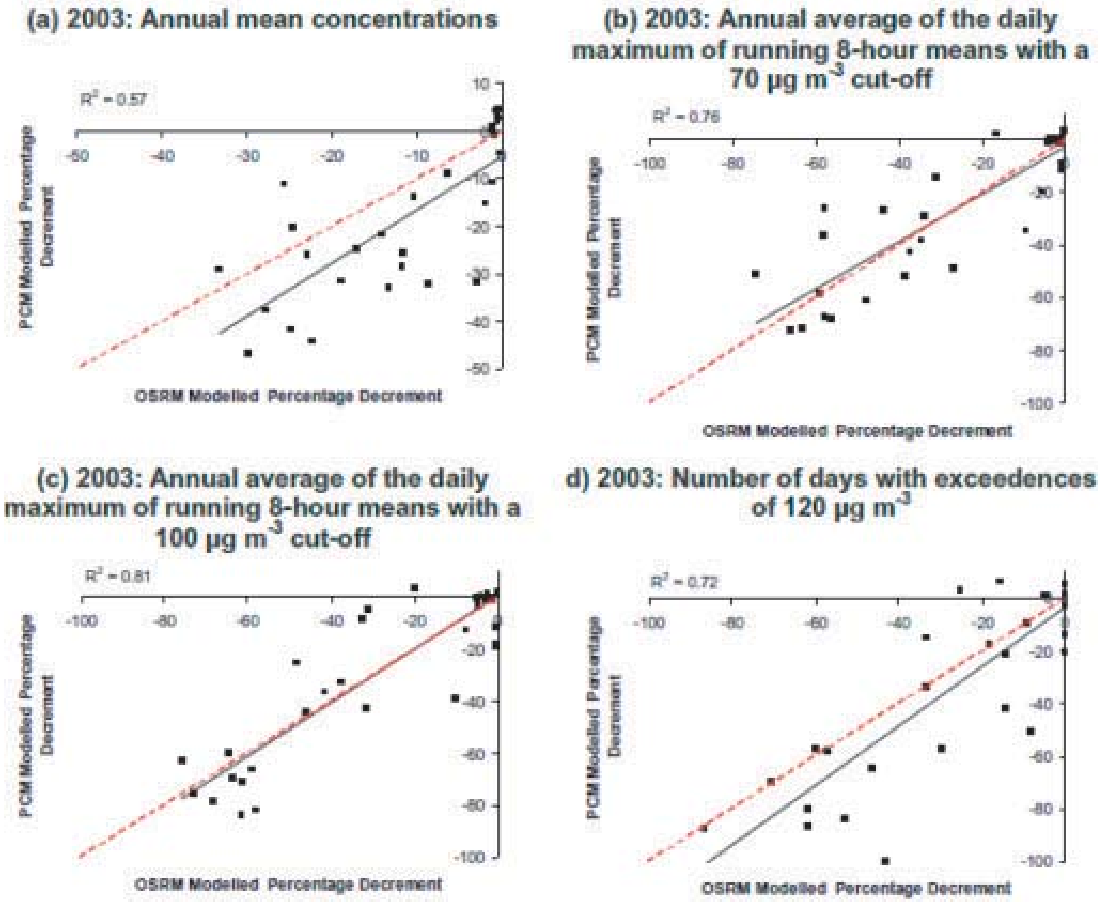


Figure A3.4 Comparison of the ozone decrements in 2003 calculated using the PCM and OSRM for four ozone metrics used to assess impacts on human health. The dotted red lines are 1:1 lines.

468. As expected, the graphs show considerable scatter but the gradients of all of the graphs are close to unity. This comparison of the two approaches suggests that the urban decrement in these ozone metrics can be assumed to be approximately linearly related to local annual mean NO_X concentrations, and hence NO_X emissions. Thus, the OSRM modelling supports the use of linear relationships with NO_X for various ozone metrics applied within the PCM models (Coyle *et al.,* 2002; Bush *et al.,* 2005; Kent *et al.,* 2006).

A3.1.2 A comparison of estimated urban decrements for a range of metrics at sites in London

469. Urban ozone decrements have also been calculated for monitoring site locations in London for various metrics using ADMS-Urban. Figure A3.5 shows a comparison of the percentage decrements plotted against the modelled NO_X concentration, calculated using the OSRM, ADMS-Urban and from ambient monitoring data using the empirical PCM approach. This analysis suggests that all three approaches estimate a similar magnitude of annual mean ozone decrement at similar NO_X concentrations at urban background locations. The decrements calculated for other metrics are also quite similar in magnitude for the different approaches. This provides additional evidence that process-based models are now available which can provide a good description of how the decrement in ozone varies spatially across urban areas. The OSRM model seems to generally predict a lesser decrement for the ozone metrics than ADMS-Urban. The PCM-derived values are based on ambient monitoring data and also incorporate additional uncertainties associated with spatial interpolation of data from rural monitoring sites and therefore show greater scatter than the process-based model estimates.

470. Table A3-1 shows a comparison of the absolute decrements (in µg m^{-3} or days) for metrics and sites for which estimates are available from all three methods. Once again there is reasonably good agreement between the different methods, with the OSRM model tending to predict smaller absolute decrements. The lower absolute decrements derived by the OSRM appear to arise from the lower rural values used to derive the decrements. While the rural values given for the ADMS and PCM models are based on measured values of the metrics (actual or interpolated), the values for the OSRM are those calculated by the model and these were generally lower than the observed values particularly for sites in the southern part of the UK.

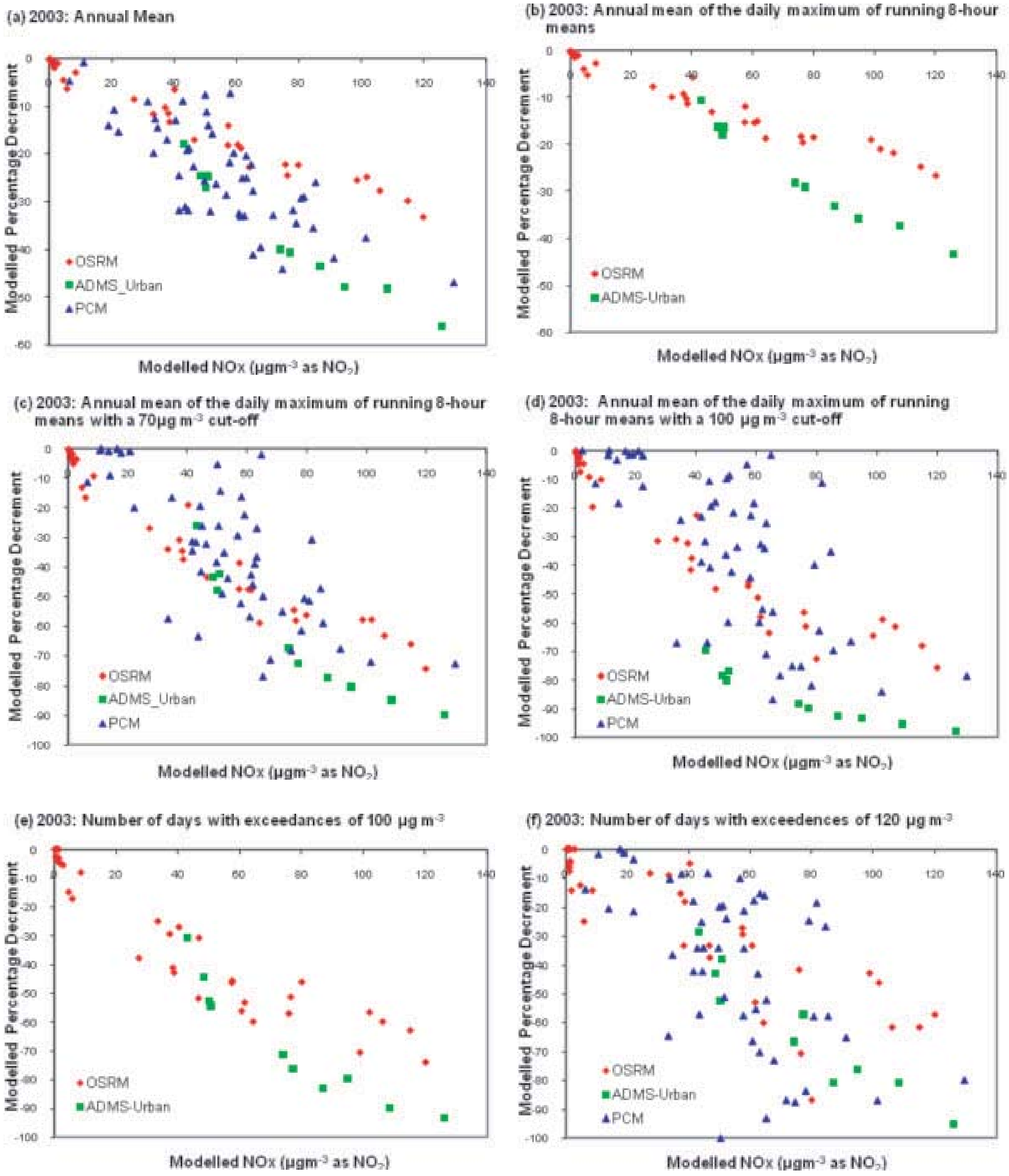


Figure A3.5 Comparison of the percentage decrements plotted against the modelled NO_X concentration, calculated using the OSRM and ADMS-Urban, and from ambient monitoring data using the empirical PCM approach for the following ozone metrics: (a) Annual mean concentration; (b) Annual average of the daily maximum running 8-hour concentration; (c) Annual average of the daily maximum running 8-hour concentration with a cut-off of 70 µg m^{-3}; (d) Annual average of the daily maximum running 8-hour concentration with a cut-off of 100 µg m^{-3}; (e) Days above 100 µg m^{-3}; and (f) Days above 120 µg m^{-3}.

Table A3-1 A comparison of the absolute decrements for metrics and sites for which values have been estimated using the OSRM and ADMS-Urban models, and from ambient monitoring data using the empirical PCM approach.

Absolute ozone decrements: ADMS				
	annual mean (μg m^{-3})	**mean >70 μg m^{-3} (μg m^{-3})**	**mean >100 μg m^{-3} (μg m^{-3})**	**days >120 μg m^{-3} (days)**
Rural value of metric	59.2	14.3	3.5	21.0
London Bloomsbury	-33.1	-12.9	-3.3	-20.0
London Teddington	-10.7	-3.7	-1.0	-6.0
London Brent	-14.6	-6.1	-1.6	-8.0
London Eltham	-16.0	-6.9	-1.9	-11.0
London Bexley	-14.6	-6.2	-1.7	-9.0
Absolute ozone decrements: PCM				
Rural value of metric	56.5	14.7	4.1	24.8
London Bloomsbury	-26.5	-10.6	-3.2	-19.8
London Teddington	-8.2	-2.2	-0.4	1.7
London Brent	-16.8	-4.3	-0.2	-2.6
London Eltham	-18.4	-7.2	-1.8	-12.6
London Bexley	-14.1	-5.5	-1.4	-7.8
Absolute ozone decrements: OSRM				
Rural value of metric	50.4	10.5	2.2	13.0
London Bloomsbury	-14.0	-5.6	-1.2	-8.0
London Teddington	-5.2	-3.2	-0.7	-2.0
London Brent	-5.7	-3.2	-0.6	-1.0
London Eltham	-4.1	-2.7	-0.7	-1.0
London Bexley	-5.4	-3.4	-1.0	-5.0

Annex 4

Additional question G supporting evidence

471. This is a technical annex to the report of the Air Quality Expert Group (AQEG) *Ozone in the United Kingdom*. It provides relevant results from the modelling undertaken for the Review of the UK Air Quality Strategy (Defra, 2007; Hayman *et al.,* 2006a), which are described in the response to question G (section 8.2.1.1). The results, presented in tabular format as population-weighted means for four ozone metrics (see below), are quoted to 2 decimal places. This is purely to illustrate the response of the different measures and is not meant to imply the calculations are accurate to this level of precision.

Table A4-1	Population-weighted annual means of daily maximum running 8-hourly ozone concentration (in μg m^{-3}) for the OSRM runs undertaken for the Review of the Air Quality Strategy.
Table A4-2	Population-weighted annual mean of the difference (in μg m^{-3}) between the daily maximum running 8-hourly ozone concentration and 70 μg m^{-3} for the OSRM runs undertaken for the Review of the Air Quality Strategy.
Table A4-3	Population-weighted annual mean of the difference (in μg m^{-3}) between the daily maximum running 8-hourly ozone concentration and 100 μg m^{-3} for the OSRM runs undertaken for the Review of the Air Quality Strategy.
Table A4-4	Population-weighted number of days when the daily maximum running 8-hourly ozone concentration exceeds 100 μg m^{-3} for the OSRM runs undertaken for the Review of the Air Quality Strategy.

Table A4-1 Summary of the population-weighted annual means of daily maximum running 8-hourly ozone concentration (in µg m^{-3}) for the OSRM runs undertaken for the Review of the Air Quality Strategy.

Run description	**Population-weighted annual means of daily maximum running 8-hourly ozone concentration (in µg m^{-3})**						
	All UK	**Scotland**	**Wales**	**Northern Ireland**	**Inner London**	**Outer London**	**Rest of England**
2003 – Current year	65.14	69.79	73.21	73.96	58.52	59.13	64.75
2010 – Base case	**69.96**	**73.03**	**76.73**	**76.42**	**65.30**	**65.51**	**69.65**
2010 – Measure A	70.00	73.04	76.76	76.43	65.35	65.56	69.69
2010 – Measure B	70.07	73.08	76.81	76.46	65.46	65.67	69.77
2010 – Measure B*	70.07	73.08	76.81	76.46	65.46	65.67	69.77
2010 – Measure C	70.08	73.09	76.81	76.46	65.48	65.68	69.77
2010 – Measure E	69.98	73.04	76.74	76.43	65.32	65.53	69.66
2010 – Measure J	70.00	73.05	76.75	76.44	65.39	65.58	69.68
2010 – Measures K & L	70.95	73.68	77.75	76.87	66.23	66.41	70.71
2010 – Measure L	69.96	73.03	76.73	76.42	65.30	65.51	69.65
2010 – Measure O	70.10	73.10	76.82	76.47	65.50	65.71	69.79
2010 – Measure P	70.08	73.09	76.81	76.46	65.48	65.68	69.77
2010 – Measure Q	70.10	73.10	76.82	76.47	65.50	65.71	69.79
2010 – Measures M & Q	70.07	73.08	76.79	76.45	65.48	65.69	69.77
2010 – Measure M	69.94	73.00	76.71	76.40	65.27	65.49	69.62
2015 – Base case	**72.58**	**74.85**	**79.01**	**77.96**	**68.40**	**68.62**	**72.34**
2015 – Measure A	73.01	75.05	79.29	78.05	69.04	69.26	72.79
2015 – Measure B	73.57	75.29	79.63	78.16	69.88	70.11	73.37
2015 – Measure B*	73.60	75.30	79.64	78.16	69.93	70.16	73.40
2015 – Measure C	73.14	75.12	79.37	78.10	69.23	69.45	72.92
2015 – Measure E	72.63	74.89	79.05	77.98	68.48	68.71	72.40
2015 – Measure J	72.68	74.93	79.06	78.00	68.65	68.81	72.43
2015 – Measures K & L	73.62	75.51	80.07	78.40	69.47	69.64	73.45
2015 – Measure L	72.66	74.90	79.07	77.98	68.54	68.74	72.43
2015 – Measure O	73.19	75.15	79.40	78.11	69.31	69.53	72.97
2015 – Measure P	73.22	75.17	79.43	78.12	69.38	69.57	73.01
2015 – Measure Q	73.28	75.20	79.46	78.13	69.46	69.65	73.06
2015 – Measures M & Q	73.25	75.17	79.43	78.11	69.44	69.62	73.03
2015 – Measure M	72.55	74.82	78.98	77.94	68.37	68.60	72.31
2020 – Base case	**74.72**	**76.46**	**81.16**	**79.39**	**70.62**	**70.88**	**74.57**
2020 – Measure A	75.53	76.76	81.61	79.50	71.87	72.14	75.40
2020 – Measure B	76.40	77.01	82.01	79.52	73.37	73.61	76.31
2020 – Measure B*	76.56	77.05	82.07	79.52	73.66	73.90	76.47
2020 – Measure C	75.57	76.78	81.64	79.51	71.94	72.20	75.45
2020 – Measure E	74.85	76.52	81.23	79.43	70.81	71.06	74.69
2020 – Measure J	74.90	76.58	81.24	79.45	71.06	71.20	74.72
2020 – Measures K & L	75.18	76.73	81.60	79.57	71.16	71.37	75.05
2020 – Measure L	74.82	76.51	81.22	79.41	70.78	71.01	74.66
2020 – Measure O	75.67	76.84	81.70	79.54	72.10	72.36	75.55
2020 – Measure P	75.66	76.83	81.70	79.53	72.11	72.33	75.54
2020 – Measure Q	75.76	76.88	81.76	79.56	72.27	72.49	75.64
2020 – Measures M & Q	75.73	76.85	81.73	79.54	72.24	72.45	75.61
2020 – Measure M	74.69	76.43	81.13	79.37	70.59	70.85	74.54

Table A4-2 Summary of the population-weighted annual mean of the difference (in μg m^{-3}) between the daily maximum running 8-hourly ozone concentration and 70 μg m^{-3} for the OSRM runs undertaken for the Review of the Air Quality Strategy.

Run description	**Population-weighted annual mean of the difference (in μg m^{-3}) between the daily maximum running 8-hourly ozone concentration and 70 μg m^{-3})**						
	All UK	**Scotland**	**Wales**	**Northern Ireland**	**Inner London**	**Outer London**	**Rest of England**
2003 – Current year	9.14	9.63	13.38	11.51	6.77	6.93	9.09
2010 – Base case	**11.10**	**11.10**	**15.07**	**12.73**	**9.26**	**9.31**	**11.08**
2010 – Measure A	11.12	11.10	15.08	12.73	9.28	9.33	11.09
2010 – Measure B	11.15	11.12	15.10	12.74	9.32	9.37	11.12
2010 – Measure B*	11.15	11.12	15.10	12.74	9.32	9.37	11.12
2010 – Measure C	11.16	11.13	15.11	12.75	9.34	9.39	11.13
2010 – Measure E	11.11	11.10	15.07	12.73	9.28	9.32	11.09
2010 – Measure J	11.12	11.11	15.08	12.74	9.30	9.34	11.09
2010 – Measures K & L	11.55	11.43	15.67	12.95	9.57	9.64	11.56
2010 – Measure L	11.10	11.10	15.07	12.73	9.26	9.31	11.08
2010 – Measure O	11.17	11.13	15.11	12.75	9.35	9.40	11.14
2010 – Measure P	11.16	11.13	15.11	12.75	9.34	9.39	11.13
2010 – Measure Q	11.17	11.13	15.11	12.75	9.35	9.40	11.14
2010 – Measures M & Q	11.15	11.12	15.09	12.73	9.34	9.39	11.12
2010 – Measure M	11.09	11.08	15.05	12.71	9.25	9.30	11.06
2015 – Base case	**12.51**	**12.21**	**16.51**	**13.73**	**10.70**	**10.80**	**12.52**
2015 – Measure A	12.67	12.28	16.60	13.72	10.93	11.04	12.69
2015 – Measure B	12.88	12.36	16.71	13.71	11.23	11.38	12.91
2015 – Measure B*	12.89	12.36	16.71	13.71	11.25	11.39	12.92
2015 – Measure C	12.74	12.32	16.65	13.75	11.03	11.14	12.76
2015 – Measure E	12.54	12.23	16.53	13.74	10.75	10.85	12.56
2015 – Measure J	12.56	12.25	16.54	13.75	10.83	10.89	12.57
2015 – Measures K & L	13.01	12.56	17.16	13.93	11.13	11.22	13.05
2015 – Measure L	12.56	12.24	16.55	13.74	10.78	10.86	12.57
2015 – Measure O	12.76	12.34	16.67	13.75	11.07	11.18	12.79
2015 – Measure P	12.78	12.34	16.69	13.75	11.11	11.21	12.80
2015 – Measure Q	12.81	12.36	16.70	13.76	11.15	11.25	12.83
2015 – Measures M & Q	12.79	12.34	16.68	13.75	11.13	11.23	12.81
2015 – Measure M	12.49	12.19	16.49	13.71	10.69	10.79	12.50
2020 – Base case	**13.82**	**13.29**	**18.02**	**14.73**	**11.87**	**12.03**	**13.88**
2020 – Measure A	14.11	13.35	18.13	14.66	12.33	12.54	14.18
2020 – Measure B	14.38	13.33	18.11	14.48	12.95	13.18	14.46
2020 – Measure B*	14.43	13.32	18.10	14.44	13.08	13.31	14.51
2020 – Measure C	14.13	13.37	18.15	14.67	12.37	12.58	14.20
2020 – Measure E	13.89	13.33	18.07	14.75	11.97	12.13	13.94
2020 – Measure J	13.90	13.36	18.07	14.76	12.10	12.20	13.95
2020 – Measures K & L	14.04	13.42	18.30	14.81	12.11	12.26	14.11
2020 – Measure L	13.87	13.32	18.06	14.74	11.95	12.09	13.93
2020 – Measure O	14.18	13.39	18.18	14.68	12.46	12.67	14.26
2020 – Measure P	14.18	13.38	18.18	14.67	12.46	12.65	14.25
2020 – Measure Q	14.23	13.41	18.21	14.69	12.55	12.74	14.30
2020 – Measures M & Q	14.21	13.39	18.18	14.67	12.53	12.72	14.28
2020 – Measure M	13.80	13.27	18.00	14.71	11.85	12.01	13.85

Table A4-3 Summary of the population-weighted annual mean of the difference (in µg m^{-3}) between the daily maximum running 8-hourly ozone concentration and 100 µg m^{-3} for the OSRM runs undertaken for the Review of the Air Quality Strategy.

Run description	Population-weighted annual mean of the difference (in µg m^{-3}) between the daily maximum running 8-hourly ozone concentration and 100 µg m^{-3})						
	All UK	Scotland	Wales	Northern Ireland	Inner London	Outer London	Rest of England
2003 – Current year	1.93	1.69	2.73	1.96	1.52	1.53	1.97
2010 – Base case	**2.14**	**1.80**	**2.89**	**2.01**	**1.92**	**1.86**	**2.18**
2010 – Measure A	2.15	1.80	2.89	2.01	1.92	1.87	2.19
2010 – Measure B	2.16	1.80	2.90	2.01	1.93	1.88	2.20
2010 – Measure B*	2.16	1.80	2.90	2.01	1.93	1.88	2.20
2010 – Measure C	2.16	1.80	2.90	2.01	1.94	1.89	2.20
2010 – Measure E	2.15	1.80	2.89	2.01	1.92	1.87	2.19
2010 – Measure J	2.15	1.80	2.89	2.01	1.93	1.87	2.19
2010 – Measures K & L	2.30	1.90	3.11	2.09	2.02	1.97	2.36
2010 – Measure L	2.14	1.80	2.89	2.01	1.92	1.86	2.18
2010 – Measure O	2.16	1.81	2.90	2.01	1.94	1.89	2.20
2010 – Measure P	2.16	1.80	2.90	2.01	1.94	1.89	2.20
2010 – Measure Q	2.16	1.81	2.90	2.01	1.94	1.89	2.20
2010 – Measures M & Q	2.15	1.80	2.89	2.00	1.94	1.89	2.19
2010 – Measure M	2.14	1.79	2.88	2.00	1.92	1.86	2.18
2015 – Base case	**2.50**	**2.06**	**3.34**	**2.29**	**2.28**	**2.23**	**2.55**
2015 – Measure A	2.55	2.08	3.38	2.30	2.34	2.28	2.60
2015 – Measure B	2.60	2.11	3.42	2.31	2.42	2.36	2.66
2015 – Measure B*	2.61	2.11	3.42	2.31	2.42	2.37	2.66
2015 – Measure C	2.57	2.09	3.39	2.30	2.37	2.31	2.62
2015 – Measure E	2.51	2.06	3.35	2.29	2.30	2.24	2.56
2015 – Measure J	2.52	2.07	3.35	2.29	2.32	2.25	2.56
2015 – Measures K & L	2.68	2.19	3.61	2.36	2.42	2.37	2.74
2015 – Measure L	2.52	2.07	3.35	2.29	2.30	2.25	2.57
2015 – Measure O	2.58	2.10	3.40	2.31	2.39	2.33	2.63
2015 – Measure P	2.58	2.10	3.41	2.31	2.40	2.34	2.63
2015 – Measure Q	2.59	2.10	3.41	2.31	2.41	2.35	2.64
2015 – Measures M & Q	2.58	2.09	3.40	2.30	2.41	2.34	2.63
2015 – Measure M	2.50	2.05	3.33	2.28	2.28	2.22	2.55
2020 – Base case	**2.95**	**2.43**	**3.96**	**2.63**	**2.66**	**2.61**	**3.01**
2020 – Measure A	3.02	2.45	3.99	2.61	2.77	2.73	3.09
2020 – Measure B	3.08	2.43	3.97	2.53	2.92	2.91	3.15
2020 – Measure B*	3.09	2.42	3.95	2.50	2.95	2.95	3.15
2020 – Measure C	3.03	2.46	4.00	2.61	2.79	2.75	3.10
2020 – Measure E	2.97	2.45	3.97	2.64	2.69	2.64	3.03
2020 – Measure J	2.97	2.45	3.97	2.64	2.73	2.65	3.03
2020 – Measures K & L	3.03	2.49	4.07	2.65	2.74	2.68	3.10
2020 – Measure L	2.97	2.44	3.97	2.63	2.69	2.63	3.03
2020 – Measure O	3.05	2.47	4.01	2.62	2.82	2.78	3.11
2020 – Measure P	3.05	2.46	4.01	2.61	2.82	2.77	3.11
2020 – Measure Q	3.06	2.47	4.02	2.62	2.85	2.80	3.13
2020 – Measures M & Q	3.05	2.46	4.01	2.60	2.84	2.79	3.11
2020 – Measure M	2.94	2.42	3.94	2.62	2.65	2.60	3.00

Table A4-4 Summary of the population-weighted number of days when the daily maximum running 8-hourly ozone concentration exceeds 100 µg m^{-3} for the OSRM runs undertaken for the Review of the Air Quality Strategy.

Run description	**Population-weighted annual mean of the difference (in µg m^{-3}) between the daily maximum running 8-hourly ozone concentration and 100 µg m^{-3})**						
	All UK	**Scotland**	**Wales**	**Northern Ireland**	**Inner London**	**Outer London**	**Rest of England**
2003 – Current year	29.73	28.16	48.34	35.88	22.03	21.66	29.77
2010 – Base case	38.99	34.11	57.89	42.98	33.71	34.28	38.96
2010 – Measure A	39.01	34.10	57.87	42.92	33.71	34.32	38.99
2010 – Measure B	39.15	34.24	57.97	42.93	33.71	34.37	39.16
2010 – Measure B*	39.15	34.24	57.97	42.93	33.71	34.37	39.16
2010 – Measure C	39.22	34.24	58.00	42.98	33.88	34.66	39.21
2010 – Measure E	39.01	34.11	57.91	43.03	33.71	34.28	38.98
2010 – Measure J	39.03	34.14	57.94	43.03	33.71	34.35	39.00
2010 – Measures K & L	41.06	35.64	61.50	43.70	35.50	35.80	41.12
2010 – Measure L	38.99	34.11	57.89	42.98	33.71	34.28	38.96
2010 – Measure O	39.25	34.25	58.10	42.89	33.88	34.66	39.24
2010 – Measure P	39.22	34.24	58.00	42.98	33.88	34.66	39.21
2010 – Measure Q	39.25	34.25	58.10	42.89	33.88	34.66	39.24
2010 – Measures M & Q	39.15	34.19	57.96	42.78	33.88	34.64	39.13
2010 – Measure M	38.92	34.03	57.74	42.92	33.71	34.19	38.89
2015 – Base case	**47.23**	**41.27**	**66.84**	**49.00**	**44.09**	**43.20**	**47.17**
2015 – Measure A	47.84	41.66	67.27	47.85	45.39	43.97	47.83
2015 – Measure B	48.35	41.90	67.32	46.72	45.39	44.56	48.49
2015 – Measure B*	48.39	41.92	67.30	46.67	45.39	44.70	48.52
2015 – Measure C	48.11	41.96	67.46	47.79	45.90	44.41	48.08
2015 – Measure E	47.38	41.44	66.97	49.00	44.31	43.40	47.31
2015 – Measure J	47.44	41.55	67.04	49.09	44.75	43.56	47.32
2015 – Measure J	47.44	41.55	67.04	49.09	44.75	43.56	47.32
2015 – Measures K & L	49.25	43.70	69.98	49.55	44.90	44.34	49.29
2015 – Measure L	47.41	41.48	67.11	48.98	44.31	43.37	47.34
2015 – Measure O	48.24	42.09	67.46	47.92	45.90	44.53	48.23
2015 – Measure P	48.33	42.22	67.65	47.77	45.90	44.76	48.30
2015 – Measure Q	48.44	42.32	67.77	47.91	46.18	44.78	48.42
2015 – Measures M & Q	48.33	42.13	67.61	47.79	45.90	44.68	48.32
2015 – Measure M	47.11	41.16	66.69	48.94	44.09	43.07	47.03
2020 – Base case	**55.14**	**49.47**	**76.92**	**55.18**	**51.04**	**49.78**	**55.17**
2020 – Measure A	56.04	49.87	76.11	52.98	51.65	51.57	56.29
2020 – Measure B	56.74	49.35	75.05	49.41	52.25	52.90	57.36
2020 – Measure B*	56.77	49.23	74.63	48.87	52.19	53.01	57.46
2020 – Measure C	56.12	49.96	76.10	52.97	51.65	51.79	56.37
2020 – Measure E	55.43	49.81	77.07	55.16	51.05	50.12	55.49
2020 – Measure J	55.49	49.78	77.06	55.21	51.46	50.48	55.51
2020 – Measures K & L	56.11	50.60	78.07	55.14	51.38	50.39	56.22
2020 – Measure L	55.36	49.56	77.11	55.13	51.05	50.03	55.43
2020 – Measure O	56.36	50.07	76.28	52.96	51.65	52.19	56.63
2020 – Measure P	56.31	50.05	76.12	52.94	51.65	52.08	56.59
2020 – Measure Q	56.58	50.26	76.38	52.93	51.81	52.64	56.85
2020 – Measures M & Q	56.42	50.04	76.22	52.90	51.65	52.51	56.69
2020 – Measure M	55.03	49.34	76.79	55.12	51.04	49.78	55.04

Abbreviations

°C	degrees Celsius
μg m^{-3}	micrograms per cubic metre of air
A1 and A2	different IPCC SRES scenarios
ACCENT	Atmospheric Composition Change: The European Network of Excellence
ADMS-Urban	Atmospheric Dispersion Models for urban areas
AMO	Atlantic (Ocean) Multidecadal Oscillation
AOT40	accumulated dose over a threshold of 40 ppb
AOT60	accumulated dose over a threshold of 60 ppb
AQEG	Air Quality Expert Group
AURN	Automatic Urban and Rural Network
B1 and B2	different IPCC SRES scenarios
BEIS	Biogenic Emissions Inventory System
BERR	Department for Business, Enterprise and Regulatory Reform
BVOC	Biogenic Volatile Organic Compounds
C	carbon
CAFÉ	Clean Air for Europe
CAMx	Comprehensive Air Quality Model with extensions
CBM-IV	Carbon Bond Mechanism version 4
CEN	European Committee for Standardisation (Comité Européen de Normalisation)
CH$_4$	methane
CHIMERE	A chemical transport model
CIAM	Centre for Integrated Assessment Modelling
CLE	Current legislation
CLRTAP	Convention on Long-Range Transboundary Air Pollution
CMAQ	Community Multiscale Air Quality modelling system
CO	carbon monoxide
CO$_2$	carbon dioxide
COMEAP	Committee on the Medical Effects of Air Pollutants
COPERT	Computer Programme to Calculate Emissions from Road Transport
CORINAIR	the air pollutant emissions section of CORINE
CORINE	CoOrdination d'Information Environmentale (environmental information gathering programme)
CRI	Common Representative Intermediates
CTM	chemical transport model

DAPPLE	Dispersion of Air Pollution and its Penetration into the Local Environment (NERC project)
Defra	Department for Environment, Food and Rural Affairs
DfT	Department for Transport
DOAS	Differential Optical Absorption Spectroscopy
DTI	Department of Trade and Industry
EDGAR	Emission Database for Global Atmospheric Research
ELMO	Edinburgh Lancaster Model for Ozone
EMEP	European Monitoring and Evaluation Programme (a Co-operative Programme under CLRTAP)
EU	European Union
EU25	the 25 countries that were members of the European Union before Romania and Bulgaria joined on 1st January 2007
EU27	the 27 member countries of the European Union after 1st January 2007
EU60	Health metric for exposure to zone, value is the number of days in the year on which the maximum running 8-hour average exceeds 60ppb (120µg m^{-2})
EURO V/VI	Vehicle emission standards
GAINS	Greenhouse Gas and Air Pollution Interactions and Synergies
GCMs	General Circulation Models
GEIA	Global Emissions Inventory Activity
GLOBEIS	Global Biosphere Emissions and Interactions System
GRS	Generalised Reaction Set chemical mechanism
HadCM3	Hadley Centre Coupled Ocean-Atmosphere Global Climate Model
HARM	Hull Acid Rain Model
HNO_3	nitric acid
HONO	nitrous acid
HO_2	Hydroperoxyl radical
HO_x	The sum of the concentration of OH and HO_2 radicals
HTAP	Hemispheric Transport of Air Pollution
IDOP	Integrated Downwind Ozone Potential
IIASA	International Institute for Applied Systems Analysis
IIASA CLE	IIASA current legislation scenario
IIASA MFR	IIAS maximum technically feasible reduction scenario
IPCC	Intergovernmental Panel on Climate Change
IS92a	an IPCC emissions scenario
kPa	kiloPascal (unit of pressure)
ktonnes	kilo tonnes
LAQN	London Air Quality Network

MCM	Master Chemical Mechanism
MEGAN	Model of Emissions of Gases and Aerosols from Nature
MFR	Maximum Technically Feasible Reduction
MOPITT	Measurements of Pollution in the Troposphere
MSC-W	Meteorological Synthesizing Centre-West in Norway
Mtonnes	Mega tonnes
NAEI	National Atmospheric Emissions Inventory
NAME	Numerical Atmospheric Dispersion Modelling Environment
NAS	US National Academy of Sciences
NATAIR	Improving and Applying Methods for the Calculation of Natural and Biogenic Emissions and Assessment of Impacts on Air Quality
NEC	National Emissions Ceiling
NECD	National Emissions Ceiling Directive
NEGTAP	National Expert Group on Transboundary Air Pollution
NERC	Natural Environment Research Council
NH_3	ammonia
NO	nitrogen monoxide, also termed nitric oxide
NO_2	nitrogen dioxide
NO_X	nitrogen oxides ($NO + NO_2$)
NO_y	total reactive nitrogen oxides
NOAA	National Oceanic and Atmospheric Administration (US)
NPP	net primary productivity
$O(^1D)$	Electronically excited state of oxygen
O_3	ozone
OH	hydroxyl radical
OSRM	Ozone Source-Receptor Model
OVOC	Other Volatile Organic Compounds, other than isoprene and terpenes
OX	oxidant
PAN	peroxyacetyl nitrate
PAR	Photosynthetically Active Radiation
PCM	Pollution Climate Model
PELCOM	Pan-European Land Use and Land Cover Monitoring
PM	particulate matter
PM_{10}	airborne particulate matter passing a sampling inlet with a 50% efficiency cut-off at 10 μm aerodynamic diameter and which transmits particles of below this size

$PM_{2.5}$	airborne particulate matter passing a sampling inlet with a 50% efficiency cut-off at 2.5 µm aerodynamic diameter and which transmits particles of below this size
POCP	Photochemical Ozone Creation Potential
POET	A global emissions inventory dataset
PORG	Photochemical Oxidants Review Group
ppb	parts per billion (1,000,000,000)
ppm	parts per million (1,000,000)
PTM	Photochemical Trajectory Model
PUMA	Pollution of the Urban Midlands Atmosphere
RAINS	IIASA Regional Air Pollution Information and Simulation model
RETRO	A global emissions inventory dataset
SCR	Selective Catalytic Reduction
SLIMCAT	a three-dimensional off-line chemical transport model
SNAP	Selected Nomenclature for Air Pollution
SO_2	sulphur dioxide
SOMO35	sum of means over 35 ppb
SRES	IPCC Special Report on Emission Scenarios
SSTs	sea surface temperatures
STOCHEM	a three-dimensional Lagrangian chemical transport model of tropospheric chemistry
TFMM	Task Force on Measurement and Modelling
TILDAS	tunable infrared laser differential absorption spectroscopy
TiO_2	titanium dioxide
TM3	an atmospheric chemical transport model
TORCH	Tropospheric ORganic CHemistry experiment (NERC campaign)
TROTREP	Tropospheric Ozone and Precursors – Trends, Budgets and Policy (EU project)
UEP21	BERR energy projection scenarios
UEP26	BERR energy projection scenarios
UNCLOS	UN Convention on the Law of the Sea
UNECE	United Nations Economic Commission for Europe
UNFCCC	United Nations Framework Convention on Climate Change
USEPA	United States Environmental Protection Agency
UTAQS	Urban Tree Air Quality Score
VOC	Volatile Organic Compound (non-methane)
WHO	World Health Organization

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