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Ground-level ozone in the 21st century: future trends, impacts and policy implications

Science Policy

REPORT 15/08

October 2008

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Cover image: Sunrise over the River Ganges in Varanasi, India showing a sunny and polluted sky, characteristic of photochemical smog. Courtesy of Dr Mhairi Coyle, CEH Edinburgh.

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RS Policy document 15/08
Issued: October 2008 RS1276

ISBN: 978-0-85403-713-1

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Design by Franziska Hinz, London
Copyedited and Typeset by Techset Composition Limited

Ground-level ozone in the 21st century: future trends, impacts and policy implications

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Executive summary

Air pollution and ground-level ozone

Clean air is a basic requirement for human health and wellbeing. However, in many countries around the world air pollution is a serious threat to both human health and the environment.

Tropospheric ozone (O_3) is a global air pollution problem and an important greenhouse gas. Although not a new issue, ground level O_3 remains one of the most pervasive of the global air pollutants, with impacts on human health, food production and the environment. At the global scale, O_3 pollution is highest in Central Europe, Eastern China, and the Eastern USA. In Europe, the highest O_3 levels occur in Central and Southern Europe. High O_3 levels also occur in the tropics and are often associated with biomass burning.

Ozone is a major constituent of photochemical smog. It is a powerful oxidant that damages human health and natural ecosystems, and reduces crop yields. It is also an important greenhouse gas, with a radiative forcing since 1750 third only to carbon dioxide (CO_2) and methane (CH_4). Unlike many other air pollutants, O_3 is not directly emitted, it is a secondary pollutant formed by sunlight driven chemical reactions involving carbon monoxide (CO), volatile organic compounds (VOC) (including CH_4) and nitrogen oxides (NO_X). These precursors arise from both natural biogenic sources and a broad range of human activities. The breadth of sources of O_3 precursors, the role of natural and physical processes in O_3 distribution, production and destruction, and complex chemistry, mean that control of O_3 is not straightforward. The only practical management strategy is to control the emissions from human activities that lead to O_3 formation.

Ozone has been recognised as a significant local and regional air quality issue for many years due to the impacts of high O_3 episodes on human health and crops. These episodes, during which O_3 concentrations may peak at 200 parts per billion by volume[1] (ppb) or more, occur in polluted regions under hot and sunny conditions. O_3 is also present at background concentrations (refer Box 1.1) which vary geographically, and throughout the year. Between the late 19th century and 1980, concentrations of background O_3 in the Northern Hemisphere mid-latitudes doubled to about 30–35 ppb and have since increased by another 5 ppb to 35–40 ppb. Impacts on human health and vegetation during high O_3 episodes are well established; however there is increasing evidence of effects at background concentrations leading to concerns about the implications of further increases in background O_3. The cause of the increase in background O_3 is not fully understood but is thought to be due mainly to increases in emissions in Northern Hemisphere countries, from poorly regulated sectors such as international shipping and aviation, and possibly also due to an increase in O_3 from the stratosphere. Ozone can no longer be considered a local air quality issue – it is a global problem, requiring a global solution.

1 Parts per billion by volume – ppb. A unit commonly used to quantify trace gas concentrations in the atmosphere.

This study assesses possible future changes in global and regional O_3 concentrations to 2050 and 2100 given changes in socioeconomic factors, trends in emissions of precursor gases and climate change projections. The effect of current O_3 precursor emissions controls on global and regional O_3 over the century is assessed. New modelling work to quantify the effects of changes in emissions and climate change on future O_3 concentrations is presented. The report evaluates the potential effects of O_3 on human health, the natural environment, agriculture, and the climate system, and considers the policy implications of these impacts.

Drivers of ground-level ozone change

The main drivers of ground-level O_3 concentrations are the emissions of the precursor gases NO_X, non-methane (nm) VOC (nmVOC), CH_4 and CO followed by climatic factors which regulate the rates of many of the processes in O_3 production and destruction.

Over this century, economic growth and an increasing global population will drive the processes that lead to emissions of O_3 precursors. Changes in climate and land use will influence the production of emissions from natural sources. Increased demand for energy, transport, food and non-food crops and other resources will influence emissions arising from human activity, and changes in patterns of consumption and production will affect the distribution of O_3. Although there will be an increase in the activities that lead to emissions, the implementation of legislation and new technologies which reduce precursor emissions, will contribute to decoupling O_3 pollution from economic growth.

Climate change is expected to significantly influence future O_3 concentrations, as the formation, destruction and transport of O_3 is strongly dependent on biogeochemical and physical processes most of which are affected by climatic factors such as temperature, rainfall and humidity. Dry deposition is one of the dominant O_3 removal processes and is strongly affected by temperature and soil moisture conditions. Climate also affects the natural production of precursors such as CH_4, other VOC such as isoprene, and NO_X from lightning and soil.

Future ozone in a changing world

Previous modelling analysis has show that with unmitigated growth in O_3 precursor emissions, O_3 will present a serious global air pollution problem by the end of the century. The results from the new scenario analysis and modelling work emphasise the important role of emissions controls for determining future O_3 concentrations. They also show that by 2050, climate change may have significant impacts on ground-level O_3 at the local and regional scale.

Considering only changes in anthropogenic emissions between 2000 and 2050, and assuming the full implementation of

current global emissions controls, mean O_3 concentrations during the maximum O_3 season are projected to reduce over much of the developed world in 2050, but increase over the developing world. In the North Eastern USA, Europe and Japan, significant reductions are projected in 2050. In regions where few legislative controls are currently in place, such as Asia and Africa, modest increases in O_3 are projected. Analysis of changes in urban O_3 concentrations suggests that O_3 will increase in many cities by 2050 despite the implementation of O_3 controls due to the reduction in the NO_X titration (refer 3.3.2) of O_3 from NO_X emission controls.

Considering only changes in climate between 2000 and 2050, mean O_3 concentrations during the maximum O_3 season are projected to increase over polluted land regions, and to decrease over the tropical oceans. When changes in both emissions and climate change are considered the largest increases in O_3 concentrations are projected for regions where there are major sources of emissions and where emissions controls are currently weakest, such as Asia and Africa. In industrialised countries, where emissions controls are relatively strong and emissions are projected to decline over forthcoming decades, O_3 concentrations will be reduced although climate change will tend to reduce the effectiveness of emission reductions. Even if emissions are reduced and background O_3 declines, it is expected that there will be an increase in the frequency of high pollution days globally due to the changes in weather expected under future climate change. In the UK and Europe there is likely to be an increase in the frequency of summer droughts, heat-wave events, and associated high O_3 episodes.

Implications of future ozone projections

Northern Hemisphere background concentrations of O_3 are now close to established levels for impacts on human health and the terrestrial environment, raising concerns about the global effects of the pollutant.

Impacts of O_3 on human health are associated with short-term acute effects on the respiratory system, chronic disease and death. Health impacts have been observed at around 35 ppb and below the current WHO guideline of 50 ppb (daily 8 hour average concentration). In the EU, 21,400 premature deaths each year are associated with O_3 (European Environment Agency (EEA) 2007). The total health impact of O_3 is now understood to be driven more by the days at which O_3 is at baseline concentrations (refer Box 1.1) than by the occasional days during which episodes occur. This has led to recognition that efforts to reduce the impacts of O_3 on human health and the environment must be focused on background as well as peak O_3. In regions of Africa, Latin America and Asia where O_3 is projected to rise due to increasing emissions, human exposure will increase. Ozone is also projected to increase in urban areas due to reduced NO_X titration and is expected to lead to higher human exposure in these locations. An increase in background O_3 will exacerbate this increase. The human health impacts of O_3 in urban areas are expected to increase with climate change due to a combination of an increase in the number of high O_3 episodes, and the reduction in NO_X titration.

The impacts of O_3 on vegetation may lead to long-term effects on ecosystem structure and function and impacts on the carbon cycle. In addition to reductions in plant growth and photosynthesis, O_3 can reduce the yield and affect the nutritional quality of major crop species, including wheat, rice and soybean. Global yields of staple crops are reduced as a consequence of current O_3 exposure, and this impact is likely to increase in some regions even with the full implementation of current legislation. In the USA in the 1980s the annual cost of loss of arable crop production due to O_3 was estimated to be $2–4 billion. In the EU in 2000 an estimated €6.7 billion was lost due to impacts to arable crops. For the same year global yield losses were estimated to be $14–26 billion for rice, soybean, maize and wheat combined. In some rapidly developing regions such as South Asia the impact of O_3 on the production of some staple crops such as wheat and rice may present a significant threat to regional food security.

Impacts of O_3 on vegetation may reduce the terrestrial carbon sink. This represents a climate effect in addition to the direct radiative forcing of O_3, and may lead to O_3 becoming a more important driver of climate change in the future. Future increases in atmospheric CO_2 concentrations may moderate this effect by reducing the sensitivity of plants to O_3, although the size of this effect varies with species and environmental conditions. The implications of O_3 impacts for biodiversity are less well known, but available evidence shows a similar range of sensitivity to O_3 in crop plants and native species.

Policy needs

In the EU, Japan and North America, abatement measures have been implemented and have been reasonably effective at reducing anthropogenic emissions of NO_X and VOC leading to reductions in peak regional concentrations and the frequency of O_3 episodes with consequent health benefits at the local and regional level. However, these policies have not controlled the background concentration of O_3, which in these regions, has increased at about 2 ppb per decade since 1980. This background is beyond the ability of any single country or region to control. Until policy frameworks are developed and implemented to address global transboundary O_3 concentrations, as well as addressing the regional and local level concentrations, national and regional level controls are unlikely to achieve their policy objectives. An international mechanism is required to globally coordinate the control of ground-level O_3.

Future O_3 concentrations will be determined mainly by emissions of O_3 precursors and it is the extent to which emissions controls are implemented over the next few decades that will determine whether O_3 is a problem in 2050. Significant sources of anthropogenic emissions not yet regulated must be integrated into the O_3 regulatory process. Of these, international shipping and aviation are the most important priorities as their emissions are projected to grow rapidly in the next decade. Existing emissions controls should be implemented as a priority, and as climate change is expected to exacerbate any future increases in O_3, a key policy objective should be to ensure O_3 precursor emissions are reduced in the

most vulnerable regions within the next few decades. Regulation of biomass burning, and stronger NO_X and CH_4 emissions controls will be required to achieve the level of reductions needed.

Unless there is a substantial increase in the capacity of many countries outside Europe, North America and Japan to assess the science and manage O_3, the success of current control efforts will be limited. New regionally focused frameworks are necessary where they are not already in place and should be designed on the basis of best practice and the experiences of other regions in developing air pollution control frameworks. The linkages between polluting sectors need to be strengthened, and emissions controls set to take into account the cumulative effects of emissions from these sectors. Capacity building will be fundamental particularly in those regions where there are limited O_3 controls in place. Innovation and technology transfer will play a crucial role.

Ozone is an important greenhouse gas, and climate is a key driver of O_3 production and destruction. Ozone is therefore no longer just an issue for air quality. Greater harmonisation between the international climate and O_3 regulatory frameworks should be a priority. Improved policy integration would maximise the benefits of air quality control measures, reduce impacts on human health and the environment, and the climate system. Prevention of deforestation and regulation of biomass burning, and reduction of CH_4 emissions would help to achieve both climate change, O_3 and other air quality objectives. Biofuels provide a good example of an emerging policy issue where better integration of energy, climate and O_3 policies would help to avoid unintended consequences.

Science and research

Recent advances in O_3 science have delivered improvements in understanding the global dimension of ground-level O_3 concentrations, responses to changes in emissions of precursors and climate change and in quantifying effects on human health and the environment. Major gaps remain however in understanding how anthropogenic and natural emissions may change in the future, the impacts of changes in climate and land-use on future O_3 concentrations, and the implications for human health, the environment and the climate system. Poor emission inventories in many countries, particularly in developing countries, and inadequate understanding of global biogenic nmVOC and NO_X emissions are a major issue. Methane emissions are another area of uncertainty as the global CH_4 budget and causes of recent changes in global concentrations are not well understood. The quality of future O_3 projections is limited by what is known about the basic processes and feedbacks between atmospheric chemistry, climate and ecosystems and the interactions between climate change and O_3. Limits in computing resources restrict the ability of climate and chemistry models to capture all of the climate sensitive processes which affect O_3 production, destruction and transport at appropriate scales.

Health impact assessments are limited by a lack of accurate baseline data in many countries, and are complicated by uncertainty regarding whether there is a threshold of effects for O_3 in humans, and by current understanding of the impacts of acute and chronic effects at the individual and population level.

Experimental investigations of the impacts of O_3 on native plant and crop species, requires a focus on effects outside of Europe and North America, in particular in regions of Asia, Latin America and Africa, where O_3 concentrations are projected to increase but there is little information on effects on crop production or ecosystem function. The effects of background O_3 also require further long-term experimental studies and mechanistic models used to assess the impacts of O_3 on vegetation will require further development and validation across a wider range of crop and wild species. The effects of O_3 on global biodiversity are largely unknown. More long-term field-based experiments are necessary to reduce the uncertainties associated with the interactions between CO_2 fertilisation and O_3 on plant productivity.

In conclusion, O_3 has become a global pollutant responsible for widespread effects on human health, crops and native plant communities. The controls implemented to date at regional scales have not been sufficient to prevent the global growth in background O_3, which has the potential to become a very serious threat to human health, food security and ecosystems later this century if insufficient control measures are implemented. The technology to solve ground-level O_3 pollution is widely available and, among the global environmental problems, ground-level O_3 is unique in being very controllable.

1 Introduction

The 20th century was a period of unprecedented change: global population increased from 1.7 billion to 6.1 billion (United Nations 2001), global GDP increased 19 fold (International Monetary Fund (IMF) 2000), and the consumption of fossil fuels rose 15 fold (Smil 2003). This enormous expansion in global production of goods and services has allowed the world to sustain much larger populations and higher standards of living than at any other time in human history. However, to support this growth, natural resources have been consumed at increasingly unsustainable rates and environmental degradation has accelerated.

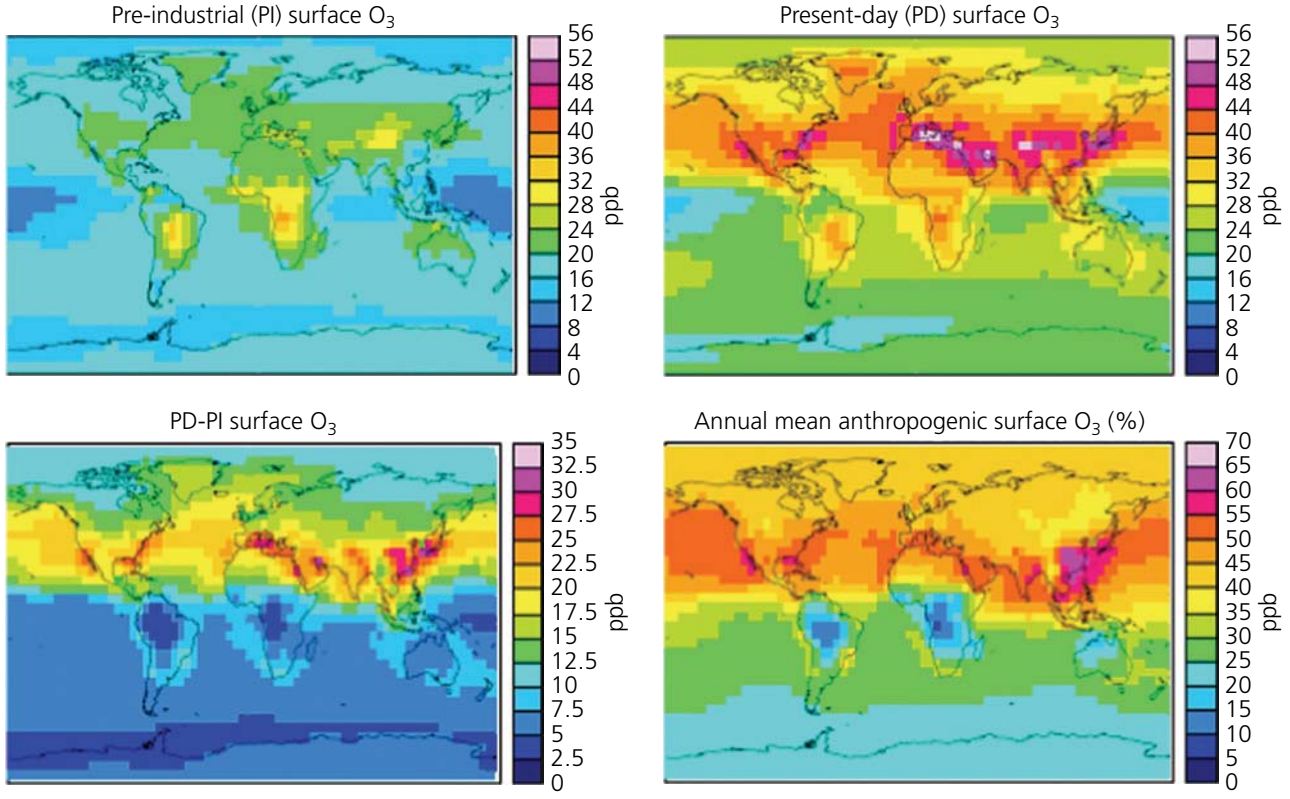
Air pollution is one of the consequences of this unprecedented change. According to the World Health Organization (WHO), more than two million premature deaths each year can be attributed to the effects of outdoor and indoor air pollution (WHO 2006). By comparison, in 2006, 1.7 million people died of tuberculosis (WHO 2008) and in 2005, 2.9 million people died from AIDS (National Aids Trust 2006). Air pollution is clearly a global problem.

The main air pollutants are particulate matter (PM), sulphur dioxide (SO_2), nitrogen dioxide (NO_2) and nitric oxide (NO) known together as nitrogen oxides (NO_x), CO, O_3 and lead (Pb). All have important human health and environmental impacts.

The focus of this study is on tropospheric O_3 (refer Box 1.1). Ozone was selected because it remains one of the most important of the global air pollutants in terms of impacts to human health, croplands and natural plant communities, and may become more important in the future. Despite efforts to control O_3, background concentrations in the northern hemisphere have more than doubled to 35–40 ppb since the industrial revolution, and peak values continue to exceed the WHO guideline values of 50 ppb in many countries, including Latin America, North America, Europe and Africa (WHO 2006). Climate change over the next century also has the potential to influence future O_3 levels by modifying the rates of O_3 production and destruction in the atmosphere and at the Earth's surface, and the processes by which O_3 and O_3 precursors are transported.

Future O_3 concentrations are quantified using chemistry–transport models operating at global, regional and local scales. Projections of future O_3 concentrations differ depending on the model parameters used. In the Northern Hemisphere, mean surface O_3 is projected under some high emission scenarios to increase by 10 to 30 ppb by 2100, with the largest increases occurring at 20–30° N (Prather *et al.* 2003) where emissions are greatest globally. As background O_3 concentrations in Europe are generally over 30 ppb currently (see Figure 1.1),

Figure 1.1 Modelled global changes in surface O_3 concentrations between pre-industrial times and the present day. Multi-model mean surface layer annual mean O_3 (ppb) is presented for pre-industrial (PI) times in the top left, and for the present day (PD) in the top right. The modelled increase in O_3 (PD–PI) is presented in the lower left figure and the percentage of annual mean O_3 attributable to anthropogenic sources in the lower right.



an increase of this magnitude would constitute a major air pollution problem and would lead to the regular exceedance of safety levels established for the protection of human health and vegetation. More recent projections of global emissions show that future O_3 is closely tied to the level of anthropogenic emissions of O_3 precursors (Dentener *et al.* 2006b), and to NO_X in particular, and is therefore controllable with the appropriate legislation.

The objective of this study was to evaluate the importance of O_3 as a pollutant through the 21st century. To set the context for the report a very brief summary is provided of the relevant policy landscape in chapter 2. This includes a short analysis of how effective past controls have been in achieving O_3 policy objectives. In chapter 3 the major processes responsible for O_3 production and destruction in the troposphere and at the Earth's surface are introduced. In chapter 4, the main emission sources of O_3 precursors and new scenario projections of changes in anthropogenic emissions of CO, NO_X and CH_4 over the next few decades are described. This is complemented by a discussion of the state of knowledge of natural emissions, with a particular focus on the influence that environmental changes will have on future biogenic nmVOC emissions into the future. These results form the basis of the new analysis presented in chapter 5 in which models are used to explore changes in O_3 concentrations in 2050 in response to changes in emissions and climate change with a focus on global scale modelling. As chemistry–transport models are limited in their ability to include all important climate or O_3 processes and their interactions at the appropriate spatial and temporal scales, chapter 6 explores these processes in more detail and provides an assessment of which may be important for future O_3 concentrations and the climate system. The implications of these future O_3 projections for human health and the environment are presented and discussed in chapters 7 and 8 respectively, and the policy implications briefly reviewed in chapter 9. A summary of the research needs identified is presented in chapter 10, and the conclusions and recommendations are provided in chapter 11.

While there are inevitable uncertainties associated with looking so far into the future, this longer term perspective is necessary for assessing the likely interactions between climate change (which is expected to be greater in the latter half of the century) and O_3. This study necessarily focuses on what may happen at the global level. However, where the appropriate information was available, regional and local scale results are discussed.

1.1 What is ground-level ozone and why does it matter?

Ozone is a secondary pollutant formed in the atmosphere by sunlight driven chemical reactions between the O_3 precursor gases: nmVOC, CO, NO_X, and CH_4 (see chapter 3). When referring to ground-level O_3, a distinction is usually drawn between baseline, hemispheric background, or peak/episodic O_3 concentrations (see Box 1.1). Background O_3 in the Northern Hemisphere is now in the range of 35–40 ppb. High O_3 concentrations occur episodically throughout the year depending on the weather. During such episodes, O_3 concentrations can reach as high as 200 ppb as experienced in France during the 2003 heat wave (Fiala *et al.* 2003). Even larger values, of up to 400 ppb, were observed during the smog episodes in Southern California in the 1960s. In some cities in the USA and Latin America (WHO 2006) and in metropolitan areas in Asia (Emberson *et al.* 2003) episodes of this magnitude are a common feature.

The long-range transport of O_3 and O_3 precursors has an important impact on O_3 concentrations at regional and local scales. For example, O_3 from Asia and Europe contributes to O_3 concentrations in North America. Similarly concentrations entering Europe from the West are substantial and range from 20 to 40 ppb through the year due to emissions of O_3 precursors elsewhere in the Northern Hemisphere, notably North America and Asia.

Ozone is also an important pollutant at the urban scale. In and around urban areas large gradients of O_3 can be observed; concentrations are generally larger in suburban and rural areas than in busy urban centres because of rapid chemical interactions between O_3 and other pollutants especially nitric oxide (NO). The reasons for this are described in more detail in chapter 3.

Ozone impacts on human health and the environment are well established. It is accepted that impacts on human health occur above 50 ppb (WHO 2006), and although it is unclear whether there is a threshold for effect in humans, impacts are thought to occur at ambient concentrations (around 35 ppb), although some individuals are more susceptible than others. Most of the human health impacts relate to the respiratory system and include reduced lung function, lung irritation and in extreme cases, mortality. In Europe it is estimated that O_3 contributes to 21,400 premature deaths annually (EEA 2007). Impacts on vegetation generally occur above 40 ppb, although this is species dependent and varies according to environmental conditions. Ozone has been shown to damage sensitive plant species, to reduce tree growth and carbon sequestration and to affect the composition of natural plant communities. Ozone also reduces the yield of staple crops. This has clear implications for future food security, as increasing O_3 concentrations may place further pressure on agricultural systems already under stress from climate change, pests and diseases, and land degradation. Ozone is also a greenhouse gas and therefore has a warming effect on the climate system.

1.2 Sources of ozone emissions and drivers of production now and in the future

Emissions of O_3 precursor gases arise from a wide range of sources that are either natural in origin or driven by human activities. Energy generation, transport, agriculture, industrial processes, biomass burning and land use changes such as deforestation are significant sources of O_3 precursor gases. Socioeconomic and environmental factors are therefore important drivers of O_3 pollution as they affect either the

Box 1.1 Useful definitions

Ozone

Ozone is a natural constituent of the atmosphere and is present in the stratosphere and troposphere. Although O_3 is transported between the stratosphere and troposphere, the two are largely separate regimes.

Stratospheric ozone

In the stratosphere, O_3 is produced following the photolysis of molecular oxygen. The 'ozone layer' acts as a protective layer filtering out dangerous UV radiation from the sun.

Tropospheric or Ground-level ozone

Ozone is present throughout the troposphere and at the Earth's surface (ground-level O_3) and is produced through very different chemical processes to stratospheric ozone. In the troposphere, ozone is a secondary pollutant generated when NO_X and VOC react in the presence of sunlight (see Chapter 3). The concentrations of ground-level O_3 currently range from a few parts per billion by volume (ppb) up to 200 ppb.

Peak ozone concentrations (or episodes)

Peak O_3 concentrations occur when high O_3 precursor emissions coincide with appropriate meteorological conditions (sunshine and high temperatures) resulting in high surface O_3 concentrations, often in excess of 100 ppb and sometimes more than 200 ppb. The number of O_3 episodes occurring each year depends strongly on the weather.

Hemispheric background ozone

Hemispheric background O_3 is the remaining concentration once the emissions of anthropogenic ozone precursors from within a region are switched off. Hemispheric background O_3 comprises O_3 produced from natural sources of O_3 precursors within a region, together with O_3 imported into the region derived from all sources (anthropogenic and natural, including the stratosphere). The value varies seasonally and with latitude.

Baseline ozone

Baseline O_3 is the average measured concentration of O_3 within a region and is made up of both the anthropogenic emissions produced within a region and the background concentration of O_3.

production of natural and anthropogenic O_3 precursor emissions, or O_3 production or destruction rates. Underpinning these drivers are population growth and economic development as they lead to increases in demand for resources such as land and fossil fuels, and polluting activities such as energy production, agriculture and transport. The world's population is expected to reach 9.2 billion in 2050 (UNFPA 2007) and increased urbanisation, particularly in rapidly developing countries, and the associated growth in transport, industrial infrastructure and energy use, are expected to lead to a rapid increase in O_3 pollution unless regulatory controls and new technologies are implemented.

Increased demand for land to support this increasing population and economic growth is also likely to be a significant driver of future O_3 precursor emissions. Between 1950 and 1980 there was an unprecedented change in land use with more land converted to cropland than at any other time since 1750 (Millennium Ecosystem Assessment (MEA) 2005). This has led to changes in emissions of O_3 precursors due to biomass burning, changes to emission rates from natural vegetation and agricultural crops, and increases in emissions from fertiliser and fossil fuel use. The demand for food and for alternatives to fossil fuels is expected to be an important driver of land use change and therefore O_3 precursor emissions over this century.

Climate change is expected to influence the contribution of natural sources of precursor emissions to O_3 pollution as emissions from vegetation, soil and lightning are affected by sunlight, temperature, humidity and rainfall. Changes in meteorology will affect transport and mixing, and increasing concentrations of CO_2 are expected to influence the rate of uptake of O_3 by vegetation. Efforts to mitigate and adapt to climate change are also likely to have an impact on O_3 concentrations. For example a shift to production of biofuel crops could have a significant impact on VOC emissions.

1.3 Approaches to controlling ozone

The USA and Japan have had national level controls in place to address O_3 since the 1960s, and the EU since the 1970s. Controls include regulation of O_3 precursor emissions, and guidelines to alert vulnerable members of the population and polluting industries when O_3 concentrations exceed levels that may cause significant harm to human health.

There are as yet, no measures in place to address the hemispheric transport of O_3 and its precursors. The emphasis of control efforts has historically been at the country level and has focused on controlling high O_3 peaks as these were the periods during which the impacts of O_3 were considered to be greatest (Organisation for Economic Co-operation and Development (OECD) 2008). However, in the United Nations Economic Commission for Europe (UNECE) region the Convention on Long Range Transboundary Air Pollution (CLRTAP) Gothenburg Protocol provides a regional approach to O_3 control. This is unique globally. There are no other equivalent approaches to air pollution management in other regions at this scale. The Gothenburg Protocol is also unique in that it provides a multi-pollutant, multi-effect strategy for managing air pollutants. The protocol recognises the effects of control measures across the spectrum of pollutants, and potential interactions between pollutants.

Few of the potential interactions between air quality and other policy areas have yet to be recognised in policy frameworks.

Technological measures also have an important role to play in controlling O_3 pollution as they can reduce the emissions per unit of production by up to 99% and have the potential to rapidly reduce emissions in many areas, particularly in developing regions. For example, vehicle technologies such as catalytic converters, improved evaporative emissions controls and fuel improvements have led to substantial reductions in emissions over the last two decades.

1.4 Conduct of the study

The Royal Society established a working group of international experts in 2007. Chaired by Professor David Fowler FRS the group was tasked with synthesising existing knowledge of ground-level O_3 and its impacts on human health and the environment, and evaluating likely changes in O_3 concentrations through the 21st century.

A call for evidence inviting written submissions on these issues was launched in January 2007. Selected experts working on ground-level O_3 were also invited to attend a workshop held in May 2007 at which issues relating to O_3 modelling and atmospheric chemistry, O_3 and climate interactions, and impacts on human health and the environment were discussed. The written evidence received is available on the Royal Society website at: royalsociety.org/document.asp?id=1170.

The main focus of this report is on the likely global changes in O_3 between 2000 and 2050. Previous modelling analysis has provided global O_3 projections to 2030 and was considered by the Intergovernmental Panel on Climate Change (IPCC) during their fourth assessment. The objective of this study was to extend this analysis beyond 2030 to later in the century when the effects of climate change are expected to be greater. To do this, previously published emission scenarios were updated to reflect legislative changes since 2002. These were then used in modelling simulations using global, regional, and urban models which had been extended to 2050 or 2100 where possible, to evaluate the relative importance of changes in emissions from human activities and climate change for future global O_3 concentrations. Additional exploratory work was also completed to estimate potential changes in natural emissions of O_3 precursors as these were not included in the emissions scenarios. As the models used do not capture all important O_3 and climate processes and interactions at sufficient temporal and spatial scales, new analyses were also undertaken to explore the relative importance of different meteorological processes at the regional scale in determining future O_3 concentrations.

The new research presented here is based on established and peer-reviewed methodologies, has been widely reviewed within the relevant research communities and will be submitted for publication in the scientific literature. The results were also discussed in detail at the expert workshop organised in conjunction with this study. Where conclusions and recommendations based on this new work are presented, they are identified as such in the text.

The conclusions in the report are based on peer-reviewed literature, evidence submitted for consideration and the results of the new work completed for this study. The Council of the Royal Society has endorsed its findings.

2 The ground-level ozone policy context

The human health and environmental effects of O_3 during the second half of the 20th century led to the development of policies and control measures in a range of countries. In this chapter the approaches taken to controlling O_3 in Europe and the USA and their success at achieving their objectives are briefly summarised to set the context for the following chapters. A detailed analysis of the effectiveness of control measures is beyond the scope of this report; however, the main effects of the policies within the countries to which they apply and on the global distribution of O_3 is considered.

Ground-level O_3 pollution impacts were first acknowledged in California during the 1950s and were perceived to be a local and regional issue (Haagen Smit 1952). Control measures were designed to reduce O_3 precursor emissions and were focused on reducing peak concentrations as these were considered to be most damaging to human health and ecosystems (Chameides *et al.* 1997). Damaging O_3 concentrations in other parts of North America, in Europe and in Japan were gradually recognised through the 1970s, and

Box 2.1 Air quality standards and other indices used for ozone

Global measures	Human health	Vegetation
The World Health Organization (WHO) provides guidelines for human health that are based on a combination of epidemiological and human exposure evidence	50 ppb daily 8-h mean	N/A
Regional measures		
UNECE – Gothenburg Protocol EMEP[2] domain only		
	Critical level for human health 60 ppb as an 8-h average (expressed as AOT 60 for purposes of integrated assessment modelling)	*(not comprehensive)* Critical level to prevent significant yield loss in sensitive arable crops: AOT40 of 3000 ppb-h over a typical three-month growing season* Critical level to prevent significant growth reduction in young trees: AOT40 of 5000 ppb-h accumulated over a growing season
EU		
The EU Directive on Ambient Air and Clean Air for Europe, defines targets to 2010 along with longer term objectives (timeframe not defined)	Target value: 60 ppb not to be exceeded on more than 25 days per year averaged over 3 years (maximum daily 8-h mean) (to be achieved by 2010) Longer term objective: 60 ppb (maximum daily 8-h mean) within a calendar year	Target value: AOT40 calculated from 1 h values 9000 ppb-h over 3 months (May–July) averaged value over 5 years Long-term objective: AOT40 calculated from 1 h values. 3000 ppb-h over 3 months (May–July)
EU Alert threshold for O_3	Information threshold: 90 ppb per hour Alert threshold: 120 ppb	N/A
National measures		
UK[3]	50 ppb not to be exceeded more than ten times a year (8-h mean) (to be achieved by December 2005 and maintained thereafter)	Target value of 9000 ppb-h based on AOT40 (to be calculated from 1 h (May–July) and to be achieved by 2010 (averaged over 5 years). To be achieved by 2010

* Refer to Chapter 8 Box 8.1.

2 EMEP is the European Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe.
3 From UK AQ Strategy 2007.

national and regional policies to control precursor emissions developed (Box 2.1) in response. No attempt has been made to control O_3 at the global scale as global background concentrations were not seen to be an issue until relatively recently (Fiore *et al.* 2002).

2.1 Meeting the challenge – the current state of play

Policy-makers have utilised a range of different mechanisms to address ground-level O_3. These include emissions controls for O_3 precursors, standards and guidelines which define air quality targets, alarm thresholds to advise sensitive sectors of the population to move indoors and O_3 alarm plans to restrict emissions associated with land transport and industry during O_3 peak episodes. Box 2.1 compares the standards and measures in place to protect human health and vegetation globally, within the UNECE region, the EU and the UK.

Traditionally, the design of abatement measures has reflected the perception that the impacts of O_3 to human health were limited to periods of elevated O_3 concentrations. However, it is now known that human health impacts occur below these levels, potentially at levels within the ambient range (35 ppb) and well below the WHO guideline level of 50 ppb (daily 8-h average). It is also clear that the total health impact to a population is driven more by the days during which O_3 is at baseline concentrations, than by the occasional days on which episodes occur. Consequently, policy makers have recognised the need for policy measures to reduce background O_3.

At the international level, the United Nations Framework Convention on Climate Change (UNFCCC) provides a framework for management of greenhouse gases. As O_3 is a greenhouse gas, it does, in principle fall under this umbrella. However, the mechanism of implementation for the Convention, the Kyoto Protocol, does not include O_3. Consequently there is no global framework in place for the direct management of O_3. The WHO has issued guidelines to policy makers for the most common air pollutants, including O_3. These are intended to provide guidance for the development of policy measures aimed at reducing the impacts of air pollutants on human health (WHO 2006).

The only international mechanisms in place to control ground-level O_3 are regional in scope. A regional approach to controlling O_3 precursor emissions has been in place in the UNECE region[4] since 1991 when the CLRTAP Sofia Protocol to control NO_X emissions came into force. This was followed by the Geneva Protocol for the control of VOC emissions and the Gothenburg Protocol which came into force in 1997 and 2005 respectively. The Gothenburg Protocol set legally binding national emission ceilings for the anthropogenic emissions of NO_X, VOC, ammonia and sulphur to 2010 for those countries who have ratified the protocol. Now ratified by 24 Parties, this was the Convention's first multi-pollutant, multi-effect mechanism. National limits for European countries are based on scientific assessments of the thresholds of effects on natural ecosystems and human health, and the relative cost effectiveness of abatement options. The Protocol also defines critical levels,[5] which together with integrated assessment modelling, are used to inform the negotiations on national obligations. A number of parties, especially in Eastern and South Eastern Europe, have not yet ratified the Gothenburg Protocol. In North America, where integrated assessment modelling was not used to support negotiations, the USA has ratified the Protocol and set emission limits as required; Canada has not yet ratified the Protocol.

The EU, in addition to the requirements under CLRTAP, has a regulatory framework in place for controlling air pollution, including ground-level O_3. The Clean Air for Europe (CAFE) Strategy specifies EU air pollution objectives and proposes measures to achieve them by 2020 (European Commission 2005). The Directive on Ambient Air Quality and Clean Air for Europe integrates previous Directives concerned with ambient air quality and sets limit values for air pollutants (SO_2, NO_2, NO_X, PM, Pb, Benzene, CO), target values and objectives for protecting vegetation, and provides for information exchange between member states. It also contains target values and long-term objectives for the concentration of O_3 in ambient air and new objectives for fine particles ($PM_{2.5}$). National targets for emissions are set in the National Emissions Ceilings (NEC) Directive. Due to be revised in 2008/2009 this sets national targets for SO_2, NO_X, VOC, $PM_{2.5}$ and ammonia to reduce acidification, O_3 and eutrophication. Also relevant for O_3 control is the Integrated Pollution, Prevention and Control (IPPC) Directive. The Directive imposes a requirement for stationary sources of pollutants from new and existing industrial and agricultural sources with high polluting potential to be permitted only where certain environmental conditions are met. This includes sources associated with the energy industries, production and processing of metals, mineral industry, chemical industry, waste management and livestock farming.

The USA and Canada are parties to the CLRTAP but have their own regional agreement in place for managing ground-level O_3. The Canada–United States Air Quality Agreement makes provision for NO_X and VOC reductions within a pollution emission management area (PEMA) designated within each country.[6] In Canada, O_3 is regulated at the federal level under the Canadian Environmental Protection Act (1999). The federal government has also established Canada-wide standards for O_3 limits.

4 The UNECE region includes the EU, non-EU Western and Eastern Europe, South East Europe, the Commonwealth of Independent States, North America and Canada.

5 Critical level is defined in the Gothenburg Protocol as the concentration of a pollutant in the atmosphere above which direct adverse effects of receptors, such as human beings, plants, ecosystems or materials may occur according to present knowledge. This is used to assess the risk of impacts arising from ozone pollution.

6 For Canada, the area of 301, 330 km^2 that covers all of the Canadian territory south of about the 48th parallel beginning east of Lake Superior to the Ottawa River, and south of the corridor that extends from the Outaouais Region east to Quebec City. For the USA, the area comprising the states of Connecticut, Delaware, Illinois, Indiana, Kentucky, Maine, Maryland, Massachusetts, Michigan, New Hampshire, New York, New Jersey, Ohio, Pennsylvania, Rhode Island, Vermont, West Virginia and Wisconsin, and the District of Columbia.

In the USA the Los Angeles smogs of the mid 20th century led to the identification of O_3 as an important air quality issue (Haagen Smit 1952) and to the development of control strategies. Controls were first introduced in California in 1964–1966 to reduce VOC and CO emissions (Wayne 2000). Further control measures were introduced across the USA as the geographical scale of the problem became clear and were extended to NO_X emissions in the 1970s. The Clean Air Act (1970 and subsequent revisions) sets limits on air pollutants and gives the US Environmental Protection Agency (EPA) the authority to limit emissions from polluting sources. Each state has in place state implementation plans that provide regulations, programmes and policies appropriate to the region for pollutant control.

In the UK, the regulatory framework is guided by the obligations established in the CLRTAP and by the EU. The UK Air Quality Strategy for England, Scotland and Wales (Defra 2007) established a framework for ambient air quality in the UK. There is no legal requirement to meet the objectives in the Strategy except where the limits are set in EU legislation. The UK objective for human health is stricter than that prescribed by the EU (refer Box 2.1). The Environment Act (1995) requires that the Environment Agency and the Scottish EPA have regard to the Strategy when exercising their pollution control functions. These are prescribed under the Environment Protection Act 1990, the Pollution Prevention and Control Regulations 2000 and the Pollution Prevention and Control (Scotland) regulations 2000. Similar requirements exist in Northern Ireland under the Environment (Northern Ireland) Order 2002 and the Pollution Prevention and Control Regulations (Northern Ireland) 2003.

While the UNECE region is the only region in the world with a regional policy framework in place that specifically addresses O_3 and O_3 precursor emissions, other regions, in particular Asia, Africa and Latin America, are starting to take a more regional approach to managing air quality. None are yet focused specifically on reducing O_3 pollution, however O_3 levels may be affected as a consequence of the controls in place. For example in 1998, the South Asian countries signed the 1998 Malé Declaration on control and prevention of air pollution and its likely transboundary effects for South Asia. In 2002, the Environment Ministers of the Association of Southeast Asian Nations (ASEAN) group signed a legally binding agreement on transboundary haze pollution resulting from land and forest fires, and in the African region, the Air Pollution Information Network for Africa (APINA) aims to improve capacity in African countries to monitor and manage air pollution problems.

Additional regional networks, designed to facilitate cooperation in management of urban air quality management, are also in place. For example, the Clean Air Initiative for Asian Cities (CAIA) is a region-wide network which aims to improve air quality in cities by improving awareness, supporting the development and implementation of air quality standards and policies, and strengthening capacity. Similarly, the Clean Air Initiative for Latin America and the Caribbean aims to improve urban air quality in the region by taking a partnership approach to improving air quality and reducing greenhouse gas emissions.

2.2 The effects of controls to date

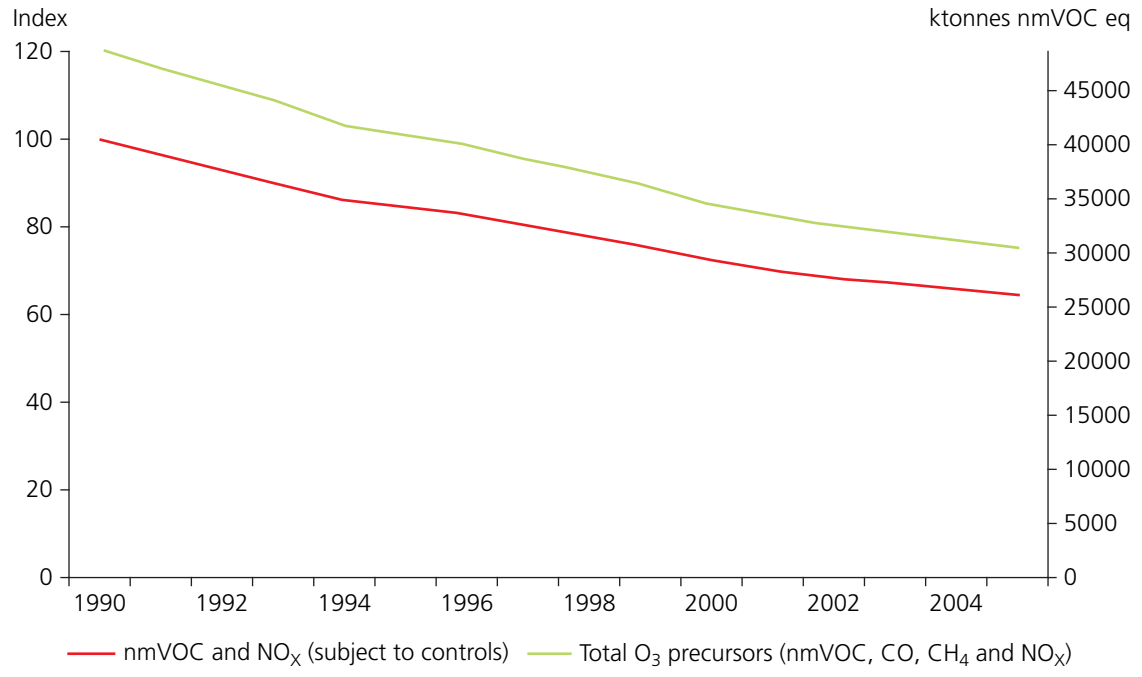
A recent review of the Gothenburg Protocol (UNECE 2007) showed that the emissions of the O_3 precursors NO_X and nmVOC have declined substantially as a result of emissions controls. In 2005, NO_X and nmVOC emissions were 30% and 38% lower than 1990 levels for the European countries within the Protocol (Figure 2.1).

The decrease in NO_X and nmVOC emissions in Europe has resulted in a reduction in the magnitude of short-term peak O_3 concentrations during episodes, with declines in daily peak concentrations of around 30 ppb. Reductions in peak O_3 concentrations have been observed widely in Europe, both in urban and rural areas. The temporal pattern of O_3 concentration however, reveals several additional changes during the period in which emissions of O_3 precursors over Europe have declined. In particular, the lower percentiles of the frequency distribution and even the mean concentration at many sites have been growing. These effects are illustrated in Figure 2.2a and 2.2b, from Jenkin (2008). This figure shows the changes in O_3 concentrations at an urban site (Leeds) and a rural site (Lullington Heath) in the UK, and show similar trends to those observed at other sites across the UK, and more widely in Northern Europe.

The concentrations observed at a site are the result of local and larger scale processes at the regional (country to continent) and importantly also at the hemispheric scale. The resulting distribution of concentrations observed at any site therefore depends on the relative influence of these three contributions at each site. For example, urban sites respond to changes in NO_X emissions in two ways: the mean and low percentiles increase as the local depletion by NO declines (see chapter 3, section 3.3.2), and the peak values decline in response to regional scale emission reductions of O_3 precursors. The growing hemispheric background contributes to the growth in the mean concentration and reduces the magnitude of the decline in the peak values. Rural sites are less influenced by effects of reductions of local NO concentrations, especially at exposed upland or coastal locations, and are more strongly coupled to regional emission changes and to the hemispheric background. The changes in O_3 concentration across the landscape of Europe have therefore responded to both the European emission reductions and to the increasing hemispheric background concentration. These two concurrent changes have changed ground-level O_3 characteristics in different ways in different locations.

Between 1996 and 2004, no change in mean O_3 concentrations was observed across Europe despite these large reductions in NO_X and VOC emissions (EEA 2007) and the substantial reduction in peak values (Solberg *et al.* 2005). The background O_3 has increased by up to 5 ppb per decade since the 1970s (CIAM 2007). No marked trend has been observed in impacts to vegetation over the period 1990–2006 and current policies are not expected to lead to a further reduction in health impacts (UNECE 2007). These trends are considered in more detail in chapter 3 (section 3.6).

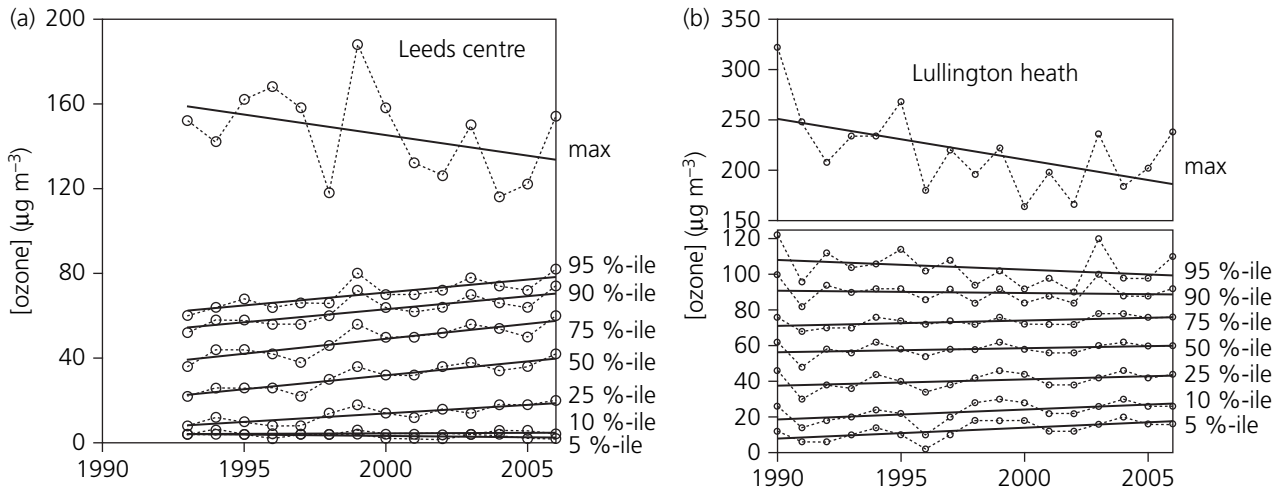
Figure 2.1 Emissions of O_3 precursors in Europe (EEA-32 member countries) from 1990–2005. Source: EEA (2007) reproduced with permission from the EEA.



Similar evidence of reductions in peak O_3 concentrations have been observed in the USA (refer to EPA 2006 for a detailed analysis). The scale of reductions in the USA of emissions of O_3 precursors by 2005 is of a similar order to those in Europe. The measured O_3 concentrations, like those in Europe, show a marked decline in hourly peak O_3 values averaged over all national monitoring stations by approximately 20% between 1980 and 2000 and increases in the lower percentiles of the frequency distribution (US EPA 2002). In recent years the contribution of the hemispheric background to ground-level O_3 concentrations has been a particular focus of the analysis of O_3 in North America (Jacob *et al.* 1999; Fiore *et al.* 2002).

The Air Quality Expert Group (AQEG 2008) have recently reviewed the progress made towards achieving UK targets for O_3 control. They report a general increase in annual mean

Figure 2.2 Changes in ground-level O_3 concentrations at (a) urban (Leeds centre) and (b) rural (Lullington Heath) sites in the UK, showing a decline in peak values and increases in the mean and lower percentiles of the distribution. Trend in hourly mean O_3 distributions based on data over the periods 1993–2006 and 1990–2006 respectively. The solid lines are linear regressions of data indicating the average trend over the period. Source: Jenkin 2008. Copyright (2008), reprinted with permission from Elsevier.



O_3 concentrations at urban sites over the last 10 years due to the reduction of NO_X emissions in urban areas, while peak O_3 concentrations have generally decreased at rural sites due to the control of regional anthropogenic VOC and NO_X emissions (AQEG 2008). Implementation of UK emission reductions are not expected to result in O_3 objectives being met for the whole country by 2020 (Defra 2006) and urban O_3 levels in the UK are expected to rise over the next two decades (Williams 2007; AQEG 2008).

2.3 International shipping and aviation

Although land-based emissions have been reduced in many countries, emissions from international aviation and shipping are rapidly increasing. Aviation emissions contribute an estimated 2% (Schumann *et al*. 2000) and international shipping an estimated 15%, of global NO_X emissions (Corbett *et al.* 2007) respectively. Emissions from these sectors are projected to increase as a result of expected growth in volume. Global passenger air travel, for example, is expected to grow by 5% per year between 2000 and 2020 (Gauss *et al.* 2005). In Europe, NO_X emissions from shipping are currently estimated to be 32% of land-based NO_X emissions (Cofala *et al.* 2007a) and are expected to exceed these sources by 2020 (European Commission 2005; Cofala *et al.* 2007a).

International aviation emissions are not currently regulated, although the International Civil Aviation Organization (ICAO) has defined NO_X emission limits for engine exhausts during the landing and take-off phases of flight. In Europe the NEC Directive regulates NO_X emissions from aircraft during landing and take-off, and emissions associated with airport and port infrastructure are subject to national air quality measures. The O_3 precursors emitted during the cruising phase of flight are not regulated but because they are emitted higher in the atmosphere have a longer lifetime and therefore disproportionate effect compared to emissions at the surface. NO_X emissions during cruising, for example, are of an order of magnitude more important than those emitted during landing and take-off (Tarrason *et al.* 2004). How important these emissions are for O_3 depends on the altitude at which they are emitted. NO_X emitted in the upper stratosphere is less important for O_3 production than when emitted in the troposphere or lower stratosphere (Gauss *et al.* 2005).

While emissions at sea are perceived to have a lower immediate impact than when emitted on land, shipping emissions provide a dominant source of urban air pollution in coastal areas and may be transported large distances (Cofala *et al.* 2007a). NO_X emissions from international shipping are limited under Annex VI of the International Convention for the Prevention of Pollution (MARPOL). However, since being adopted in 1997 and entering into force in 2005 these regulations have been widely criticised for not going far enough. For example, the European Council in 2003 called for the adoption of more ambitious measures on NO_X. This was reiterated in December 2007 when the European Commission repeated its commitment by stating that it will bring forward proposals for Community measures if the IMO fails to deliver sufficiently ambitious proposals. Proposals to strengthen the NO_X emission controls from engines and to introduce measures for VOC will be considered for adoption by the IMO in late 2008.

2.4 The role of science in meeting the ozone policy challenge

Science plays a vital role in the development and implementation of ground-level O_3 policy. Environmental, human health and economic impacts must be identified and quantified, and the mechanisms of impact understood so that appropriate policy objectives or abatement measures can be defined. In the UNECE region, EMEP coordinates national monitoring programmes which contribute to the collection, analysis and reporting of data, the International Cooperative Programmes (ICP) of the UNECE provide underpinning effects assessments, and the International Institute for Applied Systems Analysis (IIASA) provide integrated assessment to identify strategies for maximising the environmental benefits of different control options. The results of this work are then used to inform the negotiations towards development of new protocols within Europe.

The important role of science in policy can be demonstrated by the recent development of risk assessment methods in CLRTAP. Advances in understanding the mechanisms of effects on vegetation have led to the adoption of new critical levels within the CLRTAP Mapping Manual. The distribution of predicted impacts, and hence the relative benefits of emission reductions for O_3 precursors, varies geographically across Europe depending on whether the flux- or concentration-based critical levels are used (Lindskog & Sundqvist 2007; Simpson *et al.* 2007) (see chapter 8, section 8.2, Figure 8.1). For example; the use of a flux-based approach results in predictions of a more widespread spatial distribution of O_3 effects on arable crops and forests compared to the concentration-based approach which predicted O_3 impacts to be a mainly central and southern European problem. A recent synthesis of observational and experimental data within CLRTAP (Hayes *et al.* 2007b) suggests flux-based predictions provide a better prediction of observed impacts for crop species. This has implications for the economic assessment of effects and for the setting of future policy targets that may, in some regions, need to be stronger than originally defined. Similarly, efforts to improve understanding of whether there is a threshold below which O_3 is safe for human health have also led to a reassessment of policy objectives in Europe and by the WHO.

2.5 Conclusions

Despite 40 years of controls in North America, Japan and Europe, ground-level O_3 remains a serious air pollution problem. Where they have been implemented, policy measures have had demonstrable success at reducing O_3 precursor emissions with substantial reductions of NO_X and VOC and consequent declines in short-term peak O_3 concentrations of typically 20–30 ppb in Europe. At the same time, however, measurements in many regions show that the

hemispheric background has been increasing (by typically 2 ppb per decade since 1980) in the mid-latitude Northern Hemisphere over the period during which emission reductions have been made in Europe and North America. The increase in background O_3 is not fully understood, but is thought to be due mainly to emission increases in other Northern Hemisphere countries and increases in emissions from poorly regulated sectors such as international shipping and aviation. While less certain, an increase in the stratospheric source of O_3 may also have contributed to the increasing background. The trend in background O_3 is very important for two reasons: (i) it greatly reduces the benefits of the reductions in peak O_3 concentrations; and (ii) the growth in background has made ground-level O_3 a hemispheric/global issue, as well as a regional/national one.

Developments in the science have also led to recognition that efforts to reduce the impacts of O_3 on human health and the environment must target background as well as peak O_3. Regulatory frameworks are key to controlling O_3, but to be successful, these must be designed to address the specific complexities of this pollutant, and that requires measures to reduce O_3 at the hemispheric/global scales as well as regional/local scales.

3 Ozone in the troposphere: processes of ozone production and destruction

3.1 Introduction

Ozone is a natural constituent of the atmosphere and is present in the stratosphere and troposphere. In the stratosphere O_3 is produced following the photolysis of molecular oxygen. The 'ozone layer' acts as a protective layer filtering out dangerous UV radiation from the sun. Ozone is also present throughout the troposphere and at the Earth's surface (ground-level O_3) and is produced through very different chemical processes to stratospheric O_3. In the troposphere, O_3 is a green house gas. It is a secondary pollutant generated through sunlight driven chemical reactions between NO_X and VOC including CH_4, CO and many other more complex compounds (Crutzen 1974; Liu *et al.* 1980; Atkinson 2000). These gases may be present as a result of emissions within a region and also due to transport into the region of O_3 precursors. The rates of these processes and the overall O_3 budget depend on meteorological conditions such as solar intensity, temperature and pressure, and on the concentrations of O_3 precursor emissions and water vapour in the atmosphere. The other major source of O_3 in the troposphere is transport from the stratosphere.

This chapter provides an introduction to the processes which control the production and destruction of O_3. These include the chemical production and loss of O_3 within the troposphere, and the loss of O_3 through deposition to terrestrial and ocean surfaces. As there is considerable spatial variability in O_3, the chapter includes a brief description of the influences of weather, topography and other geographical features. The effects of urban scale processes on surface O_3 is discussed in some detail as it is in urban areas that the majority of the exposure of the population to O_3 occurs. The interactions between climate and O_3 are considered in more detail in chapter 6.

3.2 The lifetime and atmospheric budget of tropospheric ozone

The lifetime of O_3 is determined by removal processes (or sinks) which lead to loss of O_3 from the atmosphere. The average lifetime of O_3 in the troposphere has been estimated at 22 (±2) days (Stevenson *et al.* 2006); however, it varies with altitude and ranges from 1–2 days in the boundary layer where dry deposition (see section 3.4.1) is the major sink, to several weeks in the upper troposphere. This, in combination with the potential for O_3 to be produced from precursors long after they have been emitted, makes O_3 a global pollutant.

The amount of O_3 in the atmosphere, or O_3 budget, is determined by the rates of O_3 production and destruction. Ozone production in the atmosphere has been estimated to account for approximately 4500 Tg y^{-1}. The net flux of O_3 from the stratosphere to the troposphere is approximately 540 Tg y^{-1}. The O_3 budget is closed by two loss processes: chemical destruction and dry deposition to the Earth's surface. Chemical loss or destruction in the atmosphere accounts for approximately 4100 Tg y^{-1}, and deposition to the surface accounts for approximately 1000 Tg y^{-1} (Gettelman *et al.* 1997; Olsen *et al.* 2001, 2004; Prather *et al.* 2001; Stevenson *et al.* 2006; Denman *et al.* 2007; Wild 2007; Wu *et al.* 2007a). Integrated over the whole troposphere, chemical production and loss rates are several times larger than the influx from the stratosphere and the surface deposition flux. The budget values at a global scale are shown in Figure 3.1.

3.3 Ozone atmospheric chemistry

The key chemical processes that lead to O_3 production and destruction are driven by reaction cycles involving free-radical intermediates, which are mainly formed from the photolysis of O_3 itself (reaction (1)). At UV wavelengths shorter than about 320 nm, O_3 photolysis generates electronically excited oxygen atoms, $O(^1D)$, which can either react with water vapour to form hydroxyl (OH) radicals (reaction (2)), or reform O_3, following collision with an inert molecule (denoted "M"), most commonly nitrogen, N_2 (reactions (3) and (4)):

$$O_3 + h\upsilon \rightarrow O(^1D) + O_2 \quad (1)$$

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

$$O(^1D) + M \rightarrow O(^3P) + M \quad (3)$$

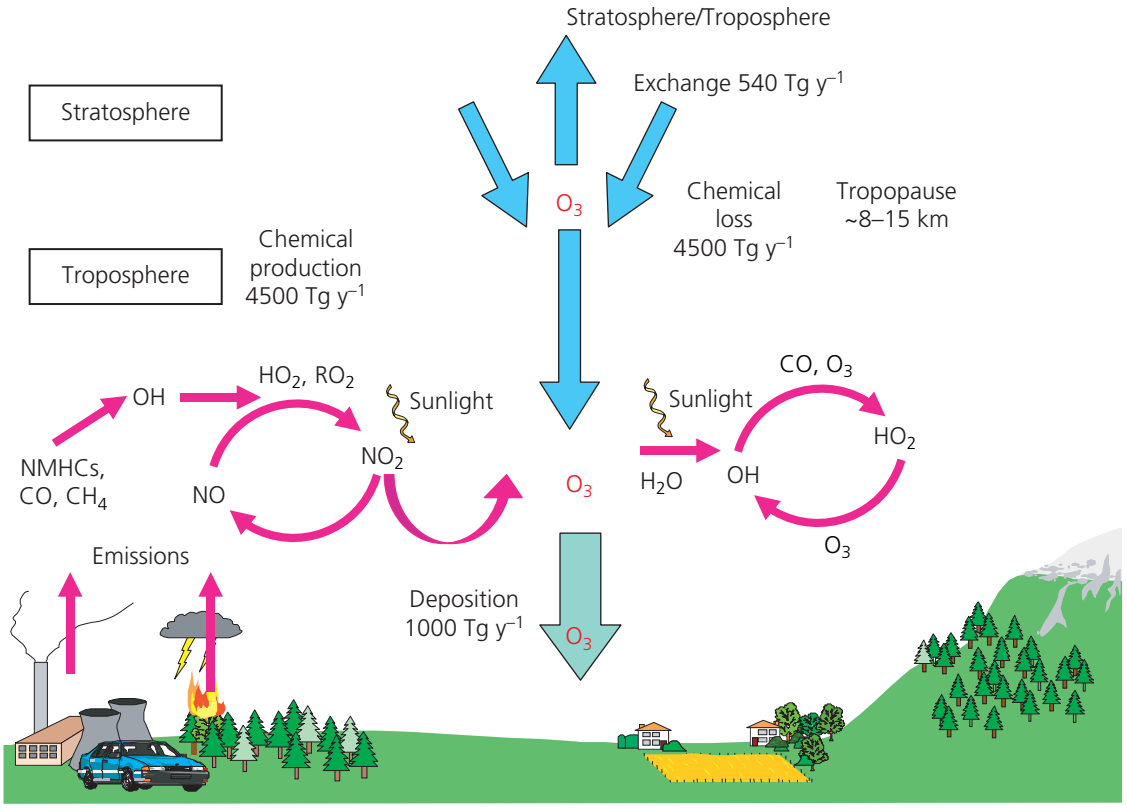
$$O(^3P) + O_2 + M \rightarrow O_3 + M \quad (4)$$

The conversion efficiency of $O(^1D)$ to OH depends on the relative rates of reactions (2) and (3). This is mainly determined by the concentration of water vapour in air which, in turn, depends on the prevailing temperature and relative humidity. For example, in air saturated with water at atmospheric pressure, the fraction of $O(^1D)$ atoms proceeding via reaction (2) increases from about 9% at 10°C to about 12% at 15°C.

OH radicals play a central role in tropospheric O_3 chemistry. As illustrated in Figures 3.2 and 3.3, OH reacts mainly with atmospheric CH_4 and CO to initiate the reaction cycles which produce and remove O_3.

Figure 3.2 provides an overview of the key chemical processes that lead to the production and removal of O_3. The concentration of NO_X is the main factor which determines whether O_3 is produced or removed in the atmosphere. The figure is divided into three regimes (I = Low NO_X, II = Intermediate NO_X, III = High NO_X), to illustrate the changing balance of the free-radical reactions, and their impact on O_3, as the level of NO_X increases. Figure 3.3 provides an example illustration of the corresponding variation of the rate of production or loss of O_3 for the three regimes, and the concentration of OH radicals, based on simple box modelling calculations appropriate to conditions in the lower troposphere.

Figure 3.1 A schematic view of the sources and sinks of O_3 in the troposphere. Annual global fluxes of O_3 calculated using a global chemistry–transport model have been included to show the magnitudes of the individual terms. These fluxes include stratosphere to troposphere exchange, chemical production and loss in the troposphere and the deposition flux to terrestrial and marine surfaces. Data source: IPCC Fourth Assessment Report Working Group I Report "The Physical Science Basis" (Denman et al. 2007).



The first regime (regime I: Figure 3.2 and 3.3) corresponds to remote regions of the atmosphere, such as the South Pacific Region which have very low NO_X levels (less than 20 parts per trillion (ppt) NO_X), and is characterised by net O_3 removal. The reactions of OH with CH_4 and CO lead to the formation of the peroxy radicals, CH_3O_2 and HO_2, which are removed by their mutual reactions to form methyl hydroperoxide, CH_3OOH, and hydrogen peroxide, H_2O_2:

$$CH_3O_2 + HO_2 \rightarrow CH_3OOH + O_2 \quad (5)$$

$$HO_2 + HO_2 \rightarrow H_2O_2 + O_2 \quad (6)$$

This chemistry constitutes a loss process for O_3, because the reaction sequence was initiated by O_3 photolysis (reaction (1)). Some additional O_3 removal also occurs because HO_2 radicals can react with O_3,

$$HO_2 + O_3 \rightarrow OH + 2O_2 \quad (7)$$

leading to the regeneration of OH radicals as part of an O_3 depleting OH–HO_2 inter-conversion cycle (see Figure 3.2, regime I).

As demonstrated in Figure 3.3, the chemistry in regime I leads to comparatively small O_3 loss rates. However, this has an important global impact because it applies to a large proportion of the troposphere. More rapid O_3 depletion rates can occur at specific locations, as a result of chemical cycles involving halogen atoms and radicals. This is described below in section 3.3.1.

The second regime (regime II: Figures 3.2 and 3.3) is characterised by net O_3 formation, the rate of which increases with increasing NO_X concentrations and corresponds to the rural areas of most industrialised countries. The reactions of the peroxy radical intermediates with NO result in the conversion of NO to NO_2 (reactions (8) and (9)) with the subsequent photolysis of NO_2 generating O_3 (reaction (10) followed by reaction (4)):

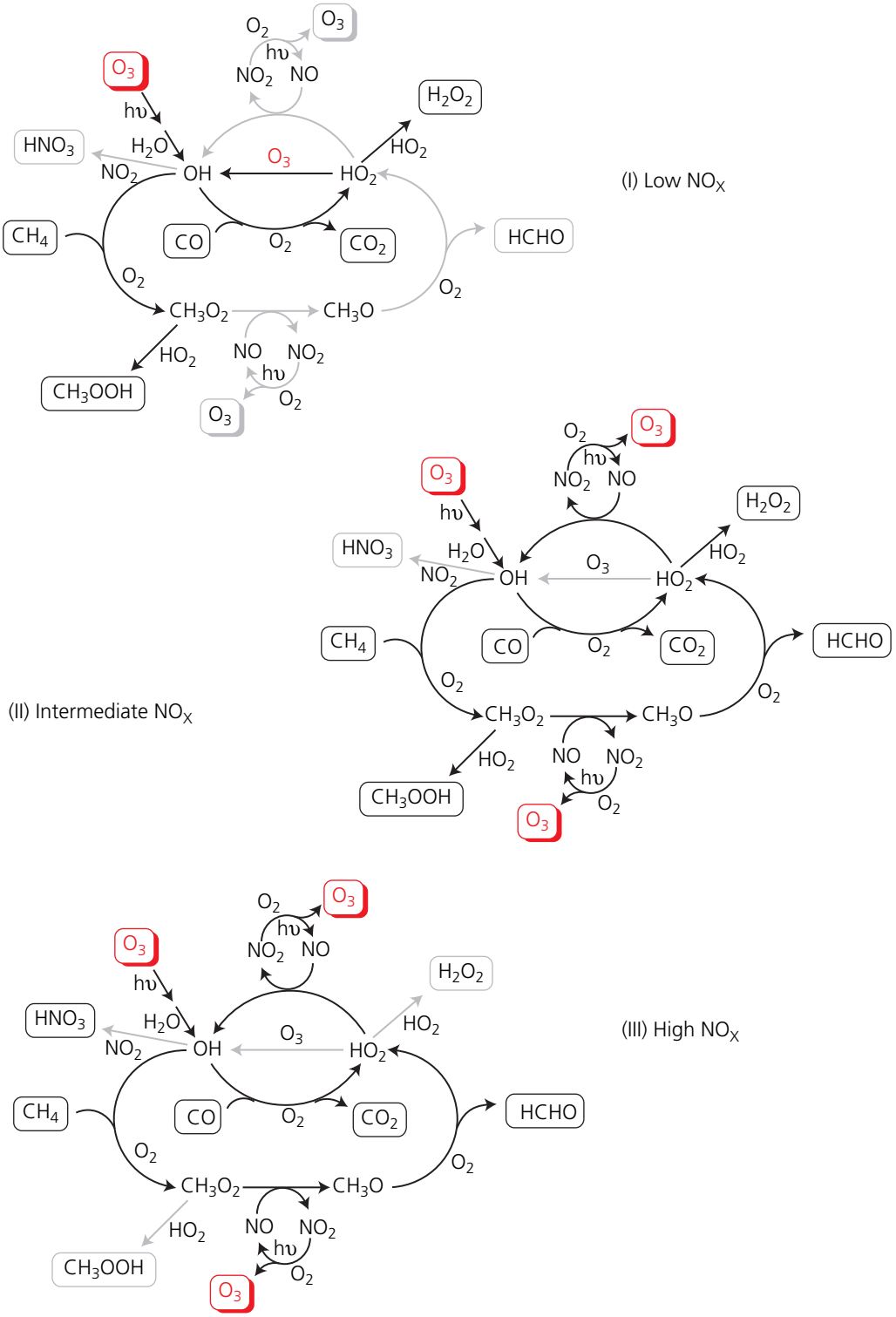
$$CH_3O_2 + NO \rightarrow CH_3O + NO_2 \quad (8)$$

$$HO_2 + NO \rightarrow OH + NO_2 \quad (9)$$

$$NO_2 + h\upsilon \rightarrow NO + O(^3P) \quad (10)$$

As shown in Figure 3.2 reactions (8) and (9) form part of the free-radical propagated O_3 forming cycles, which may occur a number of times before being halted by a radical termination reaction. Under the prevailing conditions (ie regime II), NO_X levels remain sufficiently low that peroxide formation via reactions (5) and (6) continues to be the major radical sink. Ozone formation in this regime is often referred to as 'NO_X limited' or 'NO_X sensitive', because the

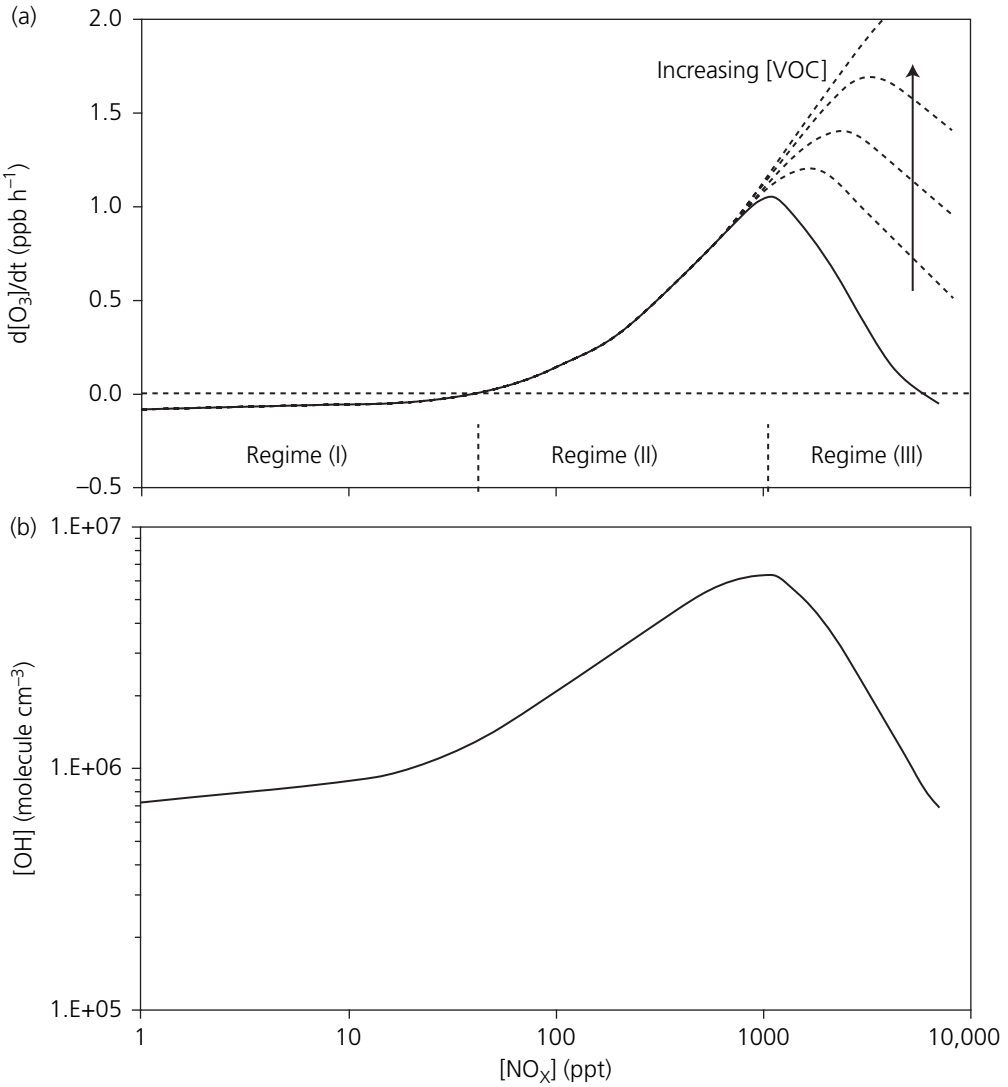
Figure 3.2 Schematic representation of O_3 production and loss processes during the free-radical mediated atmospheric oxidation of CH_4, and CO. The figure is divided into three regimes to illustrate the changing balance of the free-radical reactions as a function of NO_X level, with the dominant processes shown in black.



O_3 formation rate increases with increasing NO_X concentrations (see Figure 3.3, regime II). This is a direct consequence of the competition between reactions (8) and (5) for CH_3O_2, and reactions (9) and (6) for HO_2, such that increasing NO_X allows a greater number of free-radical propagated O_3 forming cycles to occur prior to termination.

In contrast, the O_3 formation rate is insensitive to changes in the concentration of CH_4 or CO and to inputs of other VOC. This is because OH reacts with the organics exclusively as part of the free-radical propagated O_3 forming cycles, with no competing radical termination reaction being available under the prevailing conditions.

Figure 3.3 An example illustration of the NO_X dependence of the O_3 production and loss rate, and the concentration of OH radicals, based on simple box modelling calculations appropriate to average mid-latitude conditions in the lower troposphere. The solid line in each panel corresponds to CH_4/CO oxidation. The broken curves in the upper panel illustrate the effect of additional nmVOC inputs (not based on calculation).



Continued increase of NO_X levels, however, brings the chemistry into the third regime (regime III: Figure 3.2), where reactions (8) and (9) dominate for CH_3O_2 and HO_2, but O_3 formation becomes inhibited by further increases in NO_X. This corresponds to the point at which the reaction of OH with NO_2 to form HNO_3 becomes the major termination reaction for free radicals:

$$OH + NO_2 + M \rightarrow HNO_3 + M \qquad (11)$$

Under these circumstances, increasing NO_X decreases the number of free-radical propagated O_3 forming cycles which can occur prior to termination via reaction (11), and the production rate of O_3 decreases. However, elevated emissions of CH_4 or CO allow the free-radical propagated O_3 forming cycles to compete more effectively with reaction (11), leading to an increase in the O_3 production rate. In addition to this, inputs of other more reactive nmVOC also compete for OH, leading to analogous O_3 forming cycles to that shown for CH_4 in Figure 3.2. Consequently, emissions of anthropogenic VOC (eg from road transport or solvent evaporation) and biogenic VOC (most notably isoprene) lead to a general increase in the formation rate of O_3 (as illustrated in the top panel of Figure 3.3). The prevailing conditions in regime III apply to locations which are comparatively close to pollution sources (eg the urban environment) and correspond to the conditions when intense O_3 formation rates occur for a limited period of time (high O_3 episodes). This occurs by virtue of inputs of the reactive VOC when NO_X levels are high, with such conditions often referred to as 'VOC-limited' or 'VOC-sensitive'.

The free-radical propagated oxidation of CH_4, as described above and illustrated in Figure 3.2, leads to the generation of the oxygenated product formaldehyde, HCHO. Its further

oxidation is partially initiated by photolysis, which contributes to radical generation, as follows:

$$HCHO + h\upsilon \rightarrow HCO + H \quad (12)$$

$$HCO + O_2 \rightarrow HO_2 + CO \quad (13)$$

$$H + O_2 + M \rightarrow HO_2 + M \quad (14)$$

As a result, the formation of HCHO (and analogous oxygenated products of nmVOC oxidation) has an impact on the rates of the oxidation cycles described above, through secondary radical generation. It should also be noted that HCHO oxidation generates CO, with the oxidation of CH_4 (via HCHO) being the major source of atmospheric CO.

3.3.1 Ozone atmospheric destruction by halogen-catalysed chemistry

The O_3 removal chemistry at low NO_X levels, described above leads to comparatively small O_3 loss rates. Because the chemistry is driven by O_3 photolysis (reaction (1)) the loss rate decreases as O_3 is depleted. This mechanism is therefore effectively self-inhibiting and cannot lead to severe O_3 depletion on a short timescale. However, over the marine environment a different response has been observed. This section describes recent research on halogen-catalysed O_3 destruction in marine areas, in part to bring the chemical processing section up to date but also to illustrate the fact that the understanding of some aspects of tropospheric O_3 chemistry is still developing.

Rapid O_3 depletion has been observed in numerous studies in the marine atmospheric boundary layer, particularly at high latitudes in the spring-time. This is attributed to chemical cycles involving halogen atoms and radicals, specifically the bromine (Br) and iodine (I) species.

The chemistry is initiated by the production of halogen atoms from the photolysis of both organic and inorganic bromine and iodine-containing species. These chemical species are either emitted from the oceans or released from surface reactions on sea-salt aerosol, frost flowers or other ice surfaces. The dominant tropospheric reaction for either Br or I atoms (denoted 'X') is with O_3 to form the corresponding halogen oxide radical, XO:

$$X + O_3 \rightarrow XO + O_2 \quad (15)$$

The XO radical can participate in a number of competitive reactions to propagate cycles which regenerate X atoms. Those cycles involving the reactions of XO with NO and NO_2 and (in the case of IO) direct photolysis, have no net chemistry, with the O_3 lost in reaction (15) being regenerated in a subsequent reaction step. However, if XO reacts with HO_2 or with itself, the subsequent chemistry can regenerate X atoms without regenerating O_3, therefore leading to net O_3 loss, for example as follows:

$$XO + HO_2 \rightarrow HOX + O_2 \quad (16)$$

$$HOX + h\upsilon \rightarrow OH + X \quad (17)$$

[net effect: $HO_2 + O_3 \rightarrow OH + 2O_2$ (15) + (16) + (17)]

$$XO + XO \rightarrow 2X + O_2 \quad (18)$$

[net effect: $2O_3 \rightarrow 3O_2$ 2x(15)+(18)]

The propensity of the system to maintain levels of the catalytic species, X and XO, in the gas phase is key to the sustained depletion of O_3. Alternative reactions, or reaction channels, do exist for species which participate in these O_3 depletion cycles. These lead to removal of the halogens from the gas phase. For example, the hypohalous acid, HOX (formed in reaction (17)), and molecular halogen species formed from alternative product channels of reaction (18) can transfer to the aerosol phase as part of a rich, multi-phase chemical system. This does not necessarily constitute irreversible loss of the active halogens, however, as surface and condensed phase reactions can re-release photolabile halogen species into the gas phase, allowing the reservoir of gaseous phase active halogens to be maintained. Considerable progress has been made in understanding the details of this complex, multi-phase chemistry, although it necessarily remains an active area of research. The global implications are still uncertain.

Perhaps the first demonstration of this reactivity was at Alert in the Arctic where a clear anti-correlation was seen between filterable Br and O_3 concentrations in the air, particularly in springtime in what are termed rapid surface O_3 depletion events (Barrie *et al.* 1988). Very recently Saiz-Lopez *et al.* (2007) have reported measurements of both bromine monoxide (BrO) radicals and Iodine monoxide (IO) radicals at a coastal Antarctic station, with a strong seasonal cycle, values being highest in spring and summer (the spring IO concentrations at 20 ppt are the highest ever reported in the atmosphere). However, the expected relationship with O_3 was not clear, possibly due to insufficient time for reaction to occur or due to entrainment of O_3-rich air from the free troposphere at the measurement site.

Atmospheric chemistry models have recently started to include halogen chemistry and to predict what O_3 depletions might be expected. For example, using a global model incorporating BrO chemistry, von Glasow *et al.* (2004) predict a reduction in O_3 concentration of up to 18% over widespread areas and regionally up to 40%, compared to model runs without bromine chemistry. Similarly Yang *et al.* (2005) show that inclusion of Br chemistry leads to an O_3 loss of approximately 5% in the Northern Hemisphere and up to 30% in the Southern Hemisphere. Incorporation of IO chemistry in models leads to a predicted destruction of up to 13% of available O_3 per day in marine air masses (McFiggans *et al.* 2000). Recent modelling by Saiz-Lopez *et al.* (2007) predicts that inclusion of both BrO and IO (at the high levels they observed in Antarctica) leads to a four-fold enhancement in O_3 depletion compared with the result from a model in which only BrO is considered. Finally, very recent observational data collected at the Cape Verde Atmospheric Observatory (16°N, 28°W) indicates that within the marine boundary layer significant O_3 is lost through an approximately equal combination of photolysis and halogen chemistry and that the region is a very large net sink of surface level tropospheric O_3 (Read *et al.* 2008). None of the models used for this report and described in chapter 5 incorporate any of the new halogen chemistry.

3.3.2 Ozone in the urban environment

At elevated NO_X levels, typical of the polluted urban environment, and at the upper end of the concentration range shown for regime III outlined above, O_3 levels can be severely depleted locally due to a direct reaction with emitted NO. This is known as the 'NO_X titration effect':

$$NO + O_3 \rightarrow NO_2 + O_2 \quad (19)$$

However, because O_3 is produced following the photolysis of NO_2 via reactions (4) and (10),

$$NO_2 + h\upsilon \rightarrow NO + O(^3P) \quad (4)$$

$$O(^3P) + O_2 + M \rightarrow O_3 + M \quad (10)$$

These reactions constitute a well-established chemical null-cycle which couples the chemistry of O_3, NO and NO_2 on a comparatively short timescale (eg Leighton 1961). Because of this strong chemical coupling, the term 'oxidant' is sometimes used as a collective term for NO_2 and O_3. This reaction cycle partitions NO_X between its component forms of NO and NO_2, and oxidant between its component forms of O_3 and NO_2, but conserves both NO_X and oxidant. As a result, oxidant derived from background O_3 entering the polluted urban area is partitioned between the forms of NO_2 and O_3, with a progressively greater proportion in the form of NO_2 as NO_X increases as a result of emissions.

Monitoring data from urban locations in the UK exhibit the general behaviour described above, and such data have been used to develop generic empirical expressions which describe the partitioning of oxidant into O_3 and NO_2 as a function of NO_X on an annual mean basis (Jenkin 2004). Figure 3.4 shows O_3 concentrations as a function of the NO_X and background O_3 concentrations, based on a generic partitioning expression which is broadly applicable to UK sites not in the immediate vicinity of roads. The background O_3 mixing ratio outside the urban area may be influenced by chemical processes on global and regional scales, as described above. Figure 3.4 provides a general illustration of how O_3 levels decrease with increasing NO_X levels in urban areas. This depletion of O_3 is known as the NO_X titration effect (sometimes referred to as an 'urban decrement' or 'urban deficit') but also demonstrates the potential impact of changes in background O_3 on the level of urban O_3. This empirical oxidant partitioning methodology can therefore be used to help map current urban levels of O_3, and to make predictions for the future. The oxidant partitioning model, in conjunction with NO_X emissions maps and projections, forms the basis of empirical modelling activities which assess current trends and future levels of NO_2 and O_3 in relation to UK air quality objectives, with the full modelling method referred to as a Pollution Climate Model (PCM). Chapter 5 (section 5.4) provides an illustration of future predictions for an urban centre location in London using a PCM.

Figure 3.5 shows how the PCM is able to provide a description of the variation of annual mean O_3 concentrations across the UK, for the example years 2003 and 2005, representing a warm, relatively dry and high O_3 year, and a more normal year respectively. The results clearly show the impact of NO_X-titration in reducing O_3 levels in urban areas (eg London), but also the impact of generally higher O_3 background levels in 2003, resulting from regional and global-scale effects.

Figure 3.4 Illustrative annual mean O_3 mixing ratios in UK urban areas as a function of annual mean mixing ratios of NO_X and background O_3, determined using an empirically based oxidant partitioning methodology.

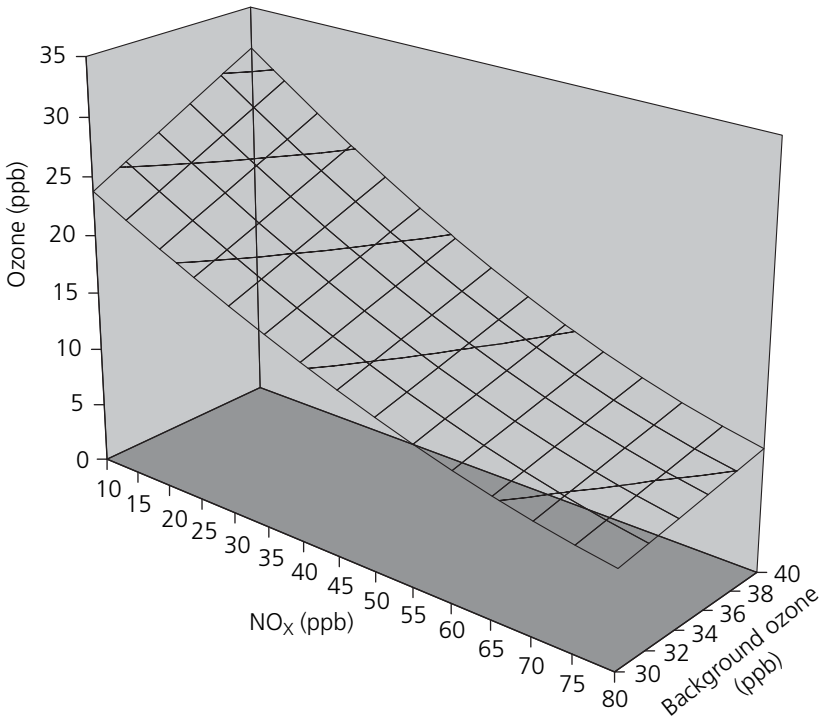
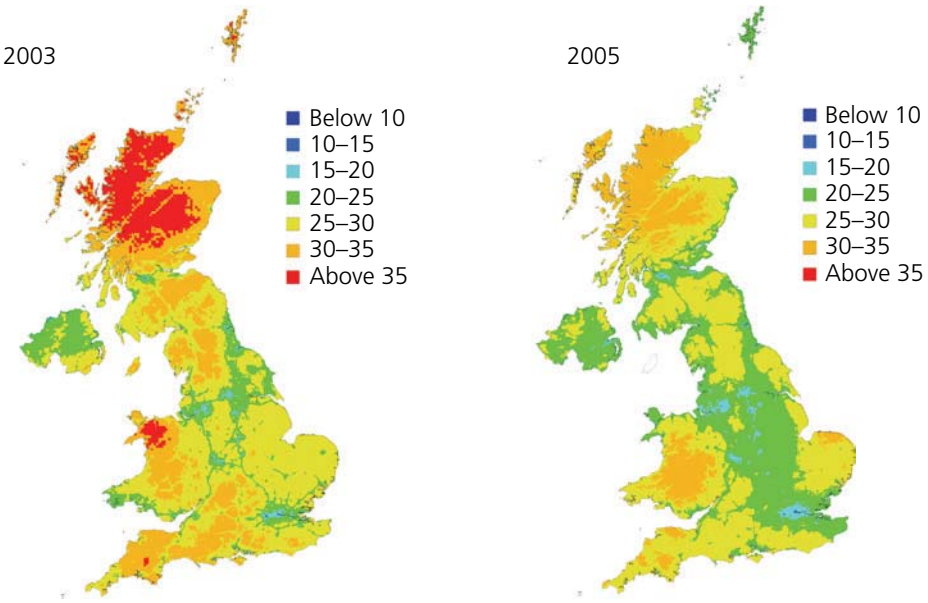


Figure 3.5 Maps of annual mean O_3 concentrations (ppb) in the UK, calculated using empirical measurements based on a Pollution Climate Model (PCM) for the years 2003 and 2005. Source: AQEG 2008.



The process of O_3 scavenging in the urban environment by titration with NO_X outlined here gradually declines as NO_X urban emissions are reduced by emission controls. Ozone concentrations in urban areas have increased as emissions of NO have declined. Further substantial increases in urban O_3 in the UK will take place as urban emissions of NO continue to decline. This is an important effect of control measures and will increase urban population exposure to O_3 in the coming decade.

3.4 Surface ozone destruction processes – dry deposition to the land and ocean

Ozone is a very reactive trace gas and is readily deposited to most surfaces including soils, vegetation and building materials. Deposition rates to water surfaces are much smaller as O_3 is not very soluble. The deposition of O_3 to terrestrial or marine surfaces is known as dry deposition (see Box 3.1 and Figure 3.6). This is the main removal process for boundary layer O_3 and limits the lifetime and maximum ground-level concentration. Rates of dry deposition to terrestrial surfaces are larger than those to ocean surfaces by typically an order of magnitude, but given that the oceans occupy approximately 70% of the Earth's surface, dry deposition to seawater is an important term in the global budget.

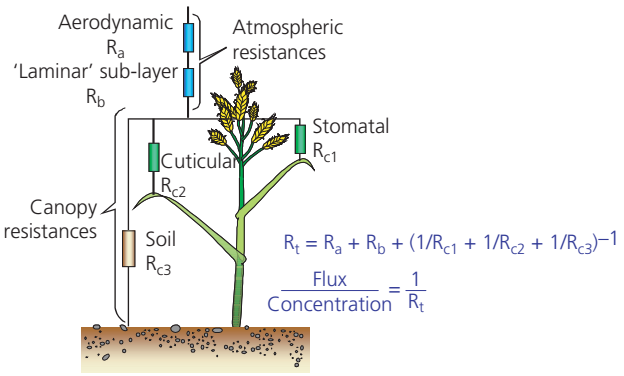
3.4.1 Surface ozone destruction – dry deposition to land

In the terrestrial environment dry deposition occurs as a result of reactions with the external surfaces of vegetation and soil and uptake by plant stomata (see Figure 3.6). Dry deposition rates range from 2 to 15 mm s^{-1} and are mainly regulated by opening and closing of stomata, wind speed and turbulence and the presence of surface water on vegetation. The aperture of the stomatal pores on the leaf surface is governed by plants in response to sunlight, temperature, humidity, soil moisture and CO_2 concentration. In drought conditions,

Box 3.1 Dry deposition

Rates of dry deposition are commonly expressed as a deposition velocity (vg) given as the (vertical flux) (ambient concentration) with dimensions of a velocity (mm s^{-1}). As ambient concentrations decline towards the surface, deposition velocity also varies with height. Rates of dry deposition are commonly parameterised using a simple resistance analogy to quantify the components in the deposition pathway from the boundary layer to sinks at the surface, as illustrated in Figure 3.6.

Figure 3.6 A simple resistance analogy for the dry deposition of O_3 to terrestrial surfaces.



the closure of stomata to protect plants against desiccation reduces stomatal conductance and reduces dry deposition of O_3. The smaller deposition rates under drought conditions are an important factor in enhancing ground-level O_3 concentrations. The stomatal flux to sites of damage within the leaf represents the main pathway leading to physiological damage to vegetation. As discussed in more detail in chapter 8, cumulative stomatal flux of O_3 above a threshold provides the most accurate basis for assessing the risk of O_3 damage to vegetation.

3.4.2 Surface ozone destruction – dry deposition to oceans

The rate of uptake of atmospheric O_3 by ocean surfaces is normally parameterised in models using a fixed deposition velocity that is independent of physical drivers such as wind speed or chemical reactions of O_3 with components at or close to the sea surface. This is however, simplistic and could result in errors in the estimation of the role of the oceans in removing O_3 from the atmosphere (Ganzeveld & Lelieveld 1995).

Deposition velocity is influenced by wind as water-side turbulence controls the exchange rate. This makes parameterisation of O_3 uptake with wind similar to that for CO_2, dimethyl sulphide and many other weakly soluble gases. However, parameterisation of the deposition velocity for these gases has uncertainties, particularly at high wind speeds where the role of bubbles is poorly understood. This leads to an uncertainty of about a factor of two in estimating the transfer velocity (Nightingale & Liss 2003).

The situation with incorporating chemical reactivity of O_3 with sea surface components introduces further and more substantial uncertainty. The two substances most likely to react rapidly with O_3 at the sea surface are iodide ions (Garland *et al.* 1980; Chang *et al.* 2004) and dissolved organic matter (McKay *et al.* 1992), however, the rate coefficient for O_3 reacting with both are poorly known. Notwithstanding this uncertainty, limited field measurements of O_3 uptake by seawater indicate that chemical reactivity appears to increase the rate of uptake by approximately a factor of 20 (Liss 1983).

3.5 Effects of weather and topography on ground-level ozone

Ozone production varies diurnally, seasonally and interannually. As the chemistry of O_3 formation requires photolysis of a range of species, and the rate of some of the reactions increase with temperature, O_3 production is generally at its maximum during warmer sunny weather. An example of the daily variability in ground-level O_3 is shown in Figure 3.7, which shows the maximum O_3 concentrations each day at a rural site in Southern England for 1995. The measurements show an annual cycle in O_3 with a spring to early summer peak at about 40 ppb and an autumn minimum at about 25 ppb. Superimposed on the annual cycle are substantial variations from day to day, with peaks of up to 200 ppb on sunny warm days and minima on cold calm winter days when dry deposition to the surface exceeds the supply of O_3 from aloft.

The strong coupling between rates of dry deposition and stomatal conductance also links weather to O_3 removal from the boundary layer. In hot, sunny conditions, production of O_3 approaches a maximum and if soils are dry, the soil water deficit leads to closure of plant stomata leading to larger surface O_3 concentrations. The very strong effect of weather on O_3 production and loss creates much of the observed

Figure 3.7 A year of surface O_3 concentration measurements in Southern England showing maximum hourly values for each day. The peak values occur during spring and summer months. An indication of the sources of observed O_3 is also shown in the figure.

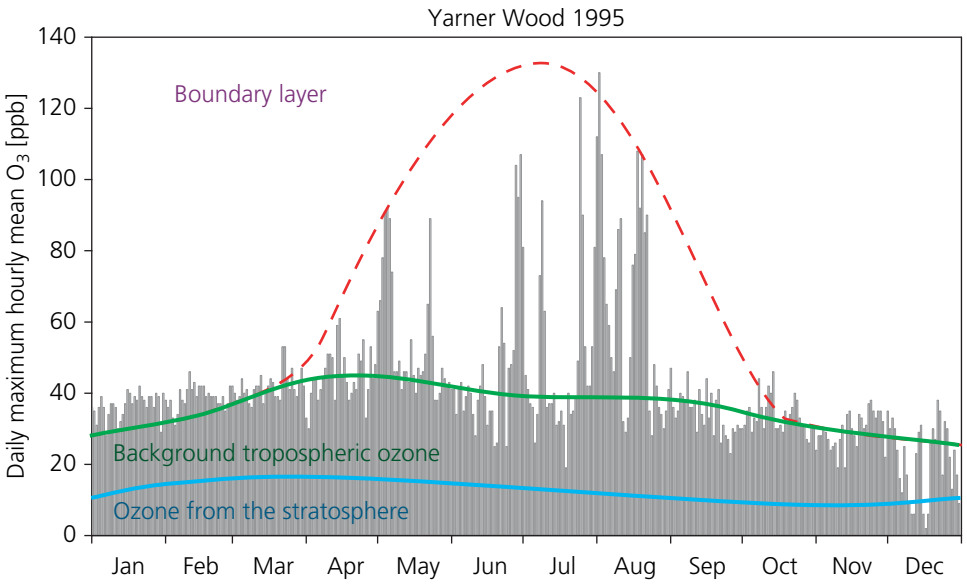
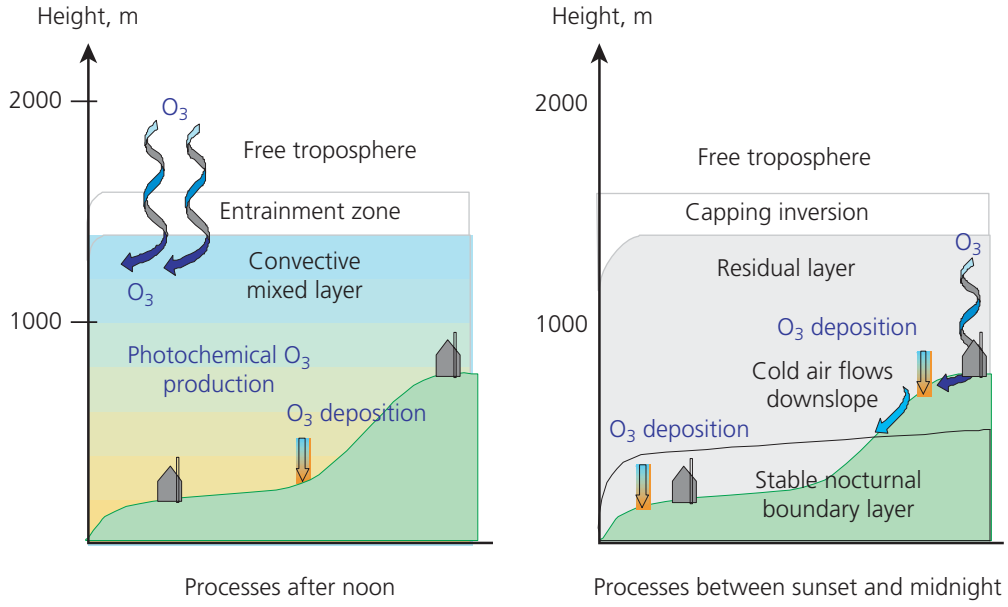


Figure 3.8 An illustration of the processes regulating the sources and sinks of boundary layer O_3 at rural locations.

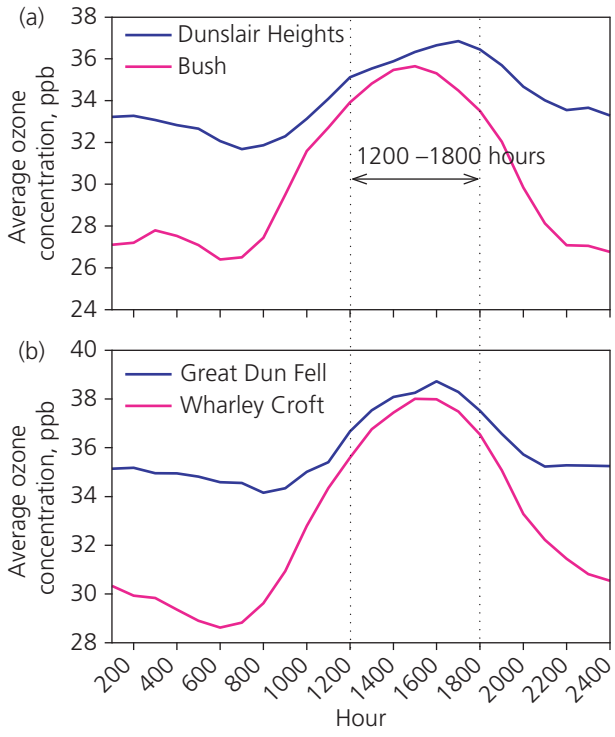


variability in concentrations with time. Over longer time scales, changes in climate due to greenhouse gas emissions are likely to influence the production and loss processes for O_3, as described in chapter 6.

The concentrations of O_3 at any specific location depend on topography and meteorology as well as background and regional sources of ozone O_3. Rural areas, hill tops and coastal areas experience larger exposure to O_3 because at these locations the supply of O_3 from higher levels in the boundary layer exceeds the rate of depletion by dry deposition. By contrast in sheltered valleys the loss by dry deposition, especially at night, often leads to a sharp decline in O_3 concentrations, in this case dry deposition is depleting O_3 faster than transport from above. It is the balance between vertical transport and surface deposition that causes the diurnal cycle of O_3 in rural areas, with small concentrations at night and large values in the daytime. The magnitude of the diurnal cycle and close link between weather and surface O_3 are illustrated in Figures 3.8 and 3.9 using monitoring data from hill top locations (Dunslair Heights and Great Dun Fell) and neighbouring locations on lower ground (Bush and Wharley Croft respectively) to show the effects of altitude on vertical mixing and transport.

The lower O_3 concentrations in urban areas are mainly due to reaction with NO, as described earlier in this chapter but of course all processes operate simultaneously and dry deposition contributes to the decline in urban O_3 concentrations at night. Wind direction is very important in determining the concentrations of O_3 precursors within a region. As an example, to the west of the UK, the Atlantic ocean represents a source of fairly uniform background O_3 and with the exception of shipping, is devoid of major sources of O_3 precursors. The breezy and often cloudy conditions associated with westerly airflow limits the magnitude of O_3 concentrations over the country in these conditions. Therefore with westerly winds over the UK, O_3 levels are generally close to the background. In contrast, easterly winds bring continental air with O_3 precursors, generally at lower wind speeds and often in sunny weather which is associated with the anticyclonic conditions. Thus the peak O_3 values illustrated in Figure 3.7 generally occur in sunny, anticyclonic weather.

Figure 3.9 The diurnal cycle in ground-level O_3 at rural sites in the UK illustrating the much larger range in the diurnal cycle at (a) low altitude and valley sites (Bush and Wharley Croft) relative to (b) hill top locations (Dunslair and Great Dun Fell).



The close link between warm sunny anticyclonic weather and episodes of elevated O_3 illustrated in Figure 3.7 is an important contributor to differences between years, and the number of O_3 episodes. Any change in the frequency of weather suitable for O_3 episodes will influence the O_3 experienced. The effects of climate change on ground-level O_3 are discussed in chapter 6.

3.6 Trends in ground-level ozone

Ground-level O_3 is currently measured throughout rural and urban areas of most European countries and in North America. Long-term records are available at a few locations from the 1970s and 1980s when most networks began. In addition, interest in atmospheric O_3 in the early 20th century provided some European ambient O_3 measurements by Schonbein (1844 cited by Staehelin & Schnadt Poberaj 2008). Others interested in these early measurements provided a substantial dataset late in the 19th century and early in the 20th century. By analysing the methodology and comparing it with more modern techniques, Volz & Kley (1988) were able to show that rural O_3 concentrations had doubled from about 10–15 ppb in rural Europe between the end of the 19th century to 20–30 ppb in the 1980s. Since the 1980s, rural O_3 concentrations have increased in many areas (Staehelin & Schnadt Poberaj 2008) with quite different rates of change at different locations.

The trend at mountain sites in the Alps (Figure 3.10) shows an upward trend of between 0.2 and 0.5 ppb y^{-1} over the period 1991–2002, in part due to increased stratospheric–tropospheric exchange (Ordóñez 2006). Trends at 13 rural sites in the UK show increases averaging 0.14 ppb y^{-1} over the period 1990–2006 (Jenkin 2008). Measurements of the vertical profile in O_3 concentration at European sonde stations show an increase in concentration in the range 5–25% per decade over the period 1970–1996 (Logan *et al.* 1999).

At Mace Head on the Atlantic coast of Ireland it is possible to examine the trends in the hemispheric background of ground-level O_3.

The data presented in Figure 3.11 reveals an increase over the monitoring period (1987–2007) at Mace Head. Ozone production varies significantly between years due to the varying rate of production and destruction, but also shows a distinct seasonal cycle, with a summer maximum as shown in Figure 3.7. The measurements from this station show an increase in the hemispheric baseline O_3 of about 5 ppb over the period, or an average annual increase of about 0.3 ppb per year, over the 20 years of data available, similar in magnitude to the trend at the mountain sites. The data also show very large intra- and inter-year variability, with peak to peak variations of about 15 ppb, caused mainly by variability in sources of O_3 precursors and weather upwind of the monitoring station. In this case having selected the westerly uncontaminated sector, eliminating air from European sources for air arriving at the site, the footprint is very large, extending westwards through a substantial fraction of the northern mid-latitudes. The data at Mace Head provide a measure of the temporal variability in the hemispheric background (causes of this variability are considered in detail by Derwent *et al.* (2008)). The longer term changes in hemispheric background O_3 are also revealed by ship cruises in the Atlantic over the period 1978–2004. These data show increases in surface O_3, with the largest increases being in the latitude range 20–40° N (Lelieveld *et al.* 2004). These examples of long-term surface O_3 concentrations show that the hemispheric background has grown substantially during the last 100 years or so. The direct

Figure 3.10 Time series of seasonal mean O_3 concentrations at three European mountain locations, Jungfraujoch, Zugspitz and Sonnblick. Source: Ordóñez 2006.

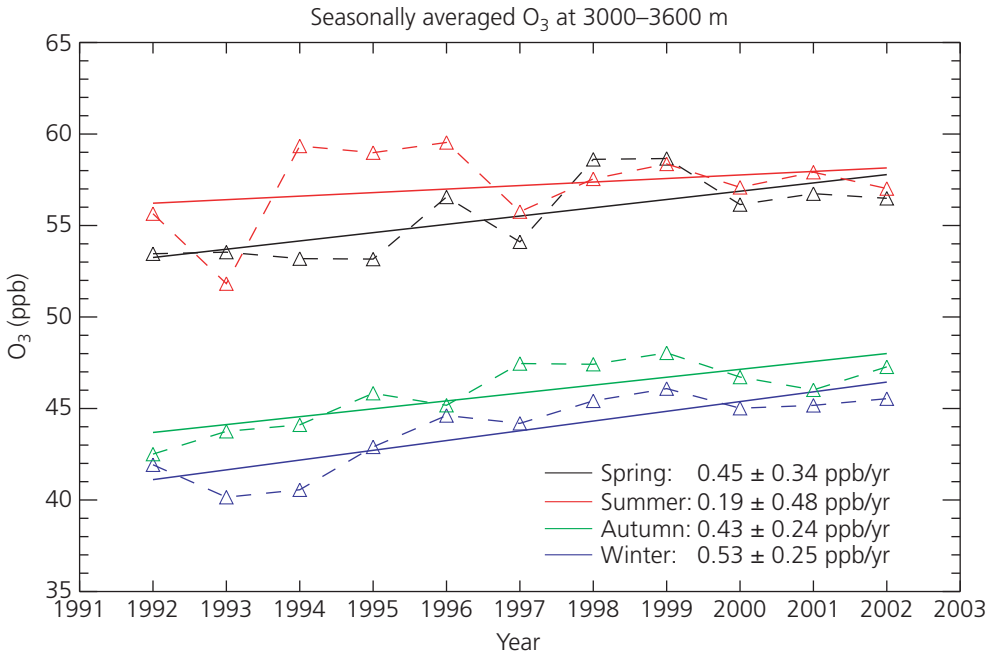
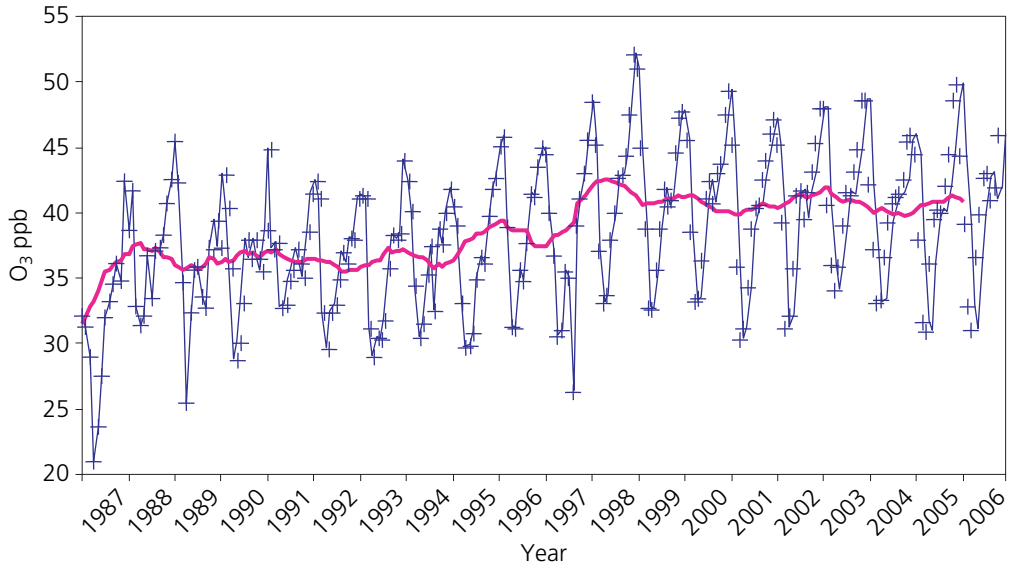


Figure 3.11 Trends in the monthly mean and 12-month running mean (solid line) baseline O_3 concentration from April 1987 to April 2007 at Mace Head, Ireland. Source: Derwent et al. 2008.



measurements show that the hemispheric background in the northern mid-latitudes approximately doubled from 10–15 to 20–30 ppb between 1900 and 1980 and since then has grown by a further 5 ppb. During the period 1980–2007 many countries have reduced emissions of O_3 precursor gases and in response the peak concentrations have declined, by typically 30 ppb in Europe, and in very polluted areas of North America, such as California, the decline in peak O_3 concentrations has been larger, as noted in chapter 2. The steady increase in hemispheric background O_3 during the last few decades has eroded the benefits of the emission reductions as average concentrations of O_3 have in many areas increased to values large enough to represent a threat to human health and the health of ecosystems, as described in chapters 7 and 8.

3.7 Conclusions

The chemical and physical processes of O_3 production and destruction are reasonably well understood and show great differences between polluted urban areas and remote clean air regions of the planet. The relatively long lifetime of O_3 and some of its precursors in the troposphere allows O_3 and O_3 precursors to be transported around bands of latitude making O_3 a hemispheric pollutant. In remote areas it is this background O_3 that provides the O_3 exposure at the surface. At the regional scale, if there are substantial sources of O_3 precursors within the region, then both the hemispheric background and the regional sources contribute. At the local scale if there are substantial sources or sinks present, then O_3 concentrations are regulated by the hemispheric background, regional production and the effects of local sources and sinks acting collectively. Chemical production (or destruction) of O_3 within the atmosphere is regulated mainly by the concentration of NO_X. At very low concentrations of NO_X (<20 ppt) O_3 is destroyed, and this is the main cause of the very low O_3 concentrations over remote oceans, although recent work shows that reactions of O_3 with halogen oxides is an important sink in some marine areas. In contrast to these remote areas, over the polluted continents, in Europe, North America and large parts of East and South Asia, Africa and South America, there is sufficient NO_X to produce O_3. In addition to chemical sinks in the atmosphere, O_3 is deposited at the Earth's surface onto soils, vegetation and water. This is the pathway leading to effects on crops and natural vegetation. Deposition exerts strong controls on ground-level O_3 such that during drought conditions, when deposition is reduced, ground-level O_3 concentrations rise appreciably.

The hemispheric background O_3 concentration in many areas is thought to have roughly doubled between 1900 and 1980 and has risen further since 1980. Rural O_3 concentrations have increased in many areas at different rates. In Northern Hemisphere mid-latitudes, where the majority of global population and food production occurs, the increase in background O_3 since 1980 was of the order of 5 ppb. The cause of the increase appears mainly to be an increase in net chemical production in the troposphere, although increases in the stratospheric source of O_3 may also have contributed.

While NO_2 is a key compound in O_3 formation, VOC are also essential reactants and in some highly polluted areas the chemistry of O_3 formation is VOC limited. VOC emission controls can therefore play an important role in O_3 reduction strategies. In the urban environment there are typically high NO_X concentrations leading to local O_3 depletion by titration. Reductions in NO_X concentrations in these areas lead to local increases in urban O_3 concentrations. Most urban areas have shown a pronounced increase in O_3 during the period in which NO_X controls have taken effect, in the UK this is clear through the period 1990–2007. Further increases in urban O_3 will occur in response to further reductions in urban NO_X concentrations.

In the following chapter projected future changes in natural and anthropogenic emissions of NO_X, VOC and CH_4 are described, followed by an analysis of changes in future O_3.

4 Natural and anthropogenic emissions of ozone precursor compounds

4.1 Introduction

To inform an assessment of how future O_3 concentrations may change in the future it is first necessary to review the sources of O_3 precursors, and to evaluate how these may change through the coming century. In this chapter the sources of emissions are briefly introduced. The important drivers of changes in anthropogenic emissions are reviewed, and the current state of knowledge regarding natural emissions is presented. Three new emission scenarios are used to inform an analysis of how anthropogenic emissions may change over the period to 2100, and a new global estimate for isoprene, one of the better-studied naturally occurring VOC species, is presented.

4.2 Sources of ozone precursors

The O_3 precursor gases NO_X, CO, CH_4, and nmVOC are emitted from a wide variety of anthropogenic (eg transport, solvents, fossil fuels) and natural (eg forests, wetlands, soils, lightning) sources.

Globally, anthropogenic sources of NO_X are estimated to produce 33 Tg y^{-1}[7] (as N) of emissions (Cofala *et al*. 2007b). Primary sources include fossil fuel combustion from stationary sources such as power generation, and mobile sources such as road transport. In 2000, road transport accounted for 33% of anthropogenic NO_X emissions, international marine shipping 19%, and energy combustion in power plants 17% (Cofala *et al*. 2007b). Deforestation, savannah burning and the burning of agricultural waste account for half of the global anthropogenic emissions from CO. Fuel combustion in the domestic sector and road transport account for the remaining emissions. The majority of CH_4 emissions are anthropogenic including coal mining, the coal and gas industry, landfill, ruminant animals, rice agriculture and biomass burning. Each of these activities contribute between 30 Tg y^{-1} and 90 Tg y^{-1}. Global anthropogenic nmVOC emissions were recorded for 2000 at 140 Tg y^{-1} (eg Olivier *et al*. 2005) however these estimates are highly uncertain.

The relative contributions of anthropogenic emission sources differ across countries, due to differences in the levels of economic development and the extent to which emissions controls have been applied. Figure 4.1 compares O_3 precursor emissions on a *per-capita* basis for the world, the EU-27 and the UK.

For NO_X, road transport constitutes the dominating share of emissions in the UK, but makes a much smaller contribution at the global level. Similarly, VOC emissions from solvents use are an important source in Europe, but less so at the global scale. For CO, the largest global contributions are from biomass burning in the domestic sector, while in the UK road transport is the largest contributor. It is important to mention that the spatial densities of O_3 precursor emissions show large spatial gradients, which in Europe are mainly driven by differences in population densities. For instance, spatial emission densities in the north-west of Europe are particularly high (Figures 4.2 and 4.3).

NO_X emissions are also produced naturally by bacterial and volcanic activity and lightning. Global estimates are highly uncertain, and range from 10 to 60 Tg N y^{-1} depending on which sources are included. Soil NO_X emissions estimates range from 7 to 20 Tg NO–N y^{-1} and are highly uncertain (Davidson *et al*. 1997; Yienger & Levy 1995). One of the most uncertain natural sources of NO_X is lightning with emissions in the range of 2–20 Tg N y^{-1} (RETRO 2007; Schumann & Huntrieser 2007). Open burning of biomass from forest fires, savannah burning and agricultural practices account for a further 20 Tg N y^{-1}. The primary natural sources of CO are vegetation, oceans (Olivier *et al.* 2005) and wild fires (biomass burning). The single largest source of CH_4 is wetlands, with an emission rate estimated to range from 100 Tg y^{-1} to 230 Tg y^{-1}. Globally, natural sources of nmVOC outweigh anthropogenic sources. A wide range of species of biogenic nmVOC are emitted to the atmosphere by many terrestrial plant species. The single most important nmVOC for O_3 production is thought to be isoprene (C_5H_8: 2-methyl-1, 3-butadiene), due to its very large global emission rate (between 500 and 750 Tg y^{-1}) and its reactivity with OH.

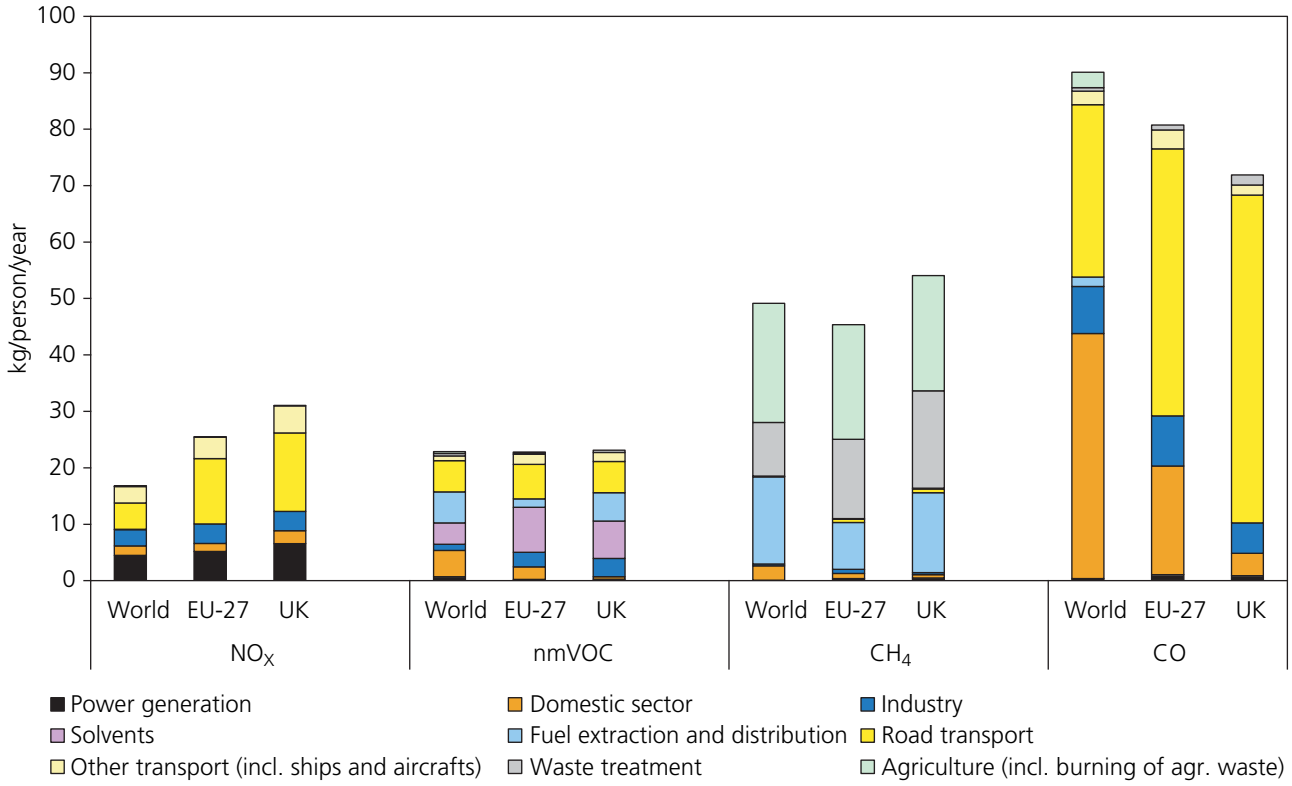
4.3 Future emissions analysis

An evaluation of future atmospheric O_3 concentrations requires knowledge of the sources and sinks of both natural and anthropogenic emissions and the likely patterns of future emissions. Ozone precursor emissions are expected to change significantly as a result of population growth, economic development, technological progress and uptake, control measures, land use change, climate and other environmental changes.

Scenarios are often used to analyse how different drivers may affect future emission rates and to assess the associated uncertainties. These are usually based on a mixture of quantitative information and expert judgment, and aim at producing an internally coherent picture of how the future could develop for a given set of explicit assumptions. Scenarios do not attempt to predict the future, since in many cases assumptions are associated with significant uncertainties that cannot be further resolved. However scenarios present a possible range of future developments constructed for a plausible range of assumptions. In terms of O_3, scenarios help to assess the relative importance of different drivers and illustrate possible changes over time. For the purposes of this

7 Emissions are normally presented as emission rates per year (in terragrams (Tg y^{-1}, (1 Tg = 10^{12} g)) to represent the yearly contribution of the emission source to the ozone budget (discussed in chapter 3).

Figure 4.1 Per-capita emissions of the O_3 precursors (NO_X, nmVOC, CH_4, CO) in the year 2000, for the world, the EU-27 and the UK.



study three scenarios have been developed to enable an analysis of the most important drivers of changes in emissions. This analysis has necessarily been restricted to consideration of anthropogenic emissions as it is not yet possible to quantify natural emission rates in the future. Section 4.3.2 outlines the main factors that are expected to be important for natural emissions, and provides a detailed analysis of current and future estimate rates for the best understood of the nmVOC; isoprene.

4.3.1 Anthropogenic emissions

The decisive factor determining the level of future anthropogenic emissions is the extent to which control measures will be implemented. The new scenarios developed for this study explore how the full implementation of legislation in place around the world in late 2006 will impact on future O_3 concentrations.

Figure 4.2 The spatially disaggregated anthropogenic emissions of nmVOC in Europe for the year 2000 including land-based and shipping emissions. Source: EMEP/Meteorological Synthesising Centre-West.

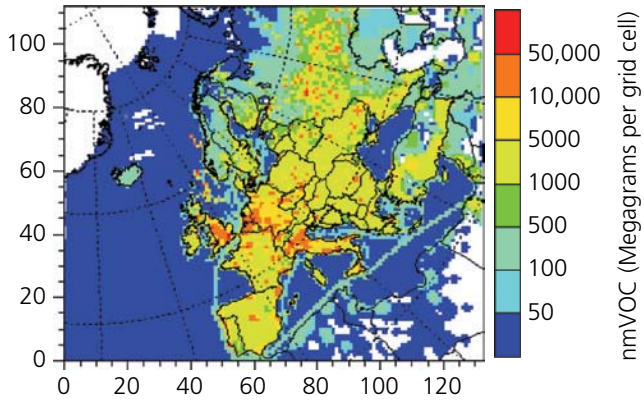


Figure 4.3 The spatially disaggregated anthropogenic NO_X emissions in Europe for the year 2000 including land-based and shipping emissions. Source: EMEP/Meteorological Synthesising Centre-West.

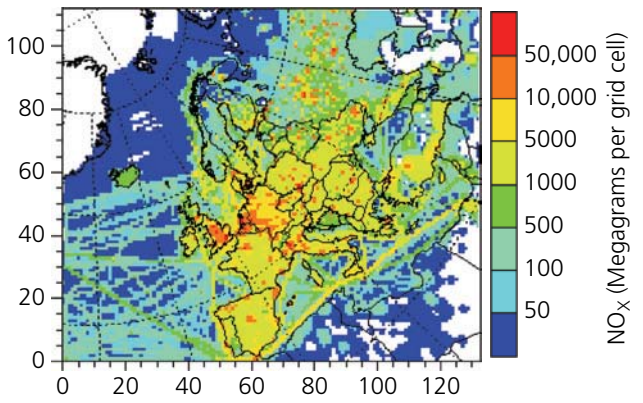


Table 4.1 Estimates of global natural and anthropogenic O_3 precursor emissions.

Estimates		
	Natural emissions	Anthropogenic emissions
CH_4	100–230 Tg y^{-1} wetlands 10 Tg y^{-1} oceans 5 Tg y^{-1} freshwater 5 Tg y^{-1} CH_4 hydrates 20 Tg y^{-1} termites	299 Tg y^{-1}
nmVOC (all)	–	139 Tg y^{-1}
nmVOC (isoprene only)	~500–600 Tg y^{-1}	–
NO_X soil	7–20 Tg NO–N y^{-1} soil	(all sources) 31 Tg NO_X–N y^{-1}
NO_X lightning	2–20 Tg N y^{-1} lightning	
NO_X from vegetation	20 Tg N y^{-1} – forest fires, savannah burning and agricultural practices	
CO	Negligible	549 Tg y^{-1}

Sources: Guenther *et al*. 1995; Yienger & Levy 1995; Davidson *et al*. 1997; IPCC 2001; EDGAR-FT2000: Olivier JGJ, Van Aardenne JA, Dentener F, Ganzeveld L and Peters JAHW (2005) (available online at *http://www.mnp.nl/edgar/*); Guenther *et al*. 2006; Lathière *et al*. 2006; Duncan *et al*. 2007; Schumann & Huntrieser 2007; Lathière, Guenther, Beerling & Hewitt (unpublished).

While numerous analyses of future emissions are available for individual countries, there are very few studies that explore the global development of air pollutant emissions for the coming decades. For O_3 precursor gases, two global scenarios have been developed by IIASA. These assess first the maximum feasible reductions (MFR) possible through the uptake of the most advanced technologies available, and secondly the effect of O_3 relevant legislation in place globally in 2002 (CLE) (Dentener *et al.* 2005; Cofala 2007b). These bottom-up scenarios are based on assumed government projections of economic development and extend only to 2030 (see Box 4.1).

Even fewer studies have investigated how emissions of air pollutants may change over the long term (100 years). The set of global scenarios for the 21st century developed by the IPCC (Nakicenovic *et al.* 2000) focus on greenhouse gas emissions to explore the long-term dynamics of the climate system. They pay little attention to the O_3 precursor gases such as NO_X and CO, although CH_4 is included. These scenarios were recently updated by Riahi *et al.* (2006) to include assumptions about current legislation to reduce greenhouse gas emissions (but exclude NO_X or CO) (see Box 4.2).

Box 4.1 IIASA air pollution scenarios to 2030

IIASA has developed two scenarios of ozone precursor emissions up to 2030 (Cofala 2007b). These scenarios assume realisation of the national expectations on economic development and implementation of the current national energy policies. They explore the implications of different assumptions on the application of emission control measures.

Maximum Feasible Reduction (MFR)

The MFR scenario explores the consequences if the most advanced technologies currently available were applied to control pollutant emissions. As the application of these technologies would involve some cost, the MFR is not expected to be the most realistic scenario; it is an example of what could be achieved under a best case scenario. It does not take account of practical limitations or the costs of new technologies or policies, or of the benefits of non-technical measures.

Current Legislation (CLE)

The CLE scenario assumes that current national legislation to control pollutant emissions (as of the end of 2002) will be enacted and adhered to. It does not however make any assumptions about what additional controls may be put in place. The CLE scenario is put forward as the most realistic estimate of future emissions change.

Box 4.2 IPCC long-term scenarios for changes in emissions over the 21st century

The IPCC, in its Special Report on Emission Scenarios (SRES) (Nakicenovic *et al*. 2000) developed four different narrative storylines to 2100 that describe the relationships between very complex dynamic systems as determined by driving forces such as demographic development, socio-economic development, and technological change. Three of these storylines (A2, B1 and B2) were recently updated by Riahi *et al*. (2006). These portray baseline developments without any greenhouse gas emissions constraints. Collectively these scenarios provide an indication of the possible range of future development, but do not include any assumptions about future emissions controls.

A1 Scenario

The A1 scenario describes a future world of very rapid economic growth, global population that peaks in mid-century and declines thereafter, and the rapid introduction of new and more efficient technologies. The scenario develops into three groups that describe alternative directions of technological change in the energy system (Fossil intensive (A1FI), non fossil energy sources (A1T), or a balance across all sources (A1B).

A2 Scenario

The revised high emission A2 scenario describes a very heterogeneous world with slow convergence of fertility patterns across regions resulting in a continuously increasing global population. Economic development is primarily regionally oriented and per-capita economic growth and technological change is more fragmented and slower than in other scenarios. The more limited rates of technological change that result from the slower rates of both productivity and economic growth translate into lower improvements in resource efficiency across all sectors. Energy supply is increasingly focused on low grade, regionally available resources (ie primarily coal), with post-fossil technologies (eg nuclear) only introduced in regions poorly endowed with resources.

B1 Scenario

The revised B1 scenario describes a convergent world with low global population growth that peaks mid-century and declines thereafter to some 7 billion people by 2100. It assumes rapid changes in economic structures towards a service and information economy, with reduction in material intensity and the introduction of clean and resource efficient technologies. The emphasis is on global solutions to economic, social and environmental sustainability, including improved equity. It is assumed that per capita GDP growth is the highest of the scenarios analysed.

B2 Scenario

The revised B2 scenario anticipates a world in which emphasis is placed on local solutions to economic, social and environmental sustainability. It is a world with continuously increasing population at a moderate rate, intermediate levels of economic development and diverse technological change. The B2 scenario is characterised by 'dynamics as usual' rates of change. This scenario sits between the A2 and B1 scenarios.

New emission scenarios

For the purposes of this study Amann *et al.* (in prep) have developed three new scenarios to evaluate possible changes in anthropogenic O_3 precursor emissions to the end of the 21st century. These are based on the socioeconomic pathways of the revised IPCC A2, B2 and B1 scenarios (Riahi *et al.* 2006) and the IIASA CLE (2030) scenario which was updated to include legislation adopted between 2002 and late 2006 (Dentener *et al.* 2005; Cofala 2007b) and extended to 2100. These new scenarios assess the future emissions of NO_X, CO and CH_4 from the human activities considered in Riahi *et al.* 2006. As there is very little information available for current rates of nmVOC emissions from anthropogenic sources at the global scale, and even less information about how these may change in the future, nmVOC emissions projections were not included in this analysis.

The three new scenarios are as follows:

IPCC SRES (Riahi 2006) B1 + IIASA CLE

IPCC SRES (Riahi 2006) B2 + IIASA CLE

IPCC SRES (Riahi 2006) A2 + IIASA CLE

The objective of this work was to gain insight into the possible evolution of O_3 precursor emissions by comparing the results of the three new scenarios. In these scenarios, assumptions are made regarding global population growth, economic development, energy consumption, technology development, emissions control measures and land use change. There are significant uncertainties about how the global economy will develop over the next century in quantitative terms. However, the available literature on long-term scenarios consistently points to strong improvements in the economic wealth of an

increasing global population, which will boost levels of economic activities.

Population growth

Population size determines the demand for economic services and other anthropogenic activities that give rise to O_3 precursor emissions. Over the next century densities and spatial patterns of populations are expected to change, for example, in developing countries increased urbanisation is predicted (UNFPA 2007).

The scenarios presented in Riahi *et al.* (2006) share the assumption that global population will grow by 45–70% from 2000 to 2050. For industrialised countries they predict an increase of less than 20% up to 2050 while in developing countries the population is expected to grow by 50–80%. For 2100, these scenarios show large variations ranging from a decline to seven billion people to an increase to 12 billion people (+100%).

Economic development

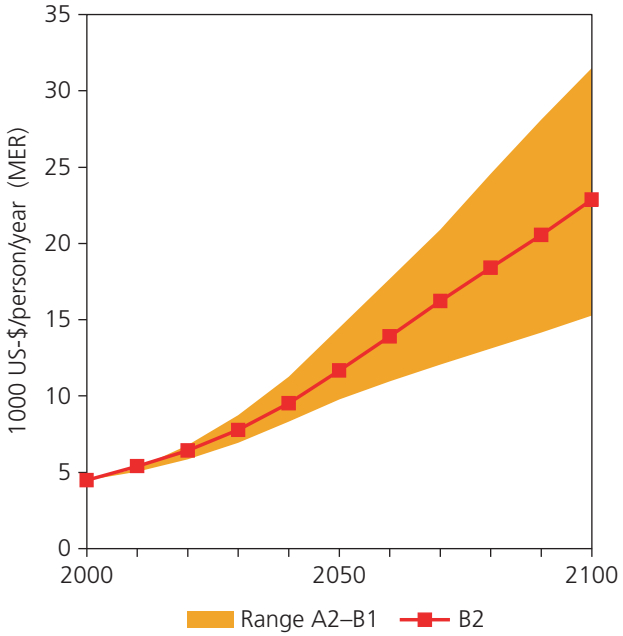
Economic development is also a major driver of trends in anthropogenic emissions although the relationship between development and population growth is not always linear. It has been observed that there is a certain threshold in economic development beyond which air pollution problems are ameliorated. This is because some of the most polluting economic activities are associated with industrial development. With economic development many of these may be replaced by other more advanced, less polluting activities, such as information technology.

The scenarios developed by Riahi *et al.* assume that personal economic wealth could increase globally in 2050 by 200–350% depending on the assumptions that are made within the scenarios. By 2100, per-capita incomes may grow between a factor of three to ten. Depending on the assumptions on global cooperation, different regions may experience different growth rates, with the possibility that by 2100 all world regions could reach the current European level of economic wealth. Together, these factors lead up to 2050 to an annual growth in the global volume of economic activities (measured in GDP) of between 2.2 and 3.2% per year, with faster growth in the beginning of the period. In 2050, GDP may be three to five times higher than now, and in 2100 seven to twelve times higher (see Figure 4.4).

Energy consumption

Historically, energy combustion has been the major source of anthropogenic O_3 precursor emissions, both in industrialised and developing countries (Cofala *et al.* 2007b). However, energy systems are continuously transforming over time. Many of these transformations occur 'autonomously' as a feature of technological progress and as a consequence of the shift in economies from focusing on energy-intensive

Figure 4.4 Range of per-capita income of the revised SRES scenarios (measured as GDP per capita in market exchange rates) (see Box 4.2 for scenarios). Source: Riahi et al. (2006). Reprinted with permission from Elsevier.



basic material industries towards new products with less material content. Modern societies have been successful at partly decoupling economic growth from energy consumption. For example, since 1990, each 1% growth in GDP has been accompanied by a 0.5% increase in primary energy consumption (International Energy Agency (IEA) 2006). The scenarios presented in Riahi *et al.* (2006) assume that this decoupling between GDP and energy use will continue at rates between 0.5 and 1.5% per year. However decoupling is unlikely to fully compensate for future economic growth and the main human activities that lead to emissions, eg power generation, transportation, industrial production and agricultural activities are likely to intensify in the coming decades (IEA 2006; Riahi *et al.* 2006). The Riahi *et al.* scenarios suggest an increase in primary energy by 130–180% by 2050, and by 160–330% for 2100 compared to 2000. This is slightly less than the 1.6% increase per year that is projected in the Reference scenario of the World Energy Outlook of the IEA up to 2030 (IEA 2006).

The three Riahi *et al.* scenarios and the recent IEA projection agree on strong growth in the demand for land and sea transport fuels with increases ranging between 150 and 250% in 2050 and up to a factor of five for the end of the century. Currently, international shipping is one of the fastest growing sectors of the global economy, expanding at roughly two times the rate of GDP (Eyring *et al.* 2005b). For the future, our new scenarios follow the assumptions in Eyring *et al.* (2005b), which suggest that under the economic growth pathway in the IPCC SRES B2 scenario, total fuel consumption for ships will almost triple between 2000 and 2050.

Technology development

Technological development will also influence the emissions resulting from a given human activity. All scenario work suggests that the autonomous structural changes in energy consumption (ie switching to cleaner fuels, reducing energy consumption through increased efficiency, etc) will not be sufficient to reduce present air pollution to levels that are safe for human health and the environment (Amann *et al.* 2007). A wide range of technological measures have been developed that can prevent emissions from being emitted into the atmosphere, and advanced technologies that could prevent 90% of air pollutants from being released to the atmosphere, are already available. It is difficult, however, to predict exactly how quickly current (and possibly even more advanced future) emission control technologies will penetrate the global market, including developing countries, during the next century as this will rely heavily on political decision making.

Emission control measures

The application of emission control measures is an important determinant of future emissions of O_3 precursors. Emission controls may be regulatory or market driven, and be of a technical or non-technical nature. Technical measures include add-on control technologies (eg selective catalytic reduction (SCR)) and also include measures that are integrated into production technology (such as low-NO_X design of burners). Non-technical measures may include the use of different fuels, which produce fewer emissions (eg natural gas instead of coal) or initiatives to modify human behaviour and consumption patterns through economic incentives, education or regulations.

The new scenario extends the previous IIASA scenarios by taking into account legislation adopted up to the end of 2006 to control NO_X, CO and CH_4 emissions. CO and NO_X controls for mobile sources in developing countries in Asia and Latin America, in addition to the equivalent legislative status of Europe and North America are shown in Table 4.2 and Figure 4.5.

For stationary emission sources the new scenarios include the current legal requirements for emission controls as adopted in the European Union, North America and Japan. NO_X controls for stationary sources are currently required mainly in the European Union and Japan and to a lesser extent in North America, but not in the developing world.

Table 4.2 Emissions control measures assumed in the new B2 + CLE scenario. For each world region the table lists the emissions control measures that are required by legislation as at the end 2006.

NO_X controls		
European Union	Stationary sources	• Large plants: Emission limit values according to the EU Large Combustion Plant directive • Industrial processes: EU IPPC directive • Smaller sources: EU IPPC Directive
	Mobile sources	• Emission limit values of the EURO-standards for road and non-road sources as decided by mid-2007
Russia	Stationary sources	• Existing combustion plants and industrial processes: No controls • New large combustion plants: Low NO_X burners
	Mobile sources	• Road vehicles: Legislation according to current governmental plans (ie step-by-step phasing-in of controls up to Euro 4 for light-duty and Euro IV for heavy-duty vehicles) • No controls for non-road sources
Other European countries	Stationary sources	• Existing combustion plants and industrial processes: No controls • New large combustion plants: Low NO_X burners
	Mobile sources	• Legislation according to current governmental plans
China	Stationary sources	• Existing combustion plants and process sources: no controls • New large combustion plants: Low NO_X burners • Limited application of SCR on large power plants in heavily-polluted areas

	Mobile sources	• Step-by-step phasing-in of China-wide emission limit values for road sources as decided by mid-2007 (up to EURO 4 for light-duty vehicles, and Euro V for heavy-duty vehicles) • Faster phase-in of controls in some mega cities • Controls for non-road sources in agriculture and construction (equivalent of Stage 1 and 2 limits in the EU)
India	Stationary sources	• Existing combustion plants and industrial processes: No controls • New large combustion plants: Low NO_X burners
	Mobile sources	• For on-road vehicles: National legislation for road sources (currently up to Euro 3/III) • For selected cities earlier phase-in of the standards than in the rest of the country • For off-road vehicles and other sources: No control
Japan	Stationary sources	• Large combustion plants: SCR • Primary measures for all industrial sources
	Mobile sources	• Japanese emission standards for road- and non-road sources
South Korea	Stationary	• Existing combustion plants and industrial processes: Combustion modification • New large combustion plants: Low NO_X burners
	Mobile sources	• National standards on road- and non-road sources
Other Asia	Stationary sources	• Existing combustion plants and industrial processes: No controls • New large combustion plants: Low NO_X burners
	Mobile sources	• National standards for on-road vehicles (ie in most countries Euro 3/III or 4/IV) • Off-road sources: No controls
Middle East	Stationary	• No controls on existing combustion plants and process sources. New large combustion plants equipped with low NO_X burners by default
	Mobile	• National standards on road sources (if in force). Non-road sources remain uncontrolled
Africa	Stationary	• No controls on existing combustion plants and process sources. New large combustion plants equipped with low NO_X burners by default
	Mobile	• No controls for road and non-road sources
Latin America	Stationary	• Limited implementation of primary measures on existing power plants. Other combustion and process sources remain uncontrolled. New large combustion plants equipped with low NO_X burners by default
	Mobile	• National standards for road sources (in the majority of countries implementation of Euro 3/III and even Euro 4/IV within the next five years). Non-road sources remain uncontrolled
USA	Stationary	• National standards, including controls on large sources resulting from the Clean Air Interstate Rule
	Mobile	• Strict national standards on road sources and non-road sources
Canada	Stationary	• Primary measures on existing sources, SCR on selected new large combustion plants. Other new large plants equipped with low NO_X burners by default

	Mobile	• Standards as in the USA, implemented with up to five years delay
Australia and New Zealand	Stationary	• No controls on existing combustion plants and process sources. New large combustion plants equipped with low NO_X burners by default
	Mobile	• National standards on road sources (up to Euro 4/IV). Non-road sources remain uncontrolled
CO controls		
All regions	Stationary	• No CO-specific add-on control measures. Effects of implementing more efficient coal and biomass stoves in residential sector are included. Side effects from implementing controls for other pollutants (eg SCR for NO_X control) are considered
All regions	Mobile	• Emission limit values according to country-specific legislation on road and non-road sources
CH_4 controls		
Industrialised countries in Europe, North America and Japan		• Short-term policies (till 2030) taken from US EPA survey (US EPA, 2005). • Increased recovery of CH_4 from coal mining • Reduced pipeline leakage in the gas sector • Productivity improvements in livestock management and agricultural production • For solid waste, IPCC country-specific mass-balance methodology modified to reflect long-term trends in waste generation rates, recycling, gas recovery, etc

The new scenarios assume that industrialised countries (Europe, North America, Japan, Australia, etc) will comply with their current legislation on NO_X emission controls. In Europe, for example, it is assumed that the EURO IV/V and 4/5 standards for mobile sources, and the Large Combustion Plant (LCP) and IPPC directives for stationary sources, will be implemented. For Asia and Latin America full compliance is assumed for the recently adopted national environmental legislation for vehicles that require for example, catalytic converters and engine modifications as well as some measures for stationary sources.

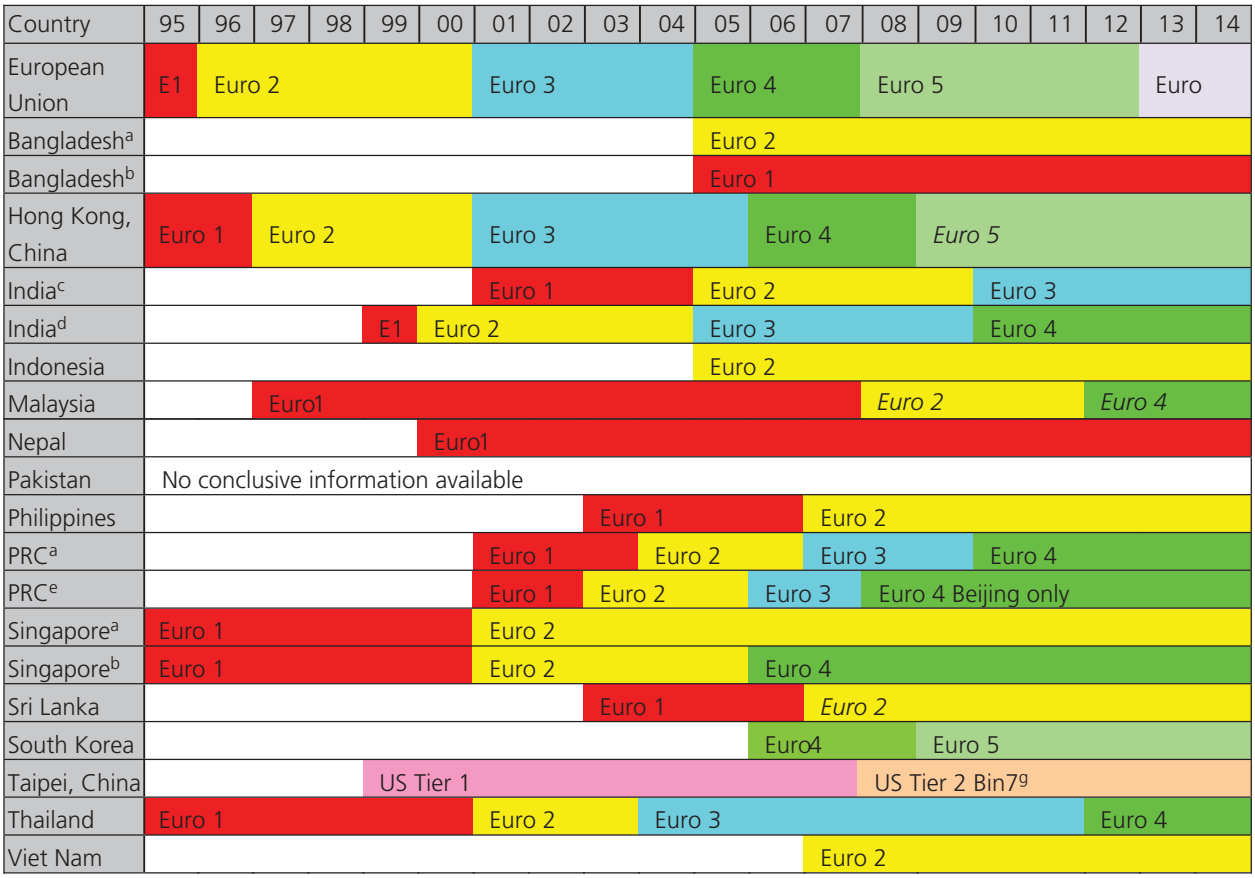
The low cost CH_4 measures currently available are now widely applied in Europe, North America and Japan but have not yet been adopted in developing countries. Where adopted, legislation requires low cost mitigation measures for waste disposal, wastewater treatment, gas distribution networks and coal mines. Such 'end-of-pipe' measures make it possible to reduce emissions substantially below historic levels even with sustained economic growth, and to approach air quality conditions that will not give rise to significant damage to human health and the environment. The extent to which such emission control measures will be applied globally in the future will critically determine prospective levels of air pollution.

The new scenarios take the conservative assumption that countries will retain and implement legislative and technological controls as adopted globally up to the end of 2006 for controlling conventional air pollutants to the end of the century (see Table 4.2). It is assumed that these measures will be phased in according to the timelines foreseen in the legislation and will remain in force up to the end of the century. The potential for strengthening of current emission standards over this period is not considered. No interventions have been assumed for other sectors and for emissions from natural sources.

Land use change

Potentially, the cause of the greatest change to biogenic emissions in the 21st century will be changes in land use due to anthropogenic activities such as population growth, shifts in crop types and management systems, and as a result of climate change. Deforestation and conversion of land to food and biofuel crops will occur in some parts of the world, but afforestation or reforestation (for amenity and biofuel production) will also occur. Land-use changes will influence the release of nmVOC and CH_4 emissions from vegetation, NO_X from the soil, CH_4 from agricultural practices, and CO and CH_4 from biomass burning (including the promotion of NO emissions from the soil due to the loss of the plant sink of inorganic nitrogen). The revised Riahi *et al.* scenarios consider future competition over land for agricultural and forestry purposes and project future land-use patterns as a function of current uses and future demand and supply interactions (Rokityanskiy *et al.* 2007). For example, the scenarios include assumptions about future changes in biomass burning, livestock numbers and manure. Riahi *et al.* (2006) and the new scenarios assume an increase of global livestock numbers in 2050 by

Figure 4.5 Level of automobile emission limits in Asian countries, compared with the European Union. Emission Standards for New Light-Duty Vehicles (as of 26 March 2008). The colour bars indicate the temporal penetration of emission control standards in the various countries. Euro-1 to Euro-4 refer to the different stages of emission standards of the European Union, while Tier 1 and 2 refer to US legislation. Source: Clean Air Initiative for Asian cities (CAI-Asia 2008).



Key: Italics – under discussion; a – gasoline; b – diesel; c – entire country; d – Delhi, Chennai, Mumbai, Kolkata, Bangalore, Hydrabad, Agra, Surat, Pune, Kanpur, Ahmedabad, Sholapur, Lucknow; Other cities in India are in Euro 2; e – Beijing and Guangzhou have adopted Euro 3 standards in September 2006 and Shanghai end of 2008; f – Euro 4 for gasoline vehicles and California ULEV standards for diesel vehicles; g – equivalent to Euro 4 emissions standards.

30–50%, and a doubling of fertiliser consumption worldwide. The effect of land use changes on natural VOC emissions from vegetation or soil NO_X emissions, are not included.

Projections of future anthropogenic emissions

The three new scenarios are analysed below in terms of the contribution of CO, NO_X and CH_4 to future O_3 to 2100. The analysis focuses on the central new B2 + CLE scenario but provides the corresponding numbers for the other two scenarios. The difference between values in 2000 is because the original IPCC figures were based on 1990's data and have since been revised upwards.

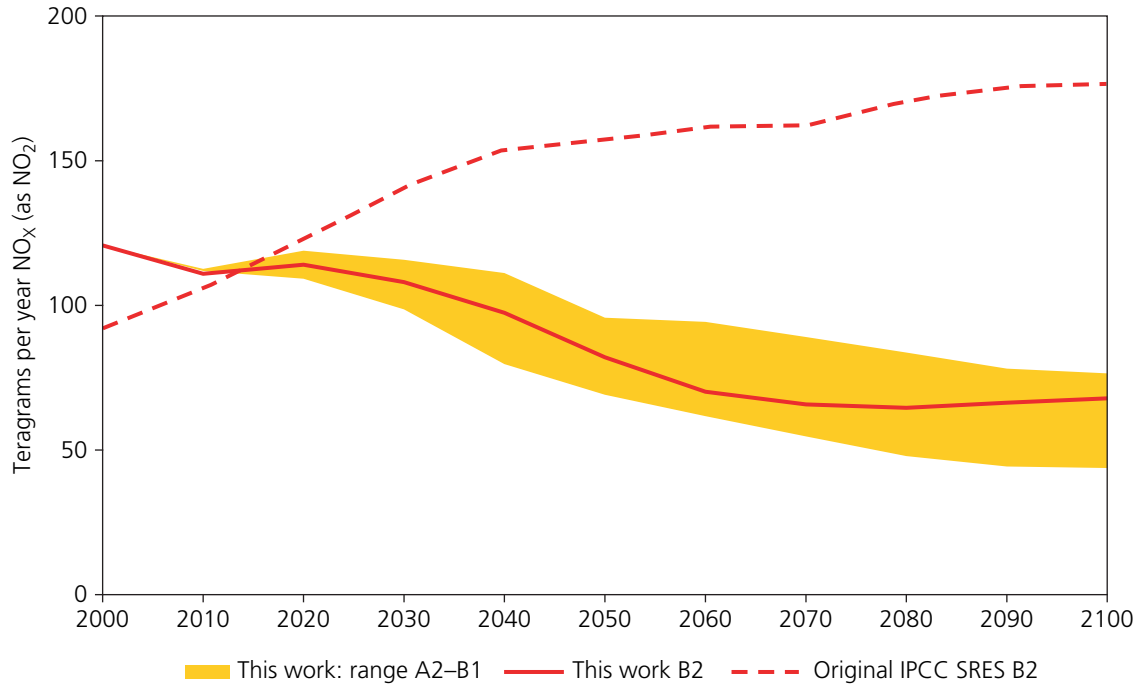
Nitrogen oxides (NO_X)

Figure 4.6 shows the envelope of possible NO_X emissions from the three new scenarios. The new B2 + CLE scenario (in red) is compared to the original 2000 IPCC SRES B2 estimate (red dotted line), which did not include emissions control legislation introduced after the year 2000. The analysis shows that while the activity levels of all important emission sources of NO_X will increase in the future, the actual level of NO_X emissions will depend critically on the extent to which emissions control legislation is implemented.

The scenario analysis suggests a declining trend in global NO_X emissions from human activities over the coming decades. Increases in Asian emissions that result from the steep growth in power generation and traffic volumes over the next few decades will be globally compensated by the decline in emissions in industrialised countries achieved through stringent emission controls and technologies (see Figure 4.5 and Table 4.2).

An exact quantification of future emission levels depends on the underlying assumptions on economic development as indicated in Riahi *et al*. (2006) by the range for the three scenarios. It is also noteworthy that this assessment, which takes explicit account of recent national emission control legislation, arrives at significantly lower projections of global NO_X emissions than the IPCC SRES estimate (Nakicenovic *et al*. 2000). This is not unexpected as Nakicenovic *et al*. (2000) does not account for the adoption of emission control legislation after the year 2000.

Figure 4.6 Scenarios of global NO_X emissions from anthropogenic sources.



As indicated in Figure 4.7, while NO_X emissions are expected to decline at the global level in the coming century, emissions in developing countries are likely to increase further over coming decades despite the control measures that have been recently adopted. Therefore, without more stringent control measures, NO_X emissions are likely to increase in developing countries in the next two to three decades and could enhance local and regional O_3 problems in these regions.

Carbon monoxide (CO)

Figure 4.8 shows the envelope of CO emissions for the three scenarios and compares it with the original IPCC SRES B2 scenario (2000). All projections of future energy use suggest a continued and far-reaching replacement of coal and fuel wood in the domestic sector by other forms of energy production that emit less CO, particularly in developing

Figure 4.7 NO_X emissions from human sources by world region for the new scenario (B2 + CLE) compared to the IPCC SRES B2 (2000) scenario.

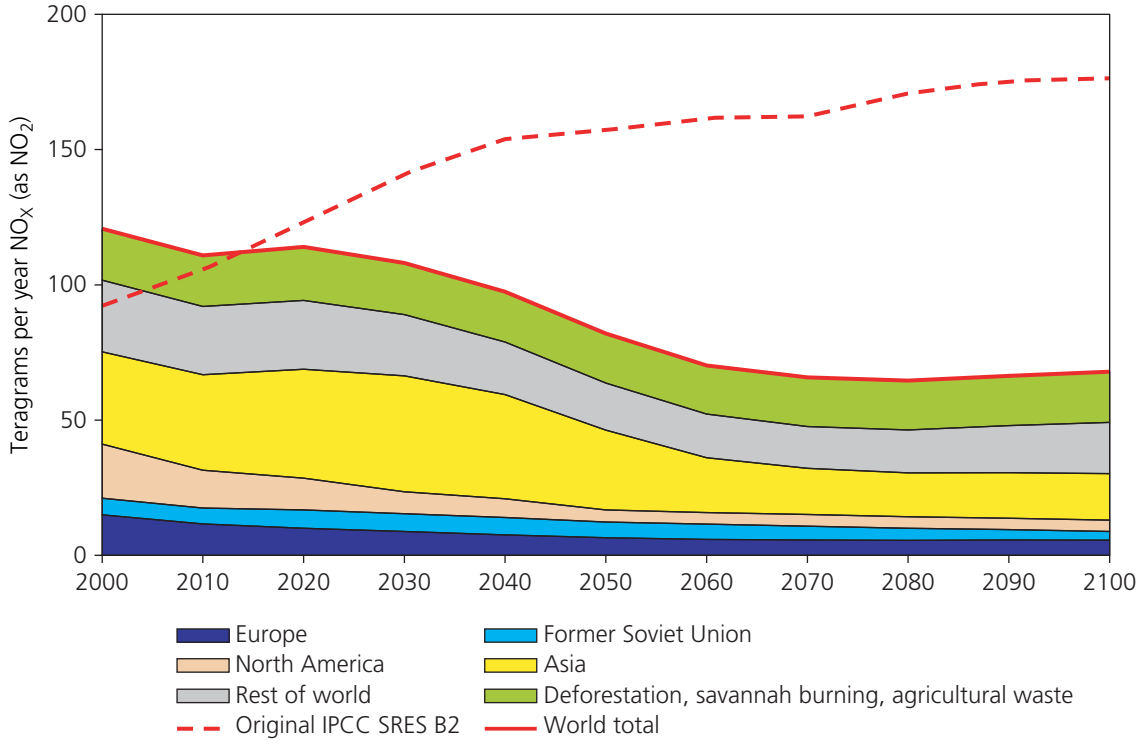
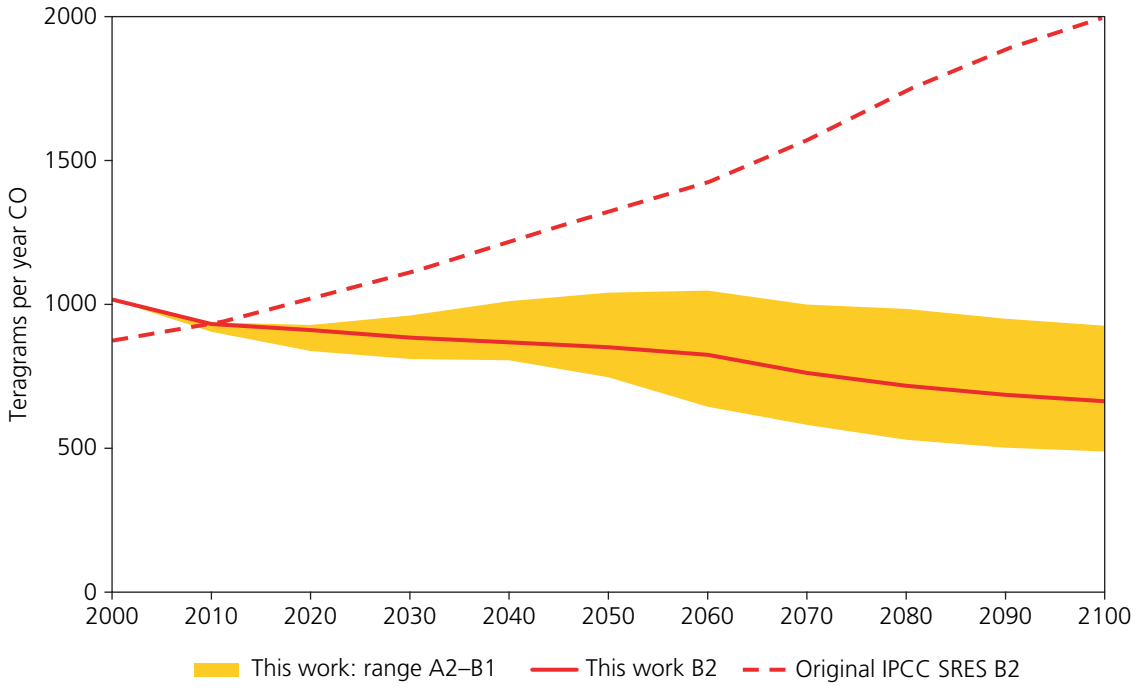


Figure 4.8 Scenarios of global CO emissions from anthropogenic sources.



countries. In addition, progressive introduction of three-way catalysts in vehicles to reduce NO_X emissions will also lead to significant reductions of CO in the transport sector. Combined, these two factors point towards a decreasing long-term trend in global CO emissions over the next century.

Figure 4.9 shows the continuing importance of deforestation and biomass burning for the global emission rate of CO. Emissions from these sources will gain in relative importance as anthropogenic emissions will decline due to technological measures and changes in fuel consumption structure (ie less burning of solid fuels in households). As for NO_X, the penetration of vehicle emission control technologies considered in the scenario analysis leads to a decrease in CO emissions over the century. This is significantly different to the trend suggested in the original IPCC SRES B2 scenario (2000) which projects an increase.

Figure 4.9 CO emissions from human sources by world region for the new scenario (B2 + CLE) compared to the original IPCC SRES B2 scenario (2000).

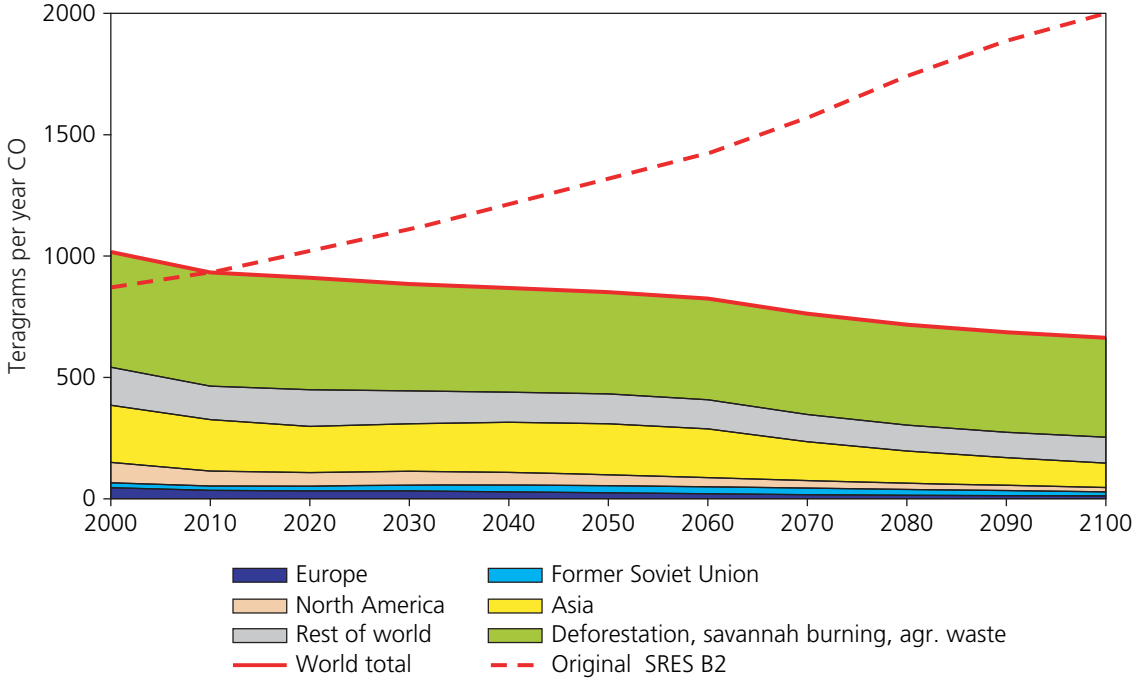
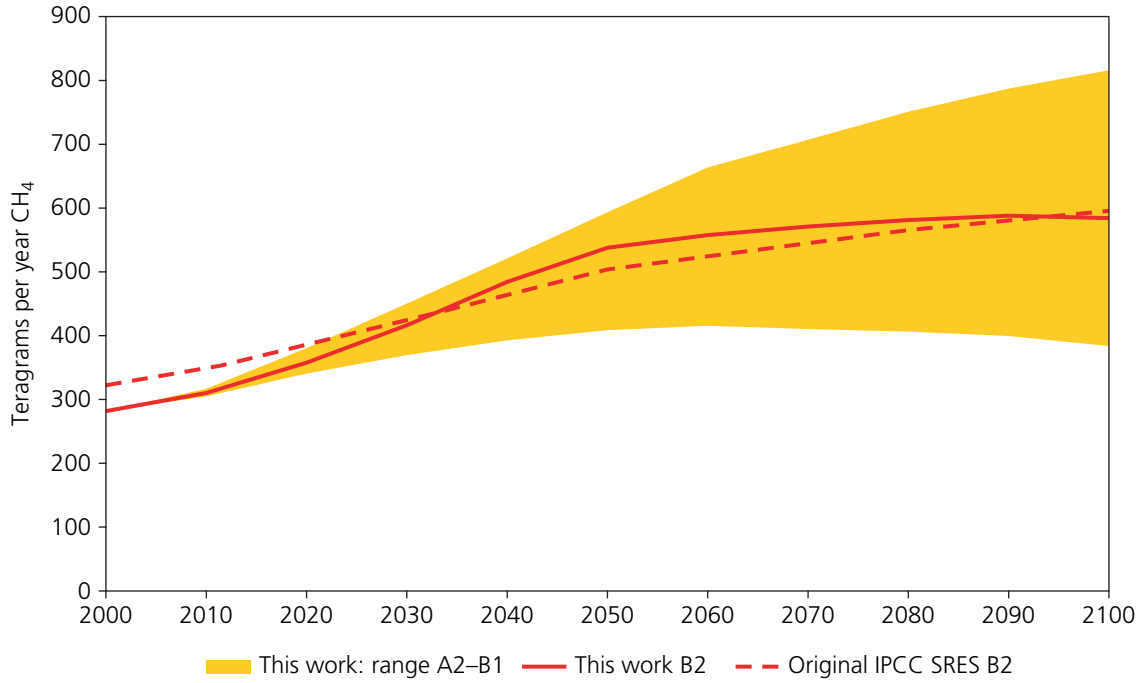


Figure 4.10 Scenarios of global CH_4 emissions from anthropogenic sources.



Methane (CH_4)

Figure 4.10 illustrates the envelope of predictions for the new scenarios, compared to the original IPCC SRES B2 (2000). To assess future trends, policies for CH_4 mitigation that have been recently adopted in North America, Japan, and the industrialised countries in Europe were taken into account (see Table 4.2) (US-EPA 2005). Consideration is given to gradual technological improvements for energy-related sources, such as reduced pipeline leakage, as well as productivity improvements in livestock management and agricultural production. For solid waste, the scenarios extrapolate long-term trends in waste generation rates, recycling and gas recovery. Higher CH_4 emissions that accompany the shift from surface to deep coal mining are predicted to be balanced by increased recovery of CH_4 in coal mines.

However, application of these mitigation measures for CH_4 will not be sufficient to prevent a global increase of CH_4 emissions throughout the century as demonstrated by Figure 4.11. Non-energy related CH_4 emissions, especially from the agricultural sector in the developing countries of Asia, Latin America and Africa, increase by a factor of two in the B2 cases and remain about constant for the second half of the century. Energy related CH_4 emissions in 2050 grow by 22% relative to the levels in 2000, and by 2100 CH_4 emissions double as compared to the base year. This increase is driven by the higher demand for fossil fuel use.

It should be noted that the uncertainties of these projections are large after 2050. In contrast to the conventional air pollutants NO_X and CO, for which stringent emissions control measures have been introduced in recent years, this assessment for CH_4 matches the IPCC SRES estimate (2000).

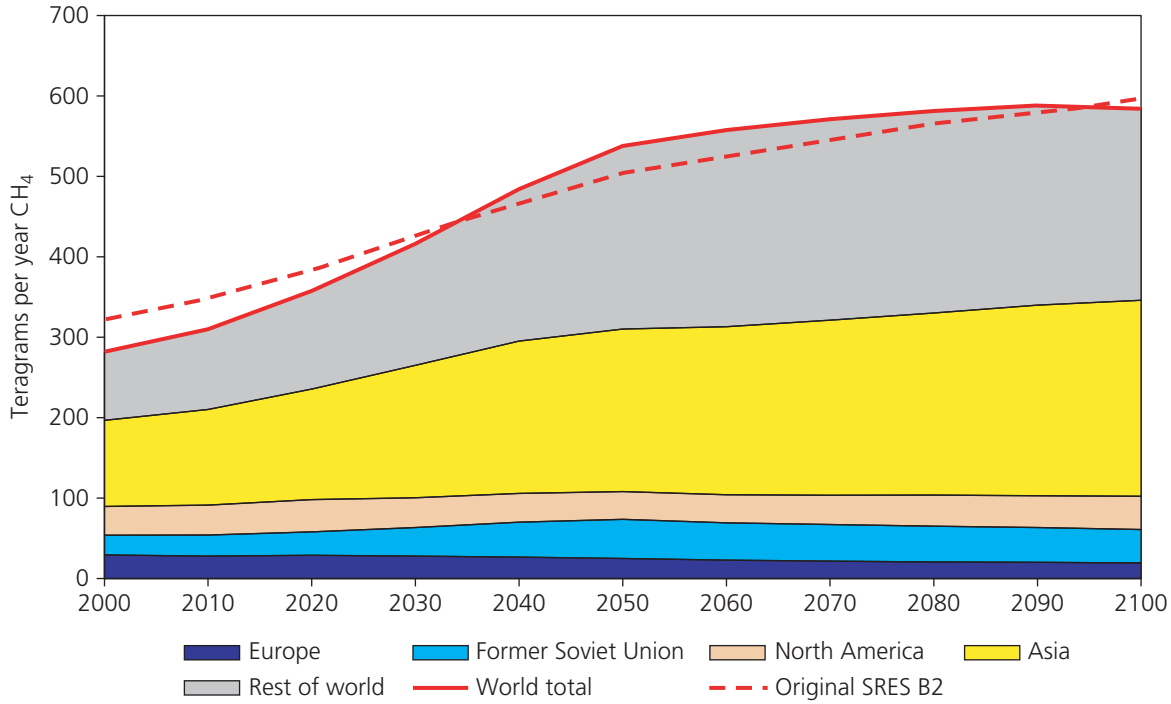
New scenario results and analysis

It can be seen from the difference in emission trends between the new B2 + CLE and the original IPCC SRES B2 scenario that the implementation of control measures is an overriding factor in determining how anthropogenic O_3 precursor emissions will change in the future. Uncertainties about future socioeconomic development (within the ranges analysed by the three scenarios) have comparably less impact on future emission levels. The results from the new scenarios suggest that recent measures to reduce emissions across the globe could reverse rising emissions of NO_X and CO globally, although it should be stressed that this will only be the case if all current measures are fully implemented. CH_4 emissions which are less stringently controlled globally are projected to continue to rise due to human activities although there is a high level of uncertainty within the envelope of future projections. More substantial controls will also be necessary across the globe if a more pronounced decrease in emissions is to be achieved.

Under these scenarios, there will be substantial increases in emissions in rapidly developing regions such as Asia over the next few decades. This suggests that more emissions controls are needed in these regions. As CH_4 is an important contributor to O_3 concentrations, and yet is not controlled in many regions, stronger controls may be necessary. Controls on the relatively unregulated biomass burning emissions will also be needed in regions where such emissions dominate (ie Africa, Asia, Latin America).

However, there are several assumptions within these scenarios which will affect the projected outcome, for example the socioeconomic pathway followed, or the rate

Figure 4.11 CH_4 emissions from human sources by world region for the new scenario (B2 + CLE) compared to the original IPCC SRES B2 scenario (2000).



of technological development. Deviations from, or even within, these assumptions will have an effect on the projection of future emissions. A major uncertainty in future emissions projections is the rate of economic growth and therefore energy demand within rapidly developing regions such as China and India. Recent estimates of economic growth in these regions significantly exceed the assumed short-term growth in the IPCC's scenarios. (IEA 2007; Rapauch *et al*. 2007). The IEA recently projected in its reference scenario (based on current legislation) that China's primary energy demand will more than double from 1742 million tonnes of oil equivalent (MToe) in 2005 to 3819 MToe in 2030 representing a 3.5% increase per year. Thereby China will become the world's largest energy consumer soon after 2010, although in the longer term demand slows. The implications of this observation on long-term development need to be further explored. For India primary energy demand is also expected to more than double by 2030 growing at 3.6% a year.

Coal will remain the most important fuel out to 2030, and use is projected to triple in this timeframe. If these trends persist throughout the coming decades and no stricter emissions controls are implemented in these countries, air pollutant emissions will grow faster in the next few years than the new scenarios predict.

The current growth in shipping emissions, a relatively unregulated industry, could also have a major effect on future global emissions. The new scenarios employ the growth trajectory developed by Eyring *et al*. (2005b) with an increase in sea trade volumes of 2.8% per year up to 2050. While this is consistent with the storyline of the IPCC B2 scenarios, it is somewhat lower than the 3.3% growth rate of the last two decades. In addition, the new analysis adopts the 'moderate technology' scenario TS2 of Eyring *et al*. (2005b), which assumes a 70% cut in NO_X emission factors for ships for 2050. As these reductions are more ambitious than those currently being discussed in the International Maritime Organization (IMO), the scenarios represent an optimistic view of future ship emissions.

The effect on anthropogenic emissions of a major shift in land-use, for example towards biofuel production, could have a significant effect on biogenic nmVOC, NO_X and CH_4 emissions, but has not been considered fully in the new scenarios. Similarly, significant changes in natural emissions resulting from climate change are not considered in these scenarios and could also substantially affect future O_3 concentrations (see section 4.5 for more detail).

It is, however important to balance these uncertainties against the possibility that emissions reductions could also be greater than projected in the new scenarios, due to the conservative assumption that no additional control measures will be implemented over the century. The implementation of future climate change policies will also have a significant effect.

4.3.2 Natural emissions

While methods have been developed for estimating global emissions of trace gases from all major natural sources, uncertainties remain substantial and many sources are poorly quantified. Quantifying future trends in natural emissions of NO_X, CH_4, CO or nmVOC is therefore a speculative exercise with large uncertainty. Natural emissions from vegetation, soil, and lightning, are strongly influenced by a range of

environmental factors, including temperature, light, humidity, meteorology, vegetation cover, leaf size or plant age. By exploring how these may change in the future, it is possible to evaluate which interactions may be most important.

Some model studies show an increase in lightning (and associated NO_X production) as the climate warms (Price and Rind 1994; Toumi *et al.* 1996; EC 2003; Brasseur *et al.* 2006; Schumann & Huntrieser 2007), with increases of up to 60% per degree K of global mean surface warming (Lamarque *et al.* 2005). Soil NO_X emissions, produced from bacterial activity, are also likely to increase as the land warms (Granier *et al.* 2003; Hauglustaine *et al.* 2005; Liao *et al.* 2006; Wu *et al.* 2007b). For example a 10ºC rise in soil temperature has been shown to produce a 2–5 fold increase in NO emission rates (EC 2003). However, emissions of NO from soil are highly uncertain and the underlying quality of these emission estimates has improved little in the last decade. Carbon monoxide emissions from biomass burning are expected to change in the future as they are closely associated with land-use change and human activities. However, an absence of long-term monitoring in many regions, and inadequate satellite observations means that it is not yet possible to predict the direction or magnitude of potential changes. In the case of CH_4 emissions, although subject to significant uncertainties, the direction of future change appears to be upwards. Many studies show that natural emissions of CH_4 from wetlands are sensitive to temperature. Typical responses from peat wetlands show an increase in CH_4 emission of a factor of two for an increase in temperature of 4°C (Hargreaves & Fowler 1998). Given the likely scale of the increases in temperature in the high latitude peatland areas, substantial increases in CH_4 are possible. Temperature changes will be important, as will changes in the hydrological cycle, which in the case of CH_4 emissions, regulates the water table and therefore controls whether soils are a sink or a source of CH_4. Emissions of nmVOC are strongly dependent on environmental conditions. Isoprene for example, is strongly influenced by the intensity of photosynthetically active light (PAR) and temperature (Tingey *et al.* 1979), and emission rates may therefore increase significantly during the 21st century.

To illustrate the potential changes in natural emissions that could be possible over the next century due to climate change and land-use change, the following sections focus on isoprene, as this is the best understood of the nmVOC. New current and future emission rate estimates based on analysis of the recent scientific evidence are provided.

Future isoprene emissions

Isoprene is probably the single most important natural nmVOC with respect to the formation of O_3 due to its very large global emission rates, and reactivity with OH. Table 4.3 illustrates the range of factors important for controlling the emission rates of VOC from vegetation. In addition to PAR and temperature, emissions are also strongly influenced by physiological factors. For example, isoprene emission rates vary with leaf age (Guenther *et al.* 1999) and it has recently been shown that emissions from some plants are under strong

Table 4.3 Parameters which can influence biogenic VOC emissions.

Factor	Effects included in latest emissions estimates for natural nmVOC for this study (Lathiere *et al.* (unpublished data)
Light	Yes
Temperature (current and previous)	Yes
Ambient CO_2 concentration	Yes
Losses within the canopy by reaction and/or deposition	Yes
Leaf age	Yes
Biomass	Yes
Plant species (usually by functional type)	Yes
Drought	Yes
O_3	No
Herbivory	No
Wind damage	No
Fire	No
Logging	No
Nutrient status	No
Circadian control	No

circadian control (Wilkinson *et al.* 2006), leading to hour-to-hour variations in emission rates that are not accounted for in current emissions models. Other biotic and abiotic stresses that may potentially affect VOC emission rates, but which are not currently accounted for in emissions estimates, include herbivory, wind damage, fire, logging, drought and nutrient status.

Changes in environmental conditions over the 21st century such as increases in mean ambient temperature and the frequency and magnitude of high temperature events may increase isoprene emission rates significantly (Pyle *et al.* 2007). For example, a 1°C increase in the July average temperature has been estimated to increase isoprene emissions from Great Britain for that month by 14%, and a 3°C increase by about 50% (Stewart *et al.* 2003) (see Figure 4.12). The effects of increasing frequency and magnitude of short-term extreme temperatures on isoprene emissions have not been quantified, but are likely to be significant with respect to summertime O_3 episodes. This effect is accounted for in emissions estimates (see Table 4.3).

However, isoprene emission rates are significantly reduced when soil moisture falls to a point at which plants begin to wilt (Tingey *et al*. 1981; Sharkey & Loreto 1993; Fang *et al*. 1996; Pegoraro *et al*. 2004; Funk *et al*. 2005; Brilli *et al*. 2007) and this may restrict the role of isoprene in O_3 formation in the future. As precipitation patterns and intensity change in the coming century in response to increasing CO_2 concentrations, emission rates may change in some regions of the world, for example in Southern Europe where reduced rainfall is predicted (see chapter 6, section 6.2.1).

Some studies have observed an increase in isoprene emission rates with increased CO_2 as a result of the effect of enhanced CO_2 on plant biomass (eg Wilkinson & Broadmeadow in prep). However, experiments have also shown that increasing CO_2 concentrations may decrease isoprene synthesis rates, and hence emission rates in some plant species (Figure 4.13, Centritto *et al.* 2004; Pegoraro *et al.* 2004; Possell *et al.* 2004, 2005). Because of the conflicting direct and indirect effects of increasing CO_2 concentrations on isoprene emission rates, it is difficult to predict with certainty how these will change over the next century. However it is possible that they may not increase globally in the way predicted by earlier modelling studies (Monson *et al.* 2007).

Isoprene emission rates may also be affected by O_3. These interactions are complex and are not yet adequately accounted for in emissions estimates. In some plant species isoprene emission rates have been shown to decrease in response to O_3 (Velikova *et al*. 2005; Fares *et al*. 2006; Calfapietra *et al*. 2007), while in others, O_3 exposure has been shown to enhance isoprene emission rates (Fares *et al*. 2006). Elevated O_3 may also lead to a reduction of net primary productivity (Felzer *et al*. 2004), which will in turn decrease biogenic emissions.

Although many of the above factors could lead to an increase in isoprene emissions in many regions, changes in vegetation cover due to either natural or anthropogenic activities (eg loss of the Amazon rainforest) may increase or decrease emissions, depending on the nature of the replacement ground cover. Land-use change projections are missing from current models and most isoprene estimates. Dynamic vegetation models project significant but uncertain natural vegetation changes

Figure 4.12 Temperature dependency of total isoprene emissions from Mediterranean vegetation relative to emission rates at 20°C. The figure shows isoprene emission rates measured in the field from different isoprene emitting species found at various sites in the Mediterranean region (squares), and the corresponding isoprene emission rates calculated using the Guenther et al. (1995) algorithm G95 using temperature and PAR from field measurements (stars). Source: Owen et al. (1998). Copyright 2008 American Geophysical Union. Reproduced/modified by permission of American Geophysical Union.

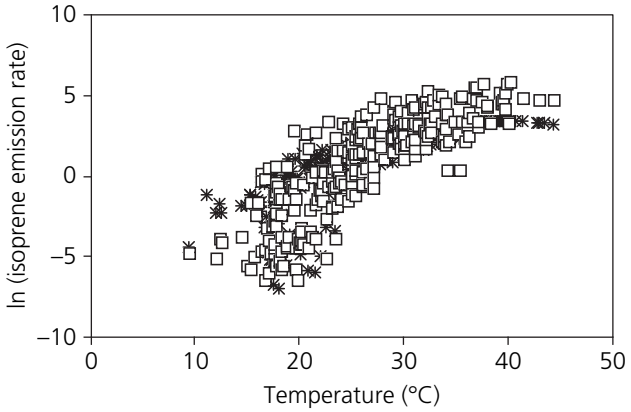
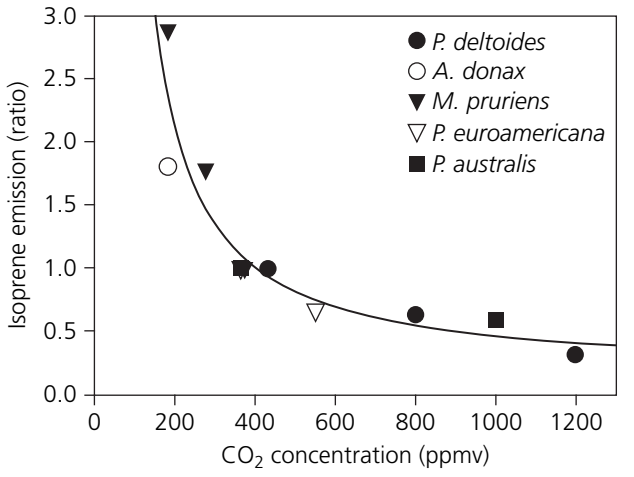


Figure 4.13 Dependence of isoprene emission rate on ambient CO_2 concentration for some plant species: Populus delatoides, Arundo donax, Mucuna pruriens, Populus euroamericana and Phragmites australis. Source: Possell et al. (2005). Reproduced with permission from Global Change Biology (Wiley-Blackwell).



in the 21st century and human land-use change may have an even larger effect on land cover. For example Steiner *et al*. (2002) estimated that the conversion of forest to rice crops in Eastern Asia has resulted in a 45% reduction in isoprene emission rates since the Industrial Revolution. Similarly Lathiere *et al*. (2005, 2006) suggest large reductions in isoprene emissions arising from past, current and future tropical deforestation. However, Owen *et al*. (unpublished data) predict substantial increases in isoprene (and other VOC) emission rates in tropical regions following logging of forest, and conversion of forest to plantations. They have speculated that the isoprene emission flux potential might change from 480 g km^{-2} h^{-1} for the current rainforest, to 1735 g km^{-2} h^{-1} for logged forest to 9410 g km^{-2} h^{-1} for oil palm plantation.

Estimating future isoprene emission rates

Attempts have been made to estimate how isoprene emission rates may change in the future. However, the uncertainties in estimating isoprene rates are considerable, and our understanding of the potential importance of factors such as CO_2 concentration, climate and land use change, is rapidly evolving. Given the nature of these uncertainties, constructing a truly accurate bottom-up emission inventory is probably an impossible task. Model evaluation using Earth observation data is essential, and preliminary steps have been taken in this direction (Guenther *et al.* 2006), suggesting that current estimates of global isoprene emissions are probably accurate to within a factor of three. An uncertainty analysis for an emission inventory for rural Great Britain concluded that the estimates had an overall relative error of a factor of four (Stewart *et al.* 2003). Biogenic VOC estimates from urban areas may be equally uncertain (Owen *et al.* 2006). Despite the relatively high resolution of the isoprene emission inventory for the UK, it has proved difficult to adequately apportion measured isoprene mixing ratios during a recent (2003) O_3 episode to sources or to adequately quantify the role of isoprene and other biogenic VOC on O_3 formation (Lee *et al.* 2006).

Guenther *et al.* (2006) predicted a doubling in global isoprene emission rates by around 2100 in response to climate change driven changes in the distribution of plant functional types and increasing temperature (Table 4.4). Using a similar approach Young (unpublished) predicts a similar increase in isoprene emission rates to 873 Tg y^{-1} for a 2100 climate, compared with 451 Tg y^{-1} for the present day. However neither of these estimates considered the possible effects of increasing CO_2 concentration on emission rates.

For the purposes of this study the effect of multiple environmental parameters (see Table 4.3), on isoprene emissions has been evaluated (Lathiere, Guenther, Beerling and Hewitt, unpublished data). The results of this analysis suggests that isoprene emissions could decrease if climate change, CO_2 effects and land use changes are all included in the emissions estimate models. However, it is recognised that the estimate presented may be superseded by more complete and better estimates in the near future. These new emissions estimates have therefore not been used in the atmospheric chemistry modelling presented in chapter 5.

Lathiere *et al.* (unpublished) applied an emission scheme, based on the MEGAN model, but incorporating the latest isoprene emissions research (including the suppression of isoprene emissions under elevated CO_2 concentrations), to the present day and future environmental conditions. Based on a present day climate (Climate Research Unit, Norwich, UK) and a potential present day vegetation generated by the Sheffield Dynamic Global Vegetation Model (SDGVM: Woodward *et al.* 1995; Woodward & Lomas 2004), future climate conditions were simulated with the Laboratoire de Meteorologie Dynamique general circulation model (LMDz) (Harzallah & Sadourny 1995) to predict potential plant functional type distributions and temperatures for 2050. These runs did not take into account the change in land cover related to change in natural vegetation. To assess the impact of crops on isoprene emissions, Lathiere *et al.* combined the potential vegetation maps generated by the SDGVM with crop distributions based on Ramankutty & Foley (1999) and Goldewijk (2001) as well as on scenarios from the IMAGE model for 2050 (De Noblet & Peterschmitt, unpublished data). In considering crop type, only one 'generic' crop type was applied.

Based on the present climate, global isoprene emissions are estimated to be 612 Tg y^{-1} by Lathiere, Guenther, Beerling & Hewitt (unpublished data). When the effects of a 2050 climate and increasing ambient CO_2 concentrations (to 532 ppm in 2050) are included, global isoprene emissions of 626 Tg y^{-1} were estimated for 2050. However, when the

Table 4.4 Estimates of global isoprene emission rates.

Study	Isoprene emission rate (Tg y^{-1})	
	Present day	Future
Lathiere, Guenther, Beerling & Hewitt (unpublished data)	A: 612, B: 532	2050 = A: 626, B: 439
Guenther *et al*. (2006)	~600	2100 = ~1200
Young (unpublished work)	451	2100 = 873

A = 'potential vegetation cover', B = actual land use (including crops).

distribution of a generic crop type is included, the estimate of global isoprene emissions is reduced to 532 Tg y^{-1} for the present-day and to 439 Tg y^{-1} for 2050. In contrast to previous estimates of future isoprene emission rates it is possible therefore that isoprene emissions may be smaller in 2050 than at the present time, and that increasing ambient CO_2 concentrations may suppress isoprene emissions globally by counteracting the increases in emissions projected to occur as a result of temperature increases over the 21st century (see Table 4.3). This is also suggested at the regional (European) scale by Arneth *et al.* (2008). More research on the relative isoprene emission rates from natural vegetation and different crop types, and interactions with CO_2 is required to inform analysis of the effects of changes in land use of future isoprene emissions.

4.4 Gaps and uncertainties

The evaluation of future changes in emissions of O_3 precursors is limited by the availability of inventory data, uncertainties in assumptions regarding drivers of emissions, and understanding of the biogeochemistry and physiology of natural emissions.

Future projections of emissions are also limited by the uncertainties inherent in the assumptions on which the analysis is based, in particular the socioeconomic pathways followed and the rate at which technological advances occur and are implemented. In this study important assumptions included the rate of economic growth in developing countries, particularly China, future coal use, growth rates in shipping emissions, the effect of a major shift in land use, for example for biofuel production, and the level of implementation of air pollution and climate change controls.

Scenario assessments are based on detailed emission inventories in different countries. However, inventories are not available for all pollutants in all countries, and where inventories are available, data collection may not be standardised. For example, inventories of anthropogenic emissions in some developed countries are of low quality for some pollutants (particularly VOC), sources (eg biomass burning) and sectors (eg international shipping). Inventories of natural emissions are poor, as globally the observational networks are patchy and there is a lack of systematic screening and analysis of such emissions. In developing countries a lack of emissions measurements and observations means that the quality of emission inventories for all pollutants is generally poor. This is partly due to a lack of expertise and institutions within these regions.

Current emission estimates for most natural O_3 precursor gases remain however the greatest area of uncertainty in the study of O_3 formation. There is limited understanding of how the complex interplay of environmental influences, such as temperature, moisture, land-use and ambient CO_2 and O_3 concentrations, affect natural precursor emissions. Emission estimates for lightning, soil NO_X, CH_4, and nmVOC from vegetation are all important uncertainties which directly influence the ability to quantify current and future O_3 concentrations. The ability to measure and estimate VOC emissions is limited by poor understanding of biogenic sources, including CH_4 (see below), and speciation of nmVOC for different plant functional types. The relative contribution of the different nmVOC to O_3 formation is also not well understood. This is despite the recent development of rapid analytical techniques, such as proton transfer reaction mass spectrometry and the use of micro-meteorologically based flux measurement techniques, which have greatly enhanced the observational base on which emission estimates are made, and the use of satellite data to verify emissions estimates.

In terms of isoprene specifically, the uncertainties associated with estimating isoprene emission rates are considerable, and may increase with model domain size as uncertainties in plant species distributions, and in the isoprene emitting capacity of individual plant species and plant functional types increase. The use of taxonomic relationships in the past to assign emission rates to unstudied species is also a source of uncertainty as this approach may give erroneous results (eg Mediterranean oaks produce monoterpenes, while oaks in other regions tend to be isoprene emitters). Understanding of how isoprene, and the other natural O_3 precursor emissions will influence future O_3 concentrations, would be improved by the development of an easily and rapidly manipulated Earth systems model which integrates biological and atmospheric processes.

There are significant uncertainties associated with estimating future trends in CH_4. Although the total emissions of CH_4 are relatively well known (500–600 Tg y^{-1}), the relative contributions to the overall flux of emissions are many and uncertain. Methane concentrations increased rapidly from the beginning of the industrial revolution to the end of the 1980s (700 to 1700–1800 ppb respectively). Since then, atmospheric concentrations have fluctuated between years but no longer show a clear trend. A detailed discussion of the competing hypotheses to explain this decrease in growth rate is provided in Denman *et al.* (2007), however there is no consensus on the cause(s) of the recent slowdown in growth.

4.5 Conclusions

Emissions of O_3 precursors, particularly NO_X, are probably the primary determinants of future O_3 concentrations. The main drivers of changes in anthropogenic O_3 precursor emissions in the future will be population growth, economic growth, land use change, new technology developments and the implementation of emissions controls. The degree to which emissions controls are implemented particularly for road transport, biomass burning including deforestation, power generation and international shipping and aviation will largely determine future anthropogenic O_3 precursor emission trends. Technology will also play an important role as emission reduction technologies are available that, if implemented, could reduce emissions by 90%. Natural O_3 precursor emissions also contribute significantly to O_3 concentrations, however it is not yet possible to quantify with any certainty how they will change in the future in response to multiple drivers including changes in land use and climate.

The new anthropogenic emission scenarios presented in this chapter make assumptions about how different country's O_3

precursor emissions will change in the future using current trends in economic development and assume that current emissions control legislation will be fully implemented. Unlike the other precursor gases, nmVOC emissions were assumed to remain constant. The results from this analysis should therefore be interpreted with these assumptions in mind.

The new scenario projections show that global anthropogenic NO_X and CO emissions will decline to 2050 provided that current emissions controls are fully implemented. Methane emissions will continue to increase to 2050. For CH_4, emissions from the agricultural sector, particularly in Asia, Latin America and Africa will be important, as will increasing demand for fossil fuel use in many regions. While emission controls for CH_4 are currently applied in Europe, North America and Japan, they have not yet been adopted in developing countries. The implementation of CH_4 emission controls in these regions could therefore lead to significant future reductions in CH_4.

In contrast to reductions from most other regions, the scenarios project an increase in NO_X emissions from Asia over the next few decades due to increases in power generation and traffic volumes, although the implementation of current emission controls for mobile sources limit further growth of emissions from this sector by 2050. NO_X emissions are also expected to increase in Africa as there are currently no NO_X controls in place for mobile or stationary sources other than for new large combustion plants. In Latin America, Africa and Asia biomass burning (deforestation, savannah burning, agricultural waste) is a significant source of CO and NO_X with emissions substantially exceeding those from the energy production sectors. Emissions are projected to remain high, or decline only slightly in these regions due to the poor regulation of biomass burning.

Several key factors could affect the scenario projections, and the predicted global decline in NO_X and CO emissions. If left unregulated and without the adoption of significant technological changes, NO_X emissions from international shipping and, to a lesser extent, international aviation will outstrip those from land-based transport sectors which would significantly increase future global emissions. Deviation from the assumed economic growth rates in China and India and other developing economies could also affect the projections. As current growth rates (eg in China) are already exceeding the assumed growth rates in the scenario this is an important area that requires further consideration.

For natural emissions of O_3 precursors it is likely that land use change will be the dominant driver of future biogenic emissions. Emissions of CH_4 and NO_X from the biosphere are likely to increase in the future in response to changes in drivers such as land use and climate. Biogenic emissions of nmVOC and isoprene in particular may increase in the future as a result of changes in climate although interactions with atmospheric CO_2 may substantially reduce the magnitude of the change. Land use change, specifically the replacement of plant species with other species of higher or lower emitting potential, will be important.

5 Model projections of future ground-level ozone at global, regional and urban scales

5.1 Introduction

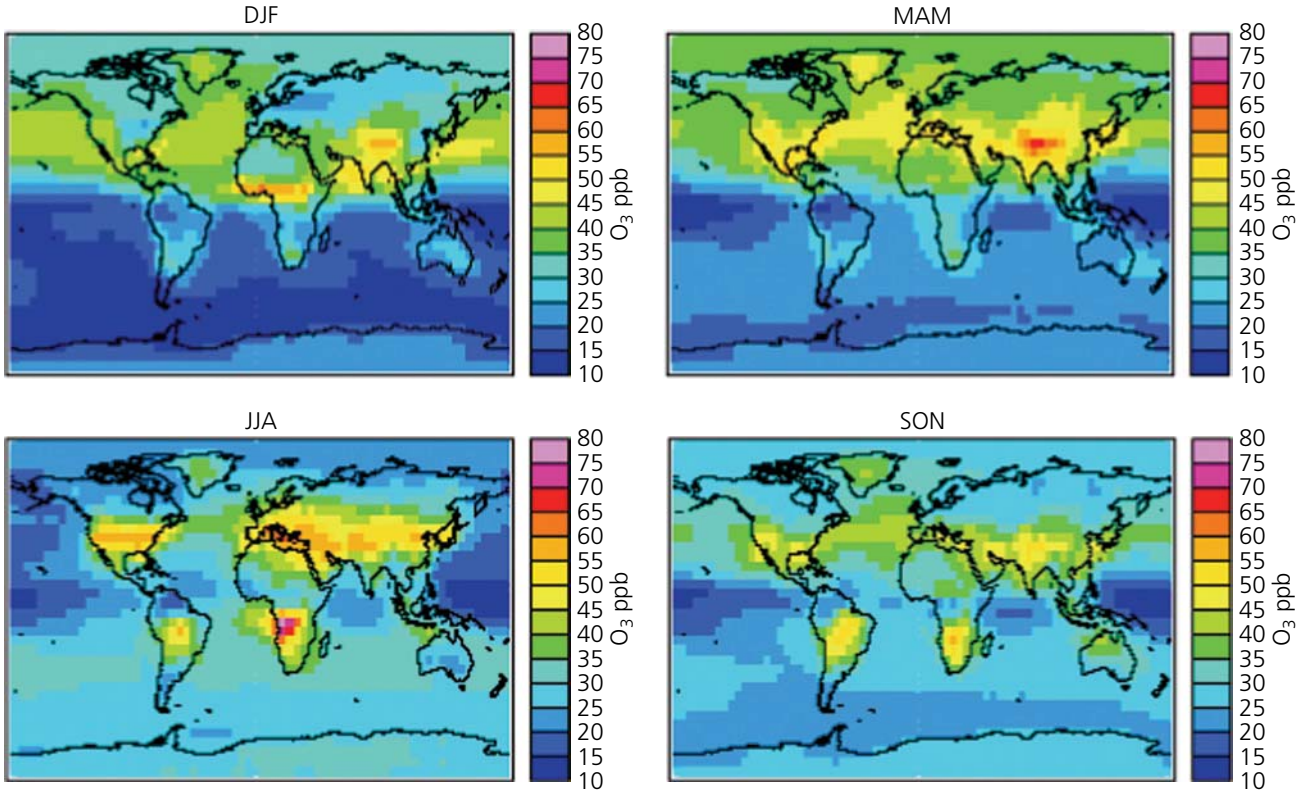
The ability to quantify changes in future O_3 depends on the application of models of atmospheric chemistry and climate, driven by future emissions projections. These models include O_3 precursor emissions, atmospheric chemistry, transport and removal processes, as described in previous chapters. Models can be used to explore interactions between the processes that control O_3 and the responses to changes in input variables, and can be validated against observations (see Box 5.1). No single model is able to realistically simulate O_3 at global, regional and urban scales. Instead, different models that span a wide range of spatial scales have been developed. Global models cover the whole troposphere (and generally the lower stratosphere) with horizontal resolutions of ~200 km. Regional models cover spatial scales of a single continent, for example, covering Europe or North America at ~50 km resolution. National scale models for smaller countries are more detailed, for example, the UK at 5 km resolution. At even finer scales, urban models (eg London) at <1 km scales, and street canyon models (~1 m) have been developed.

In this chapter projections of future ground-level O_3 from a variety of models are presented. New modelling work was completed to assess possible changes in O_3 over the period to 2050, including the effects of projected changes in both emissions and climate. The results of this new work are presented in each section, along with results from the published scientific literature. The models presented are driven by the emission scenarios discussed in chapter 4, and in some cases include projected climate effects (discussed in more detail in chapter 6). This chapter also presents several sensitivity studies, in which model parameters are changed (eg temperature) to elucidate their individual roles in O_3 formation. The chapter is divided into three main sections: global, regional and urban, with an emphasis on the global scale as this integrates the wider geographical scales and the future emission projections from chapter 4.

5.2 Global modelling

The global distribution of ground-level O_3 (Figure 5.1) will respond to changes in emissions of O_3 precursors and to

Figure 5.1 Seasonal distributions of surface (lowest ~50 m) O_3 (ppb) for the year 2000 from the ensemble mean of 26 ACCENT PhotoComp models (Dentener et al. 2006b; Stevenson et al. 2006). The top left panel shows the distribution of surface O_3 for December, January, February (DJF); the top right panel for March, April, May (MAM); bottom left for June, July, August (JJA); and bottom right for September, October, November (SON).



Box 5.1 Global model validation

Global models are usually validated by comparison with a range of observations. There is an extensive data base of O_3 measurements, both at the surface and through the troposphere. In addition a number of surface sites also monitor O_3 precursors such as CO and CH_4, and some measure surface-deposited species related to O_3 chemistry. Space-borne sensors also provide important data for validation. Many of these data sets were used to assess the models used in the IPCC Fourth Assessment Report (eg O_3 (Stevenson *et al*. 2006), CO (Shindell *et al*. 2006), column NO_2 (van Noije *et al*. 2006), and surface deposition of nitrogen compounds (Dentener *et al*. 2006a)). In addition, there are multiple campaign datasets, with many related chemical species measured simultaneously, which allows validation of some of the processes included in the models.

There are several difficulties in the assessment of model validity using observations. First, the recent multi-model papers show that there remain important differences between the different models, and that different models perform well in particular comparisons, but less well in others. Secondly, many models obtain a broadly satisfactory comparison with surface O_3 data but probably for a range of reasons. A good comparison is necessary, but not sufficient, for model validation. Furthermore, the O_3 distribution is not a good model discriminator. Campaign data on short-lived species is often difficult to compare against specific models because the exact emissions, meteorological conditions and other crucial factors, are imperfectly known. The rate of change of O_3 depends in a complex, non-linear fashion on the concentrations of NO_X and VOC. It seems clear that when assessing the future development of surface O_3, the model response to a perturbation depends critically on where the model O_3 production potential lies in this complex NO_X–VOC space. It is likely that current models, which nevertheless reproduce O_3 satisfactorily, represent this space differently. New ways of validating models for prediction need to be developed. With these caveats in mind, model projections of future O_3 include significant uncertainties. Nevertheless, the current generation of models are the best and only tools available to assess future O_3 exposure.

changes in climate. The most comprehensive study which evaluates the role of each of these drivers out to 2030 is the Atmospheric Composition Change European Network of Excellence (ACCENT) PhotoComp intercomparison. Dentener *et al.* (2006b) describe surface O_3 results from up to 26 different global models that participated in the ACCENT study. An envelope of three emissions scenarios were analysed for the year 2030: the upper case was the original IPCC SRES A2 scenario (Nakicenovic *et al.* 2000). The central case was the IPCC B2+IIASA CLE scenario, while the lower case was the IPCC B2+ IIASA MFR scenario (Dentener *et al.* 2005; Cofala *et al.* 2007b). Descriptions of these scenarios are provided in chapter 4. The scenarios were chosen to illustrate the range of possible anthropogenic emissions in the near future.

The ACCENT PhotoComp study also analysed potential impacts of climate change on surface O_3 by 2030, using a subset of 10 models (these impacts are discussed in section 5.2.2). Seasonal mean changes (see Box 5.2) in O_3 between 2000 and 2030 for June, July, August (JJA) from the ACCENT study are shown in Figure 5.2. The ACCENT study clearly shows that the evolution of anthropogenic emissions will be the main factor controlling surface O_3 in the near future and therefore that 2030 O_3 levels are largely under direct human control.

For the purposes of this study several new model simulations were performed to evaluate the relative roles of changes in emissions and changes in climate further into the future, to 2050 (Table 5.1). The ACCENT results were updated and extended to 2050 to enable analysis over the period during which the effects of climate change should be larger and therefore provide responses in the O_3 concentration field that are more easily detected.

The first simulation was a year 2000 base run (R1), using updated emission estimates with respect to the ACCENT study. These updates are detailed in Annex 3. In particular, the estimates were updated to include a more realistic treatment of ship emissions and larger emissions of Asian CO. The latest anthropogenic emissions scenario work discussed in chapter 4 was then used to simulate the effects of projected emission changes under the new B2+CLE scenario in 2050 (R2) which assumes the implementation of legislation current as at the end of 2006. This provides emission projections for NO_X and CO, but not for nmVOC. The nmVOC emissions were assumed to change in line with changes in CO. This assumption represents a significant uncertainty in the new analysis.
A further simulation was performed that included the effects of projected climate change in the 2050s combined with the new B2+CLE scenario (R3). To achieve this, the climate model components of the chemistry–climate models set:
(i) atmospheric composition to levels prescribed by the IPCC SRES A1B scenario in 2050 (Nakicenovic *et al.* 2000); and
(ii) sea-surface temperatures and sea-ice distributions to values previously calculated by the HadGEM1 climate model forced by the A1B scenario. A pre-industrial simulation (R4) was also performed together with a 2050 sensitivity study to isolate the effects of increases in global CH_4 (R5). A subset of up to six of the ACCENT global models performed these simulations. The multi-model mean results from these models are presented here. These model results are referred to in the report as the *'Royal Society 2007'* (RS07) simulations and their key characteristics are summarised in Table 5.1. It should be noted that these projections are snap-shots of the future for 2050, and are not continuous model runs for the period 2000–2050. The specifications of the RS07 simulations and participating models are detailed in Annex 3.

Figure 5.2 The mean change in surface O_3 (ppb) between 2030–2000 for the period June–July–August (JJA) from the ACCENT PhotoComp study (Dentener et al. 2006b; Stevenson et al. 2006), for three scenarios (B2+CLE; B2+MFR; and A2), and due to 2000–2030 climate change. The top left panel shows the mean change in surface O_3 under a B2+CLE scenario; the top right panel under a B2+MFR scenario; and the bottom left under an A2 scenario. The bottom right panel shows the mean change in surface O_3 due to climate change.

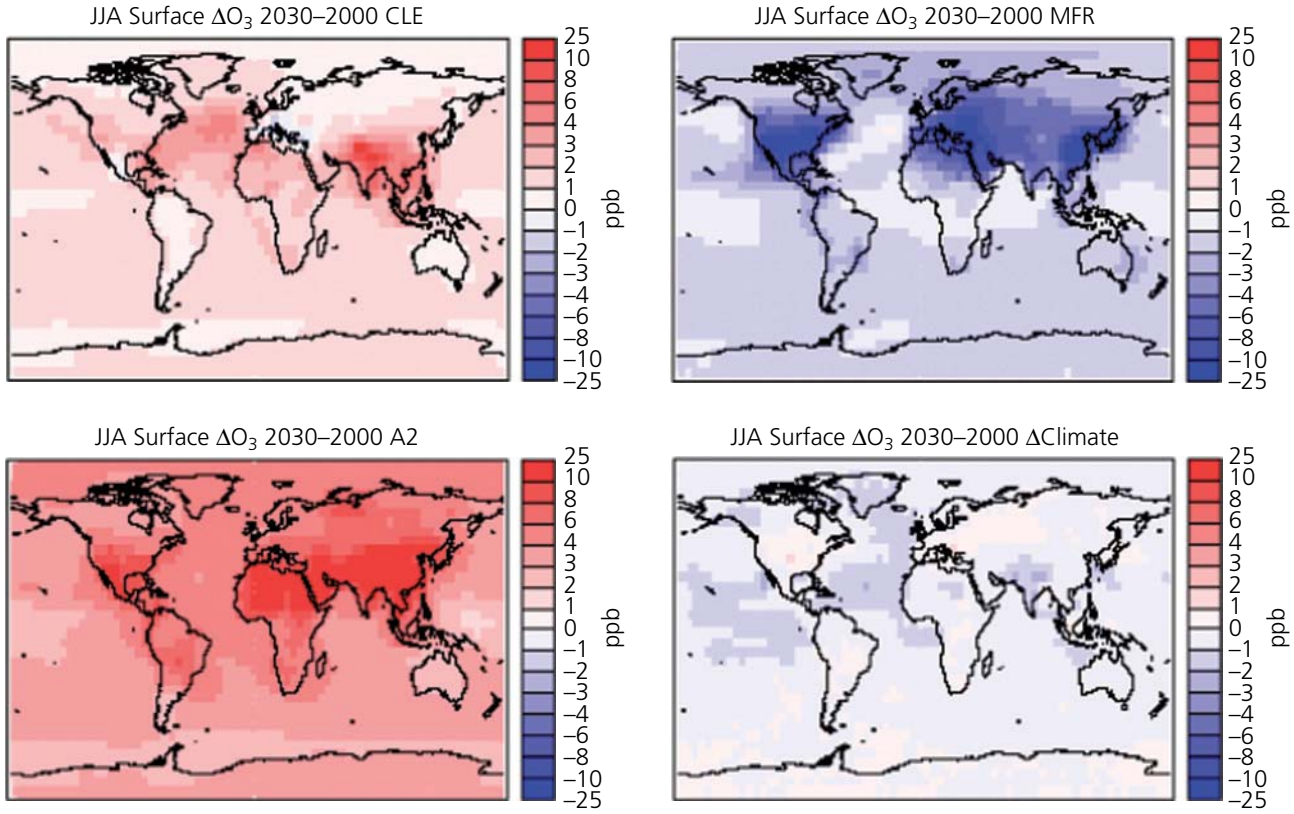


Table 5.1 'Royal Society 2007' (RS07) model simulations. 'Anthropogenic Emissions' refers to the appropriate year/scenario/amount for anthropogenic emissions (all natural sources and biomass burning are kept constant in all simulations except R3, where some models allowed climate-sensitive natural emissions to vary). In R4 anthropogenic emissions are set to zero. In all simulations except R3 a 'present-day' (or 1990s) climate is applied. The last column refers to the global mean CH_4 concentration (ppb), which is fixed for a given run.

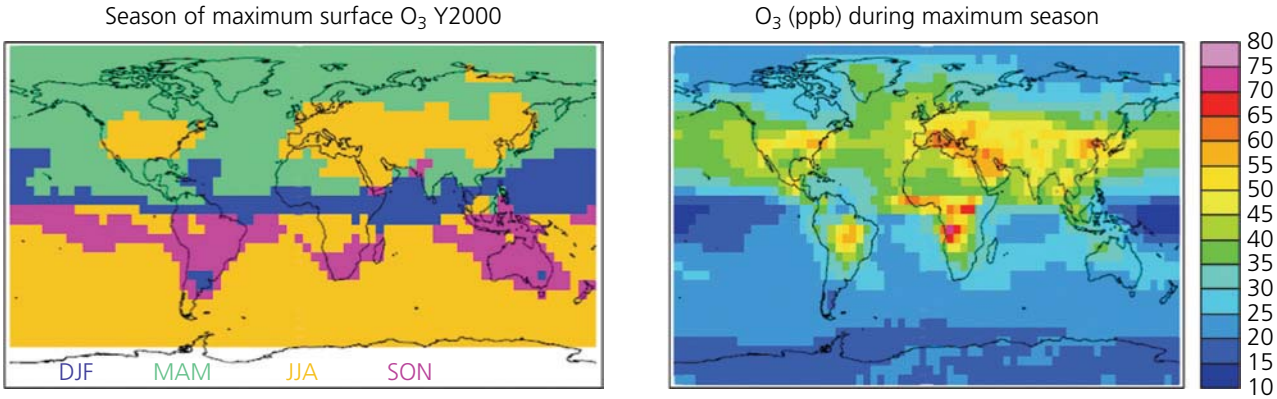
Model run	Brief description	Anthropogenic emissions	Climate	CH_4/ppb
R1	Year 2000 base case	2000	1990s	1760
R2	2050 emissions and CH_4	B2+CLE (2050)	1990s	2363 (B2)
R3	2050 emissions and CH_4; 2050s climate	B2+CLE (2050)	2050s	2363 (B2)
R4	Zero anthropogenic emissions; pre-industrial CH_4	Zero	1990s	700
R5	2050 emissions; 2000 CH_4	B2+CLE (2050)	1990s	1760

Box 5.2 When is the season of maximum surface ozone at a given location?

There is considerable spatial variation in the season of maximum surface O_3 across the globe. This is of prime interest for studies of air quality because policymakers need to know when the maximum concentrations of O_3 will occur in each region to enable an assessment of potential impact. For example, if a crop growing season in a region coincides with maximum concentrations the impacts to the crop will be larger than at other times of the year.

Figure 5.3 illustrates the season of maximum O_3 for present day conditions which is used in this analyses. As in Figure 5.2, studies often only show O_3 during June, July, August as this is generally the maximum season in polluted northern mid-latitudes. However, at other locations O_3 reaches a maximum in different seasons. This range of seasonality is due to the combination of annual cycles in photochemical production and destruction of O_3, and the strength of the stratospheric source, all of which vary spatially (eg Monks 2000). In the northern extra-tropics, surface O_3 reaches a maximum in spring over remote regions. This is in contrast to the southern extra-tropics, where O_3 almost universally reaches a maximum in austral winter. This is mainly because during summer photochemical destruction of O_3 exceeds production in the less polluted Southern Hemisphere. In much of the tropics O_3 reaches a maximum during the dry season when biomass burning emissions of O_3 precursors reach a peak.

Figure 5.3 The season of maximum surface O_3 for the year 2000. Left: season (December, January, February (DJF), March, April, May (MAM), June, July, August (JJA), and September, October, November (SON)) of maximum surface O_3. Right: corresponding maximum season surface O_3 values (ppb). Both panels use year 2000 results (see Table 5.1, R1).



5.2.1 Impact of changes in emissions on surface ozone

Previous modelling studies, including the ACCENT study have clearly shown that O_3 depends strongly on O_3 precursor emissions; in particular NO_X, but also CH_4, CO and nmVOC (Figure 5.2). The new model runs to 2050 support this conclusion. Figure 5.4 shows the difference between simulations R2 and R1, that is the change in seasonal mean surface O_3 projected under the new 2050 B2+CLE emissions scenario, relative to the year 2000. Most regions show reduced or near constant O_3 concentrations by 2050 due to lower future emissions, with improved air quality over much of the developed world. Over the North Eastern USA in summer, O_3 is projected to reduce by up to 15 ppb (an approximate reduction of 25% compared to present day levels). There are also substantial reductions over Europe and Japan. Ozone is generally reduced across most of the Northern Hemisphere mid-latitudes by about 5 ppb. One exception is during northern winter, when reduced NO_X emissions result in higher O_3 levels over Europe and North America (Figure 5.4). This is due to the reduced titration of O_3 by NO (see chapter 3). In some parts of Asia and Africa where there is very little current legislation in place and/or where significant economic growth is projected, modest increases in O_3 of up to 3 ppb (~7%) are experienced.

Figure 5.5 uses the global annual mean surface O_3 values to compare the various scenarios and time horizons considered in this study, the ACCENT PhotoComp study, and from the IPCC Third Asessment Report (Prather *et al.* 2003). Multi-model mean values are shown by the dots, while inter-model standard deviations are given by the bars, and give an indication of model uncertainty. Results from the 2050 B2+CLE scenario lie between the ACCENT 2030 B2+CLE and B2+MFR cases.

The B2+CLE and, in particular, the B2+MFR scenarios represent relatively successful futures in terms of emissions control policies. The IPCC SRES A2 scenario represents a policy fail situation which demonstrates that background O_3 levels will increase through the next century if NO_X, CH_4, CO and nmVOC emissions rise (Prather *et al.* 2003) and control legislation is not implemented. This is illustrated by the potential range of O_3 values in 2100 given in Figure 5.5 and represents the response of surface O_3 to the full range of IPCC SRES scenarios, as estimated by Prather *et al.* (2003). The high end of this range represents scenarios with very high emissions of O_3 precursors, such as A2 and A1FI. Conversely the low end represents the more optimistic SRES scenarios, such as B1 and A1T. It should be noted that the IPCC considered each scenario equally likely.

Figure 5.4 Projected change in surface O_3 for the four seasons between 2000 and 2050 due to changes in emissions following the new B2+CLE 2050 scenario (see Table 5.1). Top left panel: changes in surface O_3 for December, January, February (DJF). Top right panel: March, April, May (MAM). Bottom left: June, July, August (JJA). Bottom right: September, October, November (SON). (The results presented are the mean of 5 models.)

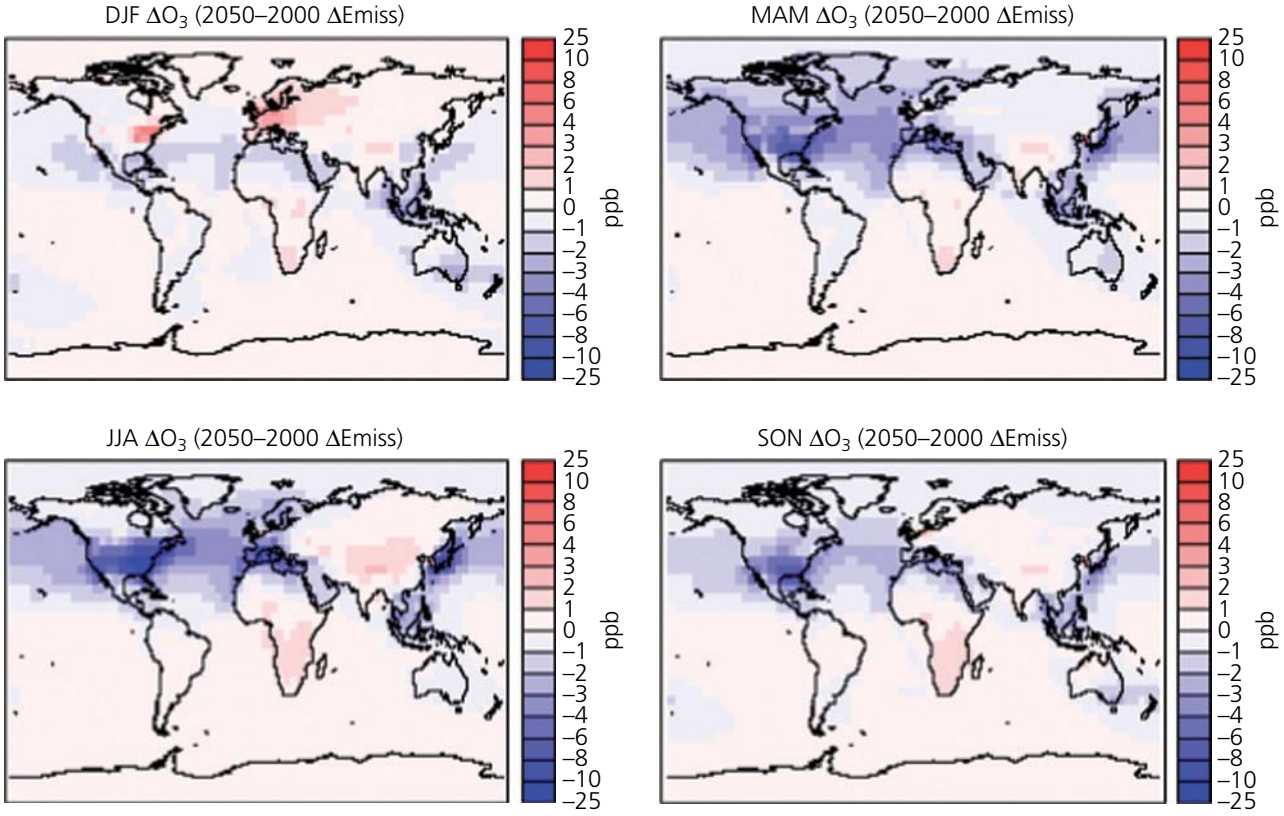
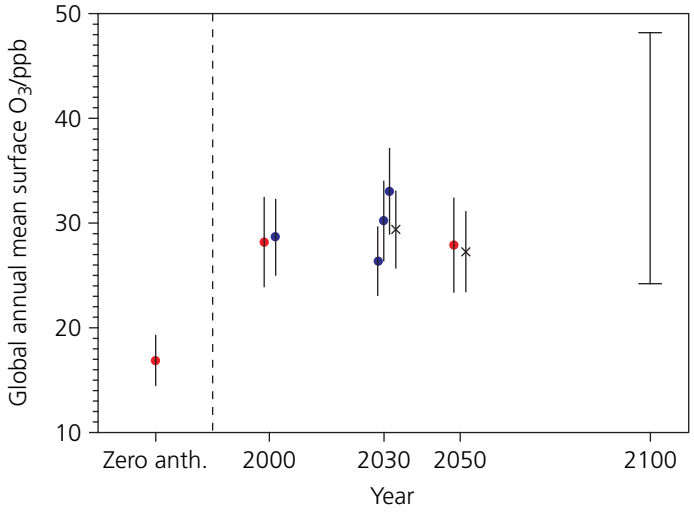


Figure 5.5 Multi-model global annual mean surface O_3 (ppb) for various scenarios from this study (RS07) (red symbols) and the ACCENT PhotoComp study (blue symbols). The bars indicate inter-model standard deviations. The three blue dots for 2030 are the B2+MFR (lowest value), B2+CLE and A2 (highest value) scenarios. The crosses are model results simulated with a future climate, as well as a future emissions scenario (the 2030 emissions scenario is the central one: B2+CLE). These show a small negative impact of climate change on global annual mean surface O_3 (see section 5.2.2). All other model simulations used a year 2000 climate. The bar shown for 2100 is the estimated range of O_3 responses to the full range of SRES scenarios (Nakicenovic et al 2000), as reported by Prather et al. (2003).



There are significant regional variations in O_3 concentrations which will have important implications for future controls. These are not evident from the annual mean O_3 concentration. Regional changes in O_3 concentrations are discussed in section 5.2.5.

5.2.2 Impact of changes in climate on surface ozone

Several global modelling studies have shown impacts of projected 21st century climate change on surface O_3 (eg Johnson *et al.* 2001; Hauglustaine *et al.* 2005; Liao *et al.* 2006; Racherla & Adams 2006). The ACCENT PhotoComp study identified a small, but statistically insignificant negative feedback of climate change on global annual mean surface O_3 in 2030 (Figure 5.5). However, the imposed climate change was small in the ACCENT model runs (global mean surface warming of ~0.7°C), and not all the modelled responses were consistent. The ACCENT results nevertheless confirm earlier studies showing a tendency for climate change to reduce O_3 over the very low NO_X environment of the tropical oceans. This was stimulated by higher absolute humidity associated with the warmer future climate (Held & Soden 2006).

Figure 5.6 shows the influence of climate change on surface seasonal mean O_3 in 2050, as revealed by the difference between RS07 simulations R3 and R2 (the combined 2050 climate and emissions run, and the 2050 emission runs respectively). The influence of climate change on future O_3 concentrations is spatially and temporally variable, but there is a general tendency for climate change to increase O_3 over polluted land regions and to reduce O_3 over the oceans with seasonal mean changes up to ±5 ppb. The causes of the increases over land are not fully understood, but may be related to increases in water vapour in comparatively high NO_X environments, which lead to enhanced net O_3 production (Isaksen *et al.* 2005). The increases may also be related to changes in meteorology, such as increased sunshine, or an increased incidence of stagnation. Increases in O_3 associated with climate change tend to coincide with tropical land regions with major emission sources from biomass burning or lightning, although the relationships are not clear. It should be noted that in these model experiments, lightning (and associated NO_X emissions) and biogenic VOC emissions (in two of the three models) respond to climate change, while biomass burning emissions remain fixed. Ozone–climate interactions are discussed in more detail in chapter 6.

5.2.3 Combined impacts of changes in emissions and climate on surface ozone

Figure 5.7 shows the combined effects of both changes in emissions and climate on surface O_3, given by the difference between simulations R3 and R1 (the combined 2050 climate and emissions run, and the base year 2000 run respectively). The figure demonstrates that emission changes are the main driver of future O_3 concentrations in most locations.

Figure 5.6 Projected change in surface O_3 for the four seasons between 2000 and 2050 due to climate change alone (see Table 5.1 (R3–R2)). Top left panel: December, January, February (DJF). Top right panel: March, April, May (MAM). Bottom left: June, July, August (JJA). Bottom right: September, October, November (SON). The results presented are the mean of three models.

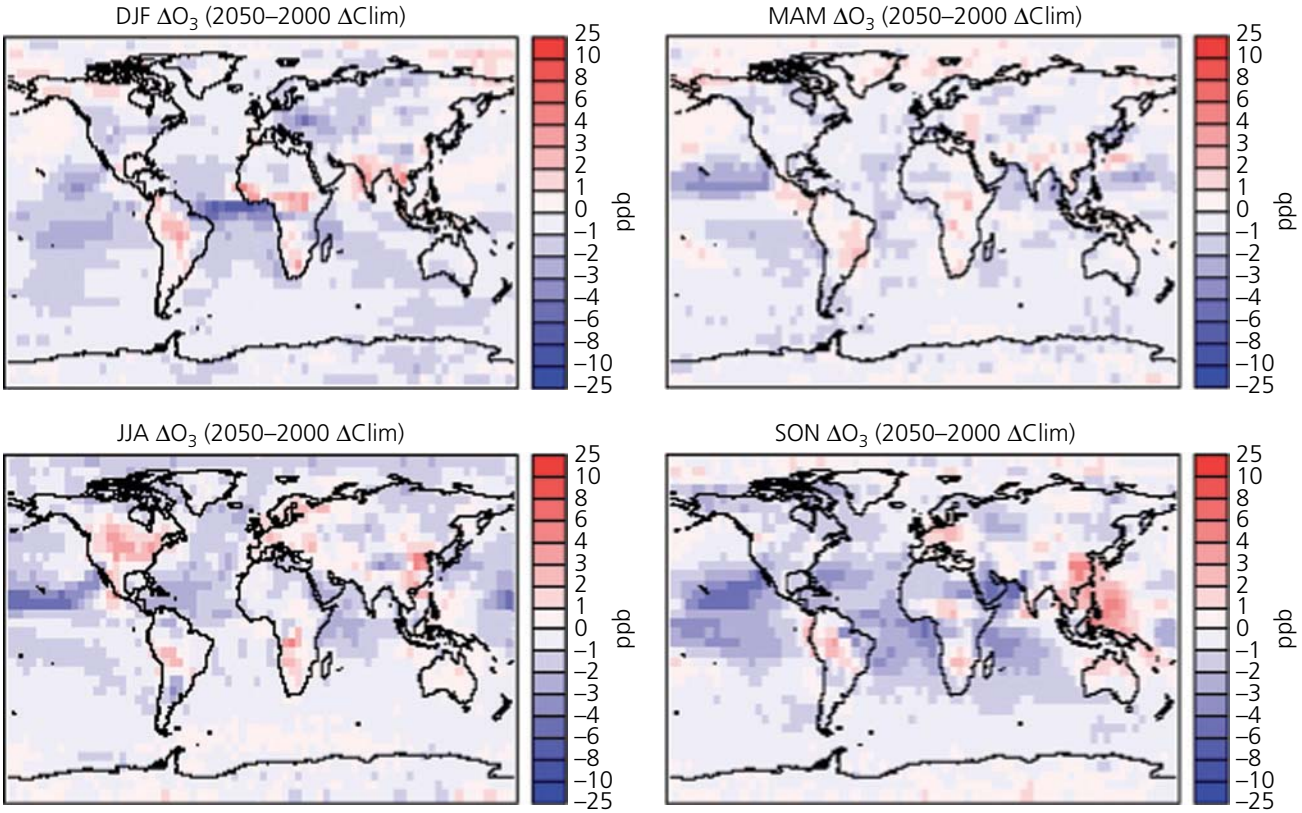


Figure 5.7 Projected changes in surface O_3 for the four seasons between 2000 and 2050 due to the combined effects of emission changes and climate change (see Table 5.1 (R3–R1)). Top left panel: December, January, February (DJF). Top right: March, April, May (MAM). Bottom left: June, July, August (JJA). Bottom right: September, October, November (SON). The results presented are the mean of three models.

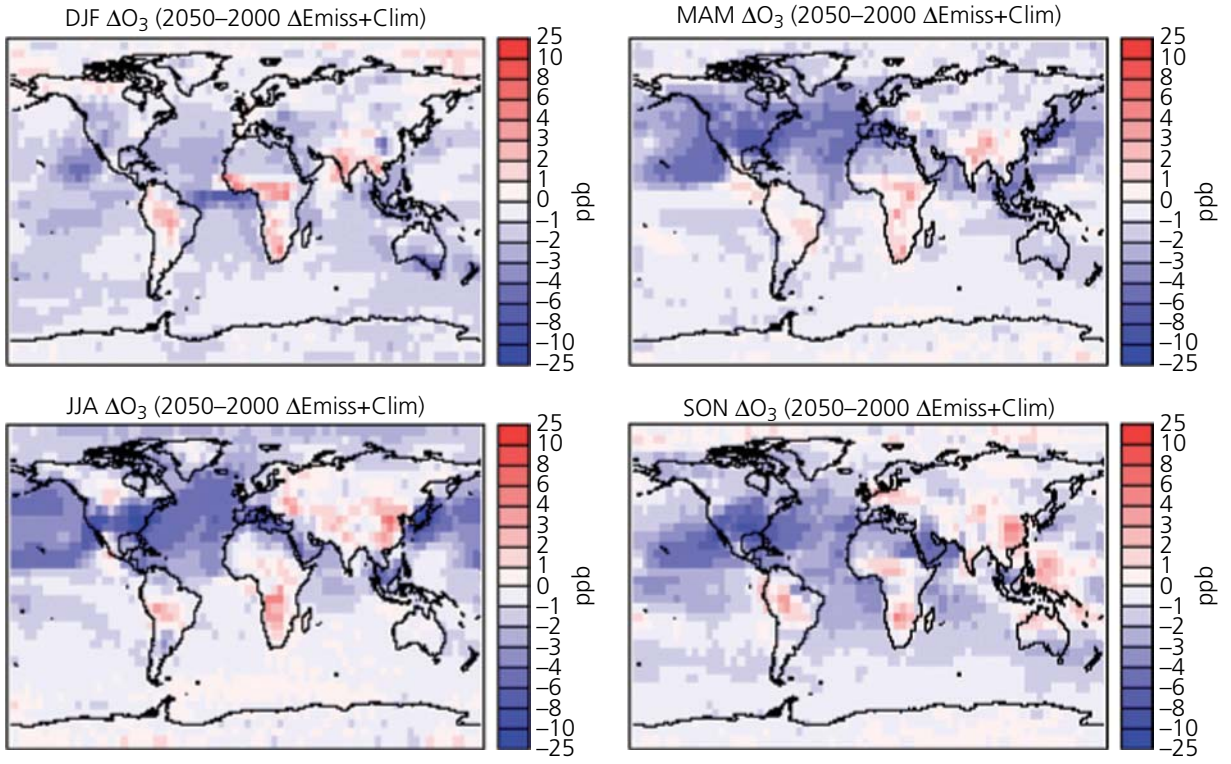
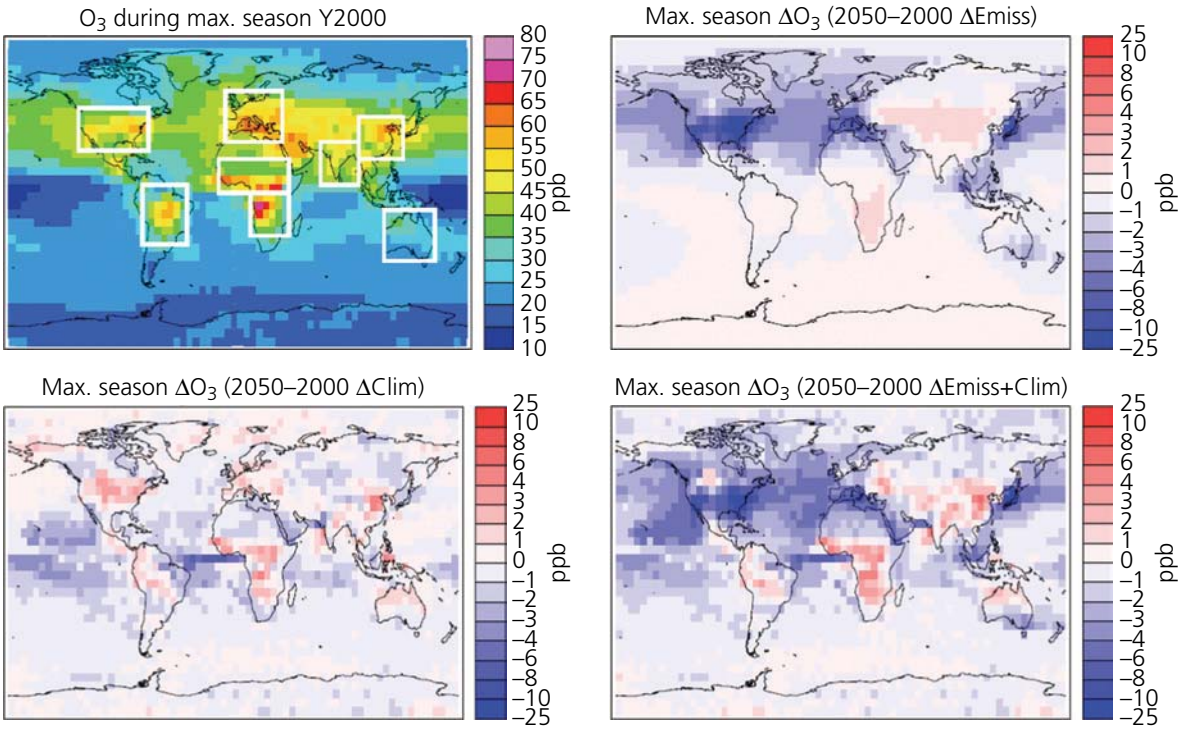


Figure 5.8 A comparison of seasonal mean surface O_3 values (ppb) during the maximum O_3 season due to changes in emissions, climate change and the combined effects of changes in emissions and climate change. Top left: seasonal mean surface O_3 values (described in Figure 5.3) during the maximum O_3 season, for the year 2000 (R1 in Table 5.1). Top right: the projected changes in O_3 (ppb) during its maximum season by 2050 due to emission changes only (R2–R1). Bottom left: due to climate changes only (R3–R2). Bottom right: due to the combined impact of both changes in emissions and climate (R3–R1). The white boxes in the first panel refer to the regions in Figure 5.9. The results presented are the mean of three models.



Where cuts in emissions are substantial, their impact dominates, for example over industrialised regions in northern mid-latitude summer. However climate change tends to reduce the efficacy of cuts in emissions. In an independent study, Wu *et al.* (2007b) found a similar result over North America.

Where projected changes in anthropogenic emissions are small, the effects of climate change on O_3 are proportionately larger. Increases in O_3 associated with climate change tend to coincide with tropical land regions as discussed in section 5.2.2.

To assess air quality impacts, it is useful to look at the changes in O_3 during the season of maximum O_3 (as discussed in Box 5.2). Figure 5.8 distils the results from Figures 5.4, 5.6 and 5.7 to show the impacts of: emissions, climate change and emissions and climate change combined, respectively, on surface O_3 during its maximum season. Many of the impacts of emission changes and climate change are increased during the maximum O_3 season. Figure 5.8 shows that many developing world regions, especially large parts of Africa and Asia, which already have high ground-level O_3 concentrations are projected to experience larger O_3 concentrations and therefore poorer air quality up to 2050, due largely to climate change. The climate change signal in these regions is more pronounced simply because changes in emissions are relatively small. Much of the developed world, where current legislation to reduce emissions is comparatively strong, shows improved air quality, but nevertheless incurs a 'climate change penalty' that will tend to reduce the benefits of emission reductions.

5.2.4 Impact of changes in global methane concentrations on surface ozone

The sensitivity run (R5) assessed the impact of keeping global CH_4 concentrations fixed at year 2000 levels (1760 ppb) rather than rising to the SRES B2 projected value (2363 ppb). Ozone values are 1–2 ppb lower in 2050 when CH_4 does not rise, as shown in Figure 5.9 in the upper panels, with the larger O_3 reductions occurring in the more polluted Northern Hemisphere, where NO_X levels are higher. Methane is clearly an important influence on background O_3 levels. Several studies have found similar results and have highlighted the potential co-benefits for both air quality and climate forcing that result from CH_4 controls (West & Fiore 2005; TFHTAP 2007).

It should be noted that the projected values of CH_4 for a given scenario, such as the 2363 ppb value for SRES B2 (IPCC 2001) were derived from relatively simple models. The future evolution of CH_4 depends on both sources (natural and anthropogenic emissions) and sinks. The dominant CH_4 sink is reaction with OH. The future evolution of OH itself strongly depends on the evolution of O_3 and its precursors such as NO_X and CO, so there is a strong non-linear coupling between the evolution of O_3 and CH_4. To produce more realistic simulations of this coupled system continuous runs of models driven by CH_4 emissions rather than prescribed concentrations are needed for the period 2000–2050. There are few examples of such fully coupled integrations (eg Johnson *et al*. 2001; Stevenson *et al*. 2005) however as they are computationally expensive. The future evolution of CH_4, even for a prescribed emission scenario, therefore remains a significant source of uncertainty for future O_3 levels.

5.2.5 Regional analysis of global model results

Global models provide regional information, albeit at a relatively coarse resolution (~200 km). Figures 5.9a and 5.9b show spatially averaged seasonal cycles of surface O_3 from the different simulations discussed above for various world

Figure 5.9a The seasonal cycle of modelled surface O_3 over Europe under various emission scenarios. The figure compares the new RS07 model runs with the ACCENT model runs. The lower panel shows the seasonal cycle in surface O_3 for the year 2000 (thick black line, R1), for 'pre-industrial' times (dashed green line, R4), and for the year 2050 using the new B2+CLE scenario (solid red line, R2), from the RS07 model runs. The shaded grey region shows the ACCENT 2030 results for three scenarios. The upper value is for the A2 scenario. The lower value is for the B2+MFR. The dotted white line shows the B2+CLE scenario. The upper panel shows the modelled change in O_3 relative to the 2050 B2+CLE scenario for two cases: inclusion of 2000–2050 climate change (solid cyan line, R3–R2) and for CH_4 fixed at year 2000 levels (dotted blue line, R5–R2) for the RS07 model runs.

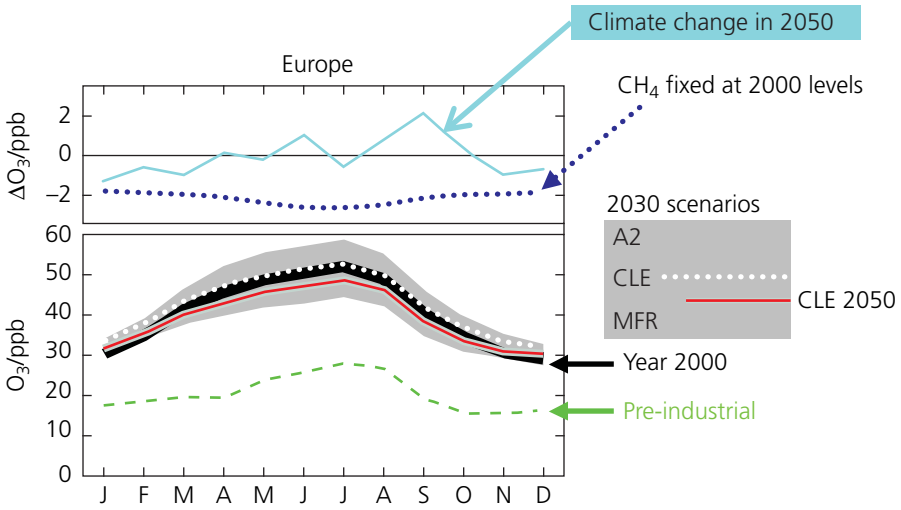
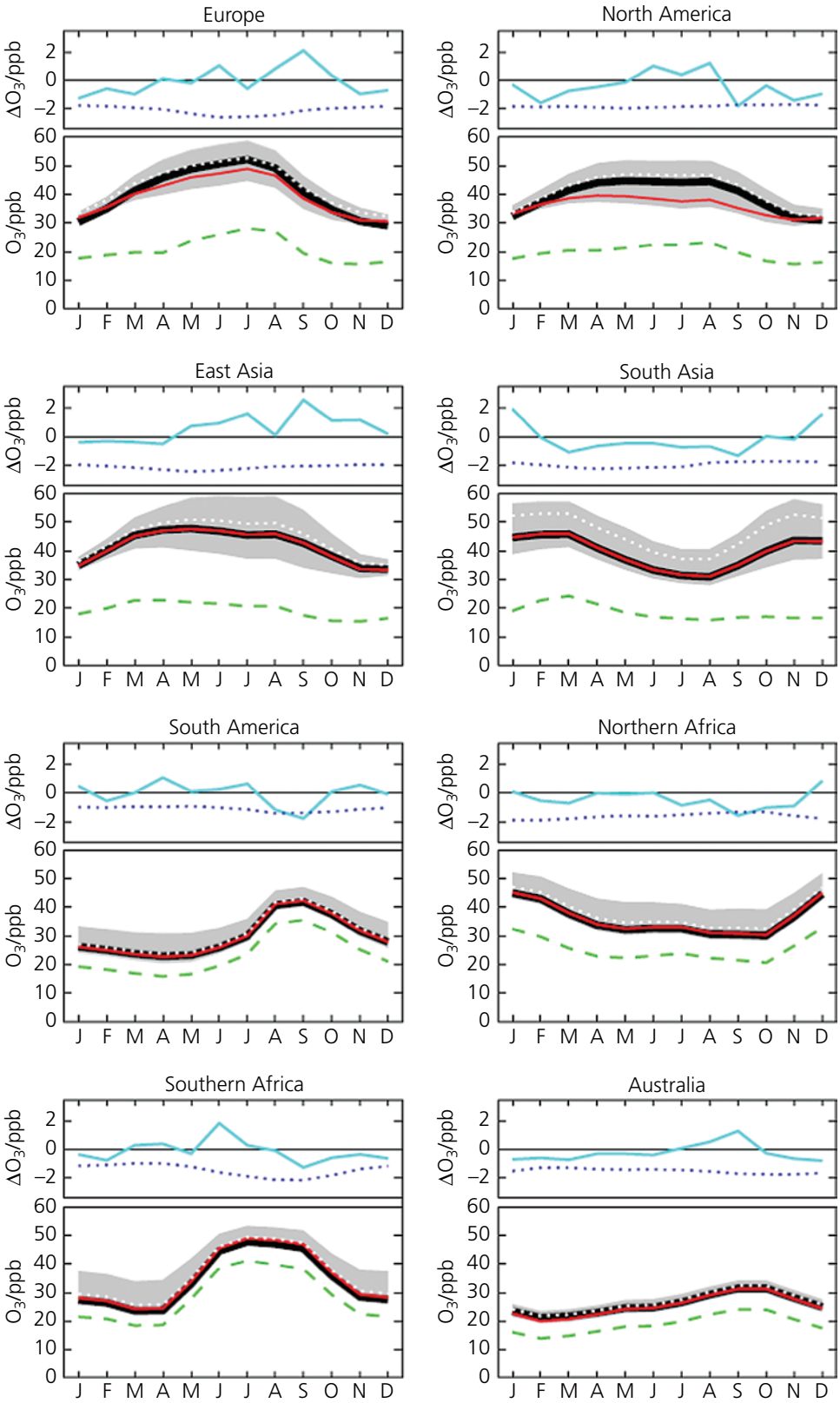


Figure 5.9b The seasonal cycle of modelled surface O_3 over a region under various emission scenarios. Each panel shows model results averaged over a different region (Europe, North America, East Asia, South Asia, South America, Northern Africa, Southern Africa and Australia). The different model results represented on the figure are described in Figure 5.9a.



regions, geographically defined by the boxes shown in Figure 5.8. By comparing year 2000 O_3 (the thick black line in Figures 5.9a and 5.9b) with levels simulated under zero anthropogenic emissions, and with CH_4 set to pre-industrial levels (the dashed green line), the magnitude of the anthropogenic impact on surface O_3 can be seen (Figures 1.2 and 5.9a and 5.9b).

For many regions a significant proportion, typically about half, or 20 ppb, of present-day (year 2000) O_3 is of anthropogenic origin and this is consistent with the observed increases in ground-level O_3 described in chapters 2 and 3. The largest anthropogenic increases occur in the industrialised regions of the Northern Hemisphere, where maximum monthly mean concentrations of O_3 currently reach up to about 50 ppb. East Asia, South Asia, and Europe, closely followed by North America and Northern/Southern Africa, are most at risk from very high O_3 levels under a 'policy fail' SRES A2-type scenario (represented by the upper bound of the grey bands in Figure 5.9b). East Asia, North America and Europe are the regions' most amenable to reductions in O_3 concentrations with strict emissions controls (the lower bound of the grey bands in Figure 5.9b, which represents the maximum reductions possible with emissions controls and technological change). The RS07 2050 new B2+CLE scenario results consistently fall within the 2030 envelope from the ACCENT PhotoComp results, with O_3 values roughly midway between the 2030 B2+CLE and B2+MFR results, although closer to the latter in North America and South Asia, due to significantly tightened legislation in the last few years in these regions. The upper panels of each plot show the relative contributions of CH_4 and climate change in 2050, as discussed above (see sections 5.2.2 and 5.2.4). The effects of holding CH_4 at year 2000 values (1760 ppb), rather than rising to 2363 ppb in 2050, is a reduction in O_3 in all regions, of 1–2 ppb with a seasonal cycle in the effect, shown as the dotted blue line in the upper panels of Figure 5.9b. The effect of climate change is also shown in the upper panels in the different regions as the solid cyan line. These model runs show enhanced warm season O_3 values in the polluted regions of 1–2 ppb O_3, and declines in the remote regions and in the cool seasons of polluted regions, of typically 1 ppb.

5.3 Regional modelling

There have been several recent regional modelling studies which have considered future O_3 levels over Europe (Langner *et al.* 2005; Szopa *et al.* 2006; Van Loon *et al.* 2006; Vautard *et al.* 2006), over North America (Hogrefe *et al.* 2004; Tagaris *et al.* 2007), and over East Asia (Yamaji *et al.* 2006). These studies typically nest regional models in global or hemispheric models that can simulate changes at the larger scale. These regional studies together with the global model results presented above, demonstrate that future changes in surface O_3 will be spatially heterogeneous, with different drivers in different regions and seasons. In some places (eg remote locations) changes in background O_3 driven by changes in CH_4 and hemispheric emissions of O_3 precursors subject to long-range transport will dominate. At other locations (eg polluted regions) changes in local anthropogenic emissions, driven by legislation and/or changes in socioeconomics will be the major control. In addition, any changes in natural emissions, chemical processing of O_3 precursors, meteorology or deposition rates associated with climate change, will exert seasonally varying influences in different regions.

Several attempts have been made to investigate the main controls on regional scale O_3 concentrations associated with changes in local emissions and climate, and how these relate to the controls at the larger scale to influence background O_3 concentrations. Model sensitivity runs, changing key emissions and climate-derived parameters (eg. temperature and water vapour) which affect the production and destruction rate of O_3, isoprene emission rates (from vegetation) or dry deposition of O_3 to the surface, are used to test the effects of each of these parameters on O_3 concentrations. For example, the rate of dry deposition of O_3 to the surface can be severely affected during heat-wave events due to the closure of plant stomata in response to temperature, soil moisture and drought, which reduces the amount of O_3 removed from the atmosphere by plants.

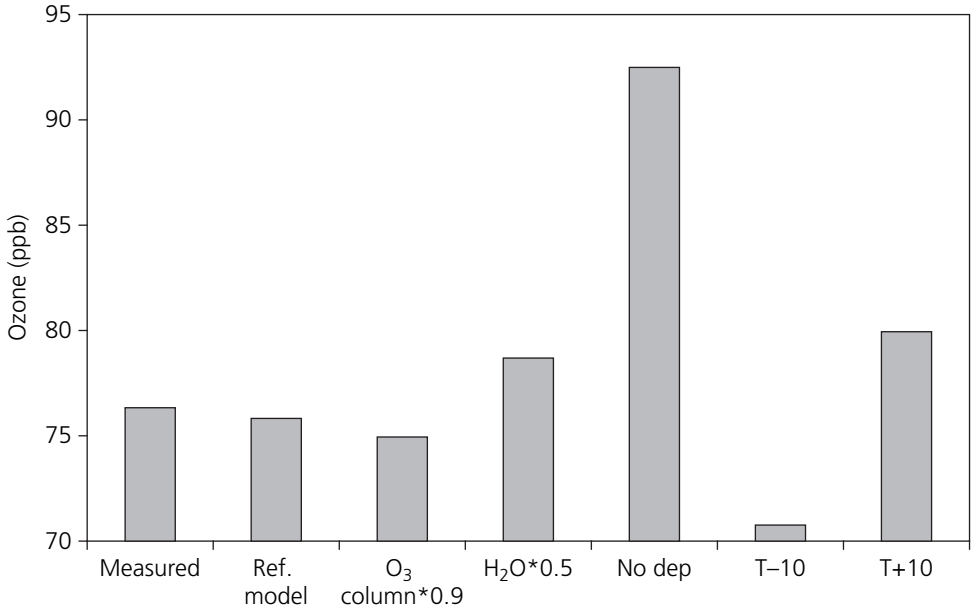
Two examples of sensitivity runs are provided here, one by Solberg *et al.* (2008) looking at climate derived parameters, and new sensitivity work completed for this study on both emissions and climate-derived parameter changes out to 2050.

Solberg *et al.* (2008) used a global chemistry–transport model to investigate the way climate related parameter changes may affect surface O_3 during high pressure weather situations with elevated O_3 concentrations, focussing on the 2003 European heat wave. Figure 5.10 shows the measured maximum hourly O_3 concentration for 8 August 2003 (at the peak of the 2003 heat wave) averaged over eight surface measurement stations in West and Central Europe. Figure 5.10 compares the measured concentrations with O_3 values calculated from a reference model run, and sensitivity runs with the O_3 column reduced by 10%, water vapour reduced by 50%, zero surface deposition and the temperature decreased and increased by 10°C inside a sub-domain of West and Central Europe (the sub-domain is confined by 10°E to 15.5°W and 36°N to 56°N and below 3.5 km height).

The effect of parameter changes on O_3 production in Solberg *et al's* work can be ranked in order of importance. Turning the dry deposition to zero produces the largest effect during heat-wave events. By setting the dry deposition to zero this sensitivity run explores the sensitivity of O_3 production to some stress-induced changes in the uptake of O_3 by plant stomata, due to drought, soil moisture or high temperatures for example (see chapters 3 and 6). Increasing the temperature by 10°C produces the next largest effect, followed by reducing the water vapour concentration by 50%.

In some areas the enhanced isoprene emission during the August 2003 heat wave (see case study in chapter 6) could have elevated the peak O_3 concentration by as much as 20 ppb, demonstrating the importance of isoprene emissions for O_3 production during heat-wave events.

Figure 5.10 Average maximum hourly O_3 concentration at eight surface measurement stations on 8 August 2003 as measured and calculated in the reference model run with the Oslo CTM2, and in sensitivity runs with the O_3 column reduced by 10%; water vapour reduced by 50%; zero surface deposition; and the temperature decreased and increased by 10°C inside the sub-domain confined by 10°E to 15.5°W and 36°N to 56°N and below 3.5 km height. Source: Solberg et al. 2008. NB: for clarity, the O_3 scale does not extend to zero, thus the largest modelled effect, due to switching off surface deposition, amounts to a 22% increase in maximum hourly O_3. Copyright (2007) American Geophysical Union. Reproduced/modified by permission of American Geophysical Union.



In this section new regional model results are presented which test the effects of climate and emissions related parameter changes on regional O_3 concentrations, over North-West Europe and the southern UK in particular. The importance of a range of different factors that influence O_3 using a photochemical trajectory model over Europe are tested along with some simple sensitivity tests. Details of the photochemical trajectory model are provided in Derwent *et al.* (1996). In this application of the model, for each day of 2005, thirty 4-day trajectories that arrive at the rural location of Harwell in Southern England in mid-afternoon were randomly selected. These 10,950 model runs sampled much of North-West Europe.

In the first sensitivity test which evaluated the effect of changes in emissions on future O_3 levels, Europe-wide O_3 precursor emissions were changed from their base case 2000 values to values appropriate to the year 2050 by simple scaling. NO_X and CO emissions were scaled to produce across-the-board reductions as indicated in the new B2+CLE scenario discussed in chapter 4. Details of the model results for maximum 1-h and annual mean of the daily maximum 1-h O_3 are presented in Figures 5.11 and 5.12, respectively.

For the sensitivity runs, a substantial decrease (~8 ppb) in episodic peak O_3 concentrations (maximum 1-h O_3) by 2050 occurs due to reductions in anthropogenic emissions of O_3 precursors (compare top two bars of Figure 5.11). The 2050 annual mean daily maximum O_3 concentration across these sensitivity runs ranges from 32.5 to 41.5 ppb. This compares to the 2050 base case of 35 ppb, and the 2000 base case of 36.5 ppb. The reduction in the annual mean daily maximum O_3 concentration (top two bars of Figure 5.12) is anticipated to be somewhat smaller (~1.5 ppb) than the reduction in episodic peak O_3 concentrations because reductions in daily maximum O_3 during the summertime are partly counteracted by increases in this metric during the spring-time. Additional sensitivity runs involved increasing or decreasing baseline O_3 levels by +0.3% per year or –0.2% per year over the time period 2000–2050. This range of baseline trends was taken from earlier global modelling work using the B2+CLE, B2+MFR and IPCC SRES A2 scenarios out to the year 2030 (Derwent *et al.* 2006), and linearly extrapolating to 2050.

Further sensitivity tests were then made, relative to the 2050 emission case to test the effects of climate derived changes on future European O_3 concentrations. These tested the influence of: a 5°C increase in temperature, switching off surface destruction (dry deposition) of O_3 due to the effects of drought on vegetation, and increasing isoprene emissions by a factor of 100. Major influences were found in both episodic peak and long-term mean O_3 concentrations from changes to the surface exchange processes involved with O_3 surface destruction (dry deposition) and isoprene emissions (lowest two bars of Figures 5.11 and 5.12). For example, switching off surface deposition caused a 31% increase in episodic peak O_3 concentrations and a 19% increase in annual mean daily maximum 1-h O_3. These results are in broad agreement with the global model results of Solberg *et al.* (2008), discussed earlier. They demonstrate that these

Figure 5.11 Results from simple sensitivity tests carried out with a photochemical trajectory model examining the changes in maximum 1-h O_3 concentrations between 2000 and 2050 at Harwell. NB: for clarity, the O_3 scale does not extend to zero, thus the largest modelled effect, due to switching off surface deposition, amounts to a 31% increase in maximum 1-h O_3.

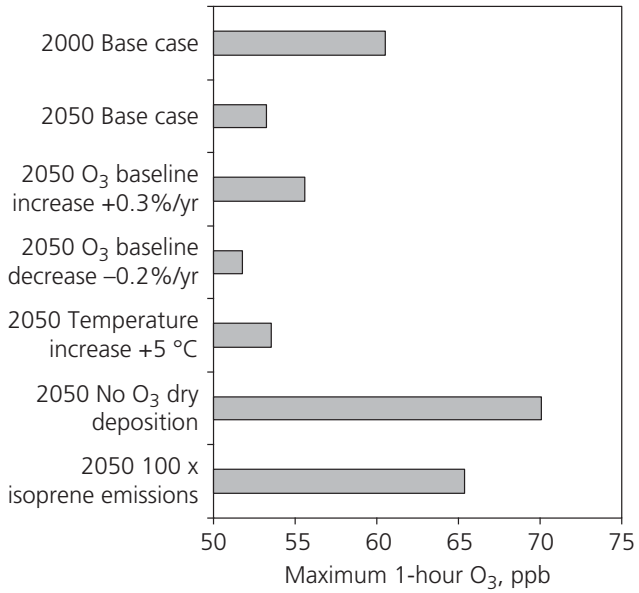
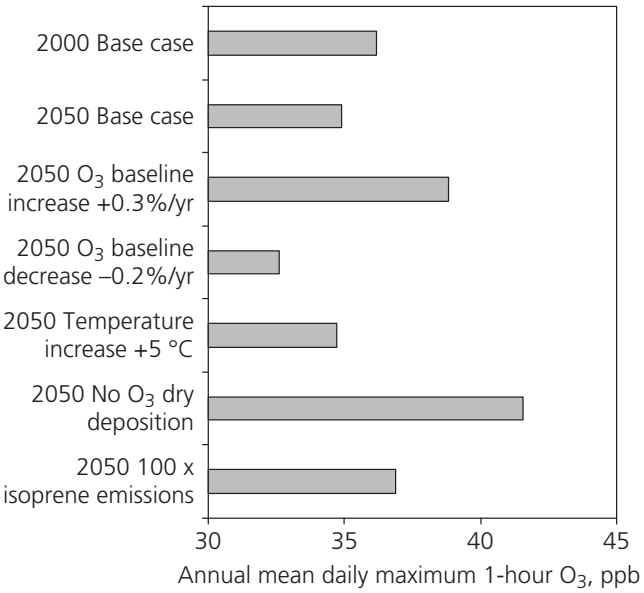


Figure 5.12 Results from simple sensitivity tests carried out with a photochemical trajectory model examining the changes in the annual mean of the daily maximum 1-h O_3 concentrations between 2000 and 2050 at Harwell. NB: for clarity, the O_3 scale does not extend to zero, thus the largest modelled effect, due to switching off surface deposition, amounts to a 19% increase in annual mean daily maximum 1-h O_3.



climate-related parameters could lead to elevated O_3 concentrations under future climate change, particularly during heat-wave events. However, it is clear that more research is needed to understand how these processes work and to predict how climate change may influence regional O_3 concentrations in the future.

5.4 Urban modelling

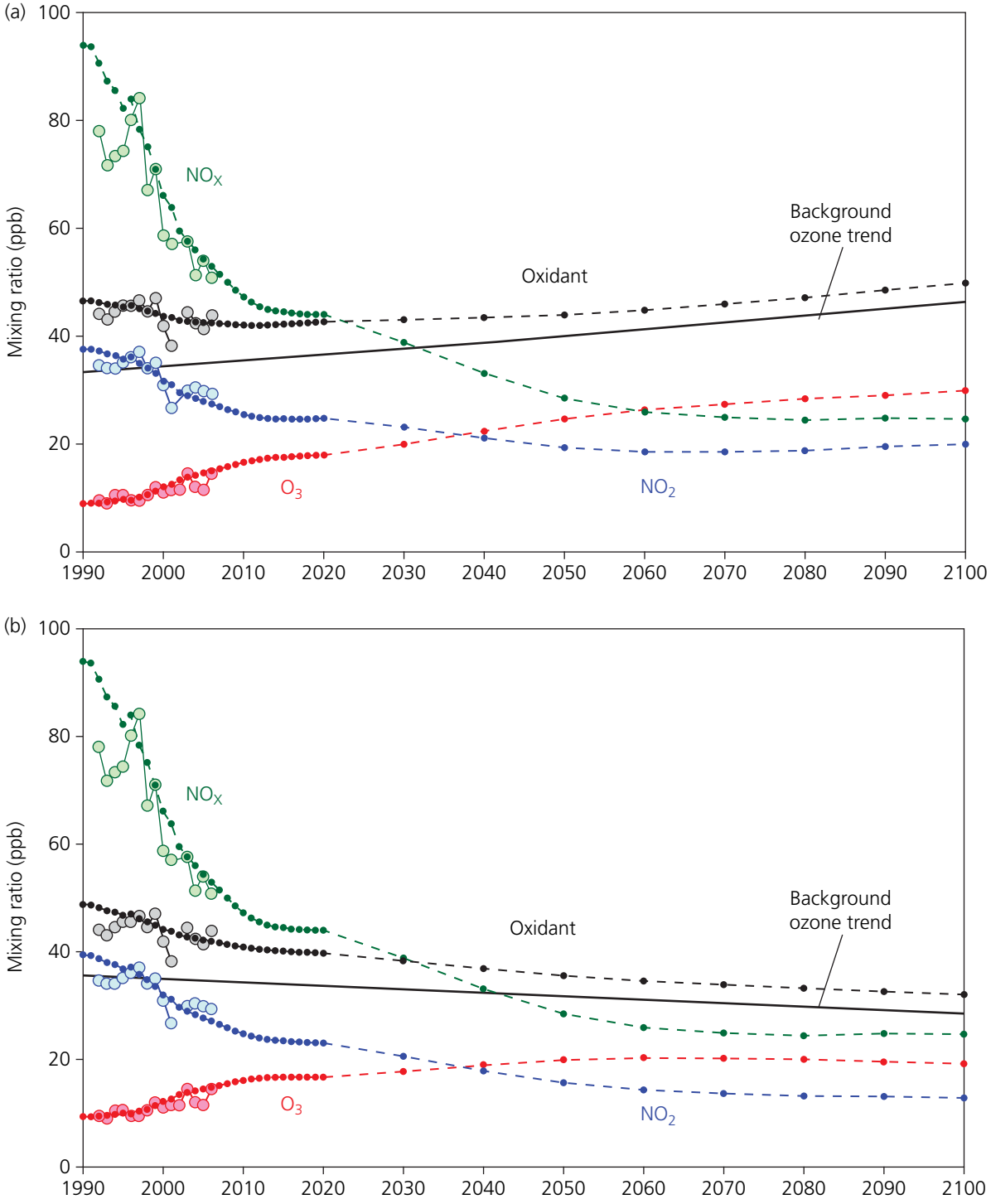
At the urban scale, projected O_3 precursor emissions controls (especially reductions in NO_X emissions) and changes in background O_3 concentrations will lead to changes in urban O_3 concentrations, with potentially large increases by the end of the century depending on the future scenario. As described in section 3.6, the changes in urban O_3 concentrations result partly from reductions in the titration of urban O_3 by reaction with emitted NO, but are also influenced by changes in background O_3 concentrations. This is because the reduction in the titration effect increases O_3 concentrations up to the background level.

Existing urban model runs for the UK only consider the 2020 time horizon (eg AQEG 2008) and these appear to be the only urban model runs available to examine possible changes to the end of the century. In this section one example of urban modelling work for London (UK) projected in the future to 2100 and completed in support of this study is presented. This work was done using a Pollution Climate Model (PCM) which, as described in section 3.3.2 and AQEG (2008), is an empirically-based model which uses an oxidant partitioning methodology, in conjunction with NO_X emissions maps and projections, to assess current trends and future levels of NO_2 and O_3 in relation to UK air quality objectives. This illustration provides example projections of annual mean O_3 for an urban centre location in central London under two scenarios representing either gradually increasing background O_3 (+0.3% per year) or gradually decreasing background O_3 (–0.2% per year). These are based on projected background O_3 trends for central and southern UK, as described in the sensitivity tests in the preceding section. It should be noted that this example is for a typical central London location, but is not necessarily representative of other locations either within London or in other urban areas. However, it does provide an indication of the main drivers of O_3 change at the urban level for an important urban centre locale and how they may change in the future under different scenarios.

As indicated above, the new urban modelling work takes account of both illustrative changes in the background concentrations of O_3 and the local NO_X titration effect over the century. Changes in annual mean O_3 concentrations (see chapter 3) have been simulated to the end of the century with local NO_X reductions based on the UK National Atmospheric Emissions Inventory (NAEI) projections up to 2020, and the IIASA B2 projections for Europe from 2020 to 2100 (see Figure 5.13). The effects of climate change on urban O_3 concentrations are not considered in this modelling work.

The results show that when either the increasing or decreasing trend in the level of background O_3 is used, reductions in local NO_X emissions from regional air pollution

Figure 5.13 Small points indicate illustrative simulated trends in annual mean O_3, NO_X and NO_2 for the London Bloomsbury site over the period 1990–2100 using an empirical measurements based Pollution Climate Model (PCM) for projected reductions in NO_X and two specified trends in background O_3 (a) +0.3% per year; (b) –0.2% per year. Large points are observed mixing ratios shown for 1992–2006.



mitigation policies progressively reduce the extent of O_3 titration over the period, leading to an increasing O_3 concentration at the urban location, and to increased population exposure to O_3. In the first case, this increase is exacerbated by the increasing background O_3, such that the simulated annual mean O_3 at the example central London location increases substantially from 12 ppb in 2000 to 30 ppb in 2100. In the second case, the decreasing background O_3 has a compensating influence, such that the increase in O_3 is effectively halted by 2050 and remains close to 20 ppb for the remainder of the century. The results are summarised in Figure 5.13 and Table 5.2.

Although these results represent only the effects in the example location, they do show that the dominant driver of urban O_3 concentrations is the extent of the NO_X titration effect and the

Table 5.2 Summary of inputs and simulated annual mean mixing ratios of O_3, NO_X and NO_2 for the London Bloomsbury site in 2000, 2050 and 2100.

	Example (a)[1]			Example (b)[1]		
	2000	2050	2100	2000	2050	2100
NO_X (ppb)[2,3]	66.0 (58.6)	28.3	24.6	66.0 (58.6)	28.3	24.6
Background O_3 (ppb)[1]	35.0	40.5	47.0	35.0	31.5	28.5
O_3 (ppb)[3]	12.0 (11.0)	25.0	30.0	12.0 (11.0)	20.0	19.0
NO_2 (ppb)[3]	31.9 (30.9)	19.4	19.9	31.9 (30.9)	15.7	12.6

Notes

[1] Applied O_3 background trend is +0.3% y^{-1} for (a) and –0.2 % y^{-1} for (b) (see text and Figure 5.11); [2]NO_X concentrations normalised to measurement in 2003. Trend inferred on the basis of the NAEI (http:www.naei.org.uk) NO_X emissions up to 2020 and IIASA B2 for Europe from 2020 to 2100; [3]Observed mixing ratios are given in brackets, where appropriate.

importance of the hemispheric background at these future dates. They show that if background concentrations of O_3 continue to increase across the globe, this will exacerbate urban O_3 concentrations. If they decrease, however, this will have a reducing influence on urban O_3 concentrations.

5.5 Uncertainties in model projections

All of the above model projections are subject to uncertainties, arising from a variety of sources. The evolution of future anthropogenic emissions will be driven by socioeconomic conditions, the strength of legislation to manage global air quality and the degree to which this legislation is enforced. Future emissions from Asia represent a major uncertainty. The rapid growth of relatively unregulated sectors such as shipping also contribute to the uncertainty surrounding the evolution of future emissions, as the future growth and regulation of these sectors is currently unclear. These key determinants of O_3 levels are inherent uncertainties for the future.

The RS07 2050 simulations assume ship NO_X emissions will fall by 50% from present-day levels by 2050, due to the implementation of significant legislation by this sector (see chapter 4; Eyring *et al.* 2005b: scenario DS2-TS2). However, this is highly uncertain. Average growth rates in total seaborne trade (in ton miles) between 2002 and 2007 have been 5.2% y^{-1}, significantly higher than in the past decades (Fearnleys 2008; Eyring *et al.* submitted). Total ship emissions are likely to have grown at a similar rate, and given the lack of current legislation for this sector, the assumed NO_X emissions reduction of 50% by 2050 may be too optimistic.

Additional uncertainties arise due to the models used to make future projections, which themselves contain many ill-constrained processes. The bars on Figure 5.5 represent one standard deviation from the inter-model results. It is unclear if such inter-model ranges represent the full range of model parameter uncertainty (Vautard *et al.* 2006), or if the outlying models that expand this uncertainty range should be considered invalid and excluded (Stevenson *et al.* 2006), which would reduce the range.

Modelled O_3 sensitivities to emissions changes typically show less variability between models compared to the abilities of models to simulate absolute levels accurately (Stevenson *et al.* 2006). As discussed above, it is unclear if this inter-model variability is a robust indicator of model uncertainty, but it is the only measure currently available. Model O_3 sensitivities to climate change span a much larger range (and are hence more uncertain).

Previous multi-model studies have failed to show a consensus on the overall sign of the climate feedback, never mind the magnitude (Stevenson *et al.* 2006). Results from this study show a greater degree of agreement, but this is partly because a smaller subset of models has been employed and partly because the imposed future climate was more tightly constrained.

This lack of consensus is unsurprising, and reflects inclusion of different parameterisations across the range of models, each with varying sensitivities to climate change, across both mainly physical processes (such as those that determine future surface temperature) and mainly chemical processes (such as those that determine future surface O_3). In addition, many of the key processes of climate–O_3 interaction are currently not included in models, or only included in very rudimentary ways (eg biogenic emission changes, biospheric couplings, land-use change). The impacts of climate change on O_3 at the urban level have not been assessed, but the feedbacks seen at the larger scale are also expected to be important at smaller scales. These gaps in our knowledge and resultant modelling uncertainties with respect to climate–O_3 interactions are explored more deeply in the next chapter.

5.6 Conclusions

Global Ozone

Models indicate that the primary influence on ground-level O_3 concentrations in 2050 will be anthropogenic emissions

of mainly NO_X, but also CH_4, CO and nmVOC. The 2050 O_3 projections presented in this chapter were based on future emissions that combined the IPCC B2 socioeconomic scenario with the assumption that emissions control legislation adopted by individual countries at the end of 2006 would be fully implemented by 2050. Under these conditions, O_3 concentrations in 2050 are projected to be near constant or reduced in most regions during the maximum O_3 season (relative to 2000). During summer, reductions of 5 ppb are projected for the Northern Hemisphere mid-latitudes. Over parts of North America reductions of up to 15 ppb (an approximate reduction of 25% compared to present-day levels) are projected. During the Northern Hemisphere winter an increase in O_3 is projected due to lower NO_X emissions and reduced titration by NO_X. During the maximum O_3 season over much of the developing world, an increase in O_3 of up to about 3 ppb (approximately 7%) is projected due to rapid economic growth in these regions.

The results show that for this particular scenario, emissions changes are generally more important for determining 2050 O_3 concentrations than the effects of climate change. Nevertheless, climate change will also have an impact. Global models indicate that climate effects on surface O_3 will be regionally variable with impacts during the maximum O_3 season in the range of ±5 ppb. The sign of the impact appears to depend on the underlying NO_X regime: climate change tends to increase O_3 production in already polluted environments, while decreasing it in clean environments. This can largely be understood in terms of the influence of higher levels of water vapour associated with a warmer atmosphere on O_3 chemistry under different NO_X regimes. The analysis shows that those developing world regions with already high O_3 concentrations, such as large parts of Africa and Asia, will experience poorer air quality in 2050 largely because of climate change. In other regions with stricter controls in place, air quality will improve, although climate change will reduce the benefits of control measures.

Global modelling also demonstrates the important role of CH_4 in determining future O_3 concentrations. While there are expectations of further increases in CH_4 emissions, uncertainty in current sources and sinks are too large to present a confident interpretation of recent trends or projections into the future. If CH_4 remains at present day levels (1760 ppb) rather than rising to levels projected by the B2 scenario in 2050 (2363 ppb) the new modelling indicates that global average surface O_3 levels will be 1–2 ppb lower. Controlling CH_4 therefore presents an obvious target for regulation.

Ozone at the regional scale

Future changes in O_3 at the regional scale (100–1000 km scale) will vary seasonally and according to the drivers of change in each region. At this scale the distribution of O_3 is controlled by the hemispheric baseline levels of O_3 in air masses arriving into the region (usually from the west), by O_3 production and destruction from O_3 precursor emissions within the region, and by local scale (1–100 km) O_3 sources and sinks.

To provide the probable scale of these changes in Europe for example, over the UK and North West Europe, reductions in O_3 concentrations by 2050 are projected under the new B2+CLE scenario as a result of NO_X and CO emission reductions. Sensitivity analyses suggest that episodic peak and long-term mean concentrations may be affected by changes in the climatic parameters important for dry deposition and O_3 production from the oxidation of isoprene. The effects of O_3 on reductions in dry deposition in particular may be large and should be further investigated.

Ozone at the urban scale

The analysis of future changes in O_3 concentrations at the urban scale suggests that the exposure of urban populations to O_3 will increase in many cities by 2050 due to reductions in local NO_X concentrations. The urban modelling shows that as NO_X emission controls are implemented and emissions decline (as recently seen in the UK for example), urban O_3 concentrations will rise towards the background concentrations. An increase in the background will further exacerbate O_3 increases at a local level, while a decrease will have a counteracting effect. In the case of a large decrease in the background, less polluted urban areas could potentially experience a net reduction in annual mean O_3 levels. The effects of climate change on urban O_3 were not evaluated explicitly and could significantly modify the long-term change in O_3 concentrations in these areas.

6 Climate–ozone interactions

6.1 Introduction

The results presented in the previous chapter suggest that future changes in climate may not have a major influence on O_3 concentrations globally, but that it is more important at the regional and local scales. However, many of the climate processes which control O_3 at these scales are not yet captured well within global atmospheric chemistry models. This chapter therefore provides a brief evaluation of which climatic processes may influence O_3 concentrations in the future. It also highlights the role of tropospheric O_3 as a greenhouse gas.

The production of O_3 is controlled by temperature, sunlight and humidity, and by the long-range transport of pollutants all of which are sensitive to changes in climate. Many of the processes creating or destroying O_3 or delivering it to ground level are influenced by synoptic[8] and local weather patterns, which also provide the pathways for the long range transport and for the ventilation of O_3 and its precursors from the boundary layer to the free troposphere.

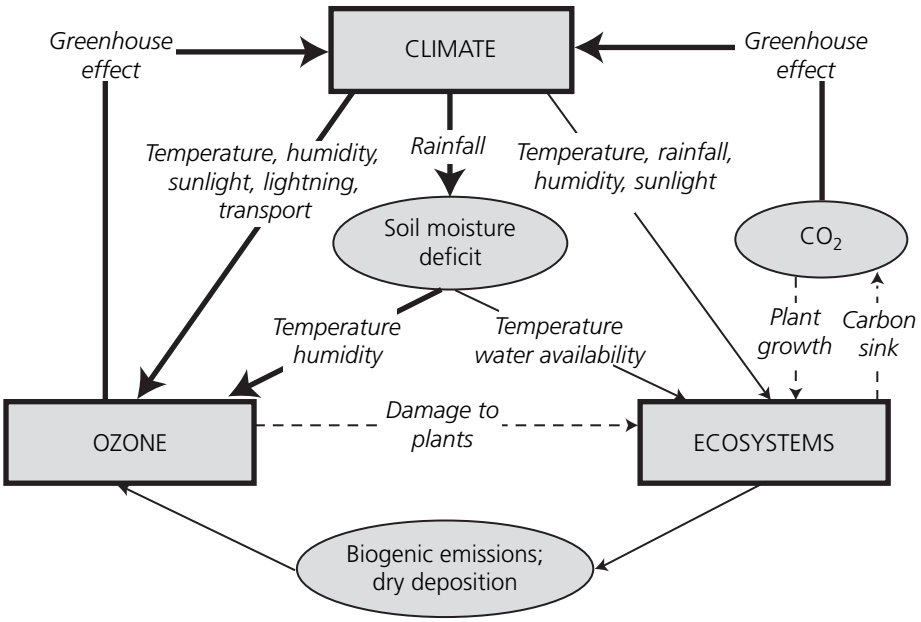
Climate change is inherently coupled to changes in regional and local meteorology, which may affect the potential for build-up of harmful levels of O_3 during episodes in regionally polluted areas (eg Western Europe). For example a change towards more frequent episodes of stationary high pressure systems (anticyclones and blocking) would suppress transport of pollutants away from the source area. Local chemistry, including O_3 surface deposition, may be affected through changes in mixing, temperature, humidity and rainfall.

This chapter presents the results of new work to examine the importance of summertime atmospheric circulation and effects on frequency of weather for O_3 formation in current and future climates. It also briefly considers how ecosystems respond to a changing climate and influence both O_3 production and destruction, and assesses possible feedback processes between surface O_3 and the climate system. The exceptionally high O_3 episode experienced over parts of Europe in summer 2003 is presented as a case study to highlight the major climate system processes and feedbacks that are critical for predicting future O_3 levels, especially high pollution episodes.

6.2 Effects of weather and climate on background ozone levels

The processes through which the climate system affects tropospheric O_3 levels are complex, and involve many interactions between the atmosphere, the land surface and ecosystems (Figure 6.1). Some of these processes are well-understood and are represented in coupled chemistry–climate models. However, those involving the terrestrial biosphere, including biogenic emissions and dry deposition, are less well understood, and are only represented simply in most models, for example carbon cycle and O_3 interactions

Figure 6.1 Schematic of the interactions between climate, ecosystems and tropospheric O_3. Thick solid lines denote processes that are generally well understood and represented in coupled chemistry–climate models. Thin solid lines denote processes that are understood but for which uncertainties exist and are only partially represented in models. Dashed lines correspond to links that are emerging as important but not generally included in model projections.



8 Synoptic refers to weather systems with scales of 100s to 1000s km typical of mid-latitude cyclones and anticyclones.

(indicated in Figure 6.1) have only recently been assessed, but may be significant (Felzer *et al*. 2004; Sitch *et al*. 2007). The importance of interannual to inter-decadal variations in climate, that occur whether or not there is human-induced climate change, can have a significant impact on background O_3 levels but are not yet captured within global models.

6.2.1 Changes in climate

Temperature, atmospheric humidity and sunshine levels influence background O_3 levels through the controls they exert on the photochemistry of net O_3 production. Higher temperatures accelerate O_3 production (depending on the NO_X regime – see chapter 3), and especially in summer, are also likely to increase biogenic VOC emissions and hence lead to higher surface O_3 concentrations in high NO_X regions (Sanderson *et al*. 2003). Over the next century increasing greenhouse gas emissions are expected to warm the planet. Temperatures are expected to increase more over the continents than the oceans, with greater increases in summer than winter, especially over Europe, and during the dry seasons of India and China. By the end of this century, it is projected that hot summers, such as that of 2003, could become an average yearly occurrence over Europe (Schär *et al*. 2004). There is a high level of confidence in modelled increases in mean temperatures at global and regional scales and hence on the implications of such changes for O_3 production (see chapter 5).

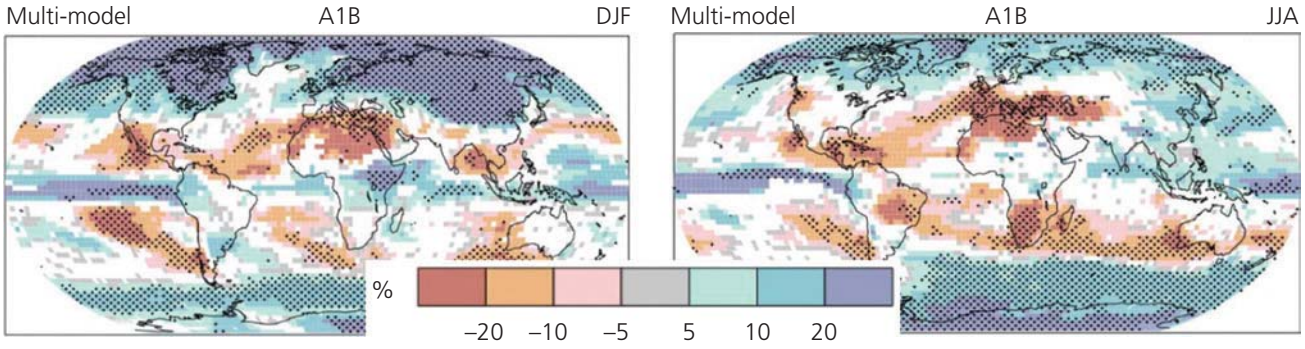
Section 3.3 discussed the chemistry of O_3 production, initiated by reactions of OH with CH_4, CO and VOC (see Figure 3.2). OH is produced following photolysis of O_3 in the presence of water vapour, therefore absolute humidity is a strong determinant of background O_3 levels, and the expected changes in atmospheric humidity associated with global warming are likely to have complex effects on surface O_3. Early studies with coupled chemistry–climate models identified humidity as a key negative climate feedback on tropospheric O_3 (Johnson *et al.* 1999, 2001). This finding has been confirmed in more recent studies (Murazaki & Hess 2006; Racherla & Adams 2006; Stevenson *et al.* 2006). Over the oceans where the relative humidity is predicted to remain constant, absolute humidity will increase with temperature and O_3 levels are likely to be reduced where NO_X levels are low. Conversely, over land, relative humidity will not remain constant and large changes in atmospheric humidity, both positive and negative, are possible, associated with shifts in major weather patterns and rainfall distributions.

Potential changes in rainfall are far less certain than temperature at regional and local levels, especially in the tropics and for major developing countries such as India and China (Figure 6.2). Nevertheless, a robust signal of reduced rainfall (and hence reduced cloudiness) across Southern Europe is predicted, which would have major implications for future O_3 levels. Shifts in rainfall patterns will influence sinks of O_3 through changes in dry deposition and increased levels of sunlight will contribute to the photochemical production of O_3 and potentially to higher biogenic emissions through higher levels of photosynthesis.

The degree of drying and the ensuing depletion of soil water reservoirs will be critical in determining how vegetation will respond and what the effects on biogenic emissions and O_3 deposition will be. For example, plants under water stress will tend to close their stomata reducing dry deposition. Summer drying will also lead to an increased incidence of forest fires which will have an impact on O_3 concentrations, regionally and globally, through the emission of NO_X, CO and nmVOC. Consequently, the distribution of rainfall is a major factor in determining future O_3 levels, although it remains a significant source of uncertainty in regional projections.

The climate controls on the long range (intercontinental) transport (LRT) of pollutants are also important in determining background O_3 levels. Winds associated with weather systems and jet streams transport pollutants over large distances, and in the last few years a better understanding of the complex nature of these transport processes and the chemical transformations that can take place (especially in stably stratified layers within the lower troposphere), has been gained (eg Fehsenfeld *et al.* 2006; Methven *et al.* 2006). The extent to which climate change will lead to changes in weather systems

Figure 6.2 Projections of likely shifts in rainfall patterns by the end of the 21st century for an A1B emission scenario. Colours show % change in rainfall where more than two-thirds of the models agree on the sign of the change. White areas denote regions where no consistent signal is predicted, eg Africa. Source: IPCC 2007b, Figure SPM 7, page 16 of the Working Group 1 Summary for Policymakers. Reproduced with permission from the IPCC and Cambridge University Press.



and shifts in the jet stream, both of which may imply an increase in trajectories that traverse areas with large precursor emissions, remains uncertain. Pinto *et al.* (2007) note that some model projections show a poleward shift of the Atlantic storm track and Lambert & Fyfe (2006) note an overall reduction in the total number of winter-time cyclonic events. In terms of summertime prevailing winds, changes over the UK and Europe will depend strongly on the incidence of anticyclonic weather (see section 6.3). In regions in which significant emission reductions are projected (eg Europe, North America, Japan), increases in background O_3 associated with long-range transport are likely to become a proportionately more important component of surface O_3 (Szopa *et al.* 2006).

As noted in chapter 3, the stratosphere provides an important source of O_3 for the troposphere via stratosphere–troposphere exchange processes. These are driven by the Brewer–Dobson circulation, a slow vertically overturning movement of air, rising at the equator and descending over the pole of the winter hemisphere, and by synoptic weather systems that create folds in the tropopause and bring stratospheric air, irreversibly, into the troposphere. Nearly all of the global models detailed in chapter 5 show an increase in the stratospheric influx of O_3 as a result of climate change, due to a stronger Brewer–Dobson circulation, a robust feature seen in many previous studies and state-of-the-art climate model projections (Butchart *et al.* 2006). In addition, climate change predictions suggest that while the troposphere will warm, the lower stratosphere will cool and slow O_3 destruction there (Zeng & Pyle 2003), allowing O_3 concentrations to rise. These factors, in conjunction with the recovery of O_3 from the influence of halogen species, will combine to increase the flux of O_3 into the troposphere. This tends to increase mid- to high-latitude upper tropospheric O_3, and in some models (eg Zeng & Pyle 2003), this effect dominates over reductions in global tropospheric O_3 from water vapour increases (Stevenson *et al.* 2006).

Tropospheric background O_3 levels are also influenced by vertical mixing in the updrafts of strong cumulus clouds, especially in the tropics. These provide important pathways taking O_3 and its precursors from the surface upwards, out of the atmospheric boundary layer and into the free troposphere (eg Hegarty *et al.* 2007) where they may be transported long distances by upper level winds. Lightning in vigorous cumulus convection is also an important source of NO_X in the free troposphere.

There is debate about whether global warming will lead to more vigorous cumulus convection, since the atmosphere will be more stable. In fact, Held & Soden (2006) report that overall, tropical convection is reduced in 21st century climate projections. Recent studies of observed rainfall intensity suggest that locally, more intense convective storms are occurring (eg Goswami *et al.* 2006), in line with results from climate model projections (eg Allen & Ingram 2002; Turner & Slingo 2008).

If more vigorous convection is an outcome of a warmer climate then this is expected to provide more mixing from the boundary layer to the free atmosphere of both O_3 and its precursors. However, the overall impact on O_3 is unclear as two modelling studies show divergent results (Lawrence *et al.* 2003; Doherty *et al.* 2005). Increased convection may also increase lightning, which potentially enhances NO_X production and may increase background tropospheric O_3, although not necessarily concentrations at ground level. Most model studies show an increase in lightning as the climate warms (eg Brasseur *et al.* 2006; Schumann & Huntreiser 2007), with increases of up to 60% per deg C of warming (Lamarque *et al.* 2005). Wu *et al.* (2007a) go as far as to suggest that increases in lightning may be the dominant climate-related driver of future tropospheric O_3 increases. However, Stevenson *et al.* (2005) found no global trend, only a geographical shift from the tropics to mid-latitudes. This shift is also observed in the new RS07 model runs.

There is currently no consensus among modelers about what the dominant climate feedback on surface O_3 globally might be, in part because different effects are likely at different spatial scales. Stevenson *et al.* (2006) found that the negative feedback associated with increases in humidity dominated over the remote tropical oceans and increased O_3 over the continents, especially where NO_X levels were largest. At the regional scale, especially over land the effects of changes in soil water availability will influence dry deposition and feedback into ground-level O_3. The increased flux of stratospheric O_3 was more important in the upper troposphere, rather than at the surface. Wu *et al.* (2007b) found that projected increases in lightning over the USA were the dominant driver up to 2050. Other studies have indicated that changes in the circulation may be important.

6.2.2 *Interactions between climate and ecosystems*

Interactions between climate and the terrestrial biosphere have an important effect on surface O_3 levels since vegetation influences both the sources and sinks of O_3 (Figure 6.1). The aerodynamic and stomatal resistance of vegetation canopies (Figure 3.6) control the dry deposition of O_3, while vegetation type determines the surface flux of isoprene to the atmosphere (see chapter 3). Changes in land-cover have the potential to change the evolution of near surface O_3 (Ganzeveld & Lelieveld 2004). Here we consider the impact of changes in natural vegetation arising from climate change and CO_2 increase.

As explained in chapter 4, biogenic emissions from many plant species are sensitive to many environmental factors including temperature and PAR and hence are strongly controlled by the local climate. There are complex sensitivities to the hydrological cycle. For example, the atmospheric vapour pressure deficit affects stomatal opening and thereby the rate of surface O_3 deposition over vegetated landscapes. The availability of soil moisture can affect plant biomass and lead to closure of stomata. As noted previously, changes in the distribution of rainfall will be critical for determining the effects of ecosystems on future O_3 levels.

There are, as yet, no global modelling studies coupling together climate, vegetation and atmospheric chemistry

in a fully interactive way. Sanderson *et al.* (2003) estimated the impact of a simulated change in natural vegetation cover on isoprene emissions and near-surface O_3 concentrations at the end of the 21st century, and showed an increase in isoprene emissions from 1990 to 2090 of between 26 and 34% depending on the land cover scenario. However, when the emission characteristics of crops is included, which are in general lower emitters of isoprene than species of semi-natural vegetation they replace, then the isoprene emission rates used to support this study show a decrease in emissions with the predicted change in land cover (see chapter 4). Given the significant dependence of isoprene emissions on plant species, modelling studies of this type must currently be seen purely as sensitivity studies. However, the suggested sensitivity of future O_3 to land-cover is indicative of an important interaction that needs to be fully integrated into climate–chemistry models.

One of the most important feedbacks with climate is through temperature induced increased emissions fluxes of O_3 precursors, including nmVOC from vegetation, as described above but also NO from soils and CH_4 from wetlands, all of which respond positively to temperature. Limited work has been reported on the latter processes, and in most cases the work is limited to an exploration of the scale of the temperature response rather than a fully interactive component of a larger climate model and must be regarded as a priority for further research.

6.2.3 Changes in climate variability

Climate variability on interannual to inter-decadal timescales may have a substantial influence on background O_3 levels by influencing surface O_3 production, stratosphere–troposphere exchange (STE) and the long-range transport of pollutants. A major question is how climate change will influence these modes of variability and what the implications are for regional surface O_3 concentrations.

On interannual timescales, the El Nino/Southern Oscillation (ENSO) is the major mode of natural climate variability, which produces large-scale shifts in the location of tropical rainfall leading to widespread droughts, shifts in weather patterns and major jet streams (Simmonds *et al.* 2004). In a modelling study, Zeng & Pyle (2005) have shown a close relationship between ENSO and STE, with anomalously large STE, affecting tropospheric O_3, following El Nino events. As yet there is no consensus among the various model projections concerning changes in ENSO in the 21st century (Guilyardi 2006; Le Treut *et al.* 2007).

ENSO also has a major impact on the monsoons of South and East Asia. The major El Nino of 1997/98 had substantial impacts on air quality across Indonesia and Southern Asia due to the very dry conditions and outbreaks of forest fires (Doherty *et al.* 2006). Recent research has suggested that there are multi-decadal changes in ENSO characteristics and ENSO–monsoon linkages, and these may be critical in determining regional pollution levels. One of the potential effects of global warming may be to change the behaviour of ENSO on multi-decadal timescales (Turner *et al.* 2007a) and thereby, its impact on key regions for surface O_3 production, such as the South and East Asian Monsoons (Turner *et al.* 2007b). Current projections of future O_3 levels tend to be based on a limited number of years and therefore do not take account of these potential changes in ENSO behaviour.

Within Europe, the North Atlantic Oscillation (NAO) strongly influences the wintertime climate, through the path and intensity of weather systems and the strength and location of the sub-tropical jet (eg Hoskins & Hodges 2002). This in turn may affect the LRT and hence the background level of O_3. Under positive NAO states the UK experiences more westerly winds in which maritime air masses prevail, and in which the transport of air from continental Europe is reduced. Hess & Lamarque (2007) and Li *et al.* (2002) show increased transport of United States emissions to North-West Europe, and increased transport of European emissions to the Arctic during the positive phase of the NAO.

Over the last three decades there has been a marked trend towards very positive NAO states which has been unprecedented at least over the last century. The effects of this have been to bring milder and wetter winters to the UK and drier more settled weather to Southern Europe. The extensive drying, currently observed in the Mediterranean region can in part be attributed to this inter-decadal change in the NAO. A major question is whether this multi-decadal shift to positive NAO states is due to climate change or part of a naturally occurring multi-decadal mode, known as the Atlantic Multi-decadal Oscillation (AMO), linked to variations in the strength of the Thermohaline Circulation (eg Sutton & Hodson 2005). Most projections show a tendency to more positive winter-time NAO states under global warming (eg Osborn 2004; Pinto *et al.* 2007). However, there are large inter-decadal variations within this increasing trend, which are not accounted for in current projections of future O_3 levels.

6.3 Effects of changes in weather on peak ozone episodes

If emissions are reduced, and O_3 concentrations decline, the potential changes in weather regimes, especially persistent summertime anticyclones and high temperature episodes, may still lead to dangerously high (regional or local) O_3 episodes. It is important that the future likelihood of these episodes is assessed because of the potential effects on human health, crop production and ecosystems in general. Following the extreme heat and high O_3 concentrations of 2003, the scientific community has been alerted to the possibility that climate change may increase the risk of such events in the future (see Box 6.1).

6.3.1 Anticyclonic weather and atmospheric blocking

Episodes of poor air quality occur predominantly in summer in mid-latitudes and during the dry season in the tropics. They tend to be strongly linked with particular weather patterns, especially atmospheric blocking and anticyclonic conditions. In mid-latitudes, atmospheric blocking events describe a disruption of the usual pattern of westerly winds and the establishment of prolonged episodes of stagnant air with low wind speeds and reduced advective transport of pollutants away from the source area. Extended dry periods associated with blocking can alter the life times and deposition pattern of a range of pollutants, including O_3 with a potentially profound influence on the frequency and severity of high pollution episodes.

Blocking occurs predominantly over the North Eastern Atlantic and Western Europe throughout the year with about 15% of the days in any one season being blocked. In summer, blocking tends to occur further east over the European sector and is generally characterised by high temperatures, dry conditions and poor ventilation of the boundary layer. In addition, anticyclonic weather (in which high pressure, light winds and dry sunny conditions prevail) can also influence surface O_3 production, through its impact on biogenic VOC emissions, as described in chapters 4 and 5. Efficient photochemical processing of emissions occurs, leading to elevated O_3 production rates, a consequent accumulation of O_3 over a period of several days, and the potential to increase regional surface O_3 levels over the UK and continental Europe to dangerous levels.

Various methodologies have been proposed to identify blocking events (see Tyrlis & Hoskins 2008 for a review), but the focus has been mostly on wintertime events. Very little research has so far been done to investigate how climate change may affect the types of weather that the UK and Europe will experience in the future, especially in summer. Here we report on a new study of atmospheric blocking and anticyclonic weather in current and future climates undertaken for this report (Barnes, Slingo, Hodges & Hoskins submitted).[9]

Based on analyses of climate change scenarios from the Max Planck Institute (ECHAM5) and Hadley Centre (HadCM3 and HadGEM1) models, Figure 6.3 suggests that summertime blocking is likely to decrease very slightly over European longitudes by the end of the 21st century (for ECHAM5 and HadGEM1) or for a doubling of CO_2 (HadCM3).

This result should be treated with caution however since there can be large decadal variations in blocking activity which arise through natural internal variability; there are also linkages between the incidence of blocking and the phases of ENSO and the NAO.

6.3.2 Heat waves and soil moisture feedbacks

As well as inhibiting the transport and dispersion of pollutants, and accelerating the chemical production of O_3, blocking events also lead to other feedbacks which may act to further increase surface O_3 levels. Summertime anticyclonic weather patterns are frequently associated with clear skies, high temperatures and low humidities. When these conditions persist, soil drying can lead to positive feedbacks that act to elevate temperatures even further, as was the case in 2003 (see Box 6.1). This provides the conditions (including reduced depositional loss of O_3 as well as increased photochemical production) for extreme pollution events.

In an analysis of a large ensemble of climate change projections with the Hadley Centre model, HadCM3, Clark *et al.* (2006) concluded that European heat waves (defined as a sequence of days with exceptionally high temperatures) will become much more intense by the end of the 21st century under a doubling of CO_2 concentrations. In the UK, heat waves lasting five days or more, as occurred for example in July 2006, currently occur approximately once every 20 years. By the end of the 21st century these may occur almost every year and could even occur several times each summer. Similarly the unprecedented high temperatures experienced during the August 2003 heatwave could occur in approximately 1 out of 10 summers by the end of the century. In agreement with other studies, Clark *et al.* also concluded that soil moisture effects are largely responsible for the change in frequency and intensity of extreme heat waves.

It is difficult to quantify the effects of climate change on weather conducive to high pollution episodes, and although the results summarised here suggest little change in the incidence of blocking, when this is linked with a general increase in temperature and declining summer rainfall, the number of days with low wind speeds and high temperatures in summer is likely to increase, leading to more high pollution episodes of the type experienced in 2003. A study of potential changes in daily maximum and minimum temperatures over Europe by Kjellstrom *et al.* (2007; Figure 6.4) suggested that maximum temperatures may increase disproportionately for more extreme events, especially for the UK, although there remains large uncertainty in the magnitude. The severity of high pollution episodes is difficult to predict, since no model adequately represents the effects of these weather events on, for example, biogenic emissions and wildfires.

9 It uses the blocking index of Tibaldi and Molteni (1990) – with a longitudinally varying central blocking latitude to represent the regional variations in the positions of the maximum synoptic scale activity (Tyrlis and Hoskins 2008) – to identify long-lived (≥5 days), synoptic scale (≥15 degrees longitude) events.

Figure 6.3 Impact of climate change on the frequency of seasonal blocking from a range of model simulations for the current and future climates.

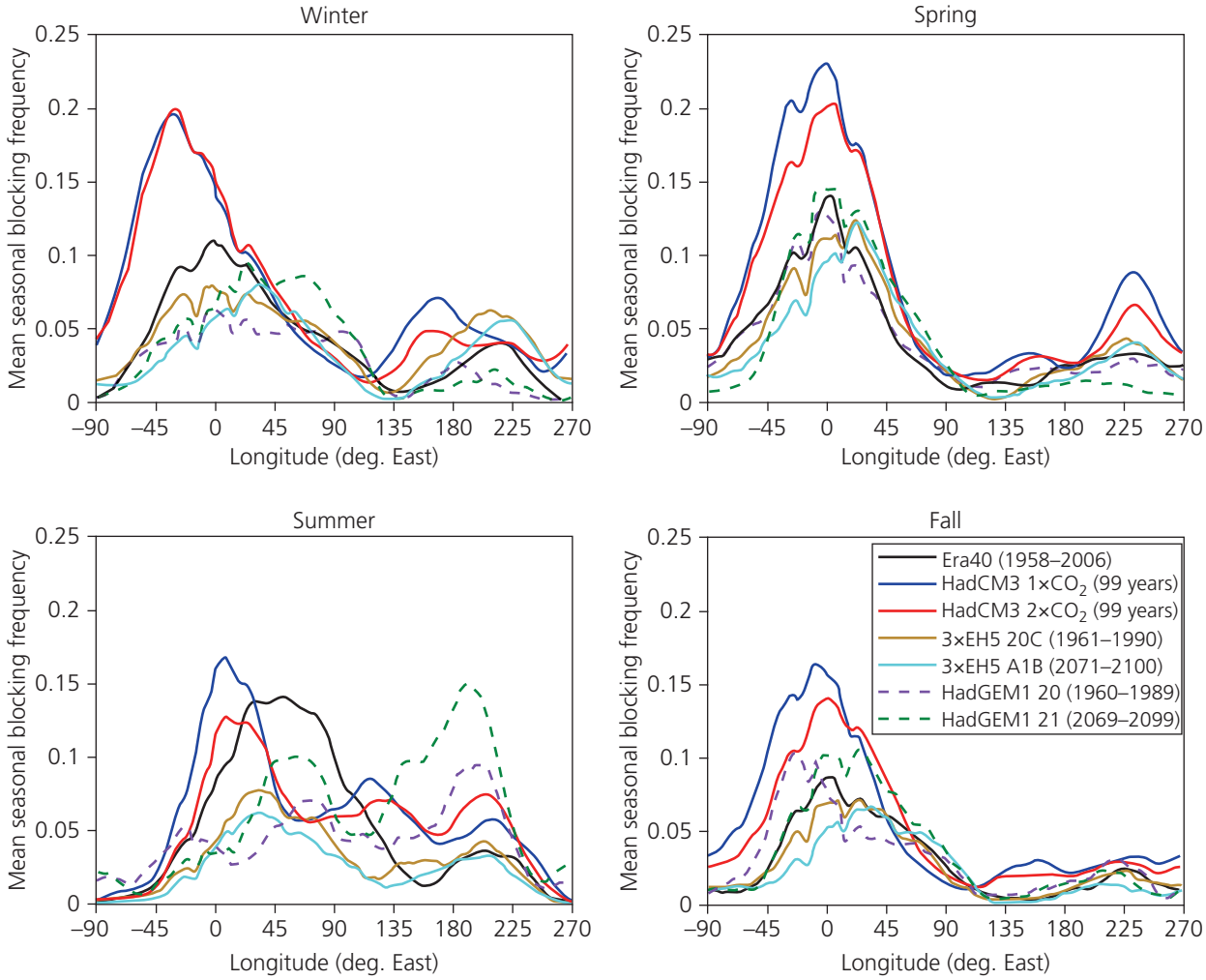
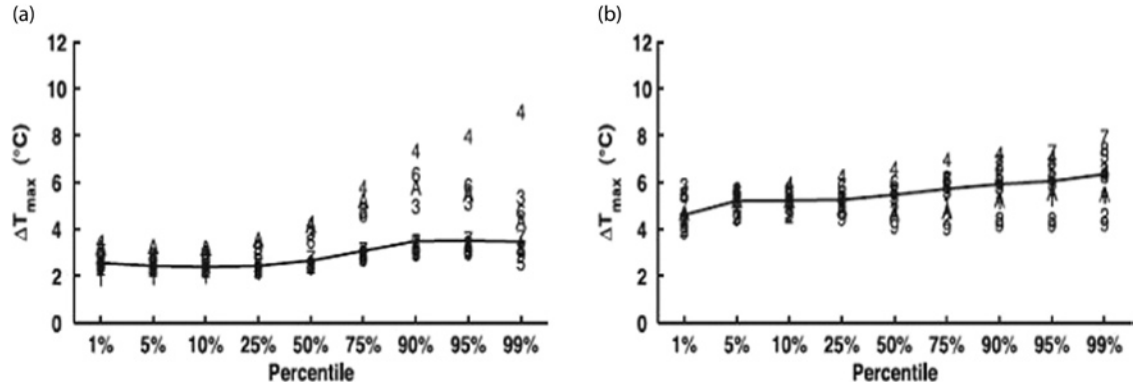


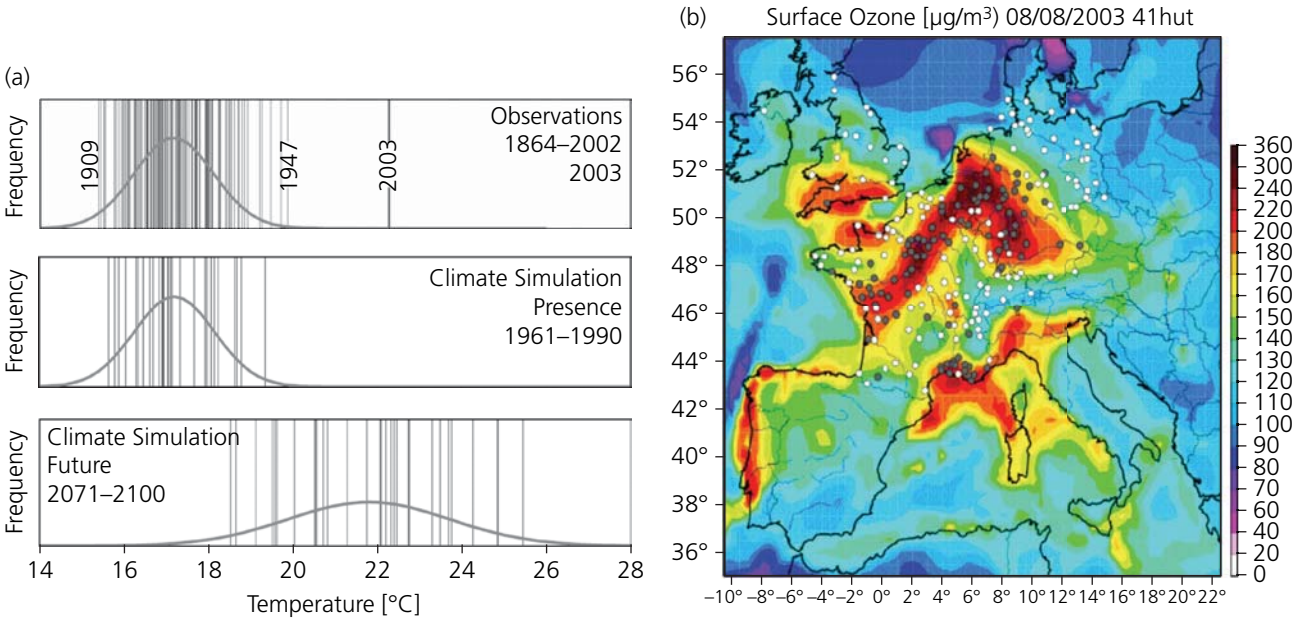
Figure 6.4 Changes in the distribution of summer (JJA) daily maximum temperatures in the British Isles (a) and in Eastern Europe (b) from a range of regional climate model simulations from 1961–1990 to 2071–2100 under the SRES A2 scenario. The horizontal axis gives the percentile of the distribution. The vertical axis gives the changes in each percentile (in degrees C) separately for the ten regional climate models. The lines show the median of the projections. Source: Christensen et al. (2007) (based on Kjellstrom et al. 2007). Reproduced with permission from the IPCC, Cambridge University Press and Springer Science and Business Media.



Box 6.1 Case study of an extreme high pollution episode: implications for the impacts of climate change on future ozone levels

In August 2003, Western Europe, especially Switzerland, France and Southern England experienced a prolonged heat wave and very high levels of surface O_3. In several locations, temperatures exceeded those previously recorded (see Box Figure 6.5a, left panel), and O_3 levels were severely elevated both in geographical extent and the duration of the episode. Ozone levels exceeded those considered harmful to human health by factors of 2 to 3 (up to 200 ppb in France (Fiala *et al*. 2003)) (see Box Figure 6.5b, right panel). It is estimated that the 2003 heat wave in Europe was responsible for at least 22,000 deaths (Schär & Jendritzky 2004) (2.5–85.3% of deaths attributed to O_3 across European cities – see chapter 8) and triggered losses of an estimated £7bn.

Figure 6.5 (a) Statistical distributions of summer temperatures in Northern Switzerland from observations and a model (using recent past 1961–1990, and a greenhouse-gas scenario for 2071–2100). Source: Schär et al. (2004). Reprinted with permission from Macmillan Publishers Ltd (copyright 2004). (b) Ground-level O_3 concentrations in Europe on the 8 August 2003. Source: EEA (2007). Reprinted with permission from the European Environment Agency. (Note: 2 µg/m³ = 1 ppb.)



The exceptionally high temperatures in summer 2003 were initiated by anticyclonic atmospheric circulation. However, strong sunshine levels and a lack of rain in the months preceding the summer contributed to a rapid loss of soil water, resulted in strongly reduced evapo-transpiration and latent cooling during the 2003 heat wave (Fischer *et al*. 2007), and highlighted the potential for soil moisture deficits to be a major factor in summertime extreme events (eg Seneviratne *et al*. 2006). A recent study by Vautard *et al*. (2007) has confirmed from the observational record that hot summers are preceded by winter rainfall deficits over Southern Europe, emphasising the critical role of the water reservoir in the soil of continental Mediterranean areas for European climate. Expected reductions in rainfall over Southern Europe under climate change are likely to increase further the potential for frequent heat waves similar to August 2003.

Many plant and tree species are high VOC emitters when placed under extreme conditions, although at the highest temperatures and conditions of drought stress biogenic VOC emissions decline significantly. The response to temperature is likely to be highly non-linear due to concomitant changes in stomatal conductance (which could also have led to a significantly reduced dry deposition loss for O_3 (see Figure 5.10). It is also highly species-dependent. During the European heat wave of August 2003 isoprene was observed at daytime concentrations in excess of 1600 pptv in South-East England, concentrations typical of tropical forested regions (Lee *et al*. 2006). Preliminary source-apportionment analysis was consistent with this isoprene, in part being of biogenic origin, although the observed levels cannot be reconciled with current estimates of isoprene emissions in that region, partly due to deficiencies in the UK emission inventory for biogenic VOC. This case has emphasised the potential for positive feedbacks between temperature, soil moisture deficits and biogenic emissions that may interact to produce highly elevated levels of surface O_3, which far exceed levels considered safe for plants and humans. It also highlights the need for better inventories of natural precursor emissions.

6.4 Tropospheric ozone changes as a driver of climate change

This chapter is predominantly concerned with the potential impact of climate change on ground-level O_3. However, O_3 increases also contribute to climate change through both direct and indirect routes (see Figure 6.1). Ozone is a greenhouse gas and future impacts of changing O_3 concentrations should therefore be considered alongside those of rising CO_2 concentrations. The IPCC estimates that tropospheric O_3 increases since pre-industrial times have contributed somewhere between 0.25 and 0.65 W m^{-2} to global radiative forcing (Forster *et al.* 2007). On the basis of this direct forcing, O_3 is ranked as the third most important anthropogenic greenhouse gas after carbon dioxide (1.49–1.83 W m^{-2}) and CH_4 (0.43–0.53 W m^{-2}).

It should be remembered that O_3 is a secondary pollutant (ie it is not directly emitted) formed from a variety of emitted precursors (see chapter 3). Control of O_3 therefore requires control of these precursors. However, the total climate impact of these precursor emissions goes beyond their effects on O_3. Emissions of NO_X tend to increase levels of OH reducing the lifetime of CH_4, and hence reducing CH_4 concentrations, producing a negative radiative forcing. The net impact on climate of NO_X emissions is believed to be a net cooling (Fuglestvedt *et al.* 1999; Derwent *et al.* 2001; Wild *et al.* 2001), although there is significant dependence on the location and season of NO_X emission (Stevenson *et al.* 2004; Naik *et al.* 2005; West *et al.* 2007; Derwent *et al.* 2008). However, emissions of CO, CH_4 and VOC tend to reduce levels of OH, increasing the CH_4 lifetime and producing an additional positive radiative forcing (Derwent *et al.* 2001; Wild *et al.* 2001; Collins *et al.* 2002a; Naik *et al.* 2005; West *et al.* 2007).

Increases in O_3 will also have an indirect effect on global warming by suppressing plant growth, reducing the land carbon sink for CO_2 and therefore increasing the rate at which CO_2 increases in the atmosphere (see Figure 6.1). Experimental studies have shown that O_3 reduces the additional carbon storage arising from increased CO_2 concentrations, but that elevated CO_2 concentrations can reduce the negative impacts of O_3 on vegetation (see chapter 8, section 8.4.2). A physiological model linking these phenomena has recently demonstrated that the indirect radiative effects of O_3 via reduced carbon sequestration could increase the total radiative forcing due to O_3 over the period 1900–2100 by at least 70% (Sitch *et al.* 2007). This recent work suggests that tropospheric O_3 increases are an even more important driver of global warming than previously assumed.

6.5 Gaps and uncertainties

Many processes and feedbacks come into play when considering the interactions between climate and O_3 (see Figure 6.1) and several global modelling studies have shown significant impacts of projected 21st century climate change on surface O_3 levels (see chapter 5). However there are significant uncertainties in the representation of many of these processes and feedbacks even in state-of-the-art coupled chemistry–climate models, as well as gaps in basic understanding. None of the current projections adequately account for decadal climate variations. Although these variations may not significantly affect the long-term average background O_3, it is possible that there may be decades with regionally enhanced or reduced O_3 and more or less frequent high pollution episodes.

Table 6.1 summarises the current state of knowledge and inclusion of processes in models. A more complete understanding of climate change effects on future O_3 levels will require longer integrations with models which include all the appropriate processes and feedbacks between atmospheric chemistry, climate and ecosystems, at sufficient resolution to represent regional scale processes, weather phenomena and ultimately urban scale modifications.

The representation of the water cycle constitutes one of the most serious uncertainties in future projections of regional climate change. Systematic biases in rainfall patterns simulated by current models compromise the projections of future changes (see Figure 6.2) and introduce uncertainties in background O_3 levels, especially over land and where NO_X levels remain high. They also limit our ability to predict changes in the incidence of heat waves and high pollution episodes.

Ecosystems are still only represented crudely in climate models and the processes through which they interact with the climate and surface O_3 levels are generally represented simplistically. Recent advances in understanding the damaging effects of O_3 on plants have not so far been included in future projections but are likely to lead to positive feedbacks that increase O_3 levels further.

Critically, many of the processes that affect O_3 production (eg detailed stratosphere–troposphere exchange processes, cumulus convection and mixing, lightning, atmospheric blocking) and exposure of terrestrial ecosystems (eg topography) operate on spatial scales which current coupled chemistry–climate models do not resolve. The computational expense of increasing model resolution is daunting and currently beyond the capability of existing resources, but our inability to represent weather systems and landscapes properly in current climate models places significant uncertainties on our current projections.

Table 6.1 Climate-related factors important for O_3 and the level of understanding in modelling of the relevant processes in coupled climate–chemistry projections.

Driver	Response to increase in driver	Increases or decreases in O_3 levels	Level of understanding	Inclusion in atmospheric chemistry projections
Temperature	Faster photochemistry	Increases (high NO_X) Decreases (low NO_X)	Good	Yes
	Increased biogenic VOC and NO emissions	Increases	Moderate	Sometimes
Atmospheric humidity	Increased O_3 destruction	Increases (high NO_X) Decreases (low NO_X)	Good	Yes
Sunlight	Increased photo-production	Increases (high NO_X), Decreases (low NO_X)	Good	Yes
	Increased stomatal opening	Decreases	Good	Yes
Drought	Decreased atmospheric humidity and higher temperatures	Increases	Good	Yes
	Plant stress and reduced stomatal opening	Increases	Good	Sometimes
	Increased frequency of wild fires	Increases	Poor	No
Blocking, anticyclonic weather	More frequent episodes of stagnant air	Increases	Poor	Yes
	Summertime/dry season heat waves	Increases	Poor	Yes
Long-range transport	Greater transport of pollutants, O_3 from remote locations	Mostly increases	Moderate	Yes
Strat–trop exchange	Increased flux of stratospheric O_3	Increases	Moderate	Yes
Cumulus convection	Increased vertical mixing of near-surface pollutants	Mostly decreases	Poor	Yes
	Increased lightning and NO_X production	Increases	Moderate/poor	Sometimes
CO_2	Less stomatal opening	Increases	Good	Yes
O_3	Less stomatal opening	Increases	Moderate	No
	Damage to plants	Increases	Poor	No
Climate variability	Greater extremes of major climate drivers, regionally	Mixed	Poor	Insufficient samples

6.6 Conclusions

The global chemistry models used in this report are not yet able to simulate all processes regulating ground-level O_3 at fine geographical scales, in part due to limitations in computing resource, but also due to weaknesses in understanding and availability of the data required. There are many processes contributing to the control of O_3 which are sensitive to climate, at a range of scales.

These global models suggest that over the 21st century, climate change is expected to decrease O_3 over the oceans, especially in the tropics; to increase O_3 over land, particularly in regions where NO_X emissions remain high; and to enhance the transfer of stratospheric O_3 into the troposphere. Consequently, projected changes in climate are likely to increase regional O_3 levels where emissions are high, which are the areas in which most people live. These increases may partially offset or even overturn some of the benefits derived from reductions in emissions.

Climate change can lead to important feedback processes which will affect: the production of O_3 precursors such as VOC from vegetation; changes in the production of lightning NO_X, the release of CH_4 from wetlands and the melting of arctic permafrost, the production of soil NO_X; and the uptake of O_3 by vegetation. Many of these processes are not routinely included in models, but they have the potential to substantially increase episodic peak and long-term mean O_3 concentrations under climate change.

Globally, an increase in the frequency of high O_3 days due to changes in weather and rainfall patterns is expected due to climate change. During the 21st century over the UK and Europe, for example, an increased frequency of summer droughts, heat-wave events and associated high O_3 episodes is anticipated. By the end of the century it is projected that hot summers, (defined in terms of the mean temperature for June, July and August), such as experienced in 2003, will become an average yearly occurrence and that the incidence of extreme hot weather of the type experienced in the heatwave of August 2003 will increase and may occur across Europe every one in ten summers by 2100. In other parts of the world, weather patterns that limit the intensity of summertime or dry season peak pollution levels are expected to be less prevalent leading to higher pollution levels.

One of the dominant processes controlling ground-level O_3 in heat-wave conditions is the dry deposition rate, which is reduced in dry conditions by stomatal closure in vegetation and was an important contributor to the very high O_3 concentrations during the 2003 heat wave in France. Such feedbacks are expected to increase as reduced soil water availability and hot summers become more common later this century.

Climate variability clearly influences ozone concentrations, through changes in atmospheric circulation for example. Inter-decadal changes in the NAO and ENSO have profound effects on regional climate and O_3 (Doherty *et al.* 2006). The projected frequency and magnitude of these very important features of climate into the future are very uncertain since current global climate models still have difficulty in capturing phenomena such as ENSO (IPCC 2007a) and it will be some time before progress in the underlying science will enable more confident projections.

Finally, tropospheric O_3 is an important greenhouse gas, ranked third behind CO_2 and CH_4 in terms of its direct radiative forcing over the period 1750–2005. Future increases in O_3 may also lead to significant indirect forcing of the climate, by reducing the global land carbon sink and therefore amplifying the rate of increase of atmospheric CO_2.

7 The effects of ozone on human health

7.1 Introduction

Ozone (O_3) has significant impacts on human health globally. In the EU 21,400 premature deaths each year are associated with O_3 (EEA 2007). This chapter provides an overview of the mechanisms by which O_3 affects human health. The final sections of this chapter discuss the current evidence of human health impacts and provide an assessment of how future background and peak O_3 concentrations may impact on human health.

7.2 Mechanisms of ozone effects – how does ozone cause damage to human health?

Ozone has been associated with a wide spectrum of human health effects most of which relate to the respiratory system (American Thoracic Society 2000; US EPA 2006; WHO 2006; US NAS *in press*). Ozone can cause direct oxidative[10] damage to cells or secondary damage by diverting energy away from primary cell functions to the production of defence mechanisms such as antioxidants.

Ozone reacts with antioxidants, such as ascorbate within the lung lining fluid (LLF), which protect the lungs from oxidative damage. In those instances when O_3 reacts with other substrates in the LLF such as protein or lipid, secondary oxidation products arise which lead to a number of cellular responses within the lung including an influx of inflammatory cells. As a consequence, the delicate blood/air barrier is damaged and the lung does not function as efficiently as it should.

Individuals react differently to O_3 exposure and many factors affect the amount of O_3 taken up by an organism, ie the dose. This makes the determination of a dose–response relationship for the assessment of human health impacts extremely complex. The mechanisms that control the balance between beneficial and detrimental interactions in the LLF compartment are not well established but these may contribute, in part, to an individual's varying sensitivity to O_3.

Adverse effects range from small effects on lung tissue through to respiratory symptoms and exacerbations of pre-existing diseases, such as asthma or chronic obstructive pulmonary disease, sufficient to cause hospitalisation or death. In the latter instance O_3 acts as one of a number of factors leading to disease exacerbation in patients with reduced lung function. The strongest evidence for effects on human health relate to acute effects but there is growing evidence that O_3 may also cause chronic effects by causing permanent damage to the lung (see Table 7.1).

7.3 Measuring and assessing the effects of ozone on human health

Measurements of the effects of O_3 on human health are based on either experimental exposure studies or real world observational studies (epidemiological studies). These measures are then used to assess the impacts of O_3 over a given area and time period through the use of models and an exposure–response coefficient to extrapolate impacts from measured effects. The limited number of monitoring sites or measured effects in different countries is a serious limitation in the use of such models.

For epidemiological studies, there are also a number of factors and processes that need to be taken into consideration when interpreting and extrapolating the observed effects of O_3.

Table 7.1 Human health and pathophysiological effects of O_3.

	Experimental studies	Epidemiological studies
Acute effects	Reduced lung function Respiratory symptoms Airway inflammation Airway hyperreactivity Increased airway permeability LLF antioxidant depletion	Reduced lung function Respiratory symptoms Increased asthma medication School absence Daily mortality – all cause, respiratory, cardiovascular Daily emergency hospital outpatient visits – respiratory Daily hospital emergency room attendance for respiratory diseases
Chronic effects	Morphological changes in the airways	Reduced lung function in children and young adults Increased incidence of asthma? Increased prevalence of asthma?

10 Oxidative damage is the extraction of electrons from target molecules which leads to a change in their chemical, physical and functional characteristics.

These include the effects of temperature and other pollutants on human health (see section 7.4.4).

Quantifying the population health impact of O_3 is important for policy and requires reliable exposure response relationships, estimates of population exposure and baseline health data. However a number of factors limit the accurate assessment of O_3 and are likely to lead to an underestimation of the effects, including: a lack of accurate baseline health data in many countries, the use of mean annual O_3 exposures to estimate population exposure which could miss important individual exposure effects, and the fact that many factors can affect the interpretation of epidemiological study results and obscure the real exposure–response relationship. This also makes it difficult to know whether there is a threshold of effects of O_3 (see Box 7.1). For this reason it is common practice to estimate health impact with and without a threshold assumption. In estimating the health impact for the EU Clean Air for Europe (CAFE) Strategy the threshold adopted was 35 ppb (European Commission 2005) and it is important to note that the air entering Europe from the Atlantic from

Box 7.1 Is there a threshold below which ozone has no effect on humans?

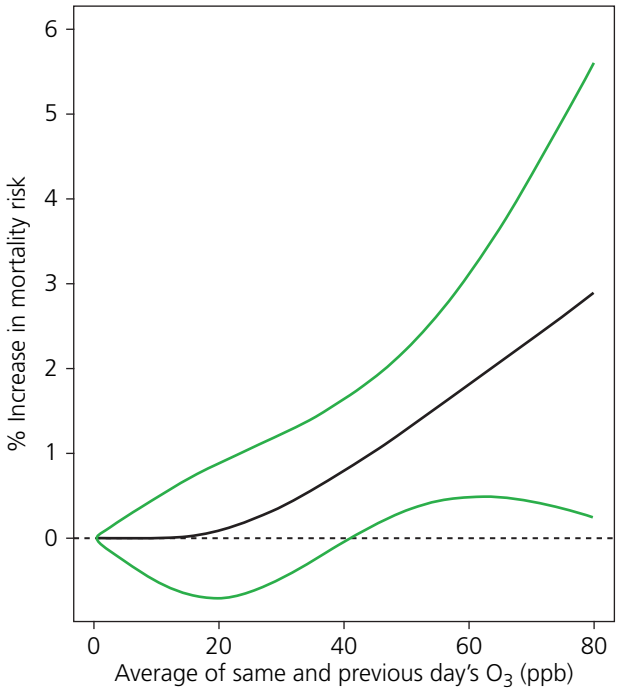
The shape of the exposure–response relationship and existence or not of a threshold for effect is critical in estimating the health impact of O_3 and has an important bearing on the number of deaths, for example, that can be attributed to O_3.

There are several reasons why epidemiological studies of daily mortality or hospital admissions might not be expected to show a threshold:

- There is variation in individual susceptibility to O_3 and in underlying disease that might be exacerbated by it.
- Actual exposure and dose to the lung are likely to vary within an area described by a community monitoring station. It has been shown that where there is a considerable difference between the mean and individually experienced exposure, epidemiological studies using community monitors are unlikely to show a threshold, even if there is one at an individual level.

A meta-analysis of 24 h (lags 0 + 1) O_3 and daily mortality based on 98 US cities (Figure 7.1) concluded that any threshold is at around background concentrations and much lower than current national air quality guidelines. This is one basis for the decision of a European working group to recommend a threshold of 35 ppb (8 h maximum) for health impact assessments. With no threshold assumption, the number of health events on peak O_3 days is outweighed by the number of events attributable to O_3 on the more numerous lower O_3 days (WHO 2003, 2006).

Figure 7.1 Summary of exposure–response curve for O_3 and daily mortality based on 98 US urban communities. Note: Percent increase in daily non-accidental mortality at various O_3 concentrations using the spline approach. The black line represents the central estimate, and the green lines represent the 95% posterior interval. Source: Bell et al. (2006). Reproduced with permission from Environmental Health Perspectives.



sources elsewhere in the Northern Hemisphere often contains larger O_3 concentrations than 35 ppb.

Irrespective of whether there is a threshold at the population level or not, the important fact is that effects are detectable in the ambient range experienced by populations worldwide and well below national air quality standards. The extension of exposure–response relationships to relatively low levels of exposure was a key factor in the recent decision of WHO to reduce the O_3 Air Quality Guideline from 120 to 100 µg/m^3 (8 h) (50ppb). The health implications of current background concentrations in many regions are therefore considerable and any further increases in background concentrations of O_3 would have a significant impact on human health (WHO 2003 and 2006).

Estimates of population exposure to O_3 in urban areas are based on community background stations. This may not adequately represent personal exposure because the population spends most of its time indoors, where O_3 levels are lower. However, it is the exposure basis of most epidemiologically derived exposure–response relationships on which impacts are based. Exposure coefficients are reported for three averaging times (1, 8 and 24 h). Given likely human exposure patterns the 8 h maximum is preferred: this is the averaging time used frequently in Europe (an estimate of 0.3% of total mortality for a 5 ppb increase in 8 h O_3 based on a meta-analysis of available European city results was used to model the mortality impact of O_3 on daily mortality for the EU CAFE Strategy) and in the revised WHO Global Guidelines (WHO 2006). For wider areas, however, it is necessary to rely on models.

Ideally, health impact assessments should use exposure–response relationships that relate to important health outcomes for which a population baseline is available. For acute effects this narrows the choice to daily mortality and daily hospital admissions. Chronic disease and loss of life years are potentially more important health impacts but the evidence is currently insufficient for reliable health impact assessments. While the baseline lung function of populations is known, the health significance of chronic reductions in lung function is uncertain (this is discussed further in section 7.4.3).

7.4 Current human health impacts

The impacts of O_3 on human health can be split into acute and chronic effects. There is substantial evidence to support the attribution of acute health effects to O_3. However, there are still considerable uncertainties in the attribution of chronic effects (Table 7.1).

7.4.1 Who is most at risk from ozone exposure?

There is evidence to suggest that children, asthmatics, and the elderly are populations that may be more susceptible or vulnerable to the effects of O_3. Various factors appear to influence an individual's susceptibility to O_3, such as: genetic susceptibility, age and pre-existing cardiorespiratory disease.

Controlled human exposure studies demonstrate a large variation in sensitivity and responsiveness to O_3 in studies of healthy subjects, but the specific factors that contributed to this variability are yet to be identified (Mudway & Kelly 2000). Other factors can also lead to enhanced vulnerability to O_3 related effects, such as heightened exposures or activity patterns leading to an increased dose. For example, studies have shown that exercising individuals are exposed to much higher doses of O_3 due to the greater volumes of air entering their lungs (Mudway & Kelly 2004).

O_3 induced differential responses in lung function and airways hyper-reactivity in people with allergic rhinitis suggest that asthmatics have greater responses than healthy people with exposure to O_3, which increase with disease severity. In addition, repeated O_3 exposure over several days has been shown to increase responsiveness to bronchial allergen challenge in subjects with pre-existing allergic airway disease, with or without asthma. Asthmatics also show significantly greater inflammatory responses than similarly-exposed healthy individuals (Scannell *et al.* 1996).

Controlled human exposure studies have shown that lung function responses to O_3 vary with age, with responsiveness generally diminishing after about 18 to 20 years of age. As the size and surface area of the airways change with age this is most likely due to higher O_3 absorption in children (Bush *et al.* 1996).

Various strains of mice and rats have demonstrated the importance of genetic make-up for O_3 susceptibility. Moreover, genetic and molecular characterisation studies in laboratory animals identified regions of the genome responsible for both sensitivity and resistance. Recent human clinical and epidemiologic studies also have shown that genetic variations for antioxidant enzymes and inflammatory genes (GSTM1, NQO1 and *TNF-α*) may modulate the effect of O_3 exposure on pulmonary function and airway inflammation (Chen *et al.* 2007).

7.4.2 Health effects of short-term (acute) exposure to ozone

There are substantial human and animal toxicological data that support health effects associated with short-term exposure to O_3 and associations have been observed with a wide range of outcomes in epidemiological studies (Table 7.1). The evidence to support the main effects such as reduced lung function, respiratory symptoms and airway inflammation are discussed below.

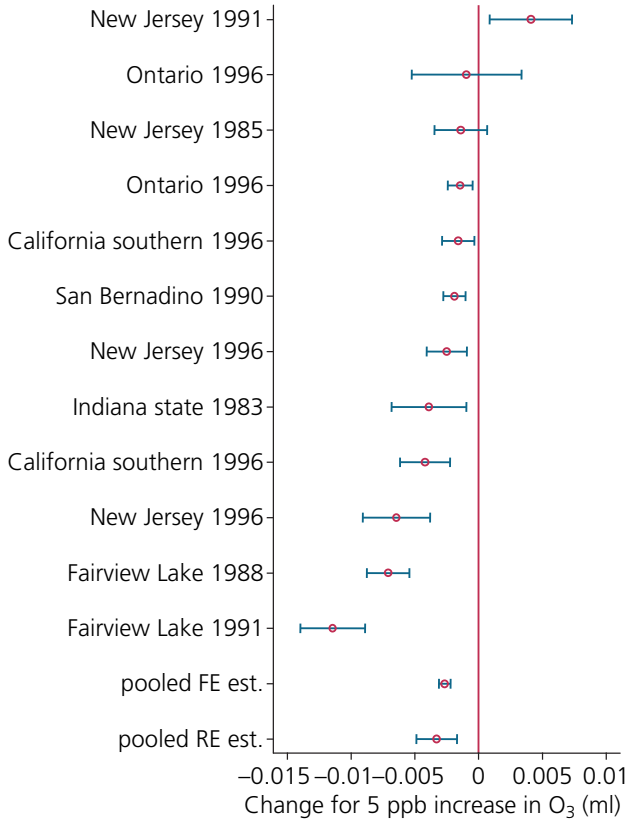
The effects of ozone on lung function

Controlled O_3 exposure studies demonstrate acute reversible reductions in lung function in 10–15% of healthy adults. However, there is considerable variability in responses between individuals, such that some may experience no symptoms at all to moderate O_3 challenge. Studies show that with repeated acute O_3 exposures over several days, lung function reductions are typically greater on the second day. However tolerance is

developed over the subsequent days of repeated exposure (Kulle *et al.* 1984).

In epidemiological studies symptoms in healthy children are rarely observed but there is good evidence from many studies that small transient decrements in lung function occur. An example from panel studies reported for 1 h O_3 from the USA is shown in Figure 7.2. The results of individual studies varied but the summary estimate for average reduction is 5 ml (less than 1%) in forced expiratory volume (this is a standard measure of lung function and represents the amount of air that can be forced out of the lung in 1 second) per 5 ppb change in O_3. These studies typically observe considerable individual variation with some subjects showing no reduction and some larger reductions, which might be sufficient to cause symptoms in subjects already on the brink of a clinical event for other reasons (such as a respiratory infection).

Figure 7.2 Effect of 1 h maximum daily O_3 on daily forced expiratory volume in one second (FEV_1) in panels of children studied during the summer period in the USA. Pooled random effects (RE) and fixed effects (FE) estimates. The unit is ml. Courtesy of Air Pollution Epidemiology Database, St George's, University of London.



Respiratory symptoms brought on by ozone

A small proportion of healthy adults exposed to O_3 exhibit symptoms of cough and pain on deep inspiration but these symptoms gradually decrease with increasing age. With repeated O_3 exposures over several days, individuals develop a tolerance however this is lost after about a week without exposure. There are also positive associations between O_3 concentrations and emergency hospital admissions or emergency room attendance for respiratory diagnoses but, overall, the evidence is mixed. More convincing is a lack of evidence for associations between O_3 and emergency admissions for cardiovascular diagnoses.

The effects on airway inflammation

Animal toxicological studies provide extensive evidence that acute (1–3 h) low dose O_3 exposures can cause lung inflammatory responses, damage to epithelial airway tissues, increases in permeability of both the lung endothelium and epithelium and increases in susceptibility to infectious diseases due to the modulation of lung host defences (Gilmour & Selgrade 1993; Leikauf *et al.* 1995). Inflammatory responses have been observed in the airway in some individuals following O_3 exposures even in the absence of O_3 induced reductions in lung function. There is also evidence that repeated O_3 exposures over several days results in a reduction in inflammation. However, evidence suggests that lung tissue damage continues during repeated exposure (Jorres *et al.* 2000).

Effects on airway responsiveness and allergic reactions

Controlled human exposure studies have found that acute O_3 exposure causes an increase in non-specific airway responsiveness. Ozone has also been shown to further increase the airway responsiveness in allergic asthmatic subjects, in response to allergen challenges. These effects are supported by studies on laboratory animals. The lung lining fluid antioxidant status is compromised in asthma and, although the cause of the decreased complement of antioxidant defences on the surface of the lung is unknown, it is likely that as a consequence these individuals will be more susceptible to an oxidative challenge. It has also been shown that O_3 induced exacerbation of airway responsiveness persists longer and decreases more slowly than the effects on lung function and respiratory symptom responses (Folinsbee *et al*. 1988; Horstman *et al*. 1990; Molfino *et al*. 1991; Peden *et al*. 1995; Jorres *et al*. 1996; Sun *et al*. 1997; Kehrl *et al*. 1999; Roux et al. 1999).

Panel studies have also observed, to a varying extent, associations between ambient O_3 on the same or prior days and symptoms of upper or lower airway irritation, reduced lung function, increased asthma symptoms and increased medication for asthma. There is considerable inconsistency between studies, however, which has not yet been explained but may relate to factors such as variations in exposure, level of activity and susceptibility.

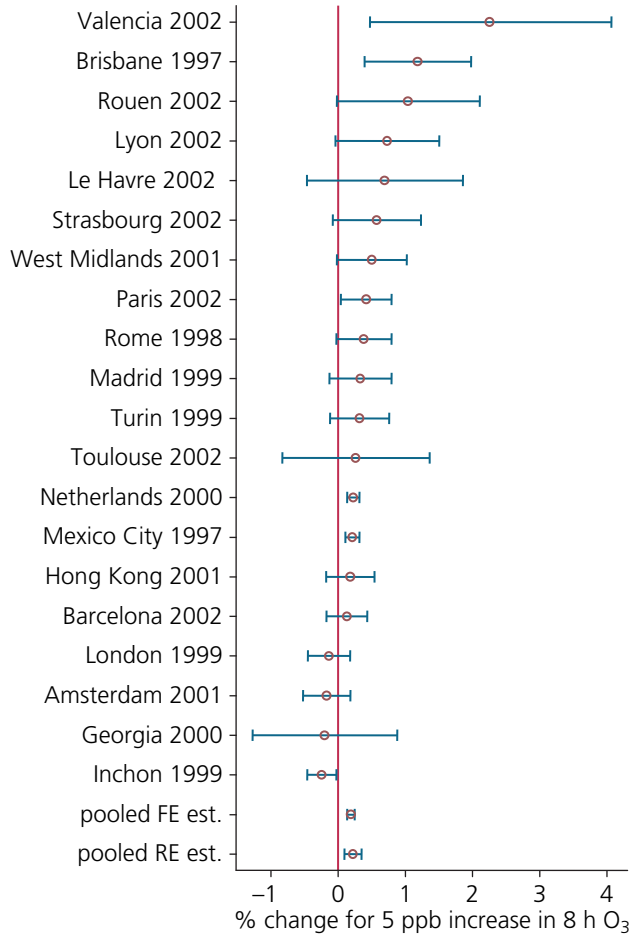
Effects of ozone on daily mortality

There is good evidence to support an association between O_3 and increases in daily mortality from time-series studies conducted in many cities throughout the world (WHO 2006). To illustrate this, Figure 7.3 shows the association between 8 h O_3 and all-cause daily mortality (total number of daily deaths in a population) for 20 individual cities published up to 2006.

The combined estimate for the 20 studies is a 0.22% increase in daily deaths for 5 ppb increase in 8 h O_3 and, while small, this is unlikely to be explained by chance. The corresponding estimates for cardiovascular and respiratory mortality are 0.38% and 0.41%, respectively. Allowing for scaling, equivalent results have been found for 1 h and 24 h averaging times (WHO 2006).

There is considerable variability between cities, some of which is explained by publication bias or *post hoc* selection of the best out of a number of model results, but much of the variability is unexplained. To reduce the variability due to the methods used, a number of coordinated multicity studies have been completed. Of 11 such studies, statistically significant positive associations with daily mortality have been observed in 7. A large European study of 23 cities, found an association only in the warm season (Gryparis *et al.* 2004). There is no direct evidence to explain the mechanism by which mortality is affected. It is postulated that these associations are explained by O_3 affecting individuals who are close to dying from some other cause. If this death is inevitable, then the role of O_3 is to bring the death forward, possibly by only a few days. If the vulnerability is temporary, it is possible that, had O_3 not had an additional effect, the individual might have recovered, in which case a more substantial loss of life may be incurred. It is also thought that O_3 influences cardiovascular mortality indirectly through placing stress on the respiratory system or indirectly through inflammatory mediators released by the lung into the blood stream (Bell *et al.* 2007).

Figure 7.3 Relative risks and 95% confidence intervals of daily mortality associated with 5 ppb increase in 8 h O_3 and daily all-cause, all-age mortality. Pooled random effects (RE) estimate 0.22% (95% CI 0.09–0.35). Courtesy of Air Pollution Epidemiology Database, St George's, University of London. FE – fixed effects, RE – random effects.



However, correlations between mortality, O_3 and various other pollutants complicate the interpretation of these associations (as discussed in section 7.4.4). Ozone tends to be positively correlated with NO_X in the warm season and negatively correlated in the cool season. In turn, NO_X, especially NO have a common emission source with PM which also has mortality effects. Most studies, however, find that the effects of O_3 are relatively independent of those of particles (WHO 2006).

7.4.3 Health effects of long-term (chronic) exposures to ozone

Animal toxicological studies show that chronic O_3 exposure causes structural changes in the respiratory tract, and simulated seasonal exposure studies in animals suggest that such exposures might have cumulative effects. The persistent nature of these changes raises the possibility of long-lasting alterations in human airways in response to chronic O_3 exposure, but it is unclear what long-term patterns of exposure or O_3 concentrations are required to produce analogous morphological changes in humans.

Chronic effects on human health can be investigated by analysing the relationship between spatial variations in long-term exposure to air pollution and health. These may be cross-sectional studies in which the data are collected at one period of time, or cohort studies in which exposed and non-exposed populations are followed over time. The chronic health effects observed so far are listed in Table 7.1.

Chronic effects on lung function and growth

The evidence that long-term O_3 exposure is associated with reductions in lung function is somewhat inconsistent. Cross-sectional studies of children in Austria and Southern Germany (Frischer 1999; Horak 2002; Ihorst 2004) have observed reductions but there was some evidence that these effects could be reversed over several years. In Southern California, however, there was no evidence that O_3 exposure was associated with reduction in the expected rate of lung function growth (Gauderman 2004).

The clinical implications of these effects are unknown. The lung has considerable excess capacity and these reductions are unlikely to cause symptoms except in individuals who have little or no reserve as a result of advanced chronic lung disease due to other causes. It has been observed in a number of population cohort studies that individuals with lower lung function have a reduced life expectancy but because the reasons for this are not known, it is not possible to predict

with any confidence whether O_3 associated reductions will have the same effects on life expectancy.

The effects of ozone on asthmatics

Asthma is one of the most important chronic diseases in childhood and there is intense interest in the possibility that it might be caused by ambient air pollution. A number of reviews have concluded that there is little or no evidence that ambient air pollution, including O_3, cause asthma, although it may play a small part in exacerbating existing disease (WHO 2005).

The most comprehensive body of evidence supporting this conclusion is based on prevalence studies of respiratory symptoms in which a number of areas, usually different towns are compared. These studies typically investigate a range of both respiratory symptoms and air pollutants which can lead to a large number of associations. About half of these studies have found associations between O_3 and at least one symptom, including asthma symptoms, but there is, overall, a lack of consistency.

Cohort studies provide direct evidence concerning the incidence of disease, but are few in number because of the logistical problems of following up large numbers of individuals over an extended period of time. A study of non-smoking adult Seven Day Adventists in California, found that long-term exposure to O_3 was associated with the incidence of doctor diagnosed asthma (Abbey *et al*. 1991). The study of Southern California children referred to above followed children without evidence of asthma over 5 years and found no increase in risk was in relation to long-term O_3 exposure. However, in a subgroup analysis, an increase in incidence was observed in both boys and girls living in high O_3 areas who played three or more team sports, suggesting that exercise rates are an important factor (McConnell *et al*. 2002). The same study also found small associations between O_3 and bronchitic symptoms in asthma subjects.

Effects on loss of life years

Potentially, perhaps the most important health effect of O_3 is a shortening of life. This is best studied by cohort studies in which subjects exposed to different long-term concentrations of O_3 are followed over time to compare mortality rates. Risks of death have been investigated in three adult cohort studies, all in the USA. The largest cohort found that all cause and cardiovascular mortality were positively associated with O_3 concentrations in the third quarter of the year (Pope *et al*. 2002). The other cohorts also found some indications of a positive effect but overall, the current evidence is insufficient to either confirm or refute an association.

7.4.4 The interaction of ozone with other pollutants and temperature

Ozone may have significant additive effects on health in combination with other environmental factors, such as pollutants, allergens or temperature. However, the mechanisms for such effects are not well understood. Animal studies have provided evidence that supports additive, synergistic and even antagonistic effects between these factors.

A few controlled human exposure and animal toxicology studies have investigated health effects associated with O_3 containing pollutant mixtures at near ambient levels and these suggest that interactions do occur between O_3 and PM with increased lung tissue injury observed with the mixture of PM + O_3 compared to either PM or O_3 alone (Bosson *et al.* 2007). Controlled human exposure studies indicate that continuous exposure of healthy human adults to SO_2 or NO_2 also increases O_3 absorption. There is some epidemiological evidence that adverse effects of particles are enhanced when O_3 concentrations are higher (Atkinson *et al.* 2001).

Controlled human exposure studies suggest that O_3 enhances the response to allergen challenge, which are supported by animal toxicology studies. However, studies in London have not been able to confirm that the effects of pollens or mould spores on hospital admissions for asthma are modified by concentrations of O_3, so it is not clear at present whether this is important for public health (Anderson *et al.* 1998).

An interaction between O_3 and temperature during hot weather is of greater potential importance in view of global warming and the predicted increase in heat wave events, as experienced in the summer of 2003 (see Box 7.2). Mortality increases in hot weather, especially in heat wave conditions where there is a sustained high temperature over several days (see section 6.3.2). These are conditions in which the rate of O_3 production also increases so that the population is exposed to extremes of both temperature and O_3. Heat wave conditions may also be associated with an increase in other pollutants such as particulates. Apart from heat stroke, the mechanisms leading to excess deaths during heat waves

Box 7.2 The summer 2003 heatwave: estimating health impacts of elevated O_3

The relative impact of O_3 and high temperature on mortality during the European 2003 heatwave was investigated in nine French cities (Bordeaux, Le Havre, Lille, Lyon, Marseilles, Paris, Rouen, Strasbourg and Toulouse) (Filleul *et al*. 2006).

The excess risk of mortality attributable to O_3 was very heterogeneous, ranging from 2.5% in Bordeaux to 85.3% in Toulouse.

Various suggested reasons for this heterogeneity include the vulnerability of the population (poverty, age, chronic disease burden), differences in the heat island effect, and errors relating community measures of temperature and O_3 to exposure of the population.

These results show that the effect of O_3 during heatwaves, while generally small in relation to the effects of temperature, is likely to constitute an additional health impact but that the size of this effect is likely to vary considerably from place to place.

in high O_3 conditions are difficult to distinguish. In such a complex and extreme situation it is not safe to assume that the linear concentration response relationships observed across the whole range of O_3 concentrations is applicable. Limited epidemiological evidence from Belgium in 1994 and France in 2003 suggest that in very hot summer conditions O_3 plays a role in increasing mortality.

7.5 Future human health impacts: how significant will the effects of ozone on human health be over the next century?

Estimating the future impact of O_3 requires the estimation of future baseline rates for mortality and morbidity, projections for future O_3 exposure levels, knowledge of the effects of O_3 on human health, and use of an appropriate dose–response relationship. Much of this information is not currently available for many locations. If changes over the next few decades are as rapid as in the last, there may be important trends over the century in life expectancy, disease prevalence and health care utilisation, but these are difficult to predict. While improvement in these health parameters will reduce the health impact of O_3, the relative importance of O_3 health effects may increase relative to other components of ambient pollution, such as PM, which are declining. Future impacts will also be influenced by a variety of lifestyle factors that can affect the dose of O_3 received by the lung. These include the amount of physical exercise, balance between time spent between indoor and outdoor environments and the way in which buildings are ventilated, including use of air conditioning.

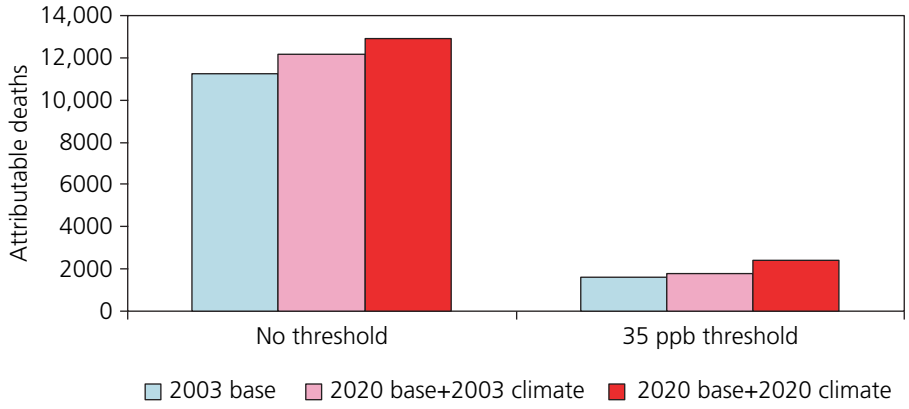
An estimate of the future impacts of O_3 in 2020 is shown in Figure 7.4 (taken from the DOH (2008) in which future O_3 concentrations were modelled using emissions and weather data). It shows the estimated number of deaths in the UK attributable to O_3 in 2003 (base) and in 2020 (with or without climate change). The overall impact is very sensitive to the threshold assumption (see Box 7.1).

If no threshold is assumed, the increase in estimated annual O_3 concentrations between 2003 and 2020 results in a 15% (11272 to 12930) increase in deaths which falls to 8% (11272 to 12140) if climate is held at 2003 conditions. If a threshold of 35 ppb is assumed, the increase is 51% (1582 to 2391), which falls to 14% (1582 to 1802) when climate is held at 2003 conditions. In the no threshold model, about half of the increase in health impact is due to changes in UK and European emissions (particularly NO_X emissions) and the remainder to climate change. In the threshold model, most of the increase is attributable to climate change. The relative importance of trends in climate vs trends in precursor emissions on future O_3 health impacts is therefore critically dependent on the threshold assumption.

Although it is not possible to accurately quantify how the impacts of O_3 will change over this century due to the uncertainties outlined above, it is possible to provide an illustrative example of the possible direction of change and the potential future health implications using the projected urban O_3 concentrations presented in chapter 5. Based on the projections for an example site in London with an increasing background O_3 concentration (shown in Table 5.2, chapter 5), it can be estimated that, with an assumption of no threshold, that daily deaths and respiratory hospital admissions attributable to O_3 will increase by 71% between 2000 and 2050 and by 95% between 2000 and 2100. If background O_3 concentrations decrease, increases in the impacts will still occur but at a reduced rate (45% and 40% respectively). This suggests that even if background concentrations of O_3 decrease the adverse effects of O_3 at urban locations will increase over the next century due to a reduction in the NO_X titration effect in many cities. This effect will be exacerbated by further increases in background O_3 concentrations.

These estimates are sensitive to exposure response and baseline risk assumptions and do not take account of the effects of climate change on future O_3 concentrations, such as increases in mean summer temperature or the projected increase in heat wave events predicted over the century (see chapter 6) which will lead to more high pollution episodes of the type experienced in 2003 (see Box 7.2). Highly elevated levels of surface O_3 during such episodes will far exceed levels considered safe for human health.

Figure 7.4 UK: deaths attributable to O_3 in 2003 and 2020 (with and without a threshold assumption, and under a current and future climate) Source: Department of Health (2008). Reproduced under the terms of the click-use license.



At the regional level, assessments of the current and future impacts of O_3 have been completed for many regions. A CAFE analysis for 25 European countries looks at O_3 health effects in 2000 and 2020 (European Commission 2005). This analysis shows that with current EU legislation, for 2000 and 2020, the number of premature deaths is estimated to be 21,400 and 20,800 respectively. The equivalent estimates for respiratory hospital admissions in the 65+ age group were 14,000 and 20,100. The relatively small change in mortality reflects the assumption that current EU legislation will be fully implemented and complied with, thereby keeping population exposure to O_3 close to 2000 levels. This estimate is likely to be conservative however as it does not include morbidity effects, and is based only on days at which O_3 concentrations are above 35 ppb.

Due to data limitations equivalent estimates are not available in regions such as Asia, Africa, and Latin America where emissions of O_3 precursors are projected to increase over the next two to three decades. In these regions higher O_3 concentrations are expected to lead to increased human exposure and therefore increased mortality and morbidity impact.

7.6 Conclusion

Ground level O_3 affects human health leading to increased mortality and respiratory problems.

Current O_3 exposure levels in North America and Europe are associated with short-term acute effects on the respiratory system resulting in increased daily mortality and morbidity in already vulnerable individuals. There is also evidence that long-term chronic exposure has adverse effects on lung function but the significance of these effects for long-term health is not yet known.

The health effects of O_3 increase with increasing concentrations. Health impacts have been observed at around ambient concentrations (approximately 35 ppb) and below the current WHO guideline of 50 ppb (daily 8 h average concentration). The total health impact is now understood to be driven more by the days at which O_3 is at baseline concentrations than by the occasional days on which episodes occur. Quantification of the effects of O_3 on human health now and in the future depends heavily on the assumption of whether there is a threshold for effects in humans. If no threshold exists, the health impacts of O_3 in the future will depend largely on trends in hemispheric baseline concentrations. If a threshold exists and is close to current ambient concentrations, then the health impact will depend more on regional and local emissions of precursors together with meteorological conditions.

In some regions (Africa, Latin America and Asia) emissions of O_3 precursors are projected to increase over the next few decades leading to increasing O_3 concentrations, and increased human exposure to O_3. In regions where emissions are projected to decrease, a reduction in the NO_X titration effect is expected to lead to increased urban O_3 concentrations as NO_X emission controls are progressively implemented. Like most urban areas, O_3 concentrations in London are expected to increase as NO_X emission controls take effect leading to increased human exposure and substantial increases in daily mortality and respiratory hospital admissions in 2100. An increase in background O_3 will exacerbate this increase while a decline in background O_3 will partially compensate for the NO_X titration effect and could lead to a decrease in human exposure.

By the end of this century, it is projected that hot summers such as that experienced in the Northern Hemisphere in 2003 and heat waves such as that experienced across parts of Europe in 2003 will become more frequent. With an increase in the frequency and magnitude of such events, mortality and morbidity are also expected to increase.

8 Impacts of ozone on the environment

8.1 Introduction

The most important and well-documented environmental effects of O_3, are those on terrestrial vegetation. O_3 has been shown to cause reductions in crop production, tree growth and carbon sequestration, and to modify species composition (Ashmore 2005; US EPA 2006).

Ozone can also have effects on many organisms other than plants, although these are less well documented. The vast majority of animal studies relate to the respiratory effects of O_3 on laboratory mammals, although recent studies suggest that O_3 can also induce secondary changes in body temperature, water balance or oxygen consumption in reptiles (Mautz & Dohm 2004). For insects, both positive and negative changes in performance have been observed, primarily as a result of changes in leaf chemistry (Valkama *et al.* 2007). Effects may be mediated through atmospheric reactions of O_3 with VOC, for example by reducing searching efficiency and interference with pheromone signalling of insects. For example, McFrederick *et al.* (2008) recently suggested that such effects could reduce the distance over which insect pollinators could detect floral scents from kilometres at pre-industrial O_3 levels to less than 500 m under present background concentrations. Both decreases and increases in plant fungal diseases have also been observed in response to O_3. The secondary effects of O_3 on both insect pests and fungal diseases could have significant implications for crop and forest productivity but are not well understood.

Although O_3 is used as a disinfectant for water, in sea water there is high reactivity of O_3 with iodide and dissolved organic matter, leading to the rapid destruction of O_3. This implies that any effect on marine organisms is likely to be minimal, except possibly for organisms living on the sea surface. However there has been little or no research on this topic.

The sections below provide an overview of the mechanisms by which O_3 affects vegetation; describe how O_3 effects are measured and assessed; and evaluate the current and potential future impacts of ground-level O_3 concentrations. Although O_3 can also have significant impacts on materials and the built environment, an assessment of these impacts is not within the remit of this report.

8.2 Mechanisms and assessment of ozone effects on vegetation

In terrestrial ecosystems, the most important direct effects of O_3 are those on leaf physiology and plant growth; many other indirect effects on ecosystems flow from these primary direct effects. A detailed review of the complex mechanisms involved in the direct impacts of O_3 on leaves is beyond the scope of this document, but two of the most important factors are the control of the flux of O_3 into the leaf and the capacity for detoxification and repair processes within the leaf (Wieser & Matyssek 2007). The flux into the leaf of O_3 from the boundary layer above the plant canopy is regulated by the atmospheric and leaf surface resistances including the stomata, illustrated in Figure 3.6. Since all these resistances vary with meteorological conditions, this flux is not constant, but varies greatly through time and space even with a constant O_3 concentration above the plant canopy.

Impacts of O_3 within the leaf also depend on the capacity for detoxification of the incoming O_3 flux, which itself may be partly under environmental control. In simple terms, there are two possible modes of action of O_3 within the leaf. At high exposures, O_3 flux may overwhelm the detoxification capacity and cause a range of direct effects. At lower exposures, O_3 may induce a range of defence reactions and gene expression, which require energy for regeneration of anti-oxidants and *de novo* synthesis, leading to decreased rates of carbon assimilation and plant growth.

A number of key observations related to interactions between O_3 and other environmental factors can be explained in terms of the processes of O_3 flux and detoxification. For example, increased concentrations of atmospheric CO_2 generally decrease stomatal conductance, decreasing the flux of O_3 into the leaf, and can also provide the plant with additional carbon for defence and detoxification. As elevated CO_2 decreases the impacts of O_3 in many species, the impacts of scenarios of future O_3 concentrations must be considered alongside the equivalent scenarios for CO_2 (Fuhrer 2003).

More generally, assessment of the impacts of O_3 during a century in which many environmental variables will change significantly requires a more process-based approach, linked to appropriate models (Ashmore 2005). However, assessment and quantification of the impacts of O_3 on vegetation still relies primarily on empirical exposure–response relationships between O_3 concentrations and changes in crop yield, or tree growth rates, derived mainly from chamber studies. There is a need to improve our mechanistic understanding of plant responses to O_3 and to further develop flux-based models to assess the full impacts of O_3 on vegetation.

The current empirical approaches used to quantify O_3 impacts summarise the variable O_3 exposures over the growing season by using an exposure index. A range of seasonal exposure indices have been proposed and used for risk assessment in both Europe and North America. In the USA, a detailed recent risk assessment by the USEPA (2006) identified that a range of adverse effects on vegetation occurred at exposures below the current standard, which was based on peak 8 h mean concentrations, and recommended a new standard based on cumulative seasonal exposure. In this section, three seasonal exposure indices have been used (see Box 8.1 below). Two of these indices include a threshold below which adverse effects are not expected, an observation that is consistent with the known capacity of plant cells to detoxify incoming O_3. However, this threshold is not fixed and varies between species and genotypes and with environmental conditions.

Box 8.1 Exposure indices for vegetation used in this chapter

Concentration-based approaches:

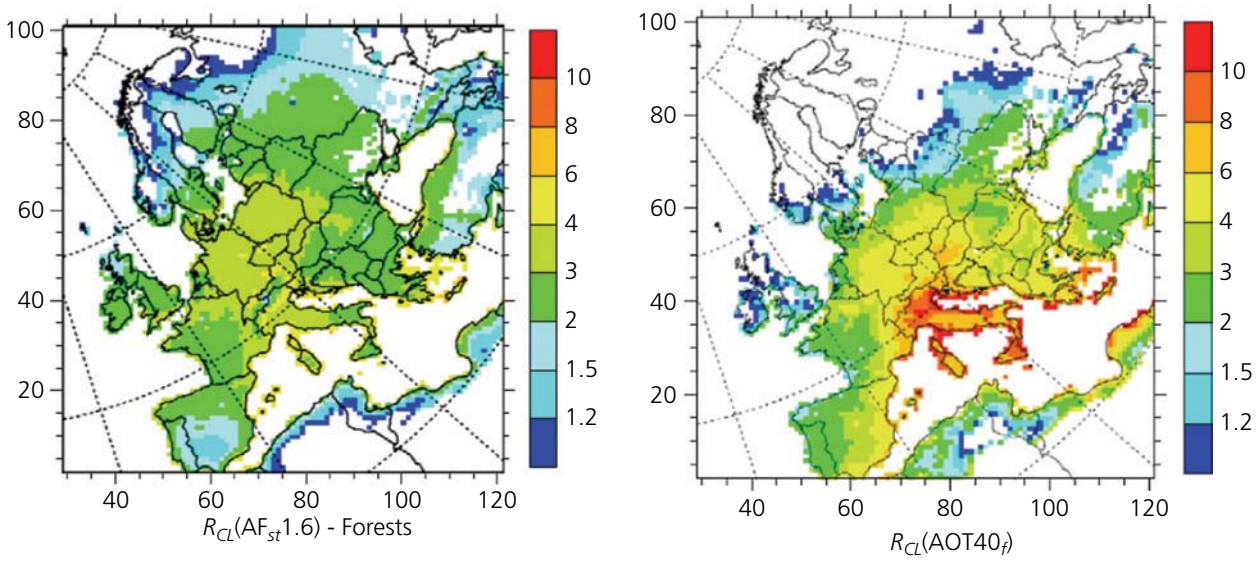
- The seasonal mean daytime concentration typically averaged over periods of 7, 8 or 12 h, originally developed from the NCLAN experimental programme in the USA (Heck *et al.* 1988). This is normally expressed as a mean concentration (ppb)
- The seasonal accumulated exposure above 40 ppb (AOT40) during daylight hours originally developed to assess data from open-top chambers in Europe (Fuhrer *et al.* 1997). This is normally expressed as a cumulative exposure (ppb h or ppm h)

Flux-based approach:

- The accumulated stomatal flux of O_3 above a critical flux threshold, more recently developed in Europe (Pleijel *et al.* 2004) to take account of the fact that O_3 effects are more closely related to O_3 flux into the leaf than to external concentrations. This is normally expressed as a cumulative uptake of O_3 into the leaf (mmol m^{-2} leaf area)

Risk assessments for O_3 effects on major arable crops and tree species in Europe show that the modelled spatial distribution of O_3 impacts in Europe is strongly dependent on the exposure index used (Simpson *et al.* 2007) (Figure 8.1). While the predicted distribution of impacts based on the AOT40 concentration index has a strong south–north gradient, reflecting the higher O_3 concentrations in the Mediterranean area, the flux-based approach shows a much more even distribution of risk which is consistent with coordinated bio-monitoring programmes of visible injury throughout Europe (Hayes *et al.* 2007a). The flux-based approach is considered to be the most appropriate of the two methods for assessment of effects, especially in the context of increasing global background concentrations, which are now close to 40 ppb (Coyle *et al.* 2003). However, its application requires flux–response relationships derived using a parameterised stomatal resistance model, which are only available for a few species. Furthermore, like other empirical models, it does not incorporate the variation in detoxification potential within the leaf, which may differ between species, with plant and leaf and age, and with environmental conditions. It should also be noted that O_3 can also affect plants through mechanisms other than those described above. In this case, a model of the effects of climate and CO_2 concentrations

Figure 8.1 A comparison of the geographical distribution of the relative risk of impacts of O_3 on deciduous forest trees in Europe using two different exposure indices. The left hand map shows the distribution using accumulated stomatal O_3 flux (AFst) above a threshold which for forests is set at 1.6 nmol O_3 m^{-2} s^{-1}, ($AF_{st}1.6$) and the right hand map shows the distribution using accumulated O_3 concentration above a threshold of 40 ppb over an assumed 6 month forest ($_f$) growth period ($AOT40_f$). For both indices R_{CL} represents the ratio of modelled flux or exposure to the relevant critical level. Source: Reprinted from Simpson et al. (2007). Copyright (2007), with permission from Elsevier.



in modifying O_3 impacts which only includes changes in stomatal flux and detoxification would be inadequate.

8.3 The impacts of ozone on vegetation

8.3.1 Impacts on crop yield and quality

There is a substantial body of evidence from North America and Europe, supported by some work in Asia, Africa and Latin America, that elevated O_3 levels cause reductions in the yield of sensitive crop species (Mauzerall & Wang 2001; Emberson *et al.* 2003), and some estimates have been made of the economic impacts of crop loss due to ambient O_3 levels. The annual cost of loss of arable crop production due to O_3 was estimated to be $2–4 billion in the USA in the 1980s, with an equivalent estimate for the EU of €6.7 billion (90% confidence interval €4.4–9.3 billion per year) in 2000 (Holland *et al.* 2006). This is equivalent to 2% of arable agricultural production, but does not account for a range of effects, including those on crop quality, visible injury, and susceptibility to pests and diseases. The greatest economic losses in Europe were predicted to be in Mediterranean countries, together with France and Germany, because the assessment used a concentration-based exposure index. Wheat, tomatoes, vegetables and potatoes were the crops with the greatest yield losses. Van Dingenen *et al.* (in press) recently provided the first global estimate of crop yield loss, for four major commodities (wheat, rice, maize, soybean), of $14–26 billion in the year 2000. This is significantly higher than present day losses to crops projected to occur as a result of climate change.

All of these estimates are primarily based on data from field chamber experiments which may under-estimate the real effects of O_3 in the field. For example, the decrease in soybean yield under open-air conditions in the Soy Free Air Concentration Enrichment (FACE) experiment, conducted in the USA, was greater than predicted by a synthesis of previous chamber studies (Morgan *et al.* 2006). In one season, this was partly because O_3 impacts increased the impact of a major defoliating hail event. More field release experiments, in which O_3 is released over a crop which is not enclosed in chambers, are therefore needed to reduce the uncertainty in future estimates of loss in crop productivity. These need to be in a range of locations and to cover different cropping systems.

The value of a crop depends on its nutritional quality as well as the yield and O_3 has also been shown to be important in this respect. Experiments on wheat in Europe suggest that O_3 has a small negative effect on protein yield per hectare (Piikki *et al.* 2007). Effects have been reported for rape seed oil and potato quality, and reductions in the nutrient content of important Indian vegetable crops, eg the iron content of spinach and the beta-carotene content of carrot (Agrawal 2007) have been observed. Ozone may also have direct effects on reproductive structures, for example, reductions in fruit quality and increased fruit abortion in peach and peppers have been reported.

There are no estimates of the economic impacts of O_3 on perennial crops. For managed pasture, studies have consistently shown that, while harvested biomass is only affected at high O_3 concentrations, reductions in the proportion of clovers in the mix are observed at moderate exposures (eg Nussbaum *et al.* 1995). This could affect the forage quality of the crop depending on the management system in place. A range of new crop species and varieties may increase in importance over the next decade with increased land devoted to biofuels. Some of the proposed biofuel crops (eg wheat, willow, poplar, oilseed rape) are known to be sensitive to O_3, but the response of many new biofuel crops (eg palm oil, panicum, miscanthus, phalaris) is not known. New work is needed to assess the extent to which O_3 may constrain future production of biofuel crops.

There is a wide intra- as well as inter-specific variation in sensitivity to O_3. Although there is evidence of cultivars bred in regions with higher O_3 concentrations being more resistant to O_3, there is little evidence that crop cultivars have become more O_3 tolerant as O_3 concentrations have risen (Barnes *et al.* 1999; Biswas *et al.* 2008). Nevertheless, the impacts of increased O_3 concentrations on crop yield, nutritional quality and food security could, in principle be reduced by the use of genotypes with a greater tolerance to O_3. There is a wide degree of genotypic variation in O_3 sensitivity and recent studies have identified some genetic loci associated with O_3 resistance in wheat, offering the potential to target certain genotypes (Barnes 2007; Ainsworth *et al.* 2008).

Much less is known about the effects of O_3 on crop yield or quality outside of Europe and North America, although significant impacts of O_3 on crops have been demonstrated in individual studies in Africa, Latin America and Asia (Emberson *et al.* 2003). The limited experimental facilities in many countries mean that it is difficult to accurately assess the impacts of O_3 for these regions. The case study below (Box 8.2) presents an assessment of the impacts of O_3 in one region – South Asia – providing both the regional food security context and a summary of the evidence of effects on yields.

Box 8.2 Case study: ozone and food production in South Asia

South Asia has more than 20% of the world's population, and 15% of the world's cropland. Agriculture remains the largest source of employment and contributes over 25% of GDP. The countries of South Asia, except Bangladesh, currently have a positive food balance, although according to the FAO, the region has the highest number of under-nourished people in the world representing about 30% of the total. The population of the region is expected to increase by about 600 million between 1995 and 2025, and this, together with other changes in income and lifestyle, will lead to large increases in demand for food and other natural resources. However, the potential for increasing the land under production is limited and there may be increased competition from biofuel production; for example India has stated it will meet 10% of the needs of its growing vehicle fleet with biofuels by 2020. Increasing the yields of existing agricultural land is likely to be the primary means by which future food supply needs will be able to be met. Reductions in productivity due to ozone pollution could compromise this potential, and may reduce the GDP growth rates for the national economies for which agriculture is a major sector.

Rice and wheat, grown in rotation, are major staple crops grown in South Asia, primarily on the Indo-Gangetic Plain. A further major production area occurs in China. This system provides staple grain for an estimated 8% of the world's population. Over the 20–30 years to 1990, production increased faster than population, primarily reflecting the impacts of the 'Green revolution'. However, yield increases have slowed, or even reversed, over the last 10–20 years. For example, Ladha *et al.* (2003) analysed the results of 33 long-term experiments on rice–wheat systems in the region, which aim to duplicate changes in farming practice, and found that yield of rice and wheat had stagnated in 72% and 85% of the experiments respectively, with 22% of the rice experiments and 6% of the wheat experiments showing a significant decline in yields. The reasons for these trends are uncertain; long-term nutrient imbalances (Ladha *et al.* 2003), climate change (Pathak *et al.* 2003), and the combined effect of global warming and atmospheric brown cloud (Auflammer *et al.* 2006) have all been proposed as possible causes.

The extent to which O_3 might have contributed to these changes in wheat and rice yield is uncertain. However, almost all of the experiments on O_3 effects in South Asia have been conducted in the Indo-Gangetic Plain and these suggest that significant yield losses of local crop cultivars can be caused by ambient O_3 concentrations. Evaluation of the impacts of O_3 on crop productivity in south Asia and the benefits of emission control measures requires regional models which can provide more reliable identification of regional areas of concern. A recent study (Engardt 2008) based on South Asian emissions inventories reported large spatial variations in modelled values of AOT40. The largest values were found in the Indo-Gangetic Plain, with values of AOT40 reaching the critical threshold for effects on crop yield of 3 ppm h throughout the year and exceeding 12 ppm h during the period March–May. This further supports the identification of this as a critical region where O_3 may have a significant impact on future food security.

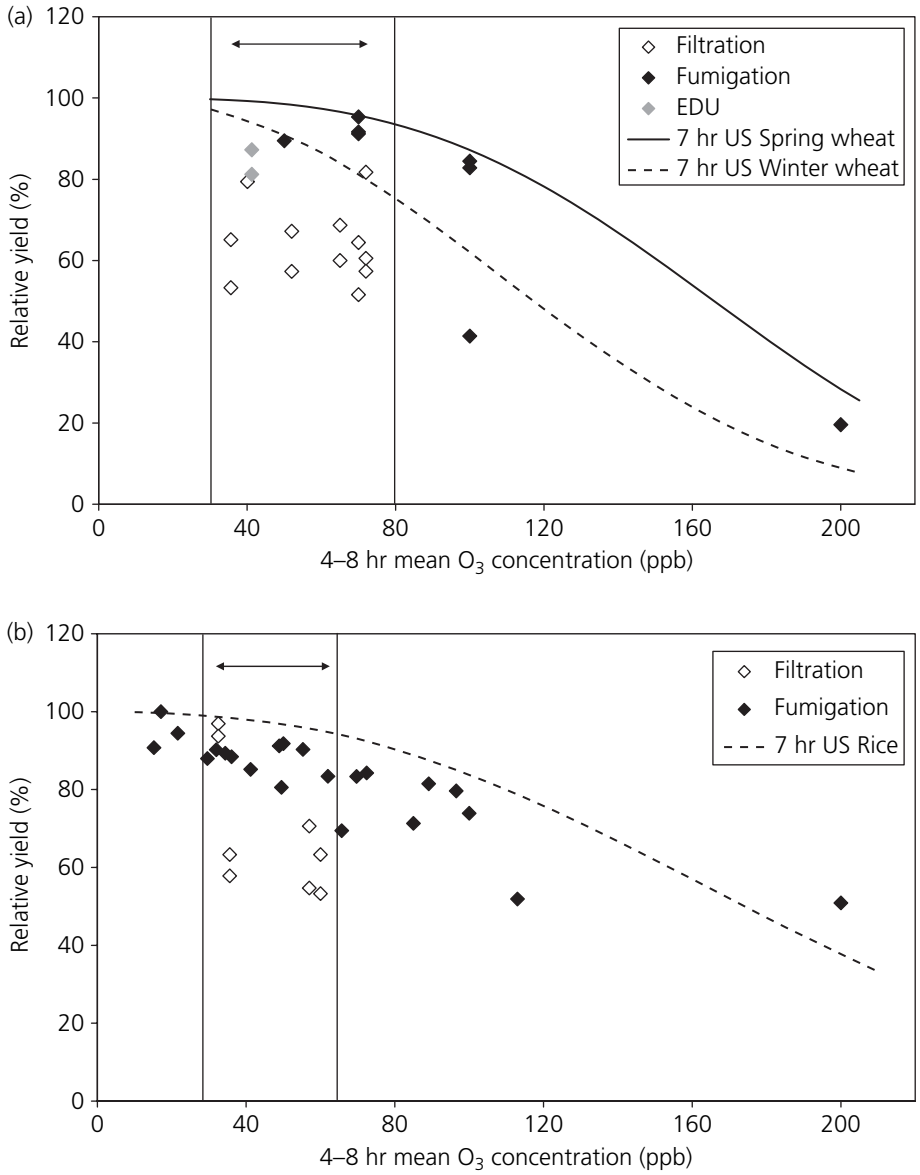
Two approaches have been taken to evaluating the impacts of O_3 on crops in South Asia:

First, a top-down approach links regional or global models of O_3 formation to exposure-response relationships to assess the effects on the yield of staple crops under different scenarios (eg Wang & Mauzerall 2004; van Dingenen in press). Recent global simulations using this approach suggest that yield losses for India overall in 2000, based on 7 h mean or 12 h mean ozone indices, were about 13% for wheat, 6% for rice and 19% for soybean (van Dingenen in press). Figure 8.2 shows examples of the exposure–response relationships used in this study, which were derived from US data for winter and spring wheat (note that in South Asia, wheat is a winter crop), and for rice.

The second approach is to rely on local experimental data, which are limited in many regions. Studies in India and Pakistan have reported yield losses due to ambient O_3 concentrations (for example 10% for rice, 13–47% for wheat, 24% for spinach and 45% for carrot) that are larger than those suggested by the 'top-down' approach. Leguminous crops such as pea and mung bean, which are vital sources of protein in the largely vegetarian diet of much of India, have shown yield losses of about 30% and 20% respectively. Figure 8.2 provides a collation of data from studies in Asia for wheat and for rice, which involve either exposure of crops to controlled O_3 concentrations, filtration of ambient air pollution, or the use of a chemical protectant against O_3 (EDU).

Although the results shown in Figure 8.2 reveal differences in the findings using the different experimental methods, there is clear evidence that the effect of O_3 in local experiments is greater than that predicted by the US-based exposure–response relationships, especially at ambient O_3 concentrations. These results imply that actual impacts of O_3 on the yield of staple crops in south Asia may be greater than those estimated in the 'top-down' approach using North American data. However, this needs more detailed examination, and it is not possible to evaluate whether this is true for other regions of the world due to the lack of experimental data.

Figure 8.2 Summary of results of experiments in South, South East, and East Asia on (a) spring and winter wheat and (b) rice, plotted alongside dose–response relationships from North American studies. Data are collated from studies exposing plants to controlled concentrations of O_3 in chambers (fumigation points) from studies comparing yield in chambers ventilated with ambient air pollution or filtered air (filtration symbols) or using the O_3 protectant chemical EDU at field sites (EDU points). The fitted lines are based on experimental data from fumigation studies in North America based on 7 h growing season mean O_3 exposures. The local ambient pollutant range is also indicated (arrowed vertical lines) based on concentrations at the Asian experimental sites. Redrawn from Emberson et al. (submitted).



8.3.2 Impacts on tree growth and carbon uptake

Ozone can cause significant effects on tree growth and rates of photosynthesis. Economic losses from the reduced yield of timber for Sweden alone are estimated to be €56 million per year (Karlsson *et al.* 2005). Studies suggest that O_3 has a cumulative effect on stem growth, although there is a lack of long-term studies to properly assess this effect. Species vary widely in their sensitivity to O_3; for example, studies on silver birch showed a 20% reduction in annual stem growth after 6 years exposure to an AOT40 exposure of 10 ppm h, while Norway spruce showed an annual stem growth rate reduction of 1% per annum in response to the same exposure (Karlsson *et al.* 2004). It has been suggested that tree species which are fast-growing pioneer species, such as birch, aspen and poplar, are relatively sensitive to O_3 compared to climax species such as beech and oak. This has implications for future O_3 effects if climax species are replaced by fast growing plantation species for agro-forestry and bio-fuel production.

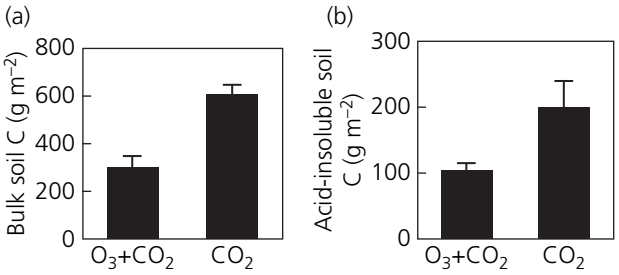
These experimental effects on the growth of different tree species can be compared with a recent meta-analysis of the effects of O_3 on photosynthesis (Wittig *et al.* 2007). In this study significant effects on broadleaved species were evident at current O_3 levels, with maximum rates of photosynthesis being reduced by about 10%, but there was no significant

effect of current levels on conifers. Hence, the size of the terrestrial carbon sink may already have been reduced by O_3, but only for sensitive broadleaved species. A further increase in O_3 concentrations would cause larger effects in a wider range of species. However, a number of factors need to be carefully considered before accepting these conclusions.

First, the effect of tree age on the level of O_3 damage is a major uncertainty, as most studies involve young trees, although negative correlations between stem growth and O_3 exposure in the field have been reported in mature trees (Braun *et al.* 1999; Karlsson *et al.* 2006). Secondly, current models assume that the primary driver of significant O_3 effects on tree growth is a reduced rate of photosynthesis which might not always be the case. For example, exposure of branches within a mature beech canopy in Germany showed that O_3 effects were greater in a strong drought year (eg 2003) than a relatively cool, wet summer (eg 2004), although stomatal conductance and O_3 flux were lower in 2003. This may reflect a reduced defence capacity of the trees under the extreme stress conditions of 2003 (Matyssek *et al.* 2007). This study showed no effect of doubling O_3 levels on photosynthesis or stem growth, but there was evidence that elevated O_3 had important physiological effects in leaves and below-ground, including induction of anti-oxidant genes, changes in the levels of key hormones, and an increase in fine root development and an increased mycorrhizal diversity. Thirdly, there is limited information on which to assess the effects of O_3 on below-ground processes (Andersen 2003) and both increases and decreases in below-ground biomass and root respiration have been reported in response to O_3.

The complex range of effects on trees caused by O_3 also highlights the importance of understanding the combined effects of O_3 and other environmental stressors. The longest running study of the effects of O_3 on trees is the open-air fumigation AspenFACE experiment, which includes an elevated CO_2 treatment, an elevated O_3 treatment and a combined elevated O_3 and CO_2 treatment. This experiment shows a 14–23% reduction in total biomass due to increased O_3 exposure after 7 years (by increasing annual mean from 37 ppb to 52 ppb) (King *et al.* 2005). Overall the elevated O_3 and elevated CO_2 treatments had opposite effects. When the two treatments were combined, elevated CO_2 generally reduced or removed the negative effects of elevated O_3 on growth and physiology. However, there were exceptions, eg the accelerated senescence and abscission of leaves caused by O_3 was not affected by elevated CO_2. Other important effects include those on bud-burst; senescence; anti-oxidant gene expression; insect herbivory; prevalence of leaf rust disease; increased water stress; and increased rates of soil respiration. The total soil carbon incorporated into these plots over four years was reduced by about 50% in the O_3 plus elevated CO_2 treatment compared with the elevated CO_2 treatment (Figure 8.3; Loya *et al.* 2003). Furthermore, a similar effect was found in the acid-insoluble fraction of carbon, which contains the compounds with the longest turnover times in soil. The wider implications of such combined effects of O_3 and elevated CO_2 for global carbon sequestration are considered in 8.4.2.

Figure 8.3 Effects of elevated CO_2 alone and elevated CO_2 and O_3 on the carbon incorporated into soils (g m^{-2}) over 4 years under aspen and aspen–birch components of the Aspen FACE experiment. (a) Total soil carbon incorporated into soils. (b) Carbon incorporated into the stable acid-insoluble fraction of soils. Source: Loya et al. 2003. Reprinted with permission from Macmillan Publishers Ltd: Nature, copyright (2003).



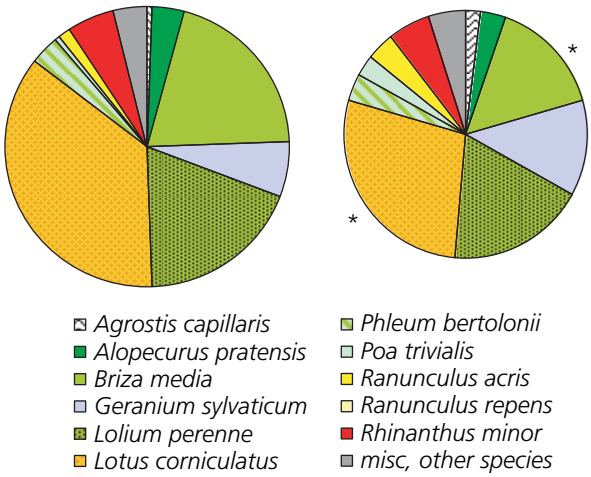
8.3.3 Ecological impacts

There is evidence from short-term studies that individual wild plants may be as sensitive to O_3 as the most sensitive known crop species (Davison & Barnes 1998). However, only a small proportion of European wild species have been studied. While faster growing species tend to be more O_3 sensitive, the most detailed meta-analysis for European species (Hayes *et al.* 2007b) showed few significant effects of growth form or habitat preference on O_3 sensitivity; the only strong effect was the greater sensitivity of leguminous species. This reflects the high sensitivity to O_3 of leguminous crops, such as beans, peas and clover.

A number of studies of real and artificial plant communities have shown that O_3 can modify the outcome of plant competition and hence species composition (eg Volk *et al.* 2005). However, there is a very limited basis on which to predict how species or communities of conservation importance may be affected by O_3. This is because only a few plant communities have been studied for only a limited duration and because many of the direct and indirect effects of other environmental variables that can modify the long-term impacts of O_3 have not been well studied. The major air pollution threat to plant biodiversity in Western Europe is anthropogenic nitrogen (N) deposition and assessing the combined impacts of these two threats to biodiversity is therefore an important priority.

The potential effects of O_3 on habitats of conservation interest in the UK can be illustrated by two recent studies. In a recent risk assessment, the O_3 exposure and sensitivity of Priority Habitats under the Biodiversity Action Plan were evaluated using national distribution maps and the classifications of species sensitivity of Hayes *et al.* (2007b). On this basis, Priority Habitats with relatively high O_3 exposures, and with O_3-sensitive positive indicator species, included species-rich acid and calcareous grasslands and lowland fens and mires (Morrissey *et al.* 2007). In a recent experimental study (Peacock *et al.* personal communication) the effects of O_3 were assessed on mesocosms of upland grassland under

*Figure 8.4 Impacts of simulated present day versus 2050 UK-upland O_3 climate on an upland mesophilic grassland community of high conservation importance. Data are presented for 2006 (third year of study), averaged across soil fertility and management treatments. Coloured segments represent the proportion of above-ground harvested biomass as different species; for those labelled * the effect of O_3 is significant. The relative sizes of the two circles indicates the effect of O_3 on total biomass. Data provided by Dr Simon Peacock, University of Newcastle.*



management regimes being used to restore biodiversity under environmental improvement schemes. The data (Figure 8.4) show that in this case O_3 reduces the proportion of the grass *Briza media* and the legume *Lotus corniculatus* which are both important amenity species. The reduction in *Lotus* is consistent with the high sensitivity of legumes highlighted by Hayes *et al.* (2007b), and may reduce nitrogen availability in this grassland system.

8.4 How significant will the global impacts of ozone be on the environment over the next century?

The model simulations discussed in chapters 5 and 6 illustrate how peak and background O_3 concentrations may change over this century, depending on the emission scenarios that are used. The long-term consequences of these changes in O_3 exposure and the associated changes in climate and CO_2 concentrations, are important elements of any assessment of the impacts of O_3 over the 21st century. This raises serious challenges as the longest recorded experiment on O_3 impacts for vegetation is 10 years and our knowledge of the long-term effects of O_3 on biodiversity, biogeochemical cycling and ecosystem services is very limited. Furthermore, the changes in O_3 exposure over this century will be accompanied by other global environment changes in, for example, temperature, CO_2 concentrations and rainfall, which will modify the impacts of O_3. The discussion below focuses on the three most significant environment impacts of O_3 for global ecosystem services and human welfare in the future: the effects of O_3 on crop production and food security; carbon sequestration; and biodiversity. Robust quantitative prediction is not possible therefore only preliminary estimates of effects on crop production, carbon sequestration, and global biodiversity are provided.

8.4.1 Crop production

To provide these preliminary estimates of the changing impacts of O_3 on the yield of staple crops, it is necessary to use relationships based on one of the metrics summarised in Box 8.1. Unfortunately, these require modelled hourly concentrations which were not available from the models used for the analysis of changes in O_3 concentrations to the middle of this century and beyond, as described in chapter 5. Predictions to the year 2030 were therefore developed using the IIASA 2030 CLE scenario (discussed at the Royal Society workshop in May 2007) to provide the preliminary estimates below (van Dingenen *et al.* in press). To do this it is first necessary to establish a global baseline of the current impacts of O_3. Figure 8.5 illustrates results obtained by van Dingenen *et al.* (in press) using the TM3 chemical transport model (Dentener *et al.* 2005) which provides hourly simulations of surface O_3 concentrations on a 1° grid. The estimates of yield loss were based on two different indices: AOT40 and seasonal mean concentration. The estimate in the bar chart is based on the mean of the two exposure indices, while the error bars indicate the range of estimates obtained using different exposure indices.

Estimated global yield losses for 2000 using seasonal mean O_3 concentrations ranged from 3% for rice to 16% for soybean, while those based on AOT40 ranged from 2% for maize to 12% for wheat. However, these global mean estimates conceal large regional differences. For example, the yield loss estimates for wheat and rice (but not maize and soybean) in India were double the global mean, and wheat in sub-saharan Africa is also particularly affected. The high uncertainty in the estimates depending on the exposure metric, together with that in modelled O_3 concentrations and the dose–response relationships, means that these yield losses can only be taken as indicative of the scale of the global and regional impacts. Furthermore, the estimates in Figure 8.5 are based on North American and European exposure–response relationships and, as noted in section 8.3.1, the impacts on sensitive crops in specific regions such as South, East and South-East Asia may in fact be greater, with significant effects on local food supply and livelihoods.

Figure 8.6 shows the modelled change in yields of these four staple crops predicted between 2000 and 2030 under the IIASA CLE scenario (van Dingenen *et al.* in press). It should be noted that this 2030 IIASA CLE scenario only assumes the full implementation of legislation adopted up to 2002[11] and

11 The scenario used in Chapter 5 included legislation adopted between 2002 and 2006.

Figure 8.5 Global and regionally aggregated relative yield losses in 2000 for rice, wheat, maize and soybean (van Dingenen et al. in press). The bars indicate the average values using two different exposure indices (AOT40 and either M7 or M12) while the error bars give the range of values using the two indices.

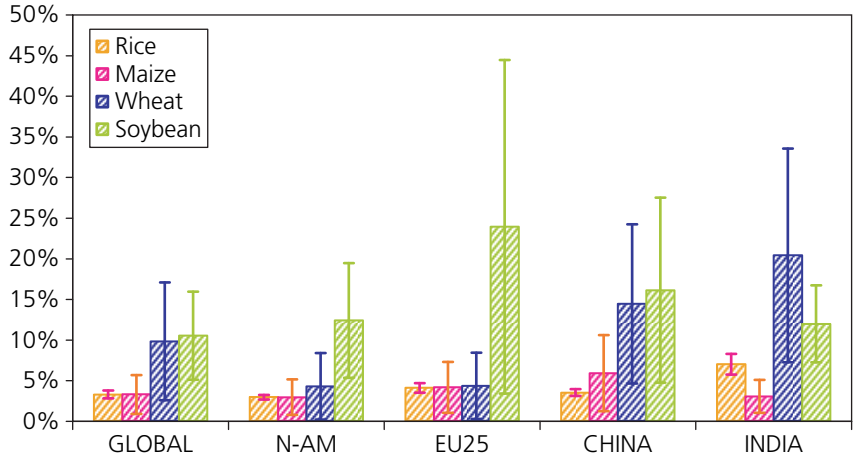
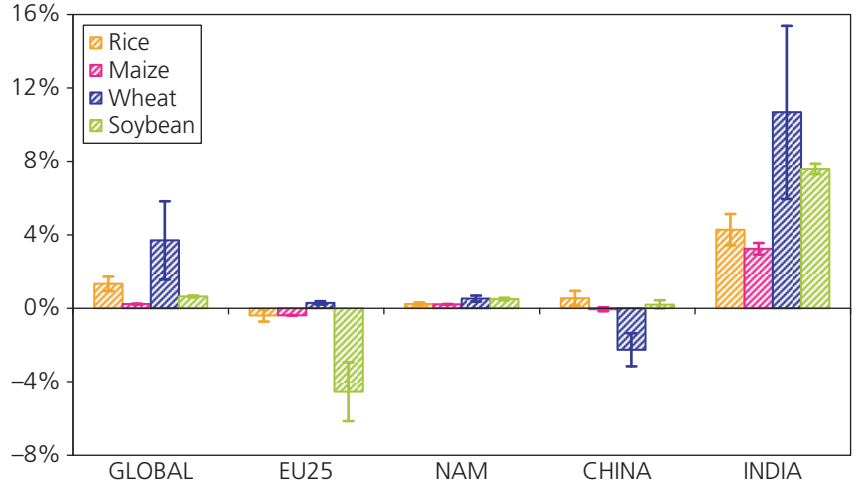


Figure 8.6 Projected changes in relative yield loss by 2030 'under implementation of current (2002) legislation' (CLE scenario) for the globe and major world regions. Negative numbers indicate a lower loss. Bars: average of AOT40 and M7/M12 based loss estimate. Error bars indicate the range between lowest and highest values.



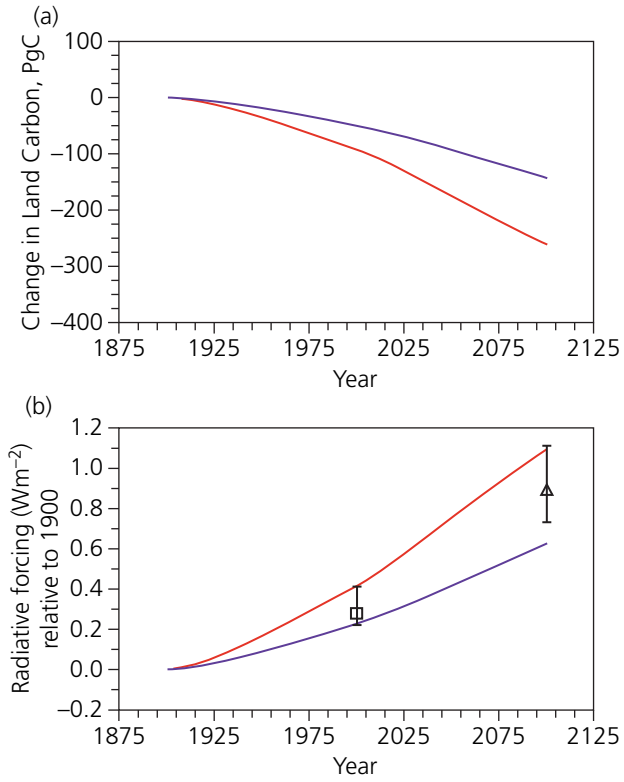
does not make any assumptions about climate change. Under this scenario, global yield losses due to O_3 increase for wheat and rice, but show little change for soybean and maize. The increase in India is particularly large whereas a smaller increase and even a decrease in relative yield loss is predicted for China. This could be due in part to the fact that this scenario assumes that only legislation adopted up to 2002 is implemented. As much of the legislation in place in India was adopted after this date it was not included in the scenario. In contrast, China had adopted stricter emissions controls by 2002 (see Chapter 4 figure 4.5). These results suggest that the full implementation of current emissions controls could partly reduce the otherwise potentially large crop losses in Asia. More detailed assessment of the impacts of O_3 further into the future would need to consider its effects in combination with key elements of climate change, such as temperature, soil moisture and CO_2 concentrations.

8.4.2 Carbon sequestration

There have been few global modelling studies of the impact of tropospheric O_3 on plant production and global land carbon storage (Denman *et al*. 2007), but a recent study suggests that O_3 feedback effects on vegetation could significantly amplify the radiative forcing due to increases in tropospheric O_3, increasing the importance of O_3 as a significant driver of 21st century climate change. Sitch *et al*. (2007) modelled the interactions between the effects of CO_2 and O_3 on plant function using an A2 scenario, which produces significant increases in surface O_3 over much of the globe[12] (Gauss *et al*. 2003). Sitch *et al*. (2007) predicted a

12 Sitch *et al*. (2007) parameterised their model for 'low' and 'high' plant ozone sensitivities; however, since their 'high' sensitivity represents the most sensitive groups of crops and trees, their 'low' sensitivity scenarios may be more realistic for global vegetation modelling, and are discussed in the body of the text here.

Figure 8.7 The simulated change in (a) land carbon storage due to O_3; (b) indirect radiative forcing of O_3 relative to 1900. The red line is for a 'high' plant sensitivity to O_3 and the blue line for a 'low' plant sensitivity. The black symbols and bars are estimate of the direct radiative forcing due to O_3 in 2000 and 2100 Source: Sitch et al. 2007. Reprinted with permission from Macmillan Publishers Ltd: Nature, copyright (2007).



reduction in land carbon storage over the period 1900–2100 of 143 PgC (Figure 8.7a), which is equivalent to a reduction of 17% of the terrestrial carbon storage projected to occur due to increasing CO_2 concentrations over this period. Such a suppression of the terrestrial carbon sink results in additional anthropogenic CO_2 emissions accumulating in the atmosphere and, therefore an indirect radiative forcing of climate change by O_3. Sitch *et al*. (2007) estimate an indirect forcing by 2100 of 0.62 W m^{-2} due to this mechanism (Figure 8.7b), which is only slightly lower that the mean direct radiative forcing due to tropospheric O_3 estimated from 11 atmospheric chemistry models, of 0.89 W m^{-2} (Gauss *et al*. 2003). Under the more moderate B2 scenario, the effects of O_3 on land carbon storage would be reduced, but the direct radiative forcing due to O_3 would also decrease. Therefore the relative importance of indirect and direct radiative forcing due to O_3 is likely to be relatively insensitive to the chosen scenario. Further work is needed to confirm the size of this indirect radiative forcing as the model used by Sitch *et al*. (2007) does not include all the relevant processes involved in O_3/CO_2 interactions, and needs further validation against experimental data.

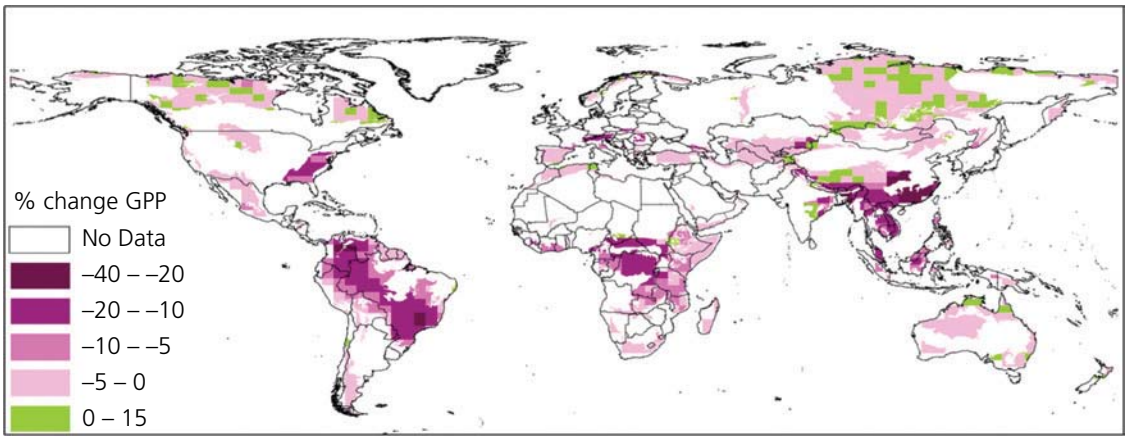
This type of large-scale modelling study has significant uncertainties, not least because model parameterisation is based on a small number of long-term experiments, which do not sample the full range of global plant functional types. In particular, the response of non-temperate species is almost unknown, even though the greatest effects on carbon uptake are predicted in these regions. Reducing the uncertainties of future predictions also requires longer-term data on the combined effects of O_3, CO_2, N deposition and climate change on land carbon storage, and an improved understanding of the mechanisms underlying O_3 damage. Nevertheless, other studies using different model approaches also suggest that there is a significant, and previously neglected, indirect effect of O_3 via the land carbon sink. For example, Felzer et al. (2005) estimated that over the 21st century O_3 will be responsible for a reduction in the land carbon sink of up to 0.4 Gt/y which would increase the cost of stabilising CO_2 at 550 ppm by 6–21%. To date, these studies assume all effects of O_3 are mediated through CO_2 fixation, although there is evidence that O_3 may also modify soil CH_4 fluxes from wetland systems (eg Rinnan *et al*. 2003).

8.4.3 Biodiversity

The impacts of O_3 on biodiversity on a global scale are unknown, largely because there are almost no studies of the sensitivity of ecosystems to O_3 outside North America and Europe. To assess the potential future impacts of O_3, the estimates of changes in global plant productivity (GPP) due to O_3 over the period 1900–2100 from Sitch *et al.* (2007) have been compared with the location of priority conservation regions around the world using a map of the Global 200 (G200) eco-regions (Olson & Dinerstein 2002). If the threat to biodiversity increases in proportion to the modelled reduction in GPP, this analysis suggests that the areas of greatest risk are in eastern North America, Central Europe, the northern half of South America, Central Africa and South-East Asia (Figure 8.8); all of these five zones contain one or more G200 eco-regions.

In total, 17 of the G200 ecoregions, covering an area of 1.4 million km^2, have a predicted decrease in GPP above 20% due to O_3. Adverse effects of O_3 on sensitive native plant species have been demonstrated both in the Appalachian mountains of North America, and in the Swiss mountains, where this value of a 20% reduction in GPP is exceeded (Figure 8.8). However, nothing is known about the sensitivity of native vegetation in the forests, grasslands and savannah eco-regions in Latin America, Africa and Asia which are identified in this analysis as having high risk of O_3 impacts, and reduction in GPP due to O_3 may not necessarily be associated with a loss of biodiversity. Comparison with a similar exercise, to identify biodiversity hotspots with the greatest rates of nitrogen deposition (Phoenix *et al.* 2006) shows that some of the hotspots at high risk from O_3 effects coincide with those at high risk from N deposition, including the forests of South-East Asia and South-West China and the Cerrado of Brazil. These findings on the risk of O_3 impacts in different ecoregions also need to be considered in the context of the major threats to biodiversity from habitat destruction and climate change.

Figure 8.8 Assessment of key biodiversity areas at high risk from O_3 impacts. The figure shows the projected percent decrease in GPP due to O_3 within the Global 200 priority conservation areas. Projected O_3 concentrations (based on SRES A2 global future simulations for the period 1901–2100) are taken from Sitch et al. (unpublished data, used for Sitch et al. (2007) and adapted from Crown copyright data supplied by the Met Office). A 'low' O_3 sensitivity is overlain with the locations of the 'Global 200' priority conservation areas (adapted from Olson & Dinerstein (2002) with permission from Missouri Botanical Garden Press). The colours indicate the predicted decrease in GPP in each eco-region; regions without any colour fall outside the G200 areas. The areas in deep purple are identified as regions where the risk of adverse effects of ozone on biodiversity is most significant at a global scale.



8.5 Conclusions

Terrestrial vegetation is directly impacted by ground-level O_3, although there are large differences in plant sensitivity both within and between species. O_3 has direct effects on leaf structure and physiology and causes visible leaf damage and reductions in growth and yield in sensitive plant species. These may lead to long-term effects on ecosystem structure and function.

The choice of exposure index can dramatically alter model predictions of O_3 impacts. In the European region the application of a deposition flux-based approach has demonstrated a more even distribution of impact than other models. This approach provides a stronger mechanistic basis for assessing O_3 impacts, especially in the context of long-term environmental change.

Assessment of O_3 effects on ecosystems should consider interactions between concentrations of O_3, CO_2 and other factors, such as climate change and N deposition. In particular, the future impacts of O_3 should be considered alongside those of increasing CO_2 concentrations as the predicted benefits of increased CO_2 concentrations in terms of increased carbon storage and increased crop yields will be offset or lost if O_3 concentrations increase sufficiently. Detrimental effects of O_3 on terrestrial carbon uptake will lead to more anthropogenic CO_2 accumulation in the atmosphere and an indirect radiative forcing of climate change that adds significantly to the direct O_3 forcing, increasing the importance of O_3 as a driver of climate change.

Global yields of staple crops are already reduced as a consequence of current O_3 exposure.

The impact of O_3 on crop production may increase over the next few decades in some rapidly developing regions, especially without the full implementation of current legislation to reduce emissions. Significantly increased impacts of O_3 are projected in South Asia where there is evidence both of substantial impacts on yield of current O_3 levels and that the local sensitivity of crops may be greater than in North America or Europe. In South Asia, and other developing regions, O_3 could present a significant threat to national food security, although it has received much less recognition than the impacts of climate change.

Evidence suggests that wild plant species in Europe and North America can be as sensitive to O_3 as the most sensitive crop species, but very few studies have assessed the long-term effects of O_3 on biodiversity or ecosystem services. An analysis of O_3 effects on global plant productivity over the 21st century shows that significant impacts on production, and hence potentially on ecosystem function and biodiversity, are predicted to occur under a high emission SRES A2 scenario in major eco-regions of high conservation priority, in Latin America, Asia and Africa, as well as in parts of Europe and North America. The long-term effects of O_3 on biodiversity are particularly uncertain at lower latitudes where almost no research has been undertaken.

9 The policy implications of projected changes in ozone concentrations over the 21st century

The preceding chapters show that O_3 remains an important air pollutant in all populated regions of the world, and despite substantial control measures taken over the last few decades to reduce emissions, continues to have significant impacts on human health and the environment. In many regions, including Europe and North America, the hemispheric background O_3 entering from sources elsewhere in the Northern Hemisphere contributes substantially to ground-level O_3 in the region, and is outside the geographical frame of current control strategies. The analysis therefore emphasises the importance of ground-level O_3 as a global air pollutant, and shows that a successful approach to O_3 management must address the hemispheric scale of the problem, as well as the regional and local scale issues.

The objective of this chapter is to highlight the policy relevant conclusions from the previous chapters. Three specific aspects of policy are also considered in this chapter: O_3 and food security, policy integration (identifying co-benefits of integrated policy development), and capacity building to increase the capacity for addressing ground-level O_3 issues at a global scale.

9.1 Emission controls

Emission controls are the primary policy instrument for managing ground-level O_3. The increasing trend in background O_3 greatly reduces the benefits of declines in peak O_3 concentrations achieved through existing emissions controls. Emissions controls must therefore be targeted at reducing both peak and background O_3, and address both the local production of emissions, and the contribution of hemispheric scale transport of O_3 and its precursors in a specific location. A new regulatory approach designed to address O_3 at the global, regional, and local scale is now required. Options for an international mechanism to provide a globally coordinated approach to air pollution and ground-level O_3 specifically, should be identified and evaluated.

The analysis shows that with the full implementation of the legislation considered in the new B2+CLE scenario, that it is possible to reduce, or keep constant seasonal mean O_3 concentrations globally by 2050 compared with 2000. Future O_3 concentrations are therefore largely under human control and depend on the action taken by national governments to implement and enforce emissions controls. The impacts of elevated O_3 concentrations on human health and vegetation are well established, and evidence now suggests that significant impacts occur even at ambient concentrations. It is probable therefore, that even with the full implementation of current controls that O_3 effects on human health and the environment will continue and may increase in some regions.

Keeping future global O_3 close to 2000 concentrations is contingent on the assumptions used in the scenario. Deviations in growth in emissions from Asia, and from shipping were identified as major uncertainties in the analysis. Recent estimates suggest that growth in China and India may be greater than that forecast in the IPCC scenarios. If this is the case, emissions will grow faster in the next few years than the scenarios predict and stronger emissions controls will be needed.

The current growth in shipping emissions is higher than that assumed in the emissions scenario, and the technology scenario adopted in this analysis is more ambitious than the controls currently being discussed by the International Maritime Organization (IMO). Projected increases in the volume of ship movements and growth in shipping emissions over forthcoming decades are expected to outweigh the benefits of existing NO_X controls (Cofala *et al.* 2007b). The IMO has demonstrated limited willingness to strengthen emission control measures to date although in early 2008, new limits for NO_X emissions from marine engines were agreed and will be submitted for adoption, along with draft guidelines for VOC emissions, later in 2008.

Aviation is an increasing source of NO_X emissions and is one of the fastest growing industrial sectors with growth between 2000 and 2020 projected to be 5% per annum (Gauss *et al.* 2005). Emission reduction technologies and practices are under development but their efficiency is currently smaller than 50%. The projected growth in air transport over the next few decades is expected to overwhelm the benefits afforded by these technological improvements.

Rapid, unregulated growth in emissions from these sectors will undermine global efforts to reduce ground-level O_3 so it is important that shipping and aviation are integrated within air quality regulatory frameworks, and that research is continued into the development of emission reduction technologies. The IMO and ICAO should be encouraged to regulate NO_X, CO and VOC emissions as far as is technically feasible and to implement agreed emissions controls. To ensure that emission reduction policies for these sectors are compatible, links should be formally established between the IMO, UNECE and UNFCCC to enable the joint consideration of air quality and climate change impacts with the objective of reviewing the state of play and identifying priorities for future work.

At the regional level, seasonal mean O_3 concentrations are projected under the new B2+CLE scenario to either increase or decrease depending on the emissions profile of the region. During summer, reductions of 5 ppb are projected for the Northern Hemisphere mid-latitudes, with declines of up to 15 ppb projected for parts of North America. In other regions such as Asia and Africa, increases in emissions will lead to higher O_3 concentrations in 2050. Stronger emissions controls will be needed if further impacts to human health and the environment are to be avoided in these regions. The analysis shows that it is the changes in mainly NO_X emissions, but also CO, nmVOC and CH_4 that will be the primary influence on O_3 concentrations in 2050.

Increases in power generation and traffic volumes are expected to lead to increases of NO_X emissions in Asia. The implementation of current emissions controls for mobile sources is expected to stabilise emissions from this sector by 2050. However stronger controls will be needed to reduce emissions from power generation. In Africa, NO_X emissions are also expected to increase as controls have not yet been adopted for mobile or stationary sources other than the installation of new large combustion plants. In Latin America, Africa and Asia, emissions from deforestation, savannah burning and agricultural waste are double those from energy sources, and are therefore significant contributors to CO and NO_X emissions. As few biomass burning emissions controls are currently in place in these regions, emissions from these sources are projected to remain high or to decline only slightly by 2050. A wide range of low-cost CH_4 control options are available for waste treatment, disposal, sewage, gas distribution, and coal mining and are now widely applied in Europe, North America and Japan. These measures have not yet been adopted in developing countries but would provide a significant contribution to the reduction of CH_4 emissions in these regions. Implementation of low cost CH_4 emission reduction technologies should be implemented in the agricultural and energy sectors in Asia, Latin America and Africa.

The exposure of urban populations to O_3 is projected to increase in many cities by 2050 due to reductions in local NO_X concentrations. As NO_X emission controls are implemented and emissions decline, as has been observed over the last decade in London, urban O_3 concentrations will increase. The trend in background will also be an important factor in determining urban O_3 concentrations, as an increasing background will contribute to higher urban O_3 concentrations while a decline will help to counteract the reduced NO_X titration effect, potentially leading to lower exposure. Control of VOC emissions is an effective option for reducing the generation of O_3 in urban plumes, and is therefore especially important as a countermeasure to compensate for reductions in the titration effect within urban areas. At the regional scale however, where O_3 concentrations are more determined by the long-term processes that form O_3, VOC controls are less efficient. The UK is currently an exception to this. In contrast to much of continental Europe, regional scale O_3 formation is VOC limited and VOC emission reductions could also provide a useful contribution for controlling O_3. Appropriately designed VOC emission control measures should therefore be included in regional and urban air quality strategies.

9.2 The implications of climate change for future ozone

Modelling results suggest that climate change will not have a major influence on ground-level O_3 in 2050 at the global scale, but the analysis suggests climate change is important at the regional and local scale. The effects of climate change should be taken into consideration when O_3 control policies and emission reduction targets are developed. In urban areas for example, emission reductions will need to be strengthened and exposure management plans reviewed, to take into account the combined effects of climate change and reduced NO_X titration, and the projected increases in frequency and magnitude of conditions conducive to high O_3 events. In those regions where O_3 is projected to decrease due to declines in emissions, climate change will reduce the benefits of these reductions and stronger emissions controls will be needed to achieve the same result. Climate change is expected to increase ground-level O_3 problems in regions where emissions are projected to rise, suggesting that emissions controls will need to be significantly strengthened if higher O_3 impacts are to be avoided.

9.3 Implications of future ozone projections for food security

Under current O_3 concentrations, significant impacts to crops in Europe and North America have been observed. Although much less information is available in other regions, significant impacts on staple crop yield and quality have also been demonstrated in experimental studies in Africa, Latin America, and Asia. As the modelling projections and scenario analysis suggests that emissions and therefore O_3 concentrations will increase in many regions of these continents to 2050, impacts to crops are also likely to increase. These findings must be placed into the wider context of future food security, globally, nationally and locally.

All of the IPCC SRES socioeconomic scenarios (except A2) predict a large decrease in numbers of people at risk of hunger over this century, due to an increase in income and food production. Climate change scenarios generally reduce the extent of this decrease in hunger, but they do not reverse it. However, these analyses do not incorporate the effects of O_3 especially in reducing or reversing the positive direct effects of increased CO_2 concentrations on crop productivity. Hence the key issue in terms of O_3 exposure is how far it may further delay achievement of critical Millennium Development Goals in terms of reducing the estimated 800 million people worldwide who are undernourished. Timescale is important in this assessment. The primary benefits of climate change mitigation measures for agriculture are likely to fall in the latter half of the century. In contrast, this report suggests that the greatest impacts of O_3 are likely to fall in the first half of the century. The negative impacts of O_3 must therefore be minimised before temperatures increase sufficiently to have significant negative impacts on global crop yields.

For climate change, and the associated effects of increased CO_2 concentrations, small beneficial effects of moderate warming are predicted for croplands in temperate regions. However even moderate temperature increases are predicted to have negative effects at low latitudes and in seasonally dry regions. Furthermore, many of these sensitive low-latitude regions have a lower capacity to adapt to adverse effects of climate change. Hence, while the analysis of Easterling *et al.* (2007) predicts that temperature increases of 1–3°C have relatively small effects on food supply worldwide, it identifies regions, such as sub-Saharan Africa, where climate change and socioeconomic conditions may lead to large negative

effects on food security. The results presented in chapter 8 suggest that the future effects of O_3 on staple food production may be greatest in developing low-latitude regions. A key question is therefore whether there are regions where both climate change and O_3 may have adverse effects on the yield of staple crops and hence on food security.

The O_3 concentrations projected for 2030 (see chapter 8) under a different version of the B2+CLE scenario are unlikely to have major additional impacts on overall global food security, although the effects of O_3 in negating the positive effects of a combination of moderate warming and increased CO_2 concentrations could be substantial if there was no control of precursor emissions. A much greater concern is the effect on food security in sensitive regions with high numbers of undernourished people where O_3 concentrations may increase for the next 20–30 years even under the new B2+CLE scenario. A similar point was made by Wang and Mauzerall (2004) who identified that countries in East Asia are on the cusp of large decreases in the yield of staple crops due to O_3.

It is possible that these impacts of O_3 on regional food security could be mitigated by measures to reduce the sensitivity of local cropping systems. The most obvious approach would be the use of genotypic variation in O_3 sensitivity especially as there is some evidence that recently introduced cultivars are more sensitive to O_3. For example, a recent study showed that an increased yield of winter wheat cultivars introduced over the last 60 years in China has been associated with an increased sensitivity to O_3, partly through higher stomatal conductance and a lower anti-oxidative capacity (Biswas *et al.* 2008). Currently, successful biotechnology applications come from engineering single genes into crops, and this approach will be much more difficult to apply to complex traits such as responses to O_3 stress (Ainsworth *et al.* 2008). Hence, key priorities for the future are to integrate O_3 tolerance into national breeding and selection programmes and into research programmes on the use of biotechnology to increase tolerance to stressors such as drought and high temperatures.

9.4 Improving policy integration

Improving the integration of O_3 policy with policies aimed at reducing other air pollutants, and mainstreaming of air quality policy into other policy areas such as food, climate and energy policy would provide an important contribution to the delivery of the emission reductions required to avoid a high O_3 future.

To demonstrate the potential benefits of improving integration the table below illustrates the interactions between different emission control strategies for O_3 reduction, and the potential implications for a range of different environmental issues.

The table demonstrates a substantial range of potential co-benefits arising from O_3 precursor controls at different scales. This is based on the relative magnitude of effects, and should be only taken as indicative of potential cross-benefits, as there are many uncertainties involved. This simple analysis highlights the following points:

- Controlling NO_X emissions has benefits for human health as a result of reducing rural O_3 concentrations and PM. There are also potential benefits to biodiversity due to reductions in both N deposition and O_3 as NO_X emission reductions will lead to a significant decrease in acidification, and N deposition is a strong factor in eutrophication of terrestrial and marine ecosystems. As NH_X deposition is not affected, the biodiversity benefits of NO_X control are medium;
- NO_X controls also have benefits for visibility by reducing PM and O_3;
- NO_X controls may have a mixed effect on carbon sequestration through reductions in N deposition and O_3; ie carbon sequestration could either decrease due to reductions in N fertilisation, or increase due to reduced O_3 impacts. However, since the long-term effects of N deposition and O_3 on carbon sequestration in different ecosystems are uncertain, and may depend on climate, soils and future levels of CO_2, quantitative comparison of these contrasting effects is very uncertain;
- VOC controls benefit human health by reducing O_3 at the urban and rural scales. There is also a benefit to C sequestration and climate change, as a result of the reduction in O_3;
- CH_4 controls benefit human health due to the effect of reducing rural O_3. Methane reduction measures also provide other environmental and economic benefits such as ground-water protection, and enhanced recovery of gas. Lower CH_4 emissions will also have climate benefits;
- The reduction of O_3 due to NO_X, VOC and CH_4 controls has benefits for human health, biodiversity, acidification and eutrophication, visibility, climate system, and carbon sequestration.

Co-benefits may not always be possible, for example, NO_X (NO_2, NO, N_2O and particles) controls are essential for protecting human health and the environment. However, NO_X controls are a primary driver of the increase in O_3 concentrations in urban areas. Integrated policy making enables the early identification of such issues, and action to reduce the potential negative consequences. In this case for example, if O_3 control is also an objective, NO_X controls must be accompanied by appropriate regional and urban VOC controls.

Integrated assessment models are useful analytical tools designed to improve the integration of air quality policies. The IIASA Regional Air Pollution Information and Simulation (RAINS) model has been widely used to identify cost effective strategies to control health impacts from $PM_{2.5}$ and O_3, and the vegetation impacts from acidification, eutrophication and O_3. Control measures from all sources of anthropogenic SO_2, NO_X, PM, NH_3, and VOC emissions in all sectors are considered (Schopp *et al.* 1999). This integrated assessment model was used in Europe to inform the development of the NEC Directive (European Community 2001), and the Gothenburg Protocol, and has also been applied in Asia with the objective of reducing SO_2 pollution. Harmonised approaches such as the RAINS model could be particularly useful to demonstrate co-benefits in regions where the development of environmental policies is still in its early stages, particularly in rapidly developing regions where

Table 9.1 Co-benefits of a far-reaching strategy to control O_3 precursor emissions. The table shows how other environmental problems (specified in the left column) are influenced by emission reductions of O_3 precursors (NO_X, VOC and CH_4, top row).

Environmental problem	NO_X control	VOC control	CH_4 control	Combined control
Human health				
NO_2 urban	Large	Negligible	Negligible	Large
O_3 urban	Medium	Medium	Small	Medium
O_3 rural	Medium	Medium	Medium	Large
PM	Medium	Small	Negligible	Medium
Biodiversity				
N deposition	Medium	Negligible	Negligible	Medium
O_3	Small	Small	Small	Small
Acidification and eutrophication				
N deposition	Medium	Negligible	Negligible	Medium
O_3	Negligible	Negligible	Negligible	Negligible
Visibility				
PM	Medium	Small	Negligible	Medium
O_3	Small	Small	Small	Small
Climate change				
O_3	Medium	Negligible	Medium	Medium
PM direct	Small	Negligible	Negligible	Small
PM indirect (cloud)	Small	Negligible	Negligible	Small
Carbon sequestration				
N deposition	Medium	Negligible	Negligible	Medium
O_3	Medium	Medium	Medium	Medium

economic growth is the overriding policy priority. This analytical framework could be extended to other policy areas where there are obvious interactions between objectives. For example, improving the integration between agricultural and air quality policy, climate change and O_3 policies would have obvious benefits. The Greenhouse Gas Air Pollution Interactions and Synergies (GAINS) model, is one such analytical tool, designed to increase the cost effectiveness of air pollution and climate strategies (Klaassen *et al*. 2004).

Climate change and O_3 are two policy areas where there are obvious co-benefits of improving integration. The interactions between climate change and tropospheric O_3 include:

- CO_2 fertilisation from atmospheric CO_2 emissions may offset negative O_3 impacts in some plant species with implications for food security, biodiversity and carbon sequestration;
- Control of O_3 precursors may have a climate impact; NO_X tend to increase OH, reducing the lifetime of methane (CH_4) and reducing CH_4 concentrations, producing a negative radiative forcing (although this is location and seasonally dependent). CO, CH_4 and VOC emissions, however, tend to reduce OH, increasing the CH_4 lifetime, producing an additional positive radiative forcing;
- O_3 impacts may reduce biomass and therefore carbon sequestration in forests, both in the timber and in soils, with implications for timber production, and the climate system;
- O_3 is a greenhouse gas with a radiative forcing (since 1750) of between 0.25 and 0.65 W m^{-2};
- Climate change impacts on O_3 production and destruction may offset the benefits of emission reductions in polluted regions;

- Climate change driven changes in weather may induce high O_3 episodes events causing significant health impacts in regional and urban areas;
- The combination of climate change and O_3 impacts on agricultural yields may reduce food (and energy) security over the long term.

The international regulatory frameworks are currently independent of each other despite the importance of O_3 as a greenhouse gas, and the role of climate in O_3 production, destruction and transport processes. For example, O_3 is not managed under the UNFCCC umbrella as the Kyoto Protocol does not include O_3 in the basket of greenhouse gases. Recently the policy and research communities (particularly within the CLRTAP) have begun to acknowledge the links and to consider whether they should be better integrated, but this work is still in its infancy.

The primary objectives of both climate and air quality policy are to reduce emissions, and as they share many of the same drivers and sources (eg agriculture, deforestation, energy production and transport), there would be a clear advantage in developing coordinated emission reduction strategies. Methane for example, is both a greenhouse gas and O_3 precursor, and so is an obvious candidate. Reductions in emissions of CO and nmVOC would also have climate benefits. Similarly, as biomass burning is such a large contributor to emissions of O_3 precursors and greenhouse gas emissions the implementation of policies to reduce or halt deforestation, wetland/peatland drainage and biomass burning, would have the dual benefit of contributing to climate change and O_3 policies. There are however examples of climate emission reduction strategies which may undermine O_3 objectives. For example, technologies to reduce NO_X emissions from aircraft engines may increase CO_2 emissions, and new technologies such as carbon capture and storage and alternative energy sources such as biofuels (Box 9.1) may provide new sources of precursor emissions. Improved policy coordination between air quality and climate policy frameworks would, however, reduce the potential for conflicting policy outcomes to occur without prior knowledge and would allow appropriate action to be taken in advance.

Greater harmonisation between the international climate and O_3 regulatory frameworks; including the UNFCCC (and Kyoto Protocol), the CLRTAP, and the EU, would have significant benefits for both climate change and air quality. The CLRTAP and UNFCCC could for example work more closely together to define coordinated long term goals (to 2050) for the reduction of emissions, and greenhouse gases (including O_3). Similarly, the European Commission in its work to revise the NEC Directive, and the development of climate change adaptation and mitigation strategies should ensure that the scientific and policy interpretation of scientific data and modelling results are harmonised to support and reinforce the work of the CLRTAP.

Box 9.1 Case study: the influence of biofuels on ozone production

Demand for biofuels is rapidly increasing as a result of instability in petroleum producing countries, the rising cost of oil, and the adoption of the Kyoto Protocol (CBD 2007). Between 2000 and 2005 bioethanol and biodiesel production increased more than three-fold. Over the last few years many governments have supported the development of biofuels to reduce emissions and improve energy security (eg the EU Biofuels Directive sets a 2010 target of 5.75%, and 10% increase in use by 2020). If produced in a sustainable manner biofuels may lead to reduced dependence on fossil fuels and climate change, and environmental and social benefits. However, biofuels may also have negative social and environmental impacts, as a result for example, of increasing competition for land to grow food crops, the need for enhanced artificial fertiliser, water and pesticide inputs, and biodiversity losses arising from the conversion of natural habitats to crop plantations.

Depending on the type of biofuel and the lifecycle of the fuel, biofuels could have substantial effects on air quality in the future. Deforestation and biomass burning occurs often as a result of clearance of land to plant new crops, particularly in the tropics, and is a major source of O_3 precursor emissions as it is not yet well regulated. Some of the preferred biofuel tree species (willow, eucalyptus, poplar and palm oil) are also high isoprene emitters (Arneth *et al*. 2007) and if planted in high NO_X environments could lead to increased regional O_3 production, particularly during heat wave events. Conversion of tropical forests to oil palm plantations, for example, could lead to substantial increases in isoprene emissions as a result of changes in isoprene flux potential from 480 g km^{-2} h^{-1} to 9410 g km^{-2} h^{-1} (Owen *et al*. unpublished data). Ozone precursors (CH_4, VOC, NO_X and $PM_{2.5}$) are also emitted along the biofuel supply chain and are affected by practices and processes including fertiliser use, agronomy, harvesting, conversion and distribution (Royal Society 2008; Crutzen *et al.* 2007).

The recent Royal Society report on sustainable biofuels (Royal Society 2008) recommends that sustainability criteria be applied to different biofuel options (and all land-uses) to ensure that the economic, social, and environmental costs and benefits are accounted for. Such criteria should be based on life cycle assessments and take into account the impacts associated with their production and consumption, including impacts to local and global air quality, as well as energy efficiency, climate change, environment, trade and development policy objectives. Such risk assessment frameworks are required internationally, and should be applied to each feed stock on a case by case basis.

9.5 Capacity building

Success in addressing the O_3 problem will be constrained unless there is a rapid increase in the capacity of countries to assess and manage O_3, particularly in those regions where precursor emissions are projected to increase over forthcoming decades. Regional frameworks for cooperation and information sharing exist in parts of Asia, Southern Africa, and Latin America, however none are focused specifically on reducing O_3 pollution. Priorities for capacity building in these regions include:

- Development of approaches for preparing national air pollutant emission inventories using a simplified and user-friendly framework that is suitable for use in different developing and rapidly industrialising countries and which is compatible with other major international emissions inventory initiatives. This approach has been developed in South Asia, and there is growing interest in applying it in other regions and linking it to regional O_3 models in South and South-East Asia;
- Use of passive monitoring of O_3 concentrations in rural areas to complement the more detailed monitoring which tends to be focussed in major urban centres;
- Development of effective and well-parameterised regional photochemical models. While extensive collaboration on model development has occurred in East Asia, there are few regional models available for application in South and South-East Asia, Africa and Latin America;
- Development of capacity for preliminary health impact assessments and the use of simple experimental studies to assess the impacts of O_3 on vegetation through bioindicator systems;
- Development of monitoring programmes to identify vulnerable ecosystems including natural vegetation and crops;
- Identification of best available technologies and prevention strategies for newly built facilities and identification of technologies and strategies that can cost effectively address air pollution and green house gases;
- Development of technical and legal support structures for the development and preparation of new protocols and agreements;
- Strengthened partnership programmes for scientific, political and economic research, and policy development.

9.6 Conclusions

Ozone policies have previously been targeted at reducing the magnitude and frequency of high O_3 episodes and the human health and environmental impacts associated with these. Emission controls are the primary policy mechanism, and have been successful at a regional scale in the USA and Europe at reducing peak O_3 concentrations. However, background O_3 has continued to rise in the mid-latitudes. Until O_3 is treated as a global pollutant and policy frameworks put in place to address the transboundary nature of O_3, national and even regional level controls are unlikely to achieve their policy objectives.

The modelling results suggest that it is possible to reduce or keep constant seasonal mean O_3 concentrations by 2050 globally. This future is however critically dependent on the actions taken by national governments to implement and enforce emissions controls over forthcoming decades. Rapid growth in emissions from unregulated sectors could undermine these efforts and all significant sources of anthropogenic emissions need to be integrated into abatement strategies. International shipping and aviation are currently poorly regulated in terms of O_3 precursors and both sectors are projected to become more important sources of emissions in the future unless regulation is strengthened and additional technology improvements are implemented.

Ozone concentrations are projected to rise between 2000 and 2050 in Latin America, Africa and Asia, due to increasing emissions and here stronger NO_X, VOC and CH_4 emissions controls will be needed to reduce emissions from mobile and stationary sources. Biomass burning is poorly regulated but is a more significant source of emissions than the energy sectors. Further controls will be required to reduce emissions from sources such as deforestation, savannah burning and agriculture. The implementation of low-cost CH_4 reduction technologies in the agriculture and energy sectors in these regions should also be a priority.

In urban locations, trends in background O_3, climate change, and reductions in the NO_X titration effect will contribute to increased urban O_3 concentrations. This combination of sources must be reflected in air quality strategies aimed at improving air quality in urban locations.

The limited evidence available suggests that under a current legislation scenario, O_3 concentrations in 2030 will not have a significant impact on global food security. There may however be significantly increased impacts on food security in vulnerable regions such as South Asia. Timescale is important; the negative impacts of O_3 on food crops must be minimised before climate change impacts significantly reduce crop yields. The integration of O_3 tolerance into national crop breeding and selection programmes, and into biotechnology research programmes should be a priority.

Climate change and O_3 are closely coupled due to the indirect and direct radiative forcing effects of O_3 on the climate system, and the role of climate in O_3 production, destruction and transport. However, O_3 is not yet considered to be a climate issue and so is not considered within the appropriate international regulatory frameworks. As a failure to implement current legislation will lead to a high O_3 future, with both direct and indirect impacts on the climate system, the integration of O_3 into this regulatory framework should be considered.

Conversely, climate change in 2050 is projected to reduce the benefits of O_3 precursor emissions controls and will need to be factored into future O_3 policies.

Ozone can therefore no longer be considered to be just an air quality issue. As O_3 and climate change share many of the same drivers and sources, the development of coordinated emission reduction strategies could make an important contribution to reducing both climate change and O_3 pollution.

Unless there is a rapid increase in the capacity of many countries outside Europe, North America and Japan to assess and manage O_3, the success of current efforts to control O_3 will be limited. Greater international collaboration, building on the capacity provided by existing regional frameworks is required.

10 Research and development

The last few years have seen improvements in scientific understanding of tropospheric O_3 production and destruction processes and impacts to human health and the environment. Advanced modelling techniques have been developed that have enabled simulation of the combined effects of climate change and emission changes on future O_3 concentrations. However there are still knowledge gaps which limit understanding of how changes in anthropogenic and natural emissions, climate and land-use will affect future O_3 concentrations and the impact that such changes will have on human health and the environment. This chapter identifies research priorities based on the needs identified within the previous chapters.

10.1 Ozone precursor emissions

Assessments of future projections of ground-level O_3 concentrations are currently limited by the availability of emission inventory data, uncertainties in assumptions in drivers of emissions and understanding of the biogeochemistry and physiology of natural emissions.

Identification of the direct and indirect drivers of changes in emissions and accurately forecasting changes into the future is not straightforward. Improved understanding of how different socioeconomic pathways, or growth in particular sectors (eg shipping and aviation) could affect O_3 precursor emissions is needed to inform future air quality projections. For example the implications of a doubling in China's energy demand for regional and global O_3 concentrations is identified in this study as an area requiring further investigation, as is the impact that the rapid growth in shipping emissions will have on future projections. Improved understanding of the lag between implementation of legislation or new technologies, and delivery of air quality benefits is needed to improve impact assessment for environment and human health and the development of air quality policy.

The main anthropogenic sources of precursors are well understood, however information is better for some gases than others, for example there is poor knowledge regarding the magnitude of global anthropogenic VOC emissions, and emission inventories are poorer than those for anthropogenic NO_X emissions. Where inventories are present, data collection may not be standardised. In developing countries the quality of emission inventories is generally insufficient to support the modelling of dispersion, transport and deposition, and effects assessment. In some developed countries emission inventories remain poor for some sectors (eg international shipping) and gases (eg nmVOC). Biomass burning lacks long-term monitoring in many regions, and there are major uncertainties in emission estimates due to inadequate knowledge of the relative contribution of open biomass burning to total emissions.

Natural emissions are much less well understood than anthropogenic emissions. Although the main sources (soils, plants, lightning) are known, the biogeochemistry is not well understood. There is limited understanding of the interactions between environmental factors such as temperature, moisture, land-use, ambient CO_2 and O_3 and effects on natural precursor emissions. There is no globally systematic analysis or screening of emissions and consequently there are large uncertainties in emission estimates. The ability to measure and estimate biogenic VOC emissions for example is currently limited by an inadequate network of observational sites globally, by poor understanding of the multitude of biogenic sources of VOC and the speciation of nmVOC for different plant functional types (only a small number of plant species emissions have been measured in major source regions such as tropical forests). The effects of biotic and abiotic stressors such as herbivory, wind damage, fire, logging, and nutrient status on VOC emission rates are not well understood. This is despite the recent development of new analytical techniques, flux-based measurement techniques, and the use of satellite data, which have greatly enhanced the observational base on which emission estimates are made.

Because of these uncertainties it is not possible to quantify how natural emissions may change in the future in response to changes in climate, land use change, or other environmental factors. The effects of major shifts in land use on local and regional air quality as a result of either replacement of plant species with high nmVOC (particularly isoprene) emitting species such as oil palm (for example) or changes in soil NO_X or CH_4 emissions requires investigation. Current understanding of which crop species are high nmVOC emitters is limited to a few species, and is particularly poor for the crops likely to be important biofuels in the future. Consequently most recent emissions estimates and modelling work do not include potential changes in natural precursor emissions.

There are substantial uncertainties associated with estimating future trends in CH_4. The total emissions are relatively well known, however the relative contributions of individual sources to the overall emission flux is uncertain and the cause of large interannual fluctuations in global CH_4 concentration over the last 15 years is unknown.

What needs to be done:

- Analysis of the implications of major deviations in energy demand from the new B2+CLE scenario in India and China for national, regional and global O_3 concentrations;
- Detailed analysis of the impacts of rapid growth in shipping emissions on future global and regional O_3 projections;
- Detailed biogeochemical analysis of the potential impacts of changes in land use on O_3 production and destruction processes. Modelling analysis to assess the impacts of these changes on local, regional and global O_3 concentrations. Case studies might include the conversion of forest to biofuels, or the effects of changes in agricultural methods such as cropping practices or fertiliser use;

- Measurements of VOC and NO emissions from managed and natural ecosystems are needed to enable the preparation of improved global emission inventories;
- Improved global inventories for natural and anthropogenic O_3 precursors are needed. Standardised data collection and reporting processes of sufficient temporal and spatial resolution are required for use in atmospheric chemistry models. Special emphasis should be placed on improving monitoring and measurement of emissions from biomass burning from domestic sources, deforestation, and agriculture, to anthropogenic and natural VOC emissions, and to international shipping emissions;
- Further research into the biogeochemistry of natural emissions, the influence of biotic and abiotic factors, and the combined effects of multiple drivers such as changes in atmospheric CO_2 concentrations, climate and land use, on emission rates is required. Efforts are needed to improve understanding of the relative contributions of natural emissions to O_3 formation;
- The IPCC in collaboration with the CLRTAP to undertake a special study on the biogeochemical interactions between climate change, ground-level O_3 and other pollutants;
- Development of global VOC emission projections for the period up to 2050;
- Establishment of a dedicated research programme aimed at identifying the causes of the changes in global CH_4 emissions over the last 15 years, and the development of an improved global emission inventory;
- Greater investment in increased capacity for observing, monitoring and reporting, modelling and assessment of O_3 impacts to human health and the environment in developing countries;
- Specific monitoring of the impacts of implementation of O_3 pollution abatement measures on emissions;
- Increased investment in the development of low cost technologies for reducing emissions for rapid implementation in regions where emissions are projected to rapidly increase, and in sectors projected to be important contributors to emissions in the future such as international aviation and shipping.

10.2 Modelling

Uncertainties in projections of future changes in O_3 are due to uncertainties inherent in the emission scenarios used to drive the model runs, and many poorly-constrained processes within the models themselves.

Improved understanding is needed of the basic processes and feedbacks between atmospheric chemistry, climate and ecosystems. Some of these interactions are complex but poorly understood at present, particularly those in the terrestrial biosphere. In the marine environment understanding of halogen-catalysed chemistry is rapidly developing, however the global and regional implications for O_3 remain unclear and few of the global models yet include this chemistry.

Many of the meteorological and ecological processes which affect O_3 production, destruction and transport are not yet well represented in coupled chemistry–climate models. Detailed stratosphere–troposphere exchange processes, deep convection and mixing, lightning and atmospheric blocking are, for example, rudimentary. Some of these processes may not have a significant impact on the long-term average concentration of O_3, but are important for O_3 at the regional scale or over shorter time scales. Similarly, interactions between climate and surface O_3 fluxes, sensitivities of future O_3 to land cover change, and impacts of O_3 and climate on the carbon cycle are either not represented or are too simplistic. The systematic biases in rainfall patterns simulated by current models compromise O_3 projections and introduce uncertainties in background O_3 levels. Improved representation of the water cycle, including soil water balance and impacts on dry deposition is necessary for improving regional climate projections and would help to improve predictions of changes in the frequency and magnitude of heat waves and high pollution episodes. Fully interactive global modelling studies which couple these climate, atmospheric chemistry and ecological processes in marine and terrestrial systems are needed to represent these basic processes and feedbacks.

What needs to be done:

- Further multi-model studies of interactions between climate change and ground-level O_3, targeted at informing a range of climate projections are required;
- Research exploring the basic processes and feedbacks between atmospheric chemistry, climate and ecosystems, particularly surface deposition processes, is needed, and the integration of this information into global climate–chemistry models is required to improve projections of O_3 at the global, regional and urban scales;
- As computing capacity develops, emphasis is needed on the development of fully interactive global modelling studies which couple climate, vegetation and atmospheric chemistry and which enable assessments of O_3–climate interactions at higher spatial resolution than is currently possible;
- Increased consideration in global chemistry modelling studies of the interactions between large scale and small scale processes; including the dispersal and early chemistry of plumes as a result of shipping and aviation emissions, and the impact of these processes on background O_3 concentrations;
- Further assessment of the impacts of halogen-catalysed chemistry on O_3 concentrations, and the integration of this chemistry into global climate–chemistry models is needed to inform the evaluation of potential impacts on regional and global O_3 concentrations;
- Further assessments are needed of the effect that climate change may have on surface exchange processes for O_3 and isoprene in the UK and Europe, and other regions as appropriate, by 2050.

10.3 Evaluating human health impacts

Assessment of the population effects of O_3 are limited by a lack of accurate baseline health data in many countries, the use of mean annual exposure which does not capture important individual exposure events, and the effect of multiple factors on the interpretation of epidemiological study results. Evaluation of the health implications of long-term exposure to ambient O_3 concentrations is complicated by uncertainty regarding whether there is a threshold for effects.

The acute impacts of O_3 are substantial and reasonably well understood, although there is variability in individual susceptibility to O_3 and variability between regions. The mechanism by which mortality is affected is unclear and is complicated by correlations between mortality, O_3 and other pollutants.

Chronic disease and loss of life-years are potentially more important for the evaluation of human health impacts than daily mortality or hospital admissions. Animal studies suggest that seasonal exposures to O_3 have cumulative effects however the implications of these results for humans are unclear. Evidence of chronic effects on reduced lung function and asthma symptoms are inconsistent, and where effects are observed, the implications for life expectancy are unknown. The information available is insufficient for reliable health impact assessments at the moment.

Ozone may have additive effects on health in combination with other environmental factors such as allergens or other pollutants such as particulates, although the importance of these interactions for human health is uncertain.

Estimates of future O_3 health impacts require the estimation of future baselines for mortality and morbidity, projections for future O_3 exposure levels, knowledge of the effects of O_3 on human health, and the use of an appropriate dose–response relationship: information which is not available for many locations.

What needs to be done:

- Improved monitoring of O_3 particularly in developing countries;
- Further research is required to improve understanding of the pathophysiological mechanisms by which O_3 affects human health;
- Further research is needed to increase understanding of the chronic effects of O_3. This should include research to quantify the effect of O_3 on human health in the O_3 concentration range 1–50 ppb, and further studies on impacts to lung function, respiratory disorders, and life expectancy;
- Improved experimental and epidemiological investigations of the exposure response function, including thresholds, is needed;
- Further research into the combined effects of O_3 and other environmental stressors, particularly temperature, on human health is required. This should include an evaluation of the impacts of climate change and O_3 on human health to 2050 in the UK, across Europe and when information permits, in Asia, Latin America and Africa.

10.4 Evaluating environmental impacts

The majority of studies undertaken to evaluate the impact of O_3 on the environment have focused on plant taxa of North America and Europe. This research has been based mainly on chamber experiments and has led to improvements in understanding of the mechanisms of direct impact in plants. More work is needed to broaden the range of species and to extend these studies to other regions. There is also a need to focus on natural vegetation, for which less information is available than for crop species.

The assessment of effects of O_3 on vegetation relies on a combination of concentration and flux-based approaches. Process models using the flux-based method allow a more accurate assessment of the local impacts of O_3 on vegetation and the regional and global effects of O_3 on carbon sequestration. However these models are at an early stage and require further development and validation across a wider range of crop and wild species before they can be used for risk assessment exercises. They also need to incorporate a wider range of effects above- and below-ground to better assess the indirect effects of O_3 on plant productivity, and to integrate O_3 impacts on the terrestrial carbon sink into assessments of climate change.

Understanding the impacts of O_3 on plant photosynthesis and carbon sequestration (in vegetation and soils) is rapidly evolving. More long-term field-based experiments are required to reduce the uncertainties associated with the combined effects of CO_2 fertilisation and O_3 impacts on plant productivity, and the effect of O_3 on the net exchange of carbon between ecosystems and the atmosphere. These studies are needed for a broader range of plant functional types from non-temperate regions and to improve estimates of the indirect radiative forcing of O_3. The results of this work should be linked to climate stabilisation research.

Work on the impacts of O_3 is critically needed in regions outside of North America and Europe. Those regions identified in this study as likely to experience increases in O_3 concentrations over the next two to three decades (eg Asia, Latin America and Africa) should be the priority. There is significant uncertainty about the effect that O_3 may have on future food production, due to a lack of experimental work on effects of O_3 in these regions, and limited capacity for monitoring and modelling of O_3 concentrations and impacts in key agricultural regions. This work should focus on testing the sensitivity of regional varieties of crops to O_3, and on evaluating the implications of these impacts for food in terms of crop yield and nutritional quality, energy security, climate change, and biodiversity. Assessments of regional vulnerability to food shortages should consider the role that O_3 impacts on crop yields and nutritional quality may play in exacerbating food insecurity.

Research is needed into mitigating the effects of O_3 on the yield of food crops, in the context of increasing competition for land, for example by identifying or selecting varieties that are less sensitive to O_3.

This report identifies the potential for significant impacts of O_3 on biodiversity, as indicated by potential impacts on GPP, in high priority regions for global conservation including eastern North America, Central Europe, northern South America, Central Africa, and South-East Asia. Future assessments are needed of the ecological impacts of O_3 in the context of other threats such as habitat destruction, N deposition and climate change. Ozone impact assessments have been focused on evaluating effects on plants. However, studies suggest that other taxa may also be affected either directly or indirectly. Understanding of the ecological impacts of O_3 is very poor, with very few studies undertaken on this issue. As with assessments of O_3 impacts on crops, most ecological studies have been conducted in Europe and North America and there is an urgent need to evaluate the impacts of O_3 on wild species in other regions.

Attempts have been made to evaluate the economic impacts of environmental O_3 effects. However, these are necessarily limited by the issues raised above. As understanding of the impacts of O_3 improves, these economic assessments will require updating.

What needs to be done:

- Experimental and field studies of the impacts of O_3 on local species should be established in key regions experiencing current O_3 problems and where O_3 levels are predicted to increase significantly over the next 20–30 years, such as South and East Asia. *In situ* studies are needed to support the development of health and environment impact assessments in these regions, and to identify effective mitigation and adaptation measures to reduce these impacts. Large scale investment in standardised facilities and experimental protocols across these regions would facilitate assessments of regional impacts of O_3 as a basis for policy interventions. International collaboration is needed urgently if these regions are to be able to respond effectively to the O_3 concentration increases projected for the next two to three decades;
- Specific research is needed by the FAO on the implications of ground-level O_3 for regional food security in Asia, Latin America and Africa. This should include a focus on identification and selection of ozone resistant local varieties, and assessments of impact on crop yield and nutritional quality. Equivalent research is needed to investigate the impacts of O_3 on non-food crops;
- Further targeted experimental studies are needed to specifically evaluate the impacts of rising background concentrations of O_3 and to compare these with the effects of episodic peak exposures;
- Long-term field studies are needed to assess the combined effects of O_3, elevated CO_2 levels, climate change and other environmental stressors both on species of economic importance and on a range of natural and semi-natural ecosystems to improve the evidence base of effects on O_3, plant productivity, the ability of ecosystems to sequester carbon, and biodiversity;
- Further development and use of process-models is needed to improve assessments of the impacts of O_3 on crop and forest production, the ability of ecosystems to sequester carbon, and biodiversity;
- Research focused on improving understanding of the impacts of O_3, CO_2 and climate change for the carbon cycle is urgently needed to inform climate change and ground-level O_3 projections;
- New research is needed into the effects of O_3 on important species for conservation, including research on the combined effects of O_3 and other environmental stressors. This research should focus on ecoregions and biodiversity hotspots where future O_3 concentrations represent the greatest threat. The results of this work should be provided to the Convention on Biological Diversity (CBD) for their consideration as a new and emerging issue of relevance to biodiversity.

10.5 Other research requirements

This report recommends a new approach to O_3 policy and identifies the need for better coordination of air quality policy at the international level. Improved understanding of the potential synergies and trade-offs between O_3 reduction policies and other policies, particularly climate change, is required.

What needs to be done:

- Options for an appropriate international mechanism aimed at providing a globally coordinated approach to air pollution issues, and O_3 specifically, should be identified and evaluated and should build on the ongoing activities of the UNECE Task Force on Hemispheric Transport of Air Pollutants (TFHTAP), and the Global Atmosphere Pollution Forum. This should be initiated at the UN level;
- Improved understanding of the potential synergies and trade-offs between O_3 reduction policies and other policies, particularly climate change, is required. The UNFCCC secretariat together with the executive body of the CLRTAP should assess the potential interactions between climate change and air quality policies, and should evaluate the interlinkages between current and future policy and technological controls on greenhouse gases and other air pollutants over the period 2010–2050 and to 2100;
- Further development of integrated policy tools for the assessment of impacts from O_3 and O_3 precursors on radiative forcing, vegetation and health with respect to future sector development, specific geographical areas and specific sources. Such tools are important for supporting life cycle analysis and strategic environmental assessments;

- Assessment by the Rio Conventions' Joint Liaison Group of the relevance of ground-level O_3 and air pollution more generally, to their respective Convention mandates;
- Evaluation by the World Trade Organization in collaboration with the FAO, of the impacts of O_3 on global food prices and trade to identify vulnerable regions over the coming decades. A key policy objective in these regions should be to ensure the negative impacts of O_3 are minimised before substantial climate change effects occur.

11 Conclusions and recommendations

11.1 Ground-level ozone as a global pollutant

Ground-level O_3 remains a serious global air pollution problem despite the implementation of emission reduction measures in industrialised countries over the last 40 years. Anthropogenic emissions of NO_X and nmVOC have declined in Europe and North America over this period leading to reductions in short-term peak O_3 concentrations of typically 20–30 ppb in Europe. Over the same period measurements have shown the hemispheric background has increased by 2 ppb per decade to 35–40 ppb in the mid-latitudes of the Northern Hemisphere. The cause of the increase in background O_3 is not fully understood, but is thought to be due mainly to precursor emission increases in Northern Hemisphere countries and increases in emissions from poorly regulated sectors including international shipping and aviation. While less certain, an increase in the stratospheric source of O_3 may also have contributed to the increasing background. This increasing trend greatly reduces the benefits of declines in peak O_3 concentrations achieved through existing emissions controls. Once considered to be a local or regional scale issue, ground-level O_3 has emerged as a global pollutant. A new regulatory approach designed to address O_3 at the global, regional, and local scale is now needed.

Recommendation 1:

Options for an international mechanism to provide a globally coordinated approach to air pollution issues, and O_3 specifically, should be identified and evaluated. This should be led by a United Nations body such as the UNECE, or UN Environment Programme (UNEP) and should build on ongoing activities such as the EMEP, including its TFHTAP, and work of the Global Atmospheric Pollution Forum.

11.2 Ground-level ozone chemistry – relevance to ozone control

The atmospheric chemistry of ground-level O_3 shows great variability between the polluted urban areas and remote relatively clean air regions of the planet. This variability is an important consideration for the design of abatement strategies for O_3:

- In remote areas, background O_3 is the primary contributor to surface O_3 concentrations;
- At the regional scale, the hemispheric background and regional sources of O_3 precursors are the main contributors;
- At the local scale, O_3 concentrations are regulated by the hemispheric background, regional production and the effects of local sources and sinks acting collectively.

NO_X concentrations play a critical role in regulating the chemical production or destruction of O_3 within the atmosphere leading to O_3 destruction in low NO_X (<20 ppt) regions and O_3 production in more polluted regions where NO_X concentrations are higher. NO_X concentrations are typically much larger in the urban environment leading to local O_3 depletion so that reductions in NO_X concentrations lead to local increases in urban O_3 concentrations. Concern about the human health impact of NO_X emissions has led to the implementation of NO_X controls in many urban centres, leading to a pronounced increase in O_3. In the UK this is clear through the period 1990–2007. This is unlikely to change in the near future and further increases in urban O_3 can be anticipated. In large urban areas O_3 formation is VOC limited and VOC emission controls can therefore be an effective option for limiting the increases in O_3 formation that occur in such areas from NO_X emission controls. In the UK, in contrast to much of continental Europe, regional scale O_3 formation is VOC limited and VOC emission reductions could also provide a useful contribution for controlling O_3. Appropriate VOC emission control measures should therefore be included in regional and urban air quality strategies.

Recommendation 2:

2.1 The UNECE, European Commission and all national governments to target emission control measures to reduce peak and background O_3 concentrations taking into account the relative importance of different emission sources at the local, regional and global scale as appropriate.

2.2 Decision making bodies responsible for local air quality to implement VOC emission controls in large urban areas where NO_X emission controls are contributing to increasing local O_3 concentrations.

2.3 UK Government to implement regional scale VOC emission controls as part of the national strategy for controlling O_3.

11.3 Ground-level ozone in the future

Future O_3 concentrations will be mainly determined by emissions of O_3 precursors and these will be driven by population growth, economic growth, land use change, new technology developments and the implementation of emissions controls. Anthropogenic emissions are largely under human control and will be determined in the future by the implementation of emission control legislation and emission reduction technologies, which if implemented, could reduce emissions by 90%.

The scenario analysis conducted in this study (based on a new B2+CLE scenario) projects the decline of anthropogenic NO_X and CO emissions globally to 2050, and an increase in global CH_4 emissions. The global modelling projections show near constant or reduced seasonal mean O_3 concentrations in 2050

relative to 2000, in most regions during the maximum O_3 season. During summer, reductions of 5 ppb are projected for the Northern Hemisphere mid-latitudes, with reductions in concentration of up to 15 ppb (or 25% relative to present day levels) projected for parts of North America. During winter an increase in O_3 concentrations is projected due to lower NO_X emissions and reduced O_3 titration by NO_X. During the maximum O_3 season over much of the developing world, a modest increase in O_3 of up to about 3 ppb (approximately 7%) is projected due to rapid economic growth in these regions.

The extent to which the legislation considered within the scenario analysis is implemented globally will be critical. The global modelling analysis shows that it is the changes in anthropogenic emissions, mainly of NO_X, but also of CH_4, CO, and nmVOC that will be the primary influence on ground-level O_3 concentrations in 2050. Projections of future trends in CH_4 are however very uncertain. If CH_4 remains at present day levels (1760 ppb) rather than rising to 2363 ppb in 2050 as projected by the B2 scenario, the new modelling indicates that global average surface O_3 levels will be 1–2 ppb lower. However, increases are expected as CH_4 emission controls are not yet in place in many developing countries and emissions from agriculture, particularly in Asia, Latin America and Africa are expected to rise. Increasing demand for fossil fuels in many regions will also contribute to higher emissions.

In contrast to most other regions, NO_X emissions from Asia are projected to rise over the next few decades due to increases in power generation and traffic volumes, although emissions from transport should stabilise by 2050 with the implementation of current legislation. Emissions are also expected to increase in Africa as there are currently no NO_X controls in place other than for new large combustion plants. In Latin America, Africa and Asia, emissions from biomass burning (deforestation, savannah burning, agricultural waste) are double those from energy sources and are a significant contributor to CO and NO_X emissions. As these sources are poorly regulated, emissions are projected to remain high, or to decline only slightly in these regions.

Modelling analysis of future changes in O_3 concentrations at the urban scale suggests that the exposure of urban populations to O_3 will increase in many cities by 2050 where reductions in local NO_X concentrations occur. As NO_X emission controls are implemented and emissions decline (as recently seen in the UK for example), urban O_3 concentrations will rise towards regional background concentrations. If background O_3 increases this will further exacerbate O_3 increases at a local level (refer recommendation 2.2).

Natural O_3 precursor emissions also contribute significantly to O_3 concentrations, however it is not yet possible to quantify with any certainty how they will change in the future in response to changes in land use and climate. Emissions of CH_4 and NO_X from the biosphere are likely to increase. Biogenic emissions of nmVOC and isoprene, in particular, may also increase but interactions with atmospheric CO_2 may substantially reduce the magnitude of the change.

Recommendation 3:

All national governments to implement current O_3 precursor emission reduction legislation as a matter of priority. Further measures to reduce precursor emissions are required, in particular:

3.1 *NO_X controls need to be strengthened for mobile sources in countries outside the EU and Japan. NO_X controls for large stationary sources should be strengthened worldwide;*

3.2 *Action is needed to reduce NO_X and CO emissions from biomass burning, particularly in Latin America, Africa and Asia where emissions are currently greatest;*

3.3 *International support should be provided by the (IEA) and UN Development Programme (UNDP) for the implementation of low-cost CH_4 emission reduction technologies in the agriculture and energy sectors in the Asian, Latin American and African regions.*

11.4 The effects of climate change on future ozone concentrations

The modelling undertaken for this study shows that for the new B2+CLE scenario, emission changes are generally more important than climate change for determining O_3 concentrations in 2050. Climate change will however also have important impacts on surface O_3, especially at the regional and local scale. The climate change effects will be regionally variable with impacts during the maximum O_3 season in the range of ±5 ppb depending on the underlying NO_X regime: O_3 production will increase in already polluted environments, and decrease in clean environments. For example in developing world regions such as large parts of Africa and Asia where emissions are projected to increase, air quality in 2050 will decline, in part due to climate change. In other regions with stricter controls in place, air quality will improve, although climate change will reduce the benefits of control measures. In the UK and NW Europe, sensitivity analysis suggests that episodic peak and long-term mean O_3 concentrations may be affected by changes in the climatic parameters important for dry deposition and O_3 production from isoprene oxidation. The effects on dry deposition in particular may be large.

Global atmospheric chemistry models are limited in their ability to simulate all of the important climate sensitive processes and climate–ozone interactions that determine surface O_3 at sufficiently high spatial and temporal scales. Important factors that influence O_3 and that may change on decadal timescales include the NAO and ENSO; production of nmVOC, particularly isoprene, from vegetation; production of NO_x from lightning and soils, release of CH_4 from wetlands and permafrost; and O_3 dry deposition rate. These processes have the potential to substantially increase episodic peak and long-term mean O_3 concentrations under climate change. For example, dry deposition is one of the dominant processes that controls surface O_3 concentrations and may be inhibited during heat waves. This was demonstrated during the 2003 European heat wave. The frequency of high O_3 episodes is expected to

increase in many areas as reduced soil water availability and hot summers become more common later this century.

Over the UK and Europe, the frequency of summer droughts, heat-wave events and associated high O_3 episodes is anticipated to increase. Across Europe by 2100 it is projected that hot summers such as that experienced in 2003, will become an average yearly occurrence and that the incidence of extreme hot weather of the type experienced in the August 2003 heatwave will increase and may occur across Europe every 1 in 10 summers by 2100. In other parts of the world, weather patterns that limit the intensity of summertime or dry season peak pollution levels are expected to be less prevalent leading to higher pollution levels.

In addition to the impacts of climate change on ground-level O_3, the impacts of O_3 on climate change may also be important in the future. O_3 is an important greenhouse gas, ranked third behind CO_2 and CH_4 in terms of direct radiative forcing of climate between 1750 and 2005. Future increases in O_3 may lead to significant indirect forcing of the climate by reducing the global land carbon sink and amplifying the rate of increase of atmospheric CO_2.

Better integration between the O_3 (UNECE CLRTAP) and climate change (UNFCCC) international regulatory frameworks is necessary. Improved coordination between ground-level O_3 and climate change policy and research initiatives is likely to have substantial co-benefits for both ground-level O_3 and climate change policy objectives. As they share many of the same drivers and sources of emissions, there is clear advantage in developing coordinated emission reduction strategies. Reducing CH_4 emissions, or emissions from deforestation, are obvious candidates. Climate change mitigation and adaptation strategies should be evaluated for their ground-level O_3 impacts, and vice versa.

Recommendation 4:

Improved coordination of O_3 and climate change policy and research frameworks is required and should include:

4.1 The UNECE, EU, and national governments to review the potential impacts of climate change on the achievement of medium (eg 2020) and long-term (eg 2050 and beyond) O_3 policy objectives and revise air quality management strategies appropriately;

4.2 An investigation into the policy interactions to be led by the UNFCCC secretariat together with the Executive Body of the CLRTAP. This should evaluate the mechanisms by which the UNFCCC and CLRTAP (and other regional air pollution bodies) objectives and targets could be better harmonised, and should include a review of the linkages between current and future policy and technological controls on greenhouse gases and other air pollutants over the period 2010–2050 and to 2100;

4.3 A review by the European Commission of how EU policy (such as the NEC Directive, and climate change adaptation and mitigation policies) can be better integrated with the objectives of the CLRTAP;

4.4 A study into the biogeochemical interactions between climate change, tropospheric O_3 and other pollutants to be undertaken by the IPCC and CLRTAP.

11.5 Impacts of ozone on human health

Current O_3 exposure levels in North America and Europe are associated with short-term acute effects on the respiratory system resulting in increased daily mortality and morbidity in already vulnerable individuals. There is also evidence that long-term chronic exposure has adverse effects on lung function but the significance of these effects for long-term health is not yet known.

Health impacts have been observed at around 35 ppb and below the current WHO guideline of 50 ppb (daily 8 h average concentration). The total health impact of O_3 is now understood to be driven more by the days at which O_3 is at baseline concentrations than by the occasional days on which episodes occur. This has led to recognition that efforts to reduce the impacts of ozone to human health and the environment must be focused on background as well as peak ozone.

Quantification of the effects of O_3 on human health now and in the future depends heavily on the question of whether there is a threshold for effects in humans which is currently unclear. If no threshold exists, the health impacts of O_3 will depend largely on trends in hemispheric baseline concentrations. If a threshold exists, the health impact will depend more on regional and local emissions of precursors together with meteorological conditions.

In regions of Africa, Latin America and Asia, where emissions of O_3 precursors are projected to increase over the next few decades, human exposure will increase. In regions where emissions are projected to decrease, human exposure is expected to increase in urban locations due to reductions in NO_X concentrations and reduced local depletion of O_3. Like many urban areas, O_3 concentrations in London are expected to increase as NO_X emission controls take effect leading to increased human exposure and substantial increases in daily mortality and respiratory hospital admissions in 2100. An increase in background O_3 will exacerbate this increase while a decline in background O_3 will partially compensate for the NO_X titration effect and could lead to a decrease in human exposure.

By 2100 the effects of climate change on the frequency and magnitude of hot summers and heat waves such as experienced in the Northern Hemisphere in 2003 are expected to lead to an increase in mortality and morbidity.

Recommendation 5:

Ozone regulatory frameworks to be strengthened to protect human health from background O_3 exposure:

5.1 The WHO, US EPA, UNECE and European Commission to examine the importance of background O_3 effects on human health, and to strengthen guidelines and standards appropriately;

5.2 The WHO and other appropriate regional organisations to ensure countries that would like to develop strategies to achieve reductions in the human health impacts of O_3 receive sufficient financial and information support to do so;

5.3 That existing systems for alerting the public when O_3 concentrations exceed certain levels be strengthened.

11.6 The environmental impacts of ozone

Among the most important impacts of ground-level O_3 on the environment are the direct impacts on terrestrial vegetation. O_3 effects leaf structure and physiology and causes visible leaf damage and reductions in growth and yield in sensitive plant species. These may lead to long-term effects on ecosystem structure and function and impacts on the carbon cycle. Impact assessments are strongly dependent on the choice of exposure index which can dramatically alter model predictions of O_3 impacts. The flux-based approach is the most appropriate method for risk assessment as it provides a stronger mechanistic basis for assessing O_3 impacts. In Europe the application of this method reveals a more even distribution of impact across the region than other methods which is consistent with observations of visible injury.

Crops and wild plant species are vulnerable to O_3 damage. In the USA in the 1980s the annual cost of loss of arable crop production due to O_3 was estimated to be $2–4 billion. In the EU in 2000 €6.7 billion were lost due to impacts to arable crops. For the same year global yield losses were estimated to be $14–26 billion for the four staple crops; rice, soybean, maize and wheat. Ozone impacts on the yield of staple crops will increase over the next few decades in some rapidly developing regions. Significantly increased impacts of O_3 are projected in South Asia where there is already evidence of substantial impacts on yield from ambient O_3 concentrations which exceed those predicted from studies in North America or Europe. O_3 could present a significant threat to national food security in South Asia, particularly over the next two to three decades when emissions are projected to increase rapidly. This may apply to other developing regions, although there is less evidence available than for South Asia. A key question is whether there are regions where both climate change and O_3 may have adverse effects on the yield of staple crops and therefore food security.

There is evidence that wild plant species in Europe and North America show a similar range of sensitivity to O_3 as crop species. However, there is very little evidence on the long-term effects of O_3 on biodiversity or ecosystem services. Ozone is projected to have significant impacts on global plant productivity and potentially ecosystem function and biodiversity under a high emission (A2) scenario. Major eco-regions in Latin America, Asia and Africa, as well as in parts of Europe and North America are projected to be most vulnerable. The long-term effects of O_3 on biodiversity are particularly uncertain at lower latitudes where there has been almost no research.

Future impact assessments will need to consider the effects of O_3 on the terrestrial carbon cycle alongside those of increasing CO_2 concentrations. In addition to the direct radiative forcing of ground-level O_3, reductions in the land carbon sink due to O_3 damage to vegetation will increase the importance of O_3 as a driver of climate change.

Recommendation 6:

6.1 Application of the flux-based O_3 exposure metric to a wide range of staple crops, food species and wild species of conservation importance, and use of this information to assist assessments of current and future O_3 impacts in Europe and other regions with high O_3 exposures. To be led by the UNECE International Cooperative Programmes (ICP) for vegetation and forests and the Global Atmosphere Pollution Forum;

6.2 That the FAO initiate an assessment of the impacts of O_3 on food and non-food crops and the implications for global and national food security, and identify and disseminate policies to reduce these impacts. Regions vulnerable to food shortages, and identified in this study as vulnerable to increasing O_3 concentrations, should be the priority for assessment. The potential combined impacts of climate change and O_3 on economically important crops should also be considered;

6.3 That the CBD secretariat undertake an assessment of the impacts of O_3 and other air pollutants on biodiversity and natural ecosystems; this should include an assessment of data requirements for an assessment of impacts to wild species of high conservation value in the major eco-regions in Latin America, Asia, Africa, and vulnerable regions of Europe and North America.

11.7 Improving ozone policy

Ozone policies have previously been targeted at reducing the magnitude and frequency of high O_3 episodes, and the human health and environmental impacts associated with these. Emission controls are the primary policy mechanism, and have been successful at a regional scale in the USA and Europe at reducing peak O_3 concentrations. As understanding has increased of the impacts from long-term low level exposure to O_3, the focus has had to shift to also considering ways to control background O_3 which presents a greater challenge due to the international nature of the problem.

Ozone is not yet treated as a global pollutant, however until policy frameworks are put in place to address the transboundary nature of O_3, national and even regional level controls are unlikely to achieve their policy objectives and the increasing trend in background O_3 is likely to continue. All significant sources of anthropogenic emissions need to be integrated into abatement strategies. International shipping and aviation are currently poorly regulated in terms of O_3. Both sectors are projected to undergo rapid increases in growth in the future and to become more important contributors to O_3 pollution unless regulation is strengthened and additional technology improvements implemented.

Ozone can no longer be considered to be just an air quality issue as the human health, environmental, economic and climate effects are now well established. Improved integration of O_3 policy with policies aimed at reducing other air pollutants and mainstreaming of air quality policy into other policy areas such as climate, food, and energy policy is needed. The indirect and direct impacts of O_3 on the climate system, and the role of climate in O_3 production, destruction and transport processes, strongly support the need for better links between the UNFCCC, CLRTAP, and European Community climate change and O_3 regulatory frameworks. Prevention of deforestation and regulation of biomass burning would make an important contribution to reducing both climate change and O_3 pollution. Reductions in CH_4, CO and nmVOC would deliver both air quality and climate change policy objectives. Where co-benefits are not possible, improved policy integration enables the early management of risk. Climate change mitigation policies are driving demand for development and implementation of biofuels which may, in some locations, contribute to poor air quality. Integrated assessment models are useful tools for improving policy integration.

Unless there is a rapid increase in the capacity of many countries outside Europe, North America and Japan to assess and manage O_3, the success of current efforts to control O_3 will be limited. Greater international collaboration focused on reducing ground-level O_3 is required.

Recommendation 7:

7.1 All significant sources of anthropogenic emissions must be added to the current mix of abatement measures. The IMO and ICAO must agree to regulate NO_X, CO and VOC emissions from international shipping and international aviation respectively as far as technically feasible, to implement agreed emissions controls, and to continue research into the development of emission reduction technologies for these sectors;

7.2 Formal links between the IMO, UNECE and UNFCCC should be established to ensure that the emission reduction policies for shipping and land-based transport emissions are compatible, so that climate change and air quality impacts are considered together. A body similar to the Joint Liaison Group of the Rio Conventions should be convened with the objective of reviewing the state of play and identifying priorities for future work;

7.3 UNECE and European Commission to review opportunities for improving O_3 policy integration into other relevant policy areas including energy, climate, food, development and biodiversity policy; this should include a review of how long-term (2050) targets for air pollutants and greenhouse gases should be defined so as to maximise benefits for air quality and climate policy objectives;

7.4 Integrated assessment frameworks and other policy options to be further developed and applied in regions where they are not yet in place, for example in Latin America, Africa and across Asia. This should be coordinated by the UNECE;

7.5 The CLRTAP to strengthen existing capacity building programmes to focus on the Eastern Europe, Caucasus and Central Asia region, and rapidly developing regions. This should be developed in coordination with the relevant regional development agencies and national aid programmes for developing countries. Programmes should include:

- *Assessessment of the economic, human health and environmental impacts of O_3 and other air pollutants in countries in Asia, Latin America and Africa;*
- *Analysis of the economic benefits of O_3 precursor controls in these regions;*
- *Guidance for the development of national emission inventory programmes, implementation of monitoring networks, and the development of regional photo-chemical models.*

11.8 Science and research needs

Despite rapid improvements over recent years in knowledge of O_3 production and destruction processes and impacts to human health and the environment, many gaps remain. Assessments are limited by the quality of emission inventory data, which are not globally available or standardised. Understanding of the biogeochemistry and physiological processes of natural emissions is not as advanced as for anthropogenic emissions and uncertainties remain regarding how drivers of emissions will change in the future. The implications of a doubling in China's energy demand for regional and global O_3 concentrations, and impacts of rapid growth in shipping emissions, on future projections require further evaluation.

Modelling techniques have been developed that have enabled the simulation of the combined effects of changes in climate and emissions on future O_3 concentrations. However, understanding of some of the basic processes and feedbacks between atmospheric chemistry, climate and ecosystems, is still developing, and limits in computing power mean that it is not yet possible to capture all of the important climate sensitive processes which affect O_3 production, destruction and transport at appropriate scales. Uncertainties in the emission scenarios and poorly constrained processes within the models are important determinants of the accuracy of future projections.

The evaluation of impacts to human health has been focused in the past on the impacts of short-term exposure to very high concentrations. However, it is now clear that health impacts are driven more by long-term exposure to baseline concentrations. Impact assessments are limited by a lack of accurate baseline data in many countries, and are complicated by uncertainty regarding whether there is a threshold of effects for O_3 in humans. Information regarding the chronic impacts of O_3 are insufficient for reliable health assessments and, although the impacts of acute effects are better understood, the mechanism by which mortality is affected is unclear.

Understanding of environmental impacts is based largely on plant species of economic value, with very little research conducted on wild species, or the ecological impacts of O_3, particularly outside of Europe and North America. The interactions between CO_2, climate change and O_3 impacts to vegetation require urgent investigation and the combined implications for food security, of O_3 and climate change need to be evaluated.

Information for developing countries is very restricted due to limited capacity to undertake emission monitoring, modelling assessments and impact evaluation.

Recommendation 8:

Significant new investment in tropospheric O_3 research is required by national governments and international agencies to improve the ground-level O_3 evidence base. This will require international cooperation and should as a priority:

8.1 Address the gaps in understanding of natural emissions and the influence of land use, climate and other global change parameters on emissions; in particular nmVOC, NO_X from soils and CH_4. This should be led by the UNECE in collaboration with the International Geosphere–Biosphere Programme;

8.2 Review anthropogenic and natural emission inventories, identify what is needed to improve modelling projections and impact assessments, particularly in developing countries, and enhance infrastructure and capacity as necessary. These activities should be regionally coordinated but led by the UNECE;

8.3 The IEA and UNDP to enhance implementation of low-cost technologies for reducing emissions in regions where emissions are projected to rapidly increase and in sectors projected to be important contributors to emissions in the future;

8.4 Focus on the urgent need to substantially increase computing power to enable the development of higher resolution, more complex models to improve the simulation of relevant climate, O_3 and ecological processes at spatial and temporal scales that resolve the time and space variability of the most important processes.

8.5 Strengthen and encourage current initiatives to measure and monitor the regional and global health burdens of O_3. This will require targeted research studies on the toxicology of O_3 and, in particular, the effects of chronic exposure; increased epidemiological research of acute and chronic effects in developing countries, and on chronic effects in other regions; and improved monitoring of ambient O_3 where required;

8.6 Establish long-term field studies for the assessment of the combined effects of O_3, elevated CO_2, climate change and other environmental stressors both on species of economic importance and on a range of natural and semi-natural ecosystems to improve the evidence base of effects on O_3, plant productivity, the ability of ecosystems to sequester carbon, important crop and forest species and biodiversity;

8.7 Initiate research into the effects of O_3 on important species for conservation, including the combined effects of O_3 and other environmental stressors. This research should focus on ecoregions and biodiversity hotspots where future O_3 concentrations represent the greatest threat. The results of this work should be provided to the CBD for their consideration as a new and emerging issue of relevance to biodiversity.

Acknowledgements

Chapter 4: Dr Peter Rafaj (IIASA) for producing the new IIASA emission scenarios. Juliette Lathiere (University of Sheffield) for the latest estimates of isoprene emissions and Alex Guenther (National Centre for Atmospheric Research, USA), Rob MacKenzie (Lancaster University), Malcolm Possell (Lancaster University) and David Beerling (University of Sheffield) for their helpful comments.

Chapter 5: Contributing modelers

Global modelling analysis:

ACCENT modelers; RS07 modelers.

- TM_4: Twan van Noije (KNMI, The Netherlands)
- FRSGC/UCI: Oliver Wild (Cambridge University; now Lancaster University)
- STOCHEM_HadGEM: Michael Sanderson and Bill Collins (UK Met Office)
- STOCHEM_HadAM3: David Stevenson and Ian MacKenzie (Edinburgh University)
- UM_CAM: Guang Zeng and John Pyle (Cambridge University)
- ULAQ: Veronica Montanaro and Gianni Pitari (Universita de L'Aquila, Italy)

Regional modelling analysis: Professor Peter Simmonds of the University of Bristol for communicating the baseline ozone observations from the Mace Head Atmospheric Research Station.

Urban modelling analysis: Mr John Stedman (AEA Technology)

Chapter 6: Elizabeth Barnes (University of Washington) for a new analysis of summertime blocking, Dr Kevin Hodges (University of Reading) for analysis of summertime anticyclones, and Professor Sir Brian Hoskins (University of Reading) for helpful discussions.

Chapter 7: Mr John Stedman for completing the health impact assessment for London.

References

Abbey DE, Mills PK, Petersen FF & Beeson WL (1991). *Long-term ambient concentrations of total suspended particulates and oxidants as related to incidence of chronic disease in California Seventh-Day Adventists*. Environmental Health Perspectives **94**, 43–50

Agrawal M (2007). *Experimental evidence of O_3 impacts on crop yield and quality in India*. Presented at a Royal Society workshop on ground-level O_3 in May 2007 Available online at http://royalsociety.org/document.asp?id=1170

Ainsworth EA, Rogers A & Leakey ADB (2008). *Targets for crop biotechnology in a future high-CO_2 and high-O_3 world*. Plant Physiology **147**, 13–19

Allen MR & Ingram WJ (2002). *Constraints on future changes in climate and the hydrologic cycle*. Nature **419**, 224–232. doi:10.1038/nature01092

Amann M, Asman W, Bertok I, Cofala J, Heyes C, Klimont Z, Schöpp W, Wagner F (2007). *Updated baseline projections for the revision of the emission ceilings directive of the European Union*. NEC Scenario Analysis Report No. 4. International Institute for Applied Systems Analysis (IIASA): Laxenburg, Austria

American Thoracic Society (2000). *What constitutes an adverse health effect of air pollution? Official Statement of the American Thoracic Society*. American Journal of Respiratory & Critical Care Medicine **161**, 2 Pt 1, 665–673

Anderson CP (2003). *Source–sink balance and carbon allocation below ground in plants exposed to O_3*. New Phytologist **157**, 213–228

Anderson HR, Ponce de LA, Bland JM, Bower JS, Emberlin J & Strachan DP (1998). *Air pollution, pollens, and daily admissions for asthma in London 1987–92*. Thorax **53**, 10, 842–848

Andreae MO and Merlet P (2001). *Emission of trace gases and aerosols from biomass burning*, Global Biogeochem. Cycles, **15**, 955–966

AQEG (2008). *Ozone in the United Kingdom. Consultation document.* The Air Quality Expert Group, UK

Arneth A, Schurgers G, Hickler T & Miller PA (2008). *Effects of species composition, land surface cover, CO_2 concentration and climate on isoprene emissions from European forests*. Plant Biology (Stuttgart) **10**, 1, 150–162

Ashmore MR (2005). *Assessing the future global impacts of O_3 on vegetation*. Plant, Cell and Environment **28**, 949–964

Atkinson R (2000). *Atmospheric chemistry of VOC and NO_X*. Atmospheric Environment **34**, 2063–2101

Atkinson RW *et al.* (2001). *Acute effects of particulate air pollution on respiratory admissions: results from APHEA 2 project. Air pollution and health: a European Approach.* American Journal of Respiratory and Critical Care Medicine **15**, 164, 10 Pt 1, 1860–1866

Auffhammer M, Ramanathan V & Vincent JR (2006). *Integrated model shows that atmospheric brown clouds and greenhouse gases have reduced rice harvests in India*. Proceedings of the National Academy of Sciences **103**, 19668–19672

Barnes J (2007). *Mechanisms of effect*. Presented at Royal Society workshop, May 2007 Available online at http://royalsociety.org/document.asp?id=1170

Barnes J, Bender J, Lyons T & Borland A (1999). *Natural and man-made selection for air pollution resistance.* Journal of Experimental Botany **50**, 1423–1435

Barnes EA, Slingo JM, Hodges KI & Hoskins BJ (submitted). *Blocking, anticyclones and climate change.* Submitted to Proc. Roy. Soc

Barrie LA, Bottenheim JW, Schnell, RC, Crutzen, PJ & Rasmussen RA (1988). *Ozone destruction and photochemical reactions at polar in the lower Arctic atmosphere.* Nature **334**, 138–141

Bell ML, Peng RD & Dominici F (2006). *The exposure–response curve for O_3 and risk of mortality and the adequacy of current O_3 regulations.* Environmental Health Perspectives **114,** 4, 532–536

Bell ML, Kim JY & Dominici F (2007). *Potential confounding of particulate matter on the short-term association between O_3 and mortality in multisite time-series studies.* Environmental Health Perspectives **115**, 11, 1591–1595

Biswas DK, Xu H, Li YG, Sunw JZ, Wangz XZ, Han XG & Jiang GM (2008). *Genotypic differences in leaf biochemical, physiological and growth responses to O_3 in 20 winter wheat cultivars released over the past 60 years.* Global Change Biology **14**, 46–59. doi: 10.1111/j.1365-2486.2007.01477.x

Bolin B & Cook RB (1983). *The Major Biogeochemical Cycles and their interactions*. SCOPE 21. Wiley: Chichester, UK

Bosson J, Pourazar J, Forsberg B, Adelroth E, Sandström T & Blomberg A (2007). *Ozone enhances the airway inflammation initiated by diesel exhaust*. Respiratory Medicine **101**, 1140–1146

Brasseur G, Schultz M, Granier C, Saunois M, Diehl T, Botzlet M, Roeckner E & Walters S (2006). *Impact of climate change on the future chemical composition of the global troposphere.* Journal of Climate **19**, 3932–3951. doi:10.1175/JCLI3832.1

Braun S, Rihm B, Schindler C & Fluckiger W (1999). *Growth of mature beech in relation to O_3 and nitrogen deposition: an epidemiological approach.* Water Air and Soil Pollution **116**, 357–364

Brilli F, Barta C, Fortunati A, Lerdau M, Loreto F & Centritto M (2007). *Response of isoprene emission and carbon metabolism to drought in white poplar (Populus alba) saplings*. New Phytologist **175**, 2, 244–254

Bush ML, Asplund PT, Miles KA, Ben-Jebria A & Ultman JS (1996). *Longitudinal distribution of O_3 absorption in the*

lung: gender differences and intersubject variability. Journal of Applied Physiology **81**, 1651–1657

Butchart N *et al.* (2006). *Simulations of anthropogenic change in the strength of the Brewer–Dobson circulation*. Climate Dynamics **27**, 727–741. doi 10.1007/s00382-006-0162-4

CAI-Asia (2008). *Emission standards for new vehicles (light duty).* Clean Air Initiative for Asian Cities. Available online at www.cleanairnet.org/caiasia/1412/articles-58969_resource_1.pdf

Calfapietra C, Wiberley AE, Falbel TG, Linsky AR, Mugnozza GS, Karnosky DF, Loreto F, Sharkey TD (2007). *Isoprene synthase expression and protein levels are reduced under elevated O_3 but not under elevated CO_2 (FACE) in field-grown aspen trees.* Plant Cell and Environment **30**, 5, 654–661

Carver GD, Brown PD and Wild O (1997). *The ASAD atmospheric chemistry integration package and chemical reaction database*. Computer Physics Communications, **105**, 197–215

CBD (2007). *New and emerging issues relating to the conservation and sustainable use of biodiversity. Biodiversity and liquid biofuel production.* Note by the Executive Secretary. UNEP/CBD/SBSTTA/12/9 Available online at http://www.cbd.int/doc/meetings/sbstta/sbstta-12/official/sbstta-12-09-en.pdf

Centritto M, Nascetti P, Petrilli L, Raschi A & Loreto F (2004). *Profiles of isoprene emission and photosynthetic parameters in hybrid poplars exposed to free-air CO_2 enrichment.* Plant, Cell and Environment **27**, 403–412

Chameides WL, Saylor RD & Cowling EB (1997). *Ozone pollution in the rural United States and the new NAAQS.* Policy Forum. Science **276**, 916

Chang WN, Heikes BG & Lee MH (2004). *Ozone deposition to the sea surface: chemical enhancement and wind speed dependence.* Atmospheric Environment **38**, 1053–1059

Chen C, Arjomandi M, Tager IB, Holland N & Balmes JR. (2007). *Effects of antioxidant enzyme polymorphisms on O_3-induced lung function changes.* European Respiratory Journal **30**, 4, 677–683

Christensen JH *et al*. (2007). *Regional climate projections (supplementary material)*. In *Climate Change 2007: The Physical Science Basis*. Contribution of Working group 1 to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change (eds Solomon SD, Qin D, Manning M, Chen Z, Marquis M, Averyt KB, Tignor M, Miller HL). Cambridge University Press: Cambridge, UK

CIAM (2007). *Review of the Gothenburg Protocol.* Task Force on Integrated Assessment Modelling of the UNECE Convention on Long-range Transboundary Air Pollution and the Centre for Integrated Assessment Modelling. CIAM report 1/2007. International Institute for Applied Systems Analysis (IIASA): Laxenburg, Austria Available online at www.iiasa.ac.at/rains/reports/CIAM1-2007.pdf

Clark R, Brown S & Murphy J (2006). *Modelling Northern Hemisphere summer heat extreme changes and their uncertainties using a physics ensemble of climate sensitivity experiments*. Journal of Climate **19**, 4418–4435

Cofala J *et al.* (2007a). *Analysis of policy measures to reduce ship emissions in the context of the revision of the national emissions ceilings directive.* Final report IIASA Contract Mp. 06-107 April 2007. International Institute for Applied Systems Analysis (IIASA): Laxenburg, Austria

Cofala J, Amann M, Klimont Z, Kupiainen K & Höglund-Isaksson L (2007b) *Scenarios of global anthropogenic emissions of air pollutants and methane until 2030.* Atmospheric Environment **41**, 8486–8499

Collins WJ, Stevenson DS, Johnson CE & Derwent RG (1997). *Tropospheric ozone in a global-scale three-dimensional Lagrangian model and its response to NO_x emission controls,* J. Atmos. Chem.*,* **26**, 223–274

Collins WJ, Derwent RG, Johnson CE & Stevenson DS (2002a). *The oxidation of organic compounds in the troposphere and their global warming potentials*. Climatic Change **52**, 453–479

Collins WJ, Derwent RG, Johnson CE & Stevenson DS (2002b). *A comparison of two schemes for the convective transport of chemical species in a Lagrangian global chemistry model.* Q. J. R. Meteorol. Soc., **128**, 991–1009

Collins WJ, Derwent RG, Garnier B, Johnson CE, Sanderson MG and Stevenson DS (2003). The effect of stratosphere-troposphere exchange on the future tropospheric ozone trend. J. Geophys. Res.*,* **108**, D12. doi: 10.1029/2002JD002617

Corbett JJ & Kohler HW (2003). *Updated emissions* from *ocean shipping*. Journal of Geophysical Research **108**, D20, 4650. doi:10.1029/2003JD003751

Corbett JJ, Winebrake JJ, Green EH, Kasibhatla P, Eyring V & Lauer A (2007). *Mortality from ship emissions: a global assessment.* Environmental Science and Technology **41**, 24, 8512–8518

Coyle M, Fowler D & Ashmore M (2003). *New directions: implications of increasing tropospheric background O_3 concentrations for vegetation*. Atmospheric Environment **37**, 153–154

Crutzen PJ (1974). *Photochemical reactions initiated by and influencing O_3 in the unpolluted troposphere*. Tellus **26**, 47–57

Crutzen PJ, Mosier AR, KA Smith & Winiwarter W (2007). N_2O *release from agro-biofuel production negates global warming reduction by replacing fossil fuels.* Atmospheric Chemistry and Physics Discussions **7**, 11191–11205

Davidson EA & Kingerlee W (1997). *A global inventory of nitric oxide emissions from soils.* Nutrient Cycling in Agroecosystems **48**, 37–50

Davison AW & Barnes J (1998). *Effects of O_3 on wild plants*. New Phytologist **139**, 135–151

DEFRA (2006). *Air Quality Strategy for England, Scotland, Wales and Northern Ireland*. A consultation document on options for further improvements in air quality. April 2006. Defra: London, UK

DEFRA (2007). *The Air Quality Strategy for England, Scotland, Wales and Northern Ireland*. Defra: London, UK Available at www.defra.gov.uk/environment/airquality/strategy/index.htm

Denman KL *et al.* (2007). *Couplings Between Changes in the Climate System and Biogeochemistry*. In *Climate Change 2007: The Physical Science Basis*. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change (eds Solomon SD, Qin D, Manning M, Chen Z, Marquis M, Averyt KB, Tignor M & Miller HL). Cambridge University Press: Cambridge, UK

Dentener F, Peters W, Krol M, van Weele M, Bergamaschi P & Lelieveld J (2003). *Interannual variability and trend of CH_4 lifetime as a measure for OH changes in the 1979–1993 time period,* J. Geophys. Res., **108**, D15, 4442. doi:10.1029/2002JD002916

Dentener F, Stevenson D, Cofala J, Mechler R, Amann M, Bergamaschi P, Raes F & Derwent R (2005). *The impact of air pollutant and methane emission controls on tropospheric O_3 and radiative forcing: CTM calculations for the period 1990–2030*. Atmospheric Chemistry and Physics **5**, 1731–1755

Dentener F *et al.* (2006a). *Nitrogen and sulphur deposition on regional and global scales: a multi-model evaluation.* Global Biogeochemical Cycles **20**, GB4003. doi:10.1029/2005GB002672

Dentener F *et al.* (2006b). *The global atmospheric environment for the next generation*. Environmental Science and Technology **40**, 3586–3594

DoH (2008). *Health effects of climate change in the UK 2008: an update of the Department of Health report.* Document number 285576. Department of Health and Health Protection Agency: London Available online at www.dh.gov.uk/en/Publicationsandstatistics/Publications/PublicationsPolicyAndGuidance/DH_080702

Derwent RG, Jenkin ME & Saunders SM (1996). *Photochemcial O_3 creation potentials for a large number of reactive hydrocarbons under European conditions.* Atmospheric Environment **30**, 181–191

Derwent RG, Collins WJ, Johnson CE & Stevenson DS (2001). *Transient behaviour of tropospheric O_3 precursors in a global 3-D CTM and their indirect greenhouse effects*, Climatic Change **49**, 463–487

Derwent RG *et al*. (2006). *External influences on Europe's air quality: Baseline methane, carbon monoxide and O_3 from 1990 to 2030 at Mace Head, Ireland*. Atmospheric Environment **40**, 844–855

Derwent RG, Stevenson DS, Doherty RM, Collins WJ, Sanderson MG & Johnson CE (2008). *Radiative forcing from surface NO_X emissions: spatial and seasonal variations.* Climatic Change **88**, 3, 385–401. doi 10.1007/s10584-007-9383-8

Doherty RM, Stevenson DS, Collins WJ & Sanderson MG (2005). *Influence of convective transport on tropospheric O_3 and its precursors in a chemistry–climate model*. Atmospheric Chemistry and Physics **5**, 3205–3218

Doherty RM, Stevenson DS, Johnson CE, Collins WJ & Sanderson MG (2006). *Tropospheric O_3 and El Nino-Southern Oscillation: influence of atmospheric dynamics, biomass burning emissions, and future climate change.* Journal of Geophysical Research **111**. doi:10.1029/2005JD006849

Duncan N, Logan JA, Bey I, Megretskaia IA, Yantosca RM, Novelli PC, Jones NB, Rinsland CP (2007). *Global budget of CO, 1988–1997: source estimates and validation with a global model*. Journal of Geophysical Research **112**, D22301. doi:10.1029/2007JD008459

Easterling WE *et al*. (2007). Chapter 5, *Food, fibre and forest products*. In *Climate Change 2007: Impacts, Adaptation and Vulnerability*. Contribution of Working Group II to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change (eds Parry ML *et al*.) 273–313. Cambridge University Press, Cambridge

EEA (1998). *Tropospheric O_3 in EU – the consolidated report*. Topic report No. 8/1998. European Environment Agency: Copenhagen Available online at http://reports.eea.europa.eu/TOP08-98/en/pahe004.html

EEA (2007). *Air Pollution in Europe 1990–2004*. EEA Report No2/2007. European Environment Agency: Copenhagen

EEA (2008). *Ozone air pollution in the summer of 2007.* Technical report 5/2008. European Environment Agency: Copenhagen

Ellingsen K *et al*. (2008). *Global O_3 and air quality: a multi-model assessment of risks to human health and crops*. Atmospheric Chemistry and Physics Discussions **8**, 2163–2223

Emberson LD, Ashmore MR & Murray F (2003). *Air Pollution Impacts on Crops and Forests – A Global Assessment*. Imperial College Press: London

Emberson LD, Büker P & Ashmore MR (2007). *Assessing the risk caused by ground-level O_3 to European forest trees: a case study in pine, beech and oak across different climate regions.* Environmental Pollution **147**, 3, 454–466

Emberson LD, Büker P, Ashmore MR, Mills G, Jackson L, Agrawal M, Atikuzzaman MD, Cinderby S, Engardt M, Jamir C, Kobayashi K, Oanh K, Wahid A (*submitted). Dose-response relationships derived in North America underestimate the effects of ozone (O_3) on crop yields in Asia.* Submitted to Atmospheric Environment.

Engardt M (2008). *Modelling of near-surface O_3 over South Asia*. Journal of Atmospheric Chemistry **59**, 1, 61–80

European Commission (2003) *Climate–O_3 interactions.* Air pollution research report No 81. EUR 20623 DG Research. European Commission: Brussels

European Commission (2005). *Thematic strategy on air pollution impact assessment, SEC (2005) 1133*. Commission staff working paper; Annex to *The Communication – on Thematic Strategy on Air Pollution and the Directive on Ambient Air Quality and Cleaner Air for Europe* [COM(2005) 446, and COM (2005) 447 final].

European Community (2001). Directive 2001/81/EC of the European Parliament and of the Council of 23 October 2001 on national emission ceilings for certain atmospheric pollutants.

Eyring V, Köhler HW, van Aardenne J, and Lauer A (2005a). *Emissions from International Shipping: 1. The last 50 Years. J. Geophys. Res.*, **110**, D17305. doi:10.1029/2004JD005619

Eyring V, Köhler HW, Lauer A & Lemper B (2005b). *Emissions from international shipping: 2. Impact of future technologies on scenarios until 2050*. Journal of Geophysical Research **110**, D17306. doi:10.1029/2004JD005620

Eyring V, Isaksen ISA, Berntsen T, Collins WJ, Corbett JJ, Endresen O, Grainger RG, Moldavana J, Schlager H & Stevenson DS (submitted). *Assessment of transport impacts on climate and ozone: shipping.* submitted to Atmospheric Environment, 2008

Fang CW, Monson RK & Cowling EB (1996). *Isoprene emission, photosynthesis, and growth in sweetgum (*Liquidambar styraciflua*) seedlings exposed to short- and long-term drying cycles*. Tree Physiology **16**, 441–446

Fares S, Barta C, Brilli F, Centritto M, Ederli L, Ferranti F, Pasqualini S, Reale L, Tricoli D & Loreto, F (2006). *Impact of high O_3 on isoprene emission, photosynthesis and histology of developing* Populus alba *leaves directly or indirectly exposed to the pollutant.* Physiologia Plantarum **128**, 456–465

Fearnleys (2008). *Fearnleys Review 2007* Available online at http://www.fearnresearch.no/index.gan?id=70

Fehsenfeld *et al*. (2006). *International Consortium for Atmospheric Research on Transport and Transformation (ICARTT): North America to Europe – Overview of the 2004 summer field study.* Journal of Geophysical Research **111**, D23S01. doi:10.1029/2006JD007829

Felzer BS, Reilly JM, Melillo JM, Kicklighter DW, Wang C, Prinn RG, Sarofim M & Zhuang Q (2004). *Past and future effects of O_3 on net primary production and carbon sequestration using a global biogeochemical model*. Report no 103. MIT Joint Program on the Science and Policy of Global Change. Massachusetts Institute of Techology: Cambridge, USA

Felzer BS, Reilly J, Melillo J, Kicklighter DW, Sarofim M, Wang C, Prinn RG, Zhuang Q (2005). *Future effects of O_3 on carbon sequestration and climate change policy using a global biogeochemical model.* Climatic Change **73**, 345–373. doi 10.1007/s10584-005-6776-4

Fiala J, Cernikovsky L, de Leeuw F & Kurfuerst P (2003). *Air pollution by O_3 in Europe in Summer 2003. Overview of exceedances of EC O_3 threshold values during the summer season April–August 2003 and comparisons with previous years*. Report to the European Commission by the European Environment Agency, European Topic Centre on Air and Climate Change based on data provided in the framework of Council Directive 92/72/EEC on air pollution by O_3 by 15 September 2003. European Environment Agency. Report 3/2003

Filleul L *et al.* (2006). *The relation between temperature, O_3, and mortality in nine French cities during the heat wave of 2003.* Environmental Health Perspectives **114**, 9, 1344–1347

Fiore AM, Jacob DJ, Bey I, Yantosca RM, Field BD, Fusco AC & Wilkinson, JG (2002). *Background O_3 over the United States in summer: origin, trend, and contribution to pollution episodes.* Journal of Geophysical Research **107**, D15. doi: 10.1029/2001JD000982

Fischer EM, Seneviratne SI, Lüthi D & Schär C (2007). *Contribution of land–atmosphere coupling to recent European summer heat waves.* Geophysical Research Letters **34** (L06707)

Folinsbee LJ, McDonnell WF & Horstman DH (1988). *Pulmonary function and symptom responses after 6.6-hour exposure to 0.12 ppm O_3 with moderate exercise.* JAPCA **38**, 1, 28–35

Forster P *et al.* (2007). *Changes in atmospheric constituents and radiative forcing.* In *Climate Change 2007: The Physical Scientific Basis*. Contribution of Working Group 1 to the Fourth Assessment Report on the Intergovernmental Panel on Climate Change (eds Solomon SD, Qin D, Manning M, Chen Z, Marquis M, Averyt KB, Tignor M, Miller HL) 129–234. Cambridge University Press, Cambridge

Frischer T, Studnicka M, Gartner C, Tauber E, Horak F, Veiter A, Spengler J, Kuhr J, Urbanek R (1999) *Lung function growth and ambient O_3: a three-year population study in school children.* American Journal of Respiratory and Critical Care Medicine **160**, 2, 390–396

Fuglestvedt JS, Berntsen T, Isaksen ISA, Mao H, Liang X, Wang W (1999). *Climatic forcing of nitrogen oxides through changes in tropospheric ozone and methane; global 3D model studies*. Atmospheric Environment **33**, 961–977

Fuhrer J (2003). *Agroecosystem responses to combinations of elevated CO_2, O_3 and global climate change.* Agriculture, Ecosystems and Environment **97**, 1–20

Fuhrer J, Skarby L & Ashmore MR (1997). *Critical levels for ozone effects on vegetation in Europe.* Environmental Pollution **97**, 91–106

Funk JL, Jones CG, Gray DW, Throop HL, Hyatt LA & Lerdau MT (2005). *Variation in isoprene emission from* Quercus rubra*: sources, causes, and consequences for estimating fluxes.* Journal of Geophysical Research – Atmospheres **110**, D04301. doi:10.1029/2004JD005229

Ganzeveld L & Lelieveld J (1995). *Dry deposition parameterization in a chemistry general circulation model and its influence on the distribution of reactive trace gases.* Journal of Geophysical Research **100**, 2135–2147

Ganzeveld L & Lelieveld J (2004). *Impact of Amazonian deforestation on atmospheric chemistry.* Geophysical Research Letters **31**, L060105. doi:10.1029/2003GL019205

Garland JA, Elzerman AW & Penkett SA (1980). *The mechanism for dry deposition of ozone to seawater surfaces.* Journal of Geophysical Research **85**, 7488–7492

Gauderman WJ, Avol E, Gilliland F, Vora H, Thomas D, Berhane K *et al.* (2004). *The effect of air pollution on lung development from 10 to 18 years of age.* New England Journal of Medicine **351**, 11, 1057–1067

Gauss M *et al.* (2003) *Radiative forcing in the 21st century due to ozone changes in the troposphere and the lower stratosphere*. Journal of Geophysical Research **108**, 4292. doi:10.1029/2002JD002624

Gauss M, Isaksen ISA, Lee DS & Sovde OA (2005). *Impact of aircraft* NO_X *emissions on the atmosphere – tradeoffs to reduce the impact.* Atmospheric Chemistry and Physics Discussions **5**, 12255–12311

Gettelman A, Holton JR & Rosenlof KH (1997). *Mass fluxes of* O_3*,* CH_4*,* N_2O*, and* CF2Cl2 *in the lower stratosphere calculated from observational data*. Journal of Geophysical Research **102**, D15, 19149–19159

Gilmour MI & Selgrade MKA (1993). *Comparison of the pulmonary defenses against streptococcal infection in rats and mice following* O_3 *exposure: differences in disease susceptibility and neutrophil recruitment.* Toxicology and Applied Pharmacology **123**, 211–218

Glickman TS (2000). *Glossary of Meteorology*, 2nd edn. American Meteorological Society: Washington, DC

Goldewijk KK (2001). *Estimating global land use change over the past 300 years: the HYDE database*. Global Biogeochemical Cycles **15**, 417–433

Goswami BN, Venugopal V, Sengupta D, Madhusoodanan MS & Xavier Prince K (2006). *Increasing trend of extreme rain events over India in a warming environment. Science* **314**, 5804, 1442–1445.

Graedel TE & Crutzen PJ (1993). *Atmospheric change: an Earth system perspective*. Freeman: New York

Granier C *et al*. (2003). *Impact of climate change on tropospheric ozone*. In *Ozone–Climate Interactions* (ed. Isaksen ISA) (chapter 5). European Commission Air Pollution Research Report No. 81, DG Research: Brussels

Gregory D and Rowntree PR (1990), *A mass flux convection scheme with representation of cloud ensemble characteristics and stability dependent closure*. Mon. Weather Rev., **118**, 1483–1506

Gryparis A *et al.* (2004). *Acute effects of ozone on mortality from the "air pollution and health: a European approach" project*. American Journal of Respiratory and Critical Care Medicine **170**, 10, 1080–1087

Guenther A *et al.* (1995). *A global model of natural volatile organic compound emissions.* Journal of Geophysical Research – Atmospheres **100**, 8873–8892

Guenther A, Baugh B, Brasseur G, Greenberg J, Harley P, Klinger L, Serca D & Vierling L (1999). *Isoprene emission estimates and uncertainties for the Central African EXPRESSO study domain.* Journal of Geophysical Research – Atmospheres **104**, D23, 30625–30639

Guenther A, Karl T, Harley P, Wiedinmyer C, Palmer PI & Geron C (2006). *Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature)*. Atmospheric Chemistry and Physics **6**, 3181–3210

Guilyardi E (2006). *El Niño–mean state–seasonal cycle interactions in a multi-model ensemble.* Climate Dynamics **26**, 329–348

Haagen Smit AJ (1952). *Chemistry and physiology of Los Angeles smog*. Industrial and Engineering Chemistry **44**, 6, 1342–1346

Hargreaves KJ & Fowler D (1998). *Quantifying the effects of water table and soil temperature on the emission of methane from peat wetland at the field scale.* Atmospheric Environment **32**, 19, 3275–3282

Harzallah A & Sadourny R (1995). *Internal versus SST-forced atmospheric variability as simulated by atmospheric general circulation model*. Journal of Climate **8**, 474–495

Hauglustaine DA, Lathiere J, Szopa S and Folberth GA (2005) *Future tropospheric ozone simulated with a climate–chemistry–biosphere model.* Geophysical Research Letters **32**, L24807. doi:10.1029/2005GL024031

Hayes F, Jones MLM, Mills G & Ashmore M (2007a). *Meta-analysis of the relative sensitivity of semi-natural vegetation species to ozone.* Environmental Pollution **136**, 754–762

Hayes F, Mills G, Harmens H and Norris D (2007b). *Evidence of widespread ozone damage to vegetation in Europe (1990–2006). ICP Vegetation.* Centre for Ecology and Hydrology: Bangor, UK

Heck WW, Taylor OC & Tingey DT (1988). *Assessment of Crop Loss from Air Pollutants*. Elsevier Applied Science: London

Hegarty J, Mao H & Talbot R (2007). *Synoptic controls on summertime surface ozone in the northeastern United States.* Journal of Geophysical Research **112**. doi:10.1029/2006JD008170

Held I & BJ Soden (2006). *Robust responses of the hydrological cycle to global warming*. Journal of Climate **18**, 5686–5699

Hess PG & Lamarque J-F (2007). *Ozone source attribution and its modulation by the Arctic oscillation during the spring months.* Journal of Geophysical Research **112**, D11303. doi:10.1029/2006JD007557

Hogrefe C, Lynn B, Civerolo K, Ku JY, Rosenthal J, Rosenzweig C, Goldberg R, Gaffin S, Knowlton K & Kinney PL (2004). *Simulating changes in regional air pollution over the eastern United States due to changes in global and regional climate and emissions*. Journal of Geophysical Research **109**, D22301. doi:10.1029/2004JD004690

Holland M, Kinghorn S, Emberson L, Cinderby S, Ashmore M, Mills G & Harmens H (2006). *Development of a framework for a probabilistic assessment of the economic losses caused by ozone damage to crops in Europe.* Centre for Ecology and Hydrology: Bangor

Horak F Jr *et al.* (2002). *Particulate matter and lung function growth in children: a 3-yr follow-up study in Austrian schoolchildren.* European Respiratory Journal **19**, 5, 838–845. Ref ID: 33

Hoskins BJ & Hodges KI (2002). *New perspectives on the Northern Hemisphere winter storm tracks.* Journal of Atmospheric Science **59**, 1041–1061

Horstman DH, Folinsbee LJ, Ives PJ, Abdul-Salaam S & McDonnell WF (1990). *Ozone concentration and pulmonary response relationships for 6.6-hour exposures with five hours of moderate exercise to 0.08, 0.10, and 0.12 ppm.* American Review of Respiratory Disease **142,** 5, 1158–1163

IEA (2006). *World Energy Outlook 2006.* International Energy Agency: France

IEA (2007). *World Energy Outlook 2007: China and India Insights*. International Energy Agency: France

Ihorst G, Frischer T, Horak F, Schumacher M, Kopp M, Forster J, Mattes J, Kuehr J (2004). *Long- and medium-term ozone effects on lung growth including a broad spectrum of exposure*. European Respiratory Journal **23**, 2, 2929

IMF (2000). *World Economic Outlook; Asset Prices and the Business Cycle.* International Monetary Fund: Washington

IPCC (2000). *Special Report on Emission Scenarios*. A Special Report of the Working Group III of Intergovernmental Panel on Climate Change. Cambridge University Press: Cambridge

IPCC (2001). *Climate Change 2001:* The Scientific Basis, Contribution of Working Group I to the Third Assessment report of the IPCC, (Eds. Houghton JT, Ding Y, Griggs DJ, Noguer M, van der Linden PJ, Dai X, Maskell K, Johnson CA) Cambridge University Press, Cambridge: UK and New York: USA

IPCC (2007). *Climate Change 2007: Impacts, Adaptation and Vulnerability. Contribution of Working Group II to the Fourth Assessment. Report of the Intergovernmental Panel on Climate Change,* ML Parry, OF Canziani, JP Palutikof, PJ van der Linden and CE Hanson, Eds., Cambridge University Press, Cambridge, UK, 976pp

IPCC (2007a). *Climate Change 2007:* The Physical Science Basis Working Group I Contribution to the Fourth Assessment Report of the IPCC (Intergovernmental Panel on Climate Change). (Eds. Solomon SD, Qin D, Manning M, Chen Z, Marquie M, Averyt KB, Tignor M and Miller HL) Cambridge University Press: Cambridge, UK and New York: USA

IPCC (2007b). *Summary for policymakers*. In *Climate Change 2007: The Physical Science Basis*. Contribution of Working Group 1 to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change (eds Solomon SD, Qin D, Manning M, Chen Z, Marquie M, Averyt KB, Tignor M & Miller HL) Figure SPM 7, p 16. Cambridge University Press: Cambridge, UK

Isaksen ISA, Zerefos C, Kourtidis K, Meleti C, Dalsøren SB, Sundet JK, Grini A, Zanis P, Balis D (2005). *Tropospheric ozone changes at unpolluted and semipolluted regions induced by stratospheric ozone changes.* Journal of Geophysical Research **110**, D02302. doi:10.1029/2004JD004618

Jacob DJ, Logan JA & Murti PP (1999). *Effect of rising Asian emissions on surface ozone in the United States.* Geophysical Research Letters **26**, 2175–2178

Jenkin ME (2004). *Analysis of sources and partitioning of oxidant in the UK. Part 1: The NO_X dependence of annual mean concentrations of nitrogen dioxide and ozone*. Atmospheric Environment **38**, 5117–5129

Jenkin ME (2008). *Trends in ozone concentration distributions in the UK since 1990: local, regional and global influences.* Atmospheric Environment **42**, 5434–5445

Johnson CE, Collins WJ, Stevenson DS & Derwent RG (1999). *The relative roles of climate and emissions changes on future oxidant concentrations.* Journal of Geophysical Research **104**, D15, 18631–18645

Johnson CE, Stevenson DS, Collins WJ & Derwent RG (2001). *Role of climate feedback on methane and ozone studied with a coupled ocean–atmosphere–chemistry model.* Geophysical Research Letters **28**, 1723–1726

Jorres R, Nowak D, Magnussen H (1996). *The effect of ozone exposure on allergen responsiveness in subjects with asthma or rhinitis.* American Journal of Respiratory and Critical Care Medicine **153**, 1, 56–64

Jorres RA *et al.* (2000) *The effect of repeated ozone exposures on inflammatory markers in brochial lavage fluid and mucosal biopsies.* American Journal of Respiratory and Critical Care Medicine **161**, 1855–1861

Karlsson PE *et al.* (2004). *New critical levels for ozone effects on young trees based on AOT40 and simulated cumulative leaf uptake of ozone.* Atmospheric Environment **38**, 2283–2294

Karlsson PE, Pleijel H, Belhaj M, Danielsson H, Dahlin B, Andersson M, Hansson M, Munthe J, Grennfelt P (2005). *Economic assessment of the negative impacts of ozone on crop yields and forest production*. A case study of the Estate Östads Säteri in Southwestern Sweden. AMBIO: A Journal of the Human Environment **34**, 1, 32–40

Karlsson PE, Orlander G, Langvall O, Uddling J, Hjorth U, Wiklander K, Areskoug B & Grennfelt P (2006). *Negative impact of ozone on the stem basal area increment of mature Norway spruce in south Sweden*. Forest Ecology and Management **232**, 146–151

Klaassen G, Amann M, Berglund C, Cofala C, Höglund-Isaksson L, Heyes C, Mechler R, Tohka A, Schöpp W & Winiwarter W (2004). *The Extension of the RAINS Model to Greenhouse Gases. IR-04-015.* International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria)

Kehrl HR, Peden DB, Ball B, Folinsbee LJ & Horstman D (1999). *Increased specific airway reactivity of persons with mild allergic asthma after 7.6 hours of exposure to 0.16 ppm ozone*. Journal of Allergy and Clinical Immunology **104**, 6, 1198–1204

King JS, Kubiske ME, Pregitzer KS, Hendrey GR, McDonald EP, Giardina CP, Quinn VS & Karnosky DF (2005). *Tropospheric O_3 compromises net primary production in young stands of trembling aspen, paper birch and sugar maple in response to elevated atmospheric* CO_2. New Phytologist **168**, 623–636

Kjellström E, Bärring L, Jacob D, Jones R, Lenderink G, Schär, C (2007). *Modelling daily temperature extremes: recent climate and future changes over Europe*. Climate Change **81**, Suppl. 1, 249–265. doi:10.1007/s10584-006-9220-5

Kulle TJ, Sauder LR, Kerr HD, Farrell BP, Bermel MS, Smith M (1984) *Duration of pulmonary function adaption to ozone*

in humans. American Industrial Hygiene Association Journal **43**, 832–837

Ladha JK *et al*. (2003). *How extensive are yield declines in long-term rice-wheat experiments in Asia?* Field Crops Research **81**, 159–180

Lamarque J-F *et al.* (2005). *Assessing future nitrogen deposition and carbon cycle feedback using a multimodel approach: analysis of nitrogen deposition*. Journal of Geophysical Research **110**, 1–21. doi:10.1029/2005JD005825

Lambert SJ & Fyfe JC (2006). *Changes in winter cyclone frequencies and strengths simulated in enhanced greenhouse warming experiments: results from the models participating in the IPCC diagnostic exercise.* Climate Dynamics **26**, 7–8, 713–728. doi:10.1007/s00382-006-0110-3

Langner J, Bergstrom R & Foltescu V (2005). *Impact of climate change on surface ozone and deposition of sulphur and nitrogen in Europe*. Atmospheric Environment **39**, 1129–1141

Lathière J, Hauglustaine DA, De Noblet-Ducoudre N, Krinner G & Folberth GA (2005). *Past and future changes in biogenic volatile organic compound emissions simulated with a global dynamic vegetation model.* Geophysical Research Letters **32**, 20. doi:10.1029/2005GL024164

Lathière J, Hauglustaine DA, Friend AD, De Noblet-Ducoudre N, Viovy N & Folberth GA (2006). *Impact of climate variability and land use changes on global biogenic volatile organic compound emissions.* Atmospheric Chemistry and Physics **6**, 2129–2146

Lawrence MG, von Kuhlmann R & Salzmann M (2003). *The balance of effects of deep convective mixing on tropospheric ozone.* Geophysical Research Letters **30**, 18. doi:10.1029/2003GL017644

Lee JD *et al*. (2006). *Ozone photochemistry during the UK heat wave of August 2003*. Atmospheric Environment **40**, 7598–7613

Leighton PA (1961). *Photochemistry of Air Pollution*. Academic Press: New York

Leikauf GD, Simpson LG, Santrock J, Zhao Q, Abbinante-Nissen J, Zhou S & Driscoll KE (1995). *Airway epithelial cell responses to ozone injury*. Environmental Health Perspectives **103**, 91–95

Lelieveld J, van Aardenne J, Fischer H, de Reus M, Williams J & Winkler P (2004). *Increasing ozone over the Atlantic Ocean.* Science **304**, 1483–1486

Le Treut H, Somerville R, Cubasch U, Ding Y, Mauritzen C, Mokssit A, Peterson T & Prather M (2007). *Historical overview of climate change*. In: *Climate Change 2007: The Physical Science Basis*. Contribution of Working Group 1 to the Fourth assessment Report of the Intergovernmental panel on Climate Change (eds Solomon S, Qin D, Manning M, Chen Z, Marquis M, Averyt KB, Tignor M & Miller HL). Cambridge University Press: Cambridge

Leonard BP, Lock AP, and MacVean MK (1995). *The NIRVANX scheme applied to one-dimensional advection, Int. J. Numer.* Methods for Heat and Fluid Flow, **5**, 341–377

Li QB, Jacob DJ & Bey I *et al.* (2002). *Transatlantic transport of pollution and its effects on surface ozone in Europe and North America.* Journal of Geophysical Research – Atmospheres **107**, D13, 4166

Liao H, Chen W-T & Seinfeld JH (2006). *Role of climate change in global predictions of future tropospheric ozone and aerosols*. Journal of Geophysical Research **111**, D12304. doi:10.1029/2005JD006852.

Lindskog R & Sundqvist G (2007). *Regulating European air: the co-production of science and policy.* In *Transboundary Air Pollution Scientific Understanding and Environmental Policy in Europe* (ed. Pleijel H). Studentlitteratur: Lund, Sweden

Liss PS (1983). *The exchange of biogeochemically important gases across the air–sea interface.* In *The Major Biogeochemical Cycles and Their Interactions* (eds Bolin B & Cook RB) 411–426. Wiley: Chichester, UK

Liu SC, Kley D, McFarland M, Mahlman JD & Levy H (1980). *On the origin of tropospheric ozone*. Journal of Geophysical Research **85**, 7546–7552

Logan J *et al.* (1999). *Trends in Vertical Distribution of Ozone: A Comparison of Two Analyses of Ozone Sonde Data.* Journal of Geophysical Research **104**, 26373–26399

Loya WM, Pregitzer KS, Karberg NJ, King JS & Giardina CP (2003) *Reduction of soil carbon formation by tropospheric ozone under increased carbon dioxide levels.* Nature **425**, 705–707

Matyssek R *et al.* (2007). *Synopsis of the CASIROZ case study: carbon sink strength of* Fagus sylvatica *L. in a changing environment – experimental risk assessment of mitigation by chronic ozone impact.* Plant Biology **9**, 163–180

Mautz WJ & Dohm MR (2004). *Respiratory effects of ozone on a lizard and a frog*. Comparative Biochemistry and Physiology, Part A **139**, 371–377

Mauzerall DL & Wang X (2001). *Protecting agricultural crops from the effects of tropospheric ozone exposure: reconciling science and standard setting in the United States, Europe and Asia*. Annual Review of Energy and the Environment **26**, 237–268

McConnell R, Berhane K, Gilliland F, London SJ, Islam T, Gauderman WJ, Avol E, Margolis HG, Peters JM (2002). *Asthma in exercising children exposed to ozone: a cohort study.* Lancet **359**, 9304, 386–391

McFiggans G, Plane JMC, Allan BJ, Carpenter L, Coe H & O'Dowd C (2000). *A modelling study of iodine chemistry in the marine boundary layer.* Journal of Geophysical Research **105**, 14,371–14,385

McFrederick QS, Kathilankal J & Fuentes JD (2008*). Air pollution modifies floral scent trails.* Atmospheric Environment **42**, 2336–2348

McKay WA, Stephens BA & Dollard DJ (1992) *Laboratory measurements of ozone deposition to sea water and other saline solutions.* Atmospheric Environment **26**, 3105–3110

McLinden CA, Olsen SC, Hannegan B, Wild O, Prather MJ, and Sundet J, *Stratospheric ozone in 3-D models: A simple chemistry and the cross-tropopause flux,* J. Geophys. Res.,**105**, D11, 14653–14666

Methven J *et al.* (2006). *Establishing Lagrangian connections between observations within air masses crossing the Atlantic during the ICARTT experiment.* Journal of Geophysical Research **111**, D23S62. doi:10.1029/2006JD007540

MEA (2005) *Millennium Ecosystem Assessment 2005 Ecosystems and human wellbeing: summary for decision makers*. Island Press: Washington DC

Molfino NA, Wright SC, Katz I, Tarlo S, Silverman F, McClean PA, Szalai JP, Raizenne M, Slutsky, AS, Zamel N (1991). *Effect of low concentrations of ozone on inhaled allergen responses in asthmatic subjects.* Lancet **338**, 8761, 199–203

Monks SP (2000). *A review of the observations and origins of the spring ozone maximum*. Atmospheric Environment **34**, 21, 3545–3561

Monson *et al.* (2007). *Isoprene emission from terrestrial ecosystems in response to global change: minding the gaps between models and observations*. Philosophical Transactions of the Royal Society **365**, 1856, 1677–1695

Morgan PB, Mies TA, Bollero GA, Nelson RL & Long SP (2006). *Season-long elevation of ozone concentration to projected 2050 levels under fully open-air conditions substantially decreases the growth and production of soybean.* New Phytologist **170**, 333–343

Morrissey T, Ashmore MR, Emberson LD, Cinderby S & Buker P (2007). *The impacts of ozone on nature conservation: a review and recommendations for research and policy.* Joint Nature Conservation Council: Peterborough

Mudway IS & Kelly FJ (2000). *Ozone and the lung: a sensitive issue*. Molecular Aspects of Medicine **21**, 1–2, 1–48

Mudway IS & Kelly FJ (2004). *An investigation of inhaled ozone dose and the magnitude of airway inflammation in healthy adults*. American Journal of Respiratory and Critical Care Medicine **169**, 1089–1095

Müller J-F, and Brasseur GP (1995). *IMAGES: A three-dimensional chemical transport model of the global troposphere,* J. Geophys. Res., **100**, 16,445–16,490

Murazaki K & Hess P (2006). *How does climate change contribute to surface ozone change over the U.S.?* Journal of Geophysical Research, **111**, ND5, D05301. doi:10.1029/2005JD005873

Naik V, Mauzerall D, Horowitz L, Schwarzkopf MD, Ramaswamy V & Oppenheimer M (2005). *Net radiative forcing due to changes in regional emissions of tropospheric ozone precursors.* Journal of Geophysical Research **110**, D24306. doi:10.1029/2005JD005908

Nakicenovic N, Swart R, Nakicenovic N, Alcamo J, Davis G, Vries Bd, Fenhann J, Gaffin S, Gregory K & Gruebler A (2000). *Special Report on Emissions Scenarios.* Working Group III of the Intergovernmental Panel on Climate Change (IPCC). Cambridge University Press: Cambridge, ISBN 0-521-80493-0

National Aids Trust (2006). *Fact Sheet 3: Global Statistics*. Available online at www.nat.org.uk/document/79

Nightingale PD & Liss PS (2003). *Gases in seawater.* In *The Oceans and Marine Geochemistry* (eds Holland HD & Turekian KK) 49–81. Elsevier: Oxford

Nussbaum S, Geissmann M & Fuhrer J (1995). *Ozone exposure–response relationships for mixtures of perennial ryegrass and white clover depend on ozone exposure patterns.* Atmospheric Environment **29**, 989–995

OECD (2008). *OECD Environmental Outlook to 2030.* Organisation for Economic Co-operation and Development: Paris

Olivier JGJ, Van Aardenne JA, Dentener F, Ganzeveld L & Peters JAHW (2005). *Recent trends in global greenhouse gas emissions: regional trends and spatial distribution of key sources*. In *Non-CO_2 Greenhouse Gases (NCGG-4)* (coord. van Amstel A) 325–330. Millpress: Rotterdam, ISBN 90 5966 043 9

Olsen SC, McLinden CA & Prather MJ (2001). *Stratospheric N_2O–NOy system: testing uncertainties in a three-dimensional framework*. Journal of Geophysical Research **106**, 28771–28784

Olsen MA, Schoeberl MR & Douglass AR (2004). *Stratosphere–troposphere exchange of mass and ozone*. Journal of Geophysical Research **109**, D24114. doi:10.1029/2004JD005186

Olson DM & Dinerstein E (2002). *The global 200: priority ecoregions for global conservation*. Annals of the Missouri Botanical Gardens **89**, 199–224

Ordóñez C (2006). *Trend analysis of ozone and evaluation of nitrogen dioxide satellite data in the troposphere over Europe*. PhD thesis ETH No. 16544, Swiss Federal Institute of Technology: Zurich

Osborn TJ (2004). *Simulating the winter North Atlantic Oscillation: The role of internal variability and greenhouse gas forcing.* Climate Dynamics **22**, 605–623

Owen SM, Boissard C, Hagenlocher B & Hewitt CN (1998). *Field studies of isoprene emissions from vegetation in the Northwest Mediterranean region.* Journal of Geophysical Research – Atmospheres **103**, D19, 25499–25511

Owen SM, MacKenzie AR, Bunce RGH, Stewart HE, Donovan RG, Stark G & Hewitt CN (2006). *Classifying urban land for stratified sampling and surveys, using principal component analysis with quantified uncertainties.* Landscape and Urban Planning **78**, 311–321

Pathak H *et al.* (2003). *Trends of climatic potential and on-farm yields of rice and wheat in the Indo-Gangetic Plains.* Field Crops Research **80**, 223–234

Peden DB, Setzer RW Jr & Devlin RB (1995). *Ozone exposure has both a priming effect on allergen-induced responses and an intrinsic inflammatory action in the nasal airways of perennially allergic asthmatics*. American Journal of Respiratory and Critical Care Medicine **151**, 5, 1336–1345

Pegoraro E, Rey A, Bobich EG, Barron-Gafford G, Grieve KA, Malhi Y & Murthy R (2004). *Effect of elevated CO_2 concentration and vapour pressure deficit on isoprene emission from leaves of Populus deltoides during drought.* Functional Plant Biology **31**, 12, 1137–1147

Phoenix GK, Hicks WH, Cinderby S, Kuylenstierna JCI, Stock WD, Dentener FJ, Giller KE, Austin AT, Lefroy RDB, Gimeno BS, Ashmore MR & Ineson P (2006). *Atmospheric nitrogen deposition in world biodiversity hotspots: the need for a greater global perspective in assessing N deposition impacts.* Global Change Biology **12**, 1–7

Piikki K, Vorne V, Ojanpera K *& Pleijel H* (2007). *Impact of elevated O_3 and CO_2 exposure on potato (*Solanum tuberosum *L. cv. Bintje) tuber macronutrients (*N, P, K, Mg, Ca*)*. Agriculture Ecosystems and Environment **118**, 104, 55–64

Pinto JG, Ulbrich U, Leckebusch GC, Spangehl T, Reyers M & Zacharias S (2007). *Changes in storm track and cyclone activity in three SRES ensemble experiments with the ECHAM5/MPI-OM1 GCM*. Climate Dynamics **29**, 2, 195–210

Pitari G, Mancini E, Rizi V, and Shindell DT (2002) *Impact of future climate and emissions changes on stratospheric aerosols and ozone.* J. Atmos. Sci., **59**, 414–440

Pleijel H, Danielsson H, Ojanperä K, De Temmerman L, Högy P, Badiani M & Karlsson PE (2004). *Relationships between ozone exposure and yield loss in European wheat and potato – a comparison of concentration – and flux-based exposure indices.* Atmospheric Environment **38**, 2259–2269

Pleijelm H (ed) (2007). *Transboundary Air Pollution; scientific understanding and environmental policy in Europe*. Studentlitteratur: Lund, Sweden

Pope CA, III, Burnett RT, Thun MJ, Calle EE, Krewski D, Ito K & Thurston GD (2002). *Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution.* Journal of the American Medical Association **287**, 9, 1132–1141

Possell M, Heath J, Hewitt NC, Ayres E and Kerstiens G (2004). *Interactive effects of elevated CO_2 and soil fertility on isoprene emissions from* Quercus robur. Global Change Biology **10**, 11, 1835–1843

Possell M, Hewitt C N and Beerling DJ (2005). *The effects of glacial atmospheric CO_2 concentrations and climate on isoprene emissions by vascular plants*. Global Change Biology **11**, 1, 60–69

Prather MJ (1986). *Numerical Advection by conservation of second-order moments,* J. Geophys. Res., **91**, 6671–6681

Prather MJ *et al.* (2001). *Atmospheric chemistry and greenhouse gases.* In *Climate Change 2001: The Scientific Basis*. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change (eds Houghton JT *et al.*) 239–287. Cambridge University Press: Cambridge

Prather M *et al*. (2003). *Fresh air in the 21st century*. Geophysical Research Letters **30**, 2, 72–74. doi:10.1029/2002GL016285

Price C & Rind D (1994). *Possible implications of global climate change on global lightning distributions and frequencies*. Journal of Geophysical Research **99**, 10823–10831

Pyle JA, Warwick N, Yang X, Young PJ & Zeng G (2007). *Climate/chemistry feedbacks and biogenic emissions.* Philosophical Transactions of the Royal Society A – Mathematical Physical and Engineering Sciences **365**, 1856, 1727–1740

Racherla PN & Adams PJ (2006) *Sensitivity of global tropospheric ozone and fine particulate matter concentrations to climate change.* Journal of Geophysical Research **111**, D24103. doi:10.1029/2005JD006939

Ramankutty N & Foley JA (1999). *Estimating historical changes in global land cover: Croplands from 1700 to 1992.* Global Biogeochemical Cycles **13**, 997–1027

Raupach MR, Marland G, Ciais P, Quéré CL, Canadell JG, Klepper G & Field CB (2007). *Global and regional drivers of accelerating CO_2 emissions*. Proceedings of the National Academy of Sciences **104**, 10288–10293. doi:10.1073/pnas.0700609104

Read KA *et al*. (2008). *Extensive halogen-mediated ozone destruction over the tropical Atlantic Ocean*. Nature **453**, 1232–1235

RETRO (2007). *REanalysis of the TROpospheric chemical composition over the past 40 years*. A long term global modelling study of tropospheric chemistry funded under the 5th EU Framework programme. EU Contract No. EVK2-CT-2002-00170 (2007) Emissions data sets and methodologies for estimating emissions. Max Planck Institute for Meteorology: Hamburg, Germany

Riahi K, Grübler A & Nakicenovic N (2006). *Scenarios of long-term socio-economic and environmental development under climate stabilization*. Technological Forecasting and Social Change **74**, 7, 887–935

Rinnan R, Impio M, Silvola J, Holopainen T & Martikainen PJ (2003). *Carbon dioxide and methane fluxes in boreal peatland microcosms with different vegetation cover – effects of ozone or ultraviolet-B exposure*. Oecologia **137**, 475–483

Rokityanskiy D, Benitez P, Kraxner F, McCallum I, Obersteiner M, Rametsteiner E & Yamagata Y (2007). *Geographically explicit global modelling of land-use change, carbon sequestration, and biomass supply*. Technological Forecasting and Social Change **74**, 7, 1057–1082

Roux E, Hyvelin JM, Savineau JP & Marthan R (1999). *Human isolated airway contraction: interaction between air pollutants and passive sensitization.* American Journal of Respiratory and Critical Care Medicine **160**, 2, 439–445

Royal Society (2008) *Sustainable biofuels: prospects and challenges.* Policy Document 01/08. The Royal Society: London

Russell GL and Lerner JA (1981). *A new finite-differencing scheme for the tracer transport equation.* J. Applied Meteorology, **20**, 1483–1498

Saiz-Lopez A, Mahajan AS, Salmon RA, Bauguitte SJ-B, Jones AE, Roscoe HK & Plane JMC (2007). *Boundary layer halogens in coastal Antarctica*. Science **317**, 348–351

Sanderson MG, Jones CD, Collins WJ, Johnson CE & Derwent RG (2003) *Effect of climate change on isoprene emissions and surface ozone levels.* Geophysical Research Letters **30**, 1936. doi:10.1029/2003GL017642

Scannell C, Chen L, Aris RM, Tager I, Christian D, Ferrando R, Welch B, Kelly T & Balmes JR (1996). *Greater ozone-induced inflammatory responses in subjects with asthma*. American Journal of Respiratory and Critical Care Medicine **154**, 1, 24–29

Schär C & Jendritzky G (2004). *Hot news from Summer 2003. The European heatwave of 2003: was it merely a rare meterological event or a first glimpse of climate change to come? Probably both, is the answer, and the anthropogenic contribution can be quantified.* Nature **432**, 559–560

Schär C, Vidale PL, Lüthi D, Frei C, Häberli C, Liniger M & Appenzeller C (2004). *The role of increasing temperature variability for European summer heat waves.* Nature **427**, 332–336

Schopp W, Amann M, Cofala J, Heyes C & Klimont Z (1999). *Integrated assessment of European air pollution emission control strategies*. Environmental Modelling and Software **14**, 1, 1–9

Schumann U, Schlager H, Arnold F, Ovarlez J, Kelder H, Hov O, Hayman G, Isaksen ISA, Staehelin J & Whitefield PD (2000). *Pollution from aircraft emissions in the North Atlantic flight corridor: overview on the POLINAT projects*. Journal of Geophysical Research – Atmospheres **105**, D3, 3605–3631

Schumann U & Huntrieser H (2007). *The global lightning-induced nitrogen oxides source.* Atmospheric Chemistry and Physics Discussions **7**, 2623–2818

Seneviratne SI, Koster RD, Guo ZC, Dirmeyer PA, Kowalczyk E, Lawrence D, Liu P, Lu C-H, Mocko D, Oleson KW & Verseghy D (2006). *Soil moisture memory in AGCM simulations: Analysis of global land–atmosphere coupling experiment (GLACE) data.* Journal of Hydrometeorology **7**, 5, 1090–1112

Sharkey TD & Loreto F (1993). *Water stress, temperature, and light effects on isoprene emission and photosynthesis of Kudzu leaves.* Plant Physiology **102**, 1, 159–159

Shindell DT *et al*. (2006). *Multimodel simulations of carbon monoxide: comparison with observations and projected near-future changes*. Journal of Geophysical Research **111**, D19306. doi:10.1029/2006JD007100

Simmonds PG, Derwent RG, Manning AL & Spain G (2004). *Significant growth in surface ozone at Mace Head, Ireland 1987–2003*. Atmospheric Environment **38**, 4769–4778

Simpson D, Emberson L, Ashmore M & Tuovinen J (2007). *A comparison of two different approaches for mapping potential ozone damage to vegetation. A model study.* Environmental Pollution **146**, 715–725

Sitch S, Cox PM, Collins WJ & Huntingford C (2007). *Indirect radiative forcing of climate change through ozone effects on the land-carbon sink.* Nature **448**, 791–794

Smil V. (2003). *Long-term trends and achievements*. In *Energy at the Crossroads. Global perspectives and uncertainties*, chapter 1. MIT Press: Cambridge, MA

Solberg S, Derwent RG, Hov O *et al.* (2005). *European abatement of surface ozone in a global perspective.* Ambio **34**, 1, 47–53

Solberg S, Hov Ø, Søvde A, Isaksen ISA, Coddeville P, De Backer H, Forster C, Oorsolini Y and Uhse K (2008). *European surface ozone in the extreme summer 2003.* Journal of Geophysical Research **113**, D07307. doi:10.1029/2007JD009098

Staehelin J & Schnadt Poberaj C (2008*). Long-term tropospheric ozone trends: a critical review.* p271–282 from: Climate Variability and Extremes during the past 100 years. Series: Advances in Global Change Research, Volume **33**. 10.1007/978-1-4020-6766-2_18; 271–282. Brönnimann S, Luterbacher J, Ewen T, Diaz HF, Stolarski RS, Neu U (Eds.). Springer: Heidelberg, Germany

Steiner A, Luo C, Huang Y & Chameides WL (2002). *Past and present-day biogenic volatile organic compound emissions in East Asia*. Atmospheric Environment **36**, 31, 4895–4905

Stevenson DS, Doherty RM, Sanderson MG, Collins WJ, Johnson CE & Derwent RG (2004). *Radiative forcing from aircraft NO_X emissions: mechanisms and seasonal dependence*. Journal of Geophysical Research **109**, D17307. doi:10.1029/2004JD004759

Stevenson DS, Doherty RM, Sanderson MG, Johnson CE, Collins WJ & Derwent RG (2005). *Impacts of climate change and variability on tropospheric ozone and its precursors*. Faraday Discussions **130**. doi:10.1039/b417412g

Stevenson DS *et al.* (2006). *Multimodel ensemble simulations of present-day and near-future tropospheric ozone*. Journal of Geophysical Research **111**, D08301. doi:10.1029/2005JD006338

Stewart HE, Hewitt CN, Bunce RGH, Steinbrecher R, Smiatek G & Schoenemeyer T (2003). *A highly spatially and temporally resolved inventory for biogenic isoprene and monoterpene emissions: model description and application to Great Britain.* Journal of Geophysical Research – Atmospheres **108**, D20, 22

Sun J, Koto H & Chung KF (1997). *Interaction of ozone and allergen challenges on bronchial responsiveness and inflammation in sensitised guinea pigs*. International Archives of Allergy and Immunology **112**, 2, 191–195

Sutton RT and Hodson DLR (2005) *Atlantic Ocean Forcing of North American and European Summer Climate.* Science **309**, 115–118. doi: 10.1126/science.1109496

Szopa S, Hauglustaine DA, Vautard R & Menut L (2006). *Future global tropospheric ozone changes and impact on European air quality*. Geophysical Research Letters **33**, L14805. doi:10.1029/2006GL025860

Tagaris E, Manomaiphiboon K, Liao KJ, Leung LR, Woo J-H, He S, Amar P & Russell AG (2007). *Impacts of global climate change and emissions on regional ozone and fine particulate matter concentrations over the United States.* Journal of Geophysical Research **112**, D14312. doi:10.1029/2006JD008262

Tarrason L, Jonson JE, Bernsten TK & Rypdal K (2004). *Study on air quality impacts of non-LTO emissions fro m aviation.* Final report to the European Commission under contract B4-3040/2002/343093/MAC/C1 Available online at www.europa.nl/environment/air/pdf/air_quality_impacts_finalreport.pdf

TFHTAP (2007). *Task Force on Hemispheric Transport of Air Pollution 2007 Interim Report* Available online at www.htap.org/activities/2007_Interim_Report.htm

Tibaldi S & Molteni F (1990). *On the operational predictability of blocking*. Tellus **42A**, 343–365

Tiedtke M (1989). *A comprehensive mass flux scheme for cumulus parameterization in large-scale models.* Mon. Wea. Rev., **117**, 1779–1800

Tingey DT, Manning M, Grothaus LC & Burns WF (1979). *Influence of light and temperature on isoprene emission rates from live oak.* Physiologia Plantarum **47**,112–118

Tingey DT, Evans R & Gumpertz M (1981). *Effects of environmental conditions on isoprene emission from live oak.* Planta **152**, 6, 565–570

Toumi R, Haigh JD & Law KS (1996). *A tropospheric ozone-lightning climate feedback.* Geophysical Research Letters **23**, 1037–1040

Turner AG, Inness PM & Slingo JM (2007a). *The effect of model basic state biases on the monsoon–ENSO system. II: Changing ENSO regimes.* Quarterly Journal of the Royal Meteorological Society **133**, 1159–1173. doi:10.1002/qj.83

Turner AG, Inness PM & Slingo JM (2007b). *The effect of doubled CO_2 and model basic state biases on the monsoon–ENSO system. I: Mean response and interannual variability.* Quarterly Journal of the Royal Meteorological Society **133**, 1143–1157. doi:10.1002/qj.82

Turner AG & Slingo JM (2008). *Subseasonal extremes of precipitation and active-break cycles of the Indian summer monsoon in a climate change scenario*. Submitted to Quarterly Journal of the Royal Meteorological Society

Tyrlis E & Hoskins BJ (2008). *Aspects of Northern Hemisphere atmospheric blocking climatology.* Journal of Atmospheric Science, **65**, 1653–1665

United Nations (2001). *World Population Monitoring 2001. Population, environment and development.* Department of Economic and Society Affairs Population Division. United Nations: New York

UNECE (2007). *Review of the 1999 Gothenburg Protocol*. Report by the Secretariat. UNECE Executive Body for the Convention on Long-Range Transboundary Air Pollution. ECE/EB.AIR/2007/10

UNFPA (2007). *State of the world population 2007 unleashing the potential of urban growth*. United Nations Population Fund (UNFPA)

US EPA (2002). *Latest findings on National Air Quality. 2001 Status and Trends*. US EPA. Document EPA 454/K-02-001 Available online at www.epa.gov/air/airtrends/aqtrnd01/summary.pdf

US EPA (2005). *Global Anthropogenic Non-CO_2 Greenhouse Gas Emissions: 1990–2020.* Office of Atmospheric Programs, Washington, DC

US EPA (2006). *Air Quality Criteria for Ozone and Related Photochemical Oxidants* (Final). EPA/600/R-05/004aF-cF. US Environmental Protection Agency: Washington, DC Available online at http://cfpub epa gov/ncea/cfm/recordisplay cfm?deid=149923

US NAS (in press). *Risk assessment of acute effects of ozone on mortality*. US National Academy of Science: Washington, DC Available online at http://nationalacademies.org/

Valkama E, Koricheva J & Oksanen E (2007). *Effects of elevated O_3, alone, and in combination with elevated CO_2, on tree leaf chemistry and insect herbivore performance: a meta-analysis.* Global Change Biology **13**, 184–201

van Amstel A (Ed) (2005). *Proceedings of the Fourth International Symposium on Non-CO_2 Greenhouse Gases: Science, Control, Policy and Implementation (NCGG-4)*. Mill Press: Rotterdam

van der Werf GR, Randerson TJ, Collatz J, and Giglio L (2003). *Carbon emissions from fires in tropical and subtropical ecosystems,* Global Change Biol., ***9***, 547–562

Van Dingenen *et al.* (in press). *The global impact of ozone on agricultural crop yields under current and future air quality legislation.* Submitted to Atmospheric Environment

Van Loon *et al.* (2006). *Evaluation of long-term ozone simulations from seven regional air quality models and their ensemble average*. Atmospheric Environment **41**, 10, 2083–2097

van Noije TPC, Eskes HJ, van Weele M, and van Velthoven PFJ (2004). *Implications of the enhanced Brewer-Dobson circulation in European Centre for Medium-Range Weather Forecasts reanalysis ERA-40 for the stratosphere-troposphere exchange of ozone in global chemistry-transport models,* J. Geophys. Res., **109**, D19308. doi:10.1029/2004JD004586

van Noije TPC *et al.* (2006). *Multi-model ensemble simulations of tropospheric NO_2 compared with GOME retrievals for the year 2000*. Atmospheric Chemistry and Physics **6**, 2943–2979

Vautard R *et al.* (2006) *Is regional air quality model diversity representative of uncertainty for ozone simulation?* Geosphysical Research Letters **33**, L24818. doi:1029/2006GL27610

Vautard R, Yiou P, D'Andrea F, de Noblet N, Viovy N, Cassou C, Polcher J, Ciais P, Kageyama M & Fan Y (2007). *Summertime European heat and drought waves induced by wintertime Mediterranean rainfall deficit.* Geophysical Research Letters **34**, L07711. doi:10.1029/2006GL028001

Velikova V, Tsonev T, Pinelli P, Alessio GA & Loreto F (2005). *Localized ozone fumigation system for studying ozone effects on photosynthesis, respiration, electron transport rate and*

isoprene emission in field-grown Mediterranean oak species. Tree Physiology **25**, 12, 1523–1532

Volk M, Bungener P, Contat F, Myrta M & Jurg F (2005). *Grassland yield declined by a quarter in 5 years of free-air ozone fumigation*. Global Change Biology **12**, 74–83. doi: 10.1111/j.1365-2486.2005.01083.x

Volz A & Kley D (1988). *Evaluation of the Montsouris series of ozone measurements made in the 19th century*. Nature **332**, 240–242

von Glasow R, von Kuhlmann R, Lawrence MG, Platt U & Crutzen PJ (2004). *Impact of reactive bromine chemistry in the troposphere.* Atmospheric Chemistry and Physics **4**, 2481–2497

Wang X & Mauzerall DL (2004). *Characterising distributions of surface ozone and its impacts on grain production in China, Japan and South Korea.* Atmospheric Environment **38**, 4383–4402

Wayne RP (2000). *Chemistry of Atmospheres,* 3rd edn. Oxford University Press: Oxford

West JJ & Fiore AM (2005). *Management of tropospheric ozone by reducing methane emissions*. Environmental Science and Technology **39**, 4685–4691

West JJ, Fiore AM, Naik V, Horowitz LW, Schwarzkopf MD and Mauzerall DL (2007). *Ozone air quality and radiative forcing consequences of changes in ozone precursor emissions.* Geophysical Research Letters **34**, L06806

WHO (2003). *Health aspects of air pollution with particulate matter, ozone and nitrogen dioxide.* Report on a WHO working group. EUR/03/5042688. WHO: Bonn

WHO (2005). *Effects of air pollution on children's health and development – a review of the evidence.* European Centre for Environment and Health. WHO Regional Office for Europe: Bonn

WHO (2006). *Air quality guidelines: global update 2005, particulate matter, ozone, nitrogen dioxide and sulphur dioxide*. WHO Regional Office for Europe: Copenhagen

WHO (2008). *Global tuberculosis control – surveillance, planning, financing.* WHO report 2008

Wieser G & Matyssek R (2007). *Linking ozone uptake and defense towards mechanistic risk assessment for forest trees*. New Phytologist **174**, 7–9

Wild O & Prather MJ (2000). *Excitation of the primary tropospheric chemical mode in a global 3-D model,* J. Geophys. Res., **105**, 24,647–24,660

Wild O, Prather MJ & Akimoto H (2001). *Indirect long-term global radiative cooling from NO_X emissions.* Geophysical Research Letters **28**, 1719–1722

Wild O, Sundet JK, Prather MJ, Isaksen ISA, Akimoto H, Browell EV, and Oltmans SJ (2003). *CTM Ozone Simulations for Spring 2001 over the Western Pacific: Comparisons with TRACE-P Lidar, ozonesondes and TOMS columns.* J. Geophys. Res., **108** (D21), 8826. doi:10.1029/2002JD003283

Wild O (2007). *Modelling the global tropospheric ozone budget: exploring the variability in current models.* Atmospheric Chemistry and Physics **7**, 2643–2660

Wilkinson MJ, Owen SM, Possell M, Hartwell J, Gould P, Hall A, Vickers C & Hewitt CN (2006). *Circadian control of isoprene emissions from oil palm (*Elaeis guineensis*).* Plant Journal **47**, 6, 960–968

Williams ML (2007). *UK air quality in 2050 – synergies with climate change policies.* Environmental Science Policy **10**, 2, 169–175

Wittig VE, Ainsworth EA & Long SP (2007). *To what extent do current and projected increases in surface ozone affect photosynthesis and stomatal conductance of trees? A meta-analytic review of the last 3 decades of experiments.* Plant Cell and Environment **30**, 1150–1162

Woodward FI & Lomas MR (2004). *Vegetation dynamics – simulating responses to climatic change*. Biological Reviews **79**, 643–670

Woodward FI, Smith TM & Emanuel WR (1995). *A global land primary productivity and phytogeography model*. Global Biogeochemical Cycles **9**, 4, 471–490

Wu S, Mickley LJ, Jacob DJ, Logan JA, Yantosca RM & Rind D (2007a). *Why are there large differences between models in global budgets of tropospheric ozone?* Journal of Geophysical Research **112**, D05302. doi:10.1029/2006JD007801

Wu S, Mickley LJ, Eric M, Leibensperger, Jacob DJ, Rind D & Streets DG (2007b). *Effects of 2000–2050 global change on ozone air quality in the United States*. Submitted to Journal of Geophysical Research

Yamaji K, Ohara T, Uno I, Tanimoto H, Kurokawa J & Akimoto H (2006). *Analysis of the seasonal variation of ozone in the boundary layer in East Asia using the Community Multi-scale Air Quality model: what controls surface ozone levels over Japan?* Atmospheric Environment **40**, 1856–1868. doi:10.1016/j.atmosenv.2005.10.067

Yang X, Cox RA, Warwick NJ, Pyle JA, Carver GD, O'Connor FM & Savage NH (2005). *Tropospheric bromine chemistry and its impacts on ozone: a model study.* Journal of Geophysical Research **110**, D23311

Yienger JJ & Levy II H (1995). *Empirical model of global soil-biogenic NO_X emissions.* Journal of Geophysical Research **100**, D6, 11447–11464

Zeng G & Pyle JA (2003). *Changes in tropospheric ozone between* 2000 *and 2100 modelled in a chemistry–climate model*. Geophysical Research Letters **30**, 7, 1392. doi:10.1029/2002GL016708

Zeng G & Pyle JA (2005). *Influence of El Niño Southern Oscillation on stratosphere/troposphere exchange and the global tropospheric ozone budget*. Geophysical Research Letters **32**, L01814

Annex 1 Glossary and abbreviations

Abscission	The natural detachment of leaves, branches, flowers, etc
ACCENT	Atmospheric Composition Change European Network of Excellence
AMO	Atlantic Multi-decadel Oscillation
Anti-cyclonic	Having a sense of rotation about the local vertical opposite to that of the Earth's rotation; that is, clockwise in the Northern Hemisphere, counter-clockwise in the Southern Hemisphere, undefined at the equator. Anticyclone – a high pressure system in which air descends to give calm conditions and clear skies. Associated with summer heat waves and winter frosts and fog.
AOT40	Accumulated exposure over a threshold of 40 ppb during daylight hours for a relevant period
AOT60	Accumulated exposure over a threshold of 60 ppb during daylight hours for a relevant period
AQEG	The UK's Air Quality Expert Group
Atmospheric blocking	Atmospheric blocking events are sustained, vertically coherent, and quasi-stationary synoptic-scale high-pressure systems whose amplitude is such that they disrupt the prevailing westerly circumpolar flow (eg Barriopedro & Garcia-Herrera 2006). The normal eastward progression of synoptic disturbances is obstructed leading to episodes of prolonged extreme weather conditions, accompanied by significant temperature and precipitation anomalies
Background O_3 (hemispheric)	Hemispheric background O_3 is the remaining concentration once the emissions of anthropogenic O_3 precursors from within a region are switched off. Hemispheric background O_3 comprises O_3 produced from natural sources of O_3 precursors within a region, together with O_3 imported into the region derived from all sources (anthropogenic and natural, including the stratosphere). The value varies seasonally and with latitude
Baseline O_3	Baseline O_3 is the average measured concentration of O_3 within a region and is made up of both the anthropogenic emissions produced within a region and the background concentration of O_3
Biogenic	Produced by the actions of living organisms
Boundary layer	The bottom layer of the troposphere that is in contact with the surface of the Earth
Br	Chemical symbol for bromine
Brewer–Dobson circulation	A slow vertically overturning movement of air, rising at the equator and descending over the pole of the winter hemisphere
bVOC	Biogenic Volatile Organic Compounds
°C	Degrees Celsius
CAFE Strategy	EU Clean Air for Europe Strategy
C_5H_8: 2-methyl-1, 3-butadiene	Chemical symbol for isoprene
CH_4	Chemical symbol for methane
CH_3OOH	Chemical symbol for methyl hydroperoxide
CH_3O_2	Chemical symbol for methyl peroxide radical
CLRTAP	Convention on Long-range Transboundary Air Pollution
CO_2	Chemical symbol for carbon dioxide
CO	Chemical symbol for carbon monoxide
Convection	In general, mass motions within a fluid resulting in transport and mixing of the properties of that fluid
Cumulus clouds	A principal cloud type (cloud genus) in the form of individual, detached elements that are generally dense and possess sharp non-fibrous outlines. These elements develop vertically, appearing as rising mounds, domes, or towers, the upper parts of which often resemble a cauliflower

Cyclonic events	Having a sense of rotation about the local vertical the same as that of the Earth's rotation: that is, as viewed from above, counterclockwise in the Northern Hemisphere, clockwise in the Southern Hemisphere, undefined at the equator; the opposite of anticyclonic
Deposition	Processes by which trace gases or particles are transferred from the atmosphere to the surface of the Earth
Dry deposition	Dry deposition is the direct deposition of gases or particles to terrestrial or marine surfaces
EMEP	European Cooperative Programme for Monitoring and Evaluation of Long-range transmission of air pollutants in Europe
Emission scenario	A plausible and often simplified description of how the future may develop based on a coherent and internally consistent set of assumptions about driving forces and key relationships. Scenarios may be derived from projections, but are often based on additional information from other sources, sometimes combined with a narrative storyline
ENSO	The El Nino-Southern Oscillation. El Nino is a warm water current which periodically flows along the coast of Ecuador and Peru. This oceanic event is associated with a fluctuation of the inter-tropical surface pressure pattern and circulation in the Indian and Pacific oceans, called the Southern Oscillation. This coupled atmospheric–ocean phenomenon is collectively known as the ENSO. During an El Nino event, the prevailing trade winds weaken and the equatorial counter current strengthens, causing warm surface waters in the Indonesian area to flow eastwards to overlie the cold waters of the Peru current. This event has great impacts on the wind, sea surface temperature and precipitation patterns in the tropical Pacific. It has climatic effects throughout the Pacific region and in many other parts of the world. The opposite of an El Nino event is called La Nina.
Epidemiology	The study of the incidence and distribution of diseases, and of their control and prevention
Episodic (or peak) O_3	Peak O_3 concentrations or episodes occur when high concentrations of O_3 precursor emissions coincide with appropriate meteorological conditions and produce large surface O_3 concentrations (often in excess of 100 ppb). The number of O_3 episodes occurring each year depends strongly on the weather
Flux	The rate of emission, absorption, transfer or deposition of a substance or energy from one reservoir to another. Often expressed as the mass or energy per unit area and per unit time
Frontal systems	Front: In meteorology, generally, the interface or transition zone between two air masses of different density
Genotype	The genetic constitution (the genome) of an individual
$h\nu$	Scientific notation for electromagnetic radiation (light), generally at a specific wavelength and specifically a photon
Halogen	Halogen elements are a series of non-metal elements comprising fluorine, F; chlorine, Cl; bromine, Br; iodine, I; and astatine At
HCHO	Chemical symbol for formaldehyde
HNO_3	Chemical symbol for nitric acid
HO_2	Chemical symbol for peroxy radical
H_2O	Chemical symbol for water
H_2O_2	Chemical symbol for hydrogen peroxide
ICAO	International Civil Aviation Organization
IMO	International Maritime Organization
IPCC	Intergovernmental Panel on Climate Change
IPPC Directive	Integrated Pollution, Prevention and Control Directive
K	Kelvin
LCP Directive	Large Combustion Plant Directive

M	Chemical symbol for an inert molecule
µg	Microgram; one millionth of a gram
µg/m^3	Micrograms per cubic metre of air. A unit for describing the concentration of air pollutants in the atmosphere, as a mass of pollutant per unit volume of air
mg m^{-2} h^{-1}	Milligrams per metre squared per hour
mm s^{-1}	Deposition rate – vertical flux/ambient concentration, with dimensions of a velocity
Mixing ratios	In a given volume of air, the ratio of the number of molecules of a particular trace gas to the number of molecules of all the other gases present
Mycorrhiza	A symbiotic association of a fungus and the roots of a plant
NAO	North Atlantic Oscillation
N_2	Chemical symbol for nitrogen gas
NEC Directive	National Emissions Ceiling Directive
NH_3	Chemical symbol for ammonia
nm	Nanometre 10^{-9} metres
nmVOC	Non-methane volatile organic compounds
NO	Chemical symbol for nitric oxide
NO_2	Chemical symbol for nitrogen dioxide
NO_X	NO_X is a generic term for mono-nitrogen oxides (NO and NO_2)
NO_X titration effect	Where high concentrations of NO locally scavenge O_3, a process leading to the formation of NO_2. Close to the sources this titration process can be considered as an O_3 sink. In addition, high NO_2 concentrations deflect the initial oxidation step of VOC by forming other products (eg nitric acid), which prevents the net formation of O_3. Because of these reactions, a decrease in NO_X can lead to an increase in O_3 at low VOC/NO_X ratios, as is the case in cities
NO_y	NO_y (reactive odd nitrogen) is defined as the sum of NO_X plus the compounds produced from the oxidation of NO_X which include nitric acid and peroxyacetyl nitrate
O_2	Chemical symbol for oxygen gas
O_3	Chemical symbol for ozone
O_3 precursor gas	Gases which lead to the formation of O_3 in the presence of sunlight: carbon monoxide (CO), volatile organic compounds (VOC) (including CH_4), and oxides of nitrogen (NO_X)
O(^{1}D)	Chemical symbol for electronically excited oxygen atoms
OH	Chemical symbol for the hydroxyl radical
Oxidant	A species having a greater oxidation potential than does molecular oxygen. Also a species that readily accepts electrons. Also a collective term for NO_2 and O_3
Oxidant partitioning	The partitioning of oxidant into O_3 and NO_2 as a function of NO_X
Oxidative damage	The extraction of electrons from target molecules which leads to a change in their chemical, physical and functional characteristics
PAR	Photosynthetically-Active Radiation
Peak O_3	See episodic O_3
Photolysis	The decomposition of molecules following the absorption of light
Photosynthesis	The process in which the energy of sunlight is used by green plants to synthesise carbohydrates from carbon dioxide and water
PM	Particulate Matter
ppb or ppbv	Parts per billion by volume. A unit commonly used to quantify trace gases in the atmosphere

ppm	Parts per million. The concentration of a pollutant in air in terms of molar ratio. A concentration of 1 ppm means that for every million (10^6) molecules in a volume of air, there is one molecule of the specified pollutant present. For practical purposes in ambient air, the molar ratio and volume ration (the volume occupied by the pollutant gas within a given volume of air) are identical
ppt	Parts per trillion (parts in 10^{12} by volume)
Radiative forcing	Radiative forcing is the change in the net, downward minus upward, irradiance (expressed in W m^{-2}) at the tropopause due to a change in an external driver of climate change, such as, for example, a change in the concentration of carbon dioxide or the output of the sun
Rhinitis	An inflammation of the mucous membrane of the nose
Scavenging	Removal of pollutants from the air
Sequestration	The process of increasing the carbon content of a carbon reservoir other than the atmosphere. Biological approaches to sequestration include direct removal of carbon dioxide from the atmosphere through land-use change, planting forests, reforestation, and practices that enhance soil carbon in agriculture. Physical approaches include separation and disposal of carbon dioxide from flue gases or from processing fossil fuels to produce hydrogen (H_2) and carbon dioxide-rich fractions and long-term storage underground in depleted oil and gas reservoirs, coal seams and saline aquifers
Sink	Any process, activity or mechanism which removes a pollutant or precursor gas from the atmosphere
SO_2	Chemical symbol for sulphur dioxide
Source	Any process, activity or mechanism which releases a pollutant or precursor gas to the atmosphere
SRES Scenarios	Special Report on Emissions Scenarios (SRES). Emission scenarios developed by the IPCC
STE	Stratosphere–Troposphere Exchange
Stomata	Tiny pores on the surface of plant leaves that can open and close to take in and give out water vapour and other gases such as CO_2 and O_2
Stratosphere	The highly stratified region of the atmosphere above the troposphere extending from about 20 km (ranging from 9 km in high latitudes to 16 km in the tropics on average) to about 50 km
Synoptic	Weather systems with scales of 100s to 1000s km typical of mid-latitude cyclones and anticyclones
T	Temperature
Tg	Teragram. 10^{12} grams. Equal to 1 million metric tons
Tg y^{-1}	Teragrams per year
Trace gas	Refers to a gas or gases which make up less than 1% by volume of the Earth's atmosphere, and therefore includes all gases except nitrogen (78.1%) and oxygen (20.9%)
Troposphere	The lowest part of the atmosphere from the surface to about 10 km in altitude in mid-latitudes (ranging from 9 km in high latitudes to 16 km in the tropics on average) where clouds and 'weather' phenomena occur. In the troposphere temperatures generally decrease with height
Tropopause	The boundary between the troposphere and the stratosphere
UNCLOS	United Nations Convention on the Law of the Sea
UNECE	United Nations Economic Commission for Europe
Urban decrement or urban deficit	Equivalent to NO_X titration, see above
UV light	Ultraviolet light
Vg	Symbol for deposition velocity
VOC	Volatile Organic Compounds
WHO	World Health Organization
X	Symbol to denote halogen
XO	Halogen oxide radical

Source: AQEG (2008), EEA (1998), Glickman (2000), Graedel and Crutzen (1993), IPCC (2007), TFHTAP (2007).

Annex 2 Call for evidence response and workshop attendees

To inform the study an open call for evidence was issued in January 2007. This was followed by a workshop for experts to discuss modelling and atmospheric chemistry and the impacts of O_3 on human health and the environment. One oral evidence session was also held. We are grateful to those who responded to this call and to those who provided additional information at the request of the working group.

Written evidence

Camilla Anderson, Magnuz Enghardt, Valentin Foltescu, Joahim Langner	Swedish Meteorological and Hydrological Institute (SMHI) and Stockholm University
Dr Almut Arneth	Lund University
Nicole Augustin	University of Bath
Professor John Ayres	Liberty Safe Work Research Centre
Simon Bareham	Joint Nature Conservancy Committee Air Pollution Network
Professor Michelle Bell	Yale University
Simon Birkett	Campaign for Clean Air in London
Dr Mark Broadmeadow	Forest Research and Forestry Commission (England)
Dr Keith Bull	UNECE
Dr Helen Clayton	Research Councils UK
Dr P Cristofanelli	Institute for Atmospheric Sciences and Climate – National Research Council (ISAC–CNR)
Dr Lisa Emberson, E Arone, P Buker, G Krugr, A Kaoya, AB Mashingaidze, V Sghitumbanuma, AM van Tienhoven, M Zunckel	Air Pollution Crop Effect Network
Dr F Fehsenfeld	National Oceanic and Atospheric Admninistration (NOAA), USA
Dr Marco Ferretti	Universita degli Studi di Siena
Professor Jurg Fuhrer	Agroscope Reckenholz-Taebikon ART
Frans Fierens and Gerwin Dumont	Belgian Interregional Environment Agency – Flemish Environment Agency (IRCEL–VMM)
Zita Ferenczi	Hungarian Meteorological Service
Dr Laurens Ganzeveld	Wageningen University
Dr Jean-Pierre Garrec	Laboratoire Pollution Atmosphérique, INRA – Centre de Recherche de Nancy
V Gerasopoulos, P Zanis and C Zerefos	National Observatory of Athens and Academy of Athens
Phil Gervat	ARMO project
Dr Rasa Girgzdiene	Institute of Physics, Lithuania
Alun Hodgson	Alun Hodgson Consulting
Professor Stephen Holgate	Southampton General Hospital
Per Erik Karlsson and Anne Lindskog, Håkan Pleijel and Jenny Sundberg, Lin Tan, Deliang Chen and Kristin Piikki	Swedish Environmental Research Institute and Gothenburg University
Professor David Karnosky	Michigan Technological University

Professor Andrew Leakey	University of Illinois
Dr Alistair Lewis, Dr L Carpenter, Dr L Lee	University of York
Dr AR MacKenzie	Lancaster University
Professor R Matyssek	Technische Universität München
Dr J McQuaid	University of Leeds
Dr Gina Mills, Dr Harry Harmens and Felicity Hayes	Centre for Ecology and Hydrology
Janice Milne	Scottish Environment Protection Agency (SEPA)
Dr Paul Monks	University of Leicester
Dr Elena Paoletti	Istituto Protezione Piante
Professor Stuart Penkett	University of East Anglia
Professor J Penuelas and Dr A Ribas	Centre de Recerca Ecologica I Aplicacions Forestals (CREAF)
Andre Prevot	Paul Scherrer Institute
Motoshi Hiratsuka	EANET (Acid Deposition Monitoring Centre in East Asia)
Dr Michael Raupach	CSIRO
Dr Riikka Rinnan	Lund University
Michael G. Sanderson, Stephen Sitch, David Thomson, William J Collins, Alison Redington and Olivier Boucher	The Met Office
Martin Shultz	Institute für Chemie und Dynamik der Geosphäre (ICG), Germany
Dr Stephen Simukanga	Air Pollution Information Network for Africa (APINA)
David Simpson	Norwegian Institute of Meteorology
Dr Cristoph Walter	Unilever
Asst Professor Jason West	University of North Carolina
Dr Oliver Wild	Cambridge University
Martin Williams	DEFRA
Paul Young	University of Cambridge

Oral evidence

Dr Keith Bull	UNECE

Workshop attendees

Professor Madhoolika Agrawal	Banaras Hindu University, India
Dr Markus Amann	IIASA, Austria
Professor Ross Anderson FMedSci	St George's Hospital University of London, UK
Professor Mike Ashmore	University of York, UK
Professor Jonathon Ayres	University of Aberdeen, UK
Professor Jeremy Barnes	University of Newcastle, UK
Dr Mark Broadmeadow	Forest Research, UK
Dr Simon Brown	UK Meterological Office, Hadley Centre

Professor Peter Cox	University of Exeter, UK
Dr Mhairi Coyle	Centre for Ecology and Hydrology, UK
Professor Michael Depledge	Peninsula Medical School, UK
Professor Dick Derwent OBE	rdscientific, UK
Dr Lisa Emberson	University of York, Stockholm Environment Institute, UK
Professor David Fowler CBE FRS	Centre for Ecology and Hydrology, UK
Professor Peringe Grennfelt	IVL Swedish Environmental Research Institute, Sweden
Professor Nick Hewitt	University of Lancaster, UK
Dr Mike Jenkin	Imperial College, London, UK
Professor David Karnosky	Michigan Technological University, USA
Professor Frank Kelly	Kings College, London, UK
Dr Andrew Leakey	University of Illinois, USA
Professor Peter Liss CBE FRS	University of East Anglia, UK
Dr Gina Mills	Centre for Ecology and Hydrology, UK
Dr Paul Monks	Leicester University, UK
Professor Mike Pilling CBE	University of Leeds, UK
Professor John Pyle FRS	University of Cambridge, UK
Dr Mike Sanderson	UK Meteorological Office
Dr Martin Schultz	Institut für Chemie und Dynamik der Geosphäre (ICG), Germany
Dr David Simpson	DNMI Norwegian Meteorological Institute, Norway
Dr Stephen Sitch	UK Meteorological Office, Hadley Centre
Dr John Stedman	AEA Technology, UK
Dr David Stevenson	University of Edinburgh, UK
Dr Rita van Dingenen	European Commission Joint Research Centre, Italy
Dr Twan Van Noije	Royal Netherlands Meteorological Institute (KNMI)
Dr Heather Walton	Health Protection Agency, UK
Dr Clare Whitfield	Joint Nature Conservation Committee, UK
Dr Oliver Wild	University of Cambridge, UK
Dr Martin Williams	UK Department for the Environment, Food and Rural Affairs
Dr Paul Young	University of Cambridge, UK
Dr Guang Zeng	University of Cambridge, UK

Annex 3 Specifications for the Royal Society 2050 chemistry–climate simulations

Background

As part of the Royal Society's study into ground-level O_3 in the 21st century, new simulations were performed to complement the ACCENT intercomparison study (Dentener *et al.* 2006b), specifically to look slightly further ahead than the 2030 timeframe, and to focus on possible interactions between climate change and O_3. Compared to the ACCENT 2030 study, the applied level of climate change in 2050 will be larger and produce a clearer signal. In addition to this focus, new global emissions of O_3 precursors are also available for 2050 (see chapter 4).

Emissions

IIASA provided ozone precursor (NO_X, CO) emissions for 11 world regions for present-day (2000) and projections at decadal intervals out to 2100, following the B2+CLE scenario, described in detail in chapter 4. For use in global models, these emission projections were further spatially disaggregated. Global gridded (1° x 1°) emissions were produced using the ACCENT year 2000 anthropogenic emission distributions as a base.

Updated year 2000 emissions were produced. The IIASA regional totals included emissions from international shipping and aviation; these were removed from the regional totals and added back later using a different approach, as in the models these emission sources need to be distributed along shipping and air routes. The base emissions were then scaled to match the adjusted IIASA regional totals. Ship emissions were included in the ACCENT base case, using EDGAR estimates, but these were updated using more comprehensive ship traffic distributions (Corbett & Kohler 2003) and emission totals (Eyring *et al.* 2005a). Ship emissions are only really significant for NO_X and SO_2; they were included in the CO and nmVOC distributions, but only for completeness.

Figure A1 compares the global NO_X emission distributions for 2000 from the ACCENT study with new emissions used here (called IIASA 2000). NO_X emissions are higher in most regions (by nearly 20 Tg(NO_2) y^{-1} in total), with the exception of the United States and Eastern Europe/Russia. The new treatment of ships spreads emissions out from the previous very localised shipping lanes, and also includes emissions on large lakes (eg the North American Great Lakes).

Figure A2 shows the equivalent plot for CO emissions, which are 15% higher globally compared to those used in the ACCENT study, with particularly strong increases in East Asia (although there are modest decreases in Eastern Europe and North Africa/Middle East). Analysis of CO in the ACCENT intercomparison showed that Northern Hemisphere emissions were probably too low (Shindell *et al.* 2006); these new emissions were considered a more accurate estimate for the year 2000.

Regional scaling factors from IIASA were similarly applied to projected 2050 emissions. For ships, projections for 2050 were taken from Eyring *et al.* (2005b) – specifically for the DS2–TS2 scenario – this corresponds to economic growth as in SRES B2, but with the assumption of quite strong technological advancements in emission control. For example, ship NO_X emissions fall by about 50%, despite an assumed GDP growth of 2.8%/y. There is little current legislation concerning emissions from international shipping, so the assumption of significant uptake of emission control technology by 2050 represents a major uncertainty. Global differences (2050–2000) in NO_X and CO emissions are shown in Figure A3.

The IIASA scenario did not specify projections for nmVOC, SO_2 or NH_3 (the latter two are not thought to be important for O_3, but were considered for completeness). The ACCENT 2000 nmVOC distribution was kept (with a minor adjustment for ships), and the IIASA CO regional scaling factors applied to the ACCENT base nmVOC distribution to project to 2050. This represents a significant uncertainty, as in many sectors, nmVOC behave differently to CO.

For SO_2, the ACCENT 2000 distribution was used, but updated with the ship emissions as described above. For 2050, the 2030 MFR distribution (Dentener *et al.* 2006a) was used, but again updating the ships. For NH_3, in the absence of better data, the ACCENT 2000 and 2030 CLE distributions were used for 2000 and 2050 respectively.

For aircraft NO_X emissions, modelers were recommended to use their present-day distribution, with totals of 0.8 Tg N y^{-1} for 2000 and 1.7 Tg N y^{-1} for 2050. The 2050 value is the 2030 value used in ACCENT.

Biomass burning produces large amounts of O_3 precursor emissions, in particular CO and NO_X, but also nmVOC. The ACCENT Photocomp work used satellite-derived monthly mean emission distributions averaged over the period 1997–2002 (van de Werf *et al.* 2003), with ecosystem-specific emission factors (Andreae & Merlet 2001) for different trace gases. This approach was also used in the Royal Society runs, with biomass burning kept fixed in all runs.

Methane emissions were not used, rather a global constant CH_4 concentration was imposed to avoid requiring models to perform spin-ups of several years. For 2000, the models used 1760 ppb; for 2050, the SRES B2 value of 2363 ppb was used.

Emissions data, at 1° x 1° resolution, are available at: http://xweb.geos.ed.ac.uk/~dstevens/Roy_Soc_AQ_WG/emissions/

Figure A1 Anthropogenic NO_x emissions for year 2000 from the ACCENT study, this study, and the difference. Units ($Gg(NO_2)$ y^{-1} 1° x 1° grid square).

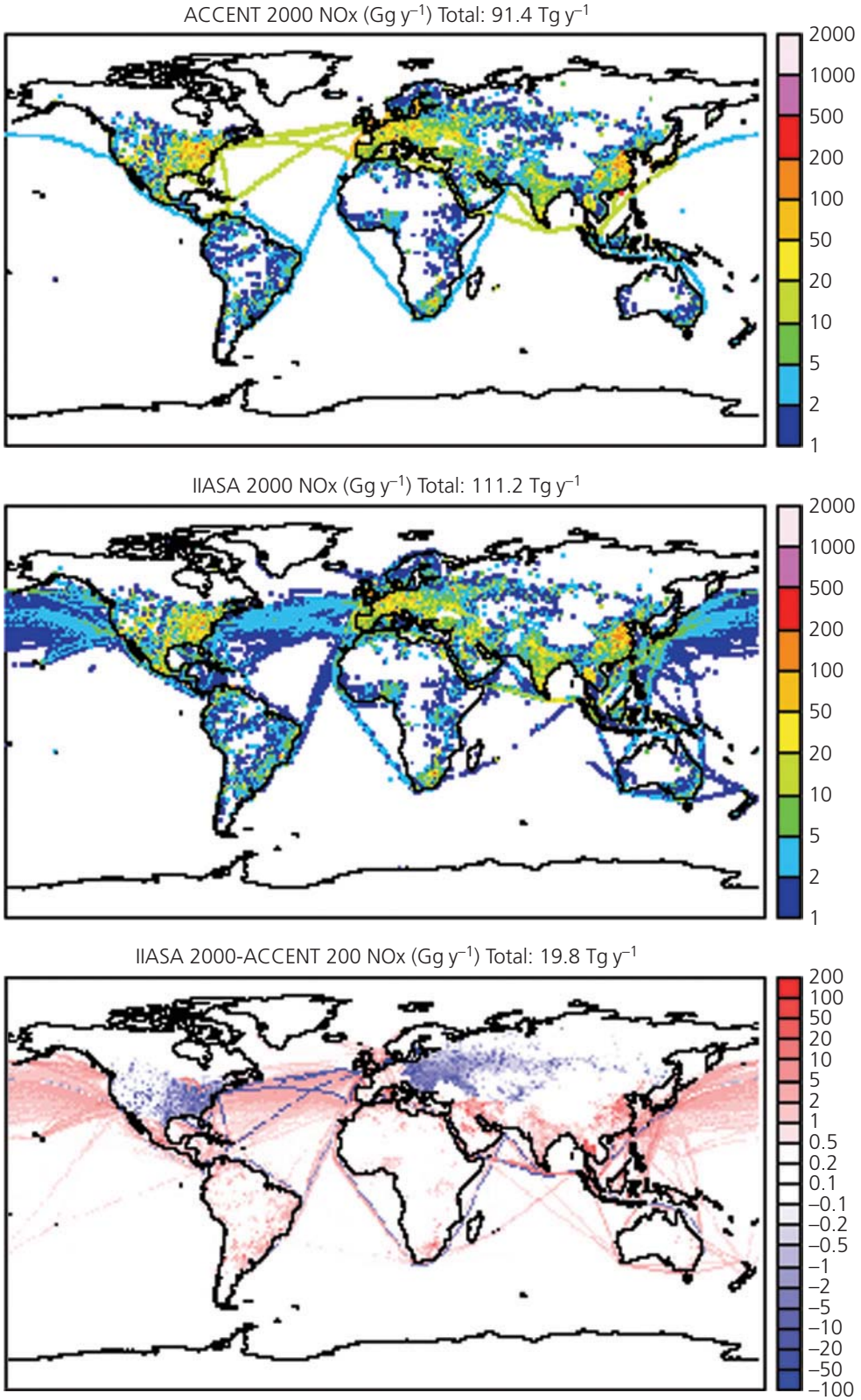


Figure A2 Anthropogenic CO emissions for year 2000 from the ACCENT study, this study, and the difference. Units (Gg(CO) y^{-1} 1° x 1° grid square).

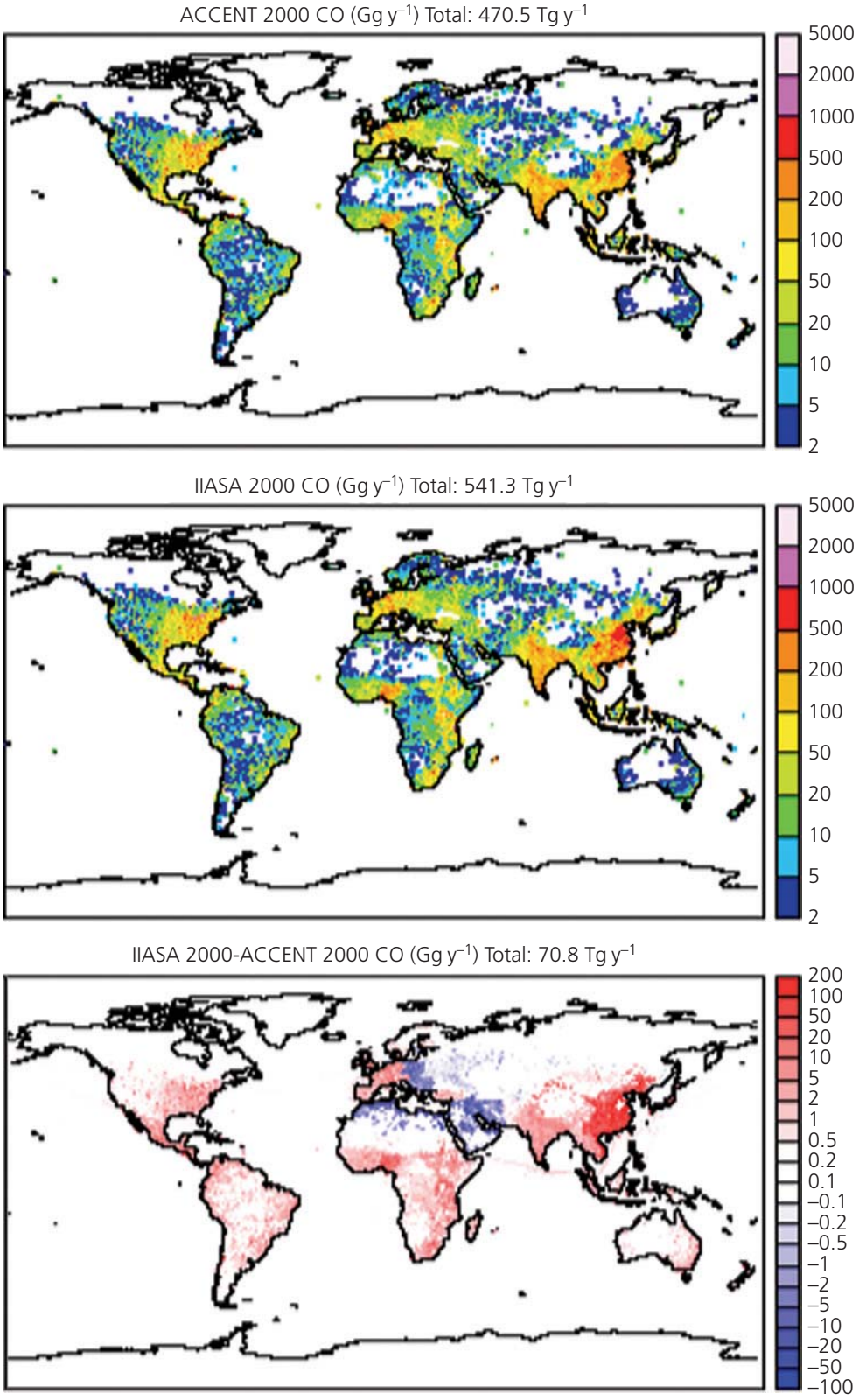
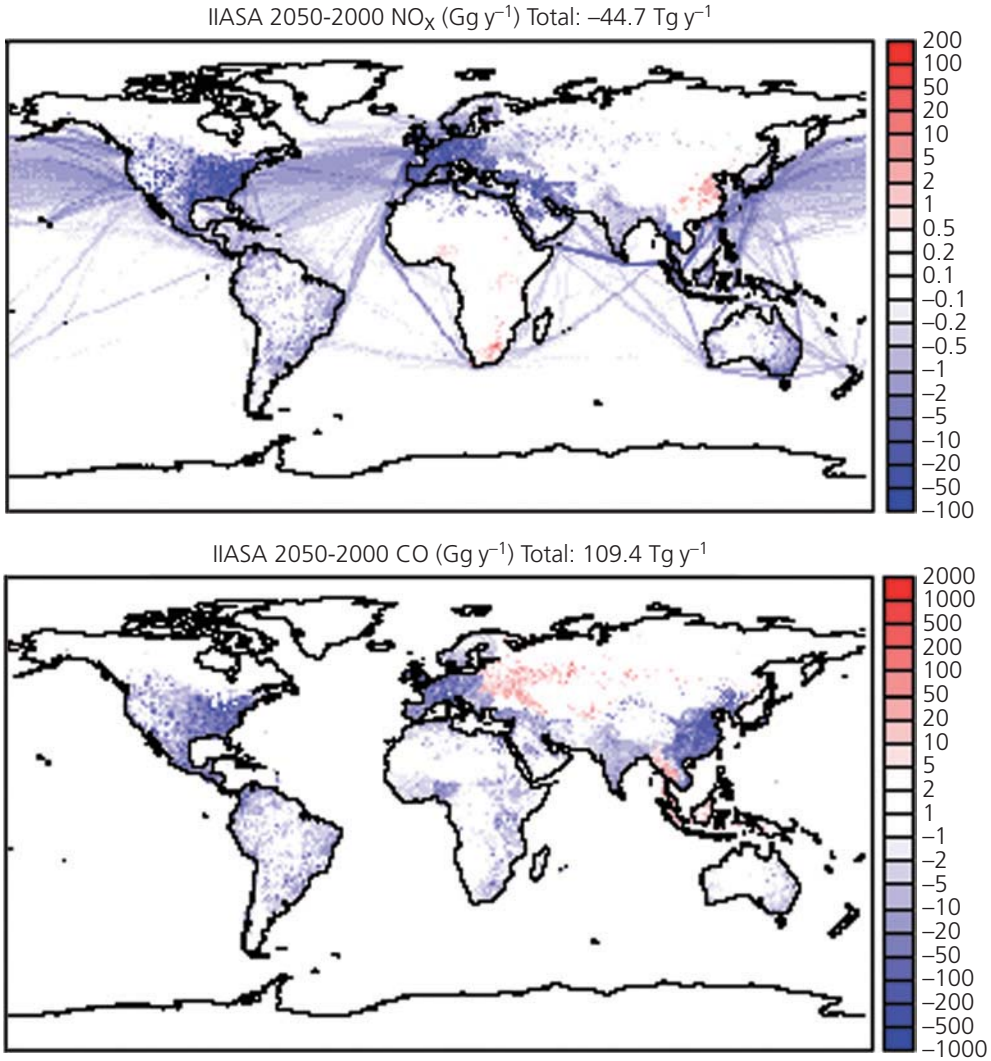


Figure A3 Projected changes in anthropogenic NO_X and CO emissions between 2000 and 2050 under the IIASA scenario. Emissions generally reduce (blue). Units as Figures A1/A2.



Simulations

Modelers performed at least 1 year spin-up in all runs, to insure that results were not strongly affected by initial conditions. The longest-lived compounds after CH_4 are species such as ethane and CO, with typical lifetimes of a few months. Table 5.1 summarises the simulations.

Participating models

Six modelling groups participated in the study (Table A1). All of these models, in essentially the same format, took part in the ACCENT Photocomp study, and further details of results and validations of individual models can be found in Dentener *et al.* (2006a,b), Stevenson *et al.* (2006), Shindell *et al.* (2006), van Noije *et al.* (2006) and Ellingsen *et al.* (2008).

Table A1: Participating models, detailing the modelers involved, model resolution, model meteorology and transport schemes, tropospheric and stratospheric chemistry schemes, and key references.

Model	Institute	Contact author	Resolution (long/lat/levels), Top level	Underlying meteorology	Advection scheme	Convection scheme	Tropospheric chemistry	Stratospheric chemistry	References
1. FRSGC/UCI	FRCGC/ JAMSTEC	Oliver Wild	2.8°/2.8°/L37 10 hPa	CTM: ECMWF–IFS pieced-forecast data for 2000	Prather (1986)	Mass fluxes taken from ECMWF–IFS fields, based on Tiedtke (1989)	35 species (27 transported), using ASAD (Carver *et al.* 1997)	LINOZ (McLinden *et al.* 2000)	Wild & Prather (2000) Wild *et al.* (2003)
2. STOCHEM-HadAM3	University of Edinburgh	David Stevenson Ian MacKenzie	5°/5°/L9 100 hPa	GCM: HadAM3 vn4.5	Collins *et al.* (1997)	Collins *et al.* (2002b)	70 species SO_X–NO_y–NH_X aerosols; interactive	Prescribed O_3 concentration gradient at 100 hPa	Collins *et al.* (1997) Stevenson *et al.* (2004)
3. STOCHEM-HadGEM	UK Met. Office	Michael Sanderson Bill Collins	3.75°/2.5°/L20 40 km	GCM: HadGEM	Collins *et al.* (1997)	Collins *et al.* (2002b)	70 species SO_X–NO_y–NH_X aerosols; interactive	Relaxed towards SPARC O_3 climatology above tropopause	Collins *et al.* (1997) Collins *et al.* (2003)
4. TM4	KNMI	Twan van Noije	3°/2°/L25 0.48 hPa	CTM: ECMWF 3–6-h operational forecasts (2000)	Russel & Lerner (1981)	Tiedtke (1989)	37 species (22 transported) SO_X–NO_y–NH_X aerosols, interactive	O_3 nudged towards climatology above 123 hPa: except 30N–30S, above 60 hPa	Dentener *et al.* (2003) van Noije *et al.* (2004)
5. ULAQ	Università L'Aquila	Veronica Montanaro Giovanni Pitari	22.5°/10°/L26 0.04 hPa	GCM: ULAQ-GCM	Eulerian flux form	Pitari *et al.* (2002) following Muller & Brasseur (1995)	Includes tropospheric aerosols	Detailed stratospheric chemistry scheme, including stratospheric aerosols	Pitari *et al.* (2002)
6. UM_CAM	University of Cambridge	Guang Zeng John Pyle	3.75°/2.5°/L19 4.6 hPa	GCM: HadAM3 vn4.5	Leonard *et al.* (1995)	Gregory & Rowntree (1990)	60 species (36 transported) No aerosols.	O_3 and NOy prescribed above 30 hPa (Tropospheric chemistry operates below 30 hPa)	Zeng & Pyle (2003, 2005)

Relevant Royal Society policy reports, statements and responses

Royal Society activities on reducing the risk of the misuse of scientific research
(6 pages, 21 Aug 2008, 17/08)

Joint science academies' statement: Climate change adaptation and the transition to a low carbon society
(2 pages, 10 Jun 2008)

Letter to Secretary of State on Carbon Capture and Storage
(2 pages, 3 April 2008)

Royal Society submission to the Efra committee call for views on Defra's scientific infrastructure
(2 pages, 27 March 2008, 13/08)

Royal Society submission to DIUS UK science and innovation strategy
(12 pages, 29 February 2008, 08/08)

Sustainable biofuels: prospects and challenges
(90 pages, 14 January 2008, 01/08)

Response to the Environmental Audit Committee's inquiry into the Millennium Ecosystem Assessment
(5 pages, December 2006, 30/06)

Response to IMOSEB consultation
(3 pages, 5 Dec 2007, 27/07)

Biodiversity–Climate Interactions: Report of a meeting held at the Royal Society
(65 pages, 6 Dec 2007, 30/07)

Joint statement by NASAC to the G8 on sustainability, energy efficiency and climate change
(2 pages, 29 Nov 2007, 28/07)

Response to the UK Climate Change Bill Consultation
(7 pages, June 2007, 18/07)

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(2 pages, March 2007 05/07)

Letter to Secretary of State for Trade and Industry regarding cuts to the Research Councils' budgets
(1 page, March 2007, 09/07)

Submission to House of Lords S&T Select Committee inquiry on radioactive waste management
(1 page, February 2007, 08/07)

Submission to the STEM Taskforce Science and Society inquiry
(6 pages, February 2007, 06/07)

Report of a Royal Society policy seminar on the scientific understanding of obesity
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(10 pages, January 2006, 02/06)

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(13 pages, October 2003, 18/03)

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ISBN: 978-0-85403-713-1
Issued: October 2008 RS1276

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ISBN 978-0-85403-713-1