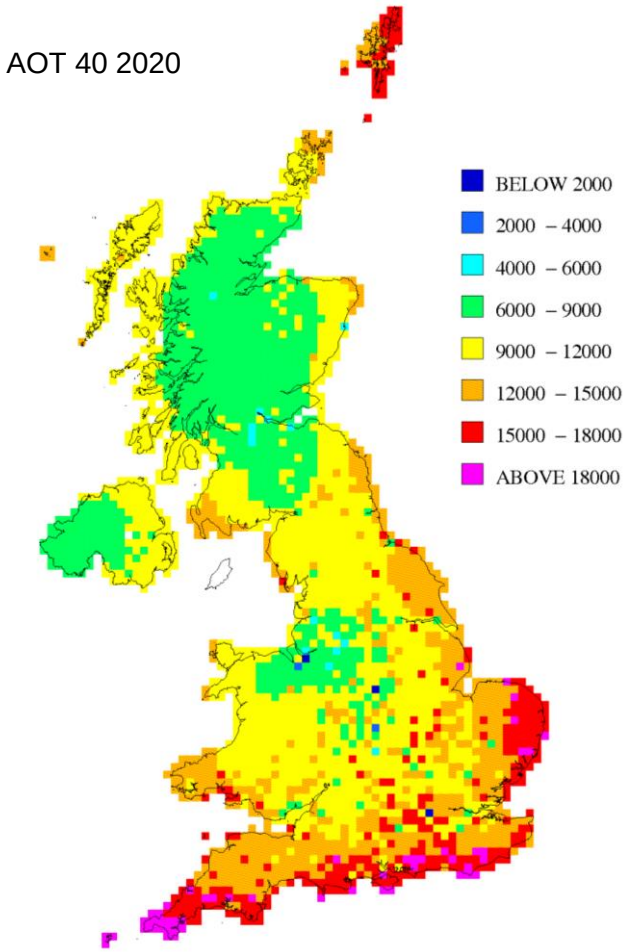
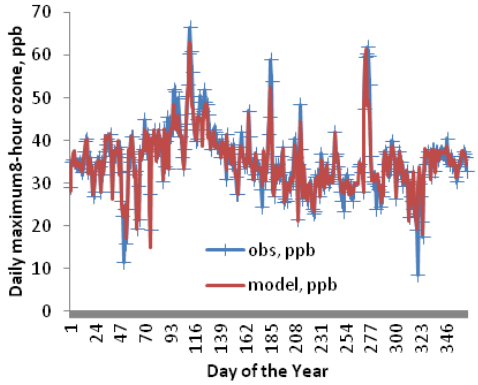
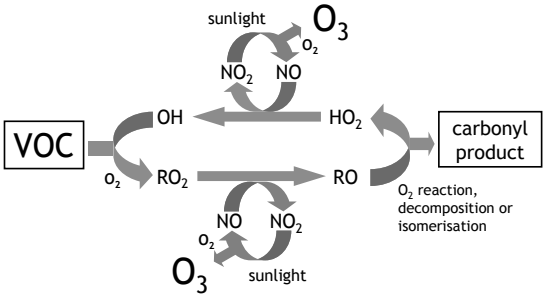
RICARDO-AEA

Modelling of Tropospheric Ozone

Annual Report 2012/13







Report for Defra

Ricardo-AEA/R/ED57616
Issue Number 3
Date 15/11/2013

Customer:

The Department for Environment, Food and Rural Affairs, Welsh Assembly Government, the Scottish Executive and the Department of the Environment for Northern Ireland

Customer reference:

AQ0722

Confidentiality, copyright & reproduction:

Ricardo-AEA reference:

Ref: ED57616- Issue Number 3

Contact:

Tim Murrells
Ricardo-AEA Ltd
Gemini Building, Harwell, Didcot, OX11 0QR
t: 0870 190 6539
e: tim.p.murrells@ricardo-aea.com
Ricardo-AEA is certificated to ISO9001 and ISO14001

Author:

Sally Cooke, Prof Dick Derwent (rdscientific), Andrea Fraser, John Abbott and Tim Murrells

Approved By:

Geoff Dollard

Date:

15 November 2013

Signed:

Executive summary

Ozone is an air pollutant that affects human health, vegetation and materials. The concentrations of ground-level ozone widely exceed environmental quality standards across the UK and Europe. Ozone is not emitted directly into the atmosphere, but is a secondary air pollutant formed in the lower atmosphere by sunlight-initiated reactions of ozone precursors - volatile organic compounds (VOCs) in the presence of nitrogen oxides (NO_x).

Concentrations are strongly influenced by meteorological conditions and emissions of the precursor gases from natural and man-made sources across the UK and rest of Europe. The non-linear nature of ground-level ozone production requires the use of sophisticated chemical transport models to understand the factors affecting its production and subsequent control.

This report describes work undertaken during 2012/13 in the project "*Modelling of Tropospheric Ozone*" funded by the Department for Environment, Food and Rural Affairs (Defra) and the Devolved Administrations (the Scottish Executive, the Welsh Assembly Government and the Department of the Environment for Northern Ireland).

This project aims to maintain two models that have been previously developed and demonstrated to quantify the rate of production and loss of ozone in the UK and to use them to support the analysis and development of Defra's policies on ozone air quality. The two models are the **Ozone Source Receptor Model (OSRM)** and the **Photochemical Trajectory Model (PTM).**

The OSRM and PTM have been used to model and interpret the UK ground level ozone concentrations for 2011. The OSRM gave the overall picture for ozone throughout the year and across the UK. The PTM was used to diagnose the nature of specific ozone episodes observed at the Aston Hill monitoring site. The models were largely consistent with the observations made.

Box 1: Ozone in the UK in 2011

- **Combined with measurements, the models showed that 2011 was a year with relatively low ozone concentrations and was unremarkable in terms of severity or number of ozone episodes.**
- **However, there were photochemical ozone episodes during 2011 including periods in April and in late September/early October, extending the normal length of the ozone season.**
- **Most of the episodes at the Aston Hill site were dominated by emissions from the rest of Europe and all but one of the episode days was NO_x-sensitive rather than VOC-sensitive.**
- **The latter finding was quite unusual because ozone episodes in the UK have a tendency to be in the VOC-sensitive category. It is suggested that this was because there was a greater than usual preponderance for southerly trajectories.**

Some improvements have been made to the efficiency and flexibility of the emission scenario pre-processor and of the treatment of shipping emissions in the OSRM.

UK ozone has been modelled for a range of different emission reduction scenarios of relevance to Defra's ozone air quality policy.

Box 2: Ozone concentrations for different emission scenarios

The OSRM was used to model UK ozone for:

- **emissions in 2008 for direct comparison with simulations previously run for Defra based on projected emissions for 2020-2030 assuming the same meteorological conditions.**
- **an arbitrary 10% reduction in UK NOx and VOC emissions. This was to provide Defra with the information to update health-, crop- and materials damage cost functions in relation to ozone.**
- **The simulations consistently show how ozone concentrations are predicted to be higher in future years when emissions are reduced and all other conditions are maintained the same.**

The PTM was used to model the impact of the 2020 emission reductions associated with the revised Gothenburg Protocol on UK ozone episodes experienced during 2011

- **The impact was found to be substantial, bringing about both increases and decreases in ozone at the different locations modelled under the 2011 episode conditions.**
- **However the overall impact of the Gothenburg Protocol emission reductions, averaging a net decrease in peak ozone of around 3.0 ± 4.7 ppb, was not sufficient to bring all the episodes, at all the sites during 2011, below the 50 ppb WHO guideline**

Results from the Defra Model Intercomparison Exercise were further analysed to compare and understand differences in the sensitivity of the model results to changes in ozone precursor emissions during episode conditions. Ozone episodes in July 2006 were assigned to either NO_x- versus VOC-sensitivity and UK- versus rest of Europe-dominance. The conclusion is that these assignments are not robust between the different days of July 2006 and between the different models.

Table of contents

1 Introduction

Ozone is an air pollutant that affects human health, vegetation and materials. In light of this the concentrations of ambient ozone near ground level are of concern.

Ozone is not emitted directly into the atmosphere, but is a secondary air pollutant formed in the lower atmosphere by sunlight-initiated reactions of ozone precursors - volatile organic compounds (VOCs) in the presence of nitrogen oxides (NO_x). These precursors are emitted from both natural and man-made sources. Formation of atmospheric ozone occurs over a large spatial scale and as such ground level concentrations of ozone experienced in the UK are the result of emissions from sources within the UK, across Europe and further afield.

Concentrations are strongly influenced by meteorological conditions. Elevated concentrations over the UK occur in the spring and summer when slow-moving, or stagnant, high pressure (anticyclonic) weather systems bring in photochemically reacting air masses from mainland Europe. Concentrations of ozone are also influenced by ozone entering the UK in the free troposphere from the north Atlantic under prevailing meteorological conditions, providing a so-called hemispheric baseline ozone concentration upon which regional contributions are superimposed.

Local effects can play a part in both removing and forming ozone. Local emissions of highly reactive VOCs can lead to rapid photochemical production of ozone under favourable meteorological conditions, however in urban areas these are usually offset by ozone removal through reaction with locally emitted NO_x from sources such as road traffic.

Recognising the transboundary nature of ozone formation, a series of European directives have been introduced to reduce emissions of ozone precursor gases. The Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone, under the Convention on Long-range Transboundary Air Pollution, set national emission ceilings for 2010 on total emissions of sulphur dioxide, nitrogen oxides, volatile organic compounds and ammonia. This was amended in 2012 to include national emission reduction commitments to be achieved in 2020 and beyond. There are similar ceilings set under the EU National Emissions Ceiling Directive (NECD). A proposal to amend the NECD is still under preparation and should also set emission ceilings to be respected by 2020. There is also a series of complementary EU Directives targeting specific sources that emit ozone precursor gases such as use of solvents, large combustion plant and road transport.

The EU Air Quality Directive sets target values and long-term objectives for ambient ozone concentrations for the protection of human health and vegetation. The two target values for ambient ozone both came into force from 1/1/2010:

- A maximum daily 8-hour mean concentration of 120 µg m^{-3}, not to be exceeded on 25 days per calendar year averaged over 3 years;
- AOT40 (calculated from 1-hr values) 18000 µg m^{-3} h averaged (May to July) over five years.

National emissions inventories show that VOC and NO_x emissions have been falling in the UK and the rest of Europe over the last two decades. In spite of this there are still exceedances of the target values and the more challenging, long term objective values.

Understanding past trends in ozone concentrations and quantifying how measures aimed at reducing emissions of ozone precursor gases are likely to affect ground level ozone concentrations in the future is challenging. The complex nature of ozone production requires the use of sophisticated models that combine meteorological effects, emissions data and descriptions of chemical processes that occur in the atmosphere in order to quantify the rate

of ozone production and loss in a moving air mass that receives precursor emissions over a wide spatial scale.

This project aims to maintain two models that have been previously developed and demonstrated to quantify the rate of production and loss of ozone in the UK and to use them to support the analysis and development of Defra's policies on ozone air quality. More specifically, the project uses these models to predict future concentrations of ozone and allow assessments of how concentrations respond to changes in precursor emissions. The results will be used to inform policy makers in the development of policies on precursor emission control and to evaluate their effects on UK ambient ozone concentrations including compliance with the EU Air Quality Directive. Importantly the models will be used to assess the effects of alternative emission reduction scenarios on UK ambient ozone concentrations in 2020 and other years as part of the review of the EU Air Quality Directive and NECD.

The two models used in this project are both Lagrangian-type models: the Ozone Source Receptor Model (OSRM) and the Photochemical Trajectory Model (PTM).

The following section provides a brief description of these models. The overall aims and objectives of the project, which started in March 2012 and runs until March 2014, are then described. These can be summarised as:

- **Objective 1:** The modelling of UK ozone in 2011 and 2012 using the OSRM and PTM.
- **Objective 2:** Policy application of the OSRM and PTM by modelling the formation and loss of ozone for alternative emission scenarios. This work is undertaken on a call-off basis;
- **Objective 3:** Maps of ozone concentration and surface flux parameters for different agricultural crops and semi-natural species;
- **Objective 4:** Optimising the use of emissions inventory information in the OSRM.

This report summarises the work undertaken for Objective 1 concerning the modelling and analysis of ozone concentrations in the UK in 2011. The specific model simulations requested by Defra and carried out under Objective 2 during 2012/13 are described. Further model development is not part of this project, but some improvements in the coding of the OSRM were undertaken in Objective 4 to enhance the efficiency in the way the OSRM uses emissions inventory information. These improvements are described in this report.

The work for Objective 3 was completed in March 2012 and was reported then.

2 The Ozone Source Receptor and Photochemical Trajectory Models

The OSRM and PTM models are both Lagrangian-type models, developed and supported by Defra over the years through previous tropospheric ozone modelling contracts. Their performance has been demonstrated in peer-reviewed scientific journals and both models have been assessed in Defra's air quality Model Intercomparison Exercise (Williams et al, 2011).

Both models have been the backbone of Defra's ozone policy development and analysis for some years, having been extensively used for formulating and testing alternative policies on precursor emission controls. These include vehicle emission and fuel quality directives and directives on biofuels, solvents and industrial emissions. They have also been used to assess the effects of domestic policies and measures on ozone including those considered in the review of the Air Quality Strategy. The models have been used to model the UK's future ground level ozone climate up to 2020 assuming different meteorology conditions. More recently, the models have been used to assess the effects of various emission reduction scenarios considered for different countries in revisions to the Gothenburg Protocol.

The Monks' review of Defra's future ozone modelling requirements (Monks et al, 2007) recommended that Defra should consider moving its ozone modelling activity to an Eulerian basis. Although Eulerian models such as EMEP4UK and CMAQ are being used in the UK for regional scale ozone modelling, Defra is still assessing their practical application for national scale modelling and for formulating and assessing ozone air quality policy. In the meantime, there is still an urgent need for efficient and tested ozone models such as the OSRM and PTM to support a range of policies currently being developed, most notably to address different future emission scenarios such as those proposed for the revisions to the Gothenburg Protocol and National Emissions Ceilings Directive as well as to the reviews of the European Air Quality Directive.

2.1 The Ozone Source Receptor Model

Details of the OSRM have been given elsewhere in project reports and publications and only a brief description of the model is given here (e.g. Hayman et al, 2010 and Murrells et al, 2012).

The OSRM simulates the photochemical production of ozone in reactive air masses as they arrive at different receptor points in the UK. Essentially, each parcel of air picks up emissions from natural and man-made sources as it moves over land and sea surfaces over a large spatial scale and these undergo a series of chemical reactions initiated by sunlight leading to the production of ozone. Gridded 1 x 1 km emissions data for the UK are taken from the NAEI[1] (Bush et al, 2010) and 50 x 50 km emissions data for the rest of Europe are taken from EMEP[2]. Emission terms to describe natural biogenic emissions from European forests and agricultural crops are derived from the European PELCOM project.

The model uses archived 96-hour back trajectory data from the Met Office NAME model providing boundary layer depth and other parameters. The chemical mechanism used to define the rate of ozone formation and loss is a modified version of the mechanisms used in the STOCHEM model, but an option is available to use the condensed CRIv2-R5 chemical scheme,

[1] National Atmospheric Emissions Inventory, http://naei.defra.gov.uk/
[2] http://www.ceip.at/webdab-emission-database/emissions-as-used-in-emep-models/

linked to the Master Chemical Mechanism [3]. Dry deposition processes are represented using a conventional resistance approach.

The OSRM calculates ozone concentrations at mid-boundary layer height at hourly intervals on a 10 x 10 km grid covering the whole of the UK. These are corrected to account for loss of ozone due to reaction with local emissions of NO_x and deposition to land and sea surfaces in order to generate concentrations at ground-level.

The OSRM is also used in conjunction with a Surface Ozone Flux Model which can be used to model the uptake of ozone by different types of vegetation species under different meteorological conditions.

In conjunction with GIS-based tools, the OSRM is used to derive population- and area-weighted means of different ozone concentration metrics to provide the information necessary to Defra policy makers for cost-benefit analysis of emission reduction policies.

Previous work had shown that the empirical modelling approach used in Defra's UK Ambient Air Quality Assessments (UKAAQA) model[4] traditionally gives results for ozone concentration metrics that, in model verification, are more representative of the measured concentrations than corresponding outputs provided by the OSRM. Hence, the UKAAQA modelling contract is used to provide the supplementary ozone modelling required for EU Air Quality Directive reporting on ozone to the European Commission each year on behalf of Defra. The OSRM, on the other hand, has a stronger role to play in scenario analysis and policy development as the OSRM can model future emission scenarios and the chemistry involved in forming and removing ozone over a large spatial scale from the emitted precursor gases, NO_x and VOCs. The OSRM is therefore maintained and evaluated each year using appropriate meteorology and emissions data and comparing calculated ozone concentrations with those from the UKAAQA empirical model and with monitoring data at specific AURN sites.

The OSRM has been used to model the UK ground-level ozone climate based on meteorological conditions and emissions from 1999 to 2010 and for forecasting ozone under future UK and European-wide emission scenarios for different meteorological conditions represented by those of previous years. The model has been optimised for computational efficiency and has been a vital policy tool for Defra routinely used in quantifying the response of the UK's ground-level ozone climate to measures aimed at reducing emissions of the precursor species.

Both the UKAAQA and OSRM modelling techniques are verified against measured data to provide confidence in their performance. The two models have been compared in previous years, most recently for 2004, 2005, 2007, 2009 and 2010, which were noted as relatively "low ozone" years (Hayman et al, 2006a, Murrells et al, 2009), 2006 which was a relatively "high ozone" year (Murrells et al, 2008) and 2008 which was a broadly moderate year for ozone concentrations (Murrells et al, 2011).

2.2 The Photochemical Trajectory Model

The PTM has been used to describe photochemical ozone formation as well as secondary inorganic and organic aerosol formation in north-western Europe. Details are given in Derwent et al (1996, 1998, 2009), Abdalmogith et al. (2006) and Johnson et al. (2006). The model describes the chemical development within an air parcel that follows a trajectory for up to 10 days. For each mid-afternoon of each day a large number of equally probable and randomly selected 96-hour air parcel trajectories are generated using the Met Office Numerical Atmospheric dispersion Model Environment (NAME) model. The PTM uses NAEI and EMEP gridded emissions data and inventories for natural biogenic emissions. Initial and background species concentrations are taken from the EMEP site at the Valentia Observatory and the atmospheric baseline station at Mace Head, Ireland. The model has the option of using different chemical mechanisms. Dry deposition processes are represented using a conventional resistance approach.

[3] http://mcm.leeds.ac.uk/MCM/
[4] Previously referred as the Pollution Climate Mapping model (PCM)

The PTM has been used for a variety of purposes to support Defra policy on ozone and secondary particulate matter (PM). These include the estimation of photochemical ozone creation potentials (POCPs) of individual VOCs (Derwent et al., 1998) and more recently to estimate secondary organic aerosol formation potentials (SOAPs, Derwent et al, 2010a). It has also been used to evaluate the effectiveness of current precursor emission controls in Europe on levels of ground-level ozone in the UK (Derwent et al, 2010b) and the effectiveness of future potential emission controls.

The PTM is similar to the OSRM but uses 3-dimensional trajectories to specified receptors and is able to use much more detailed chemical mechanisms than the OSRM, including the Master Chemical Mechanism (MCM) and its reduced derivatives. Thus the PTM complements the OSRM in being able to address the impact of chemistry on ozone formation and removal by using alternative chemical schemes to get an appreciation of the sensitivity of predicted ozone concentrations to choice of chemical scheme. By using the MCM, the PTM can also be more aligned to the detailed speciation VOC emissions inventory produced by the NAEI (Passant 2002) and can be used to address policies targeted at more specific groups of VOCs.

As a summary, the OSRM is used to model ground-level ozone concentrations (and ozone flux to vegetation) across the UK domain at 10 km resolution so is well set up to provide metrics on a national scale for the damage costs of ozone and the effectiveness of emission reduction measures to be evaluated. The outputs generated in this project are mainly as population- and area-weighted concentration metrics. The PTM is mainly used in this project to model ozone ***episodes*** at specific receptors rather than the whole of the UK like the OSRM but is used to provide a detailed picture of the sources responsible for the episodes.

3 Overview of Project Aims and Objectives

The overall aim of the project is to maintain a level of modelling support for Defra's ozone air quality policy development and assessment using the existing OSRM and PTM models.

The work is divided into four main project objectives. These are aimed at maintaining the OSRM and PTM models for predicting ground-level ozone concentrations in the UK and applying them to future emission scenarios relevant to Defra's policies on air quality and impacts on health and vegetation. The emission scenarios are worked on throughout the year on an *ad-hoc* basis.

Objective 1: Modelling of UK ozone in 2011 and 2012 using the OSRM and PTM

This Objective involves incorporating the latest meteorology and emissions inventory data to model the ground-level ozone climate in the UK for 2011 and 2012. This creates a new OSRM "Basecase" so the model is primed for predicting future ozone concentrations when emissions or meteorological conditions are changed (Objective 2). It also involves an initialisation of the concentrations relevant to the model year. The work in this reporting year has involved modelling UK ozone in 2011.

Objective 2: Policy application of the OSRM and PTM

This Objective involves the use of the OSRM and PTM to model ozone concentrations in the UK for emission scenarios specified by Defra. The modelling normally entails forecasting ozone concentrations in future years, typically 2020 or 2030, for relevant emission changes assuming meteorological conditions represented by those of a historical year. These might be years characterised by particularly high levels of ozone during summer episodes such as 2006 or years characterised by cool summers with little photochemical activity such as 2007. The work involves national scale modelling at 10 x 10 km resolution using the OSRM, producing outputs specified by Defra. The PTM would be used to assess the probability distributions of the various outcomes of the emission scenarios in terms of ozone, normally in terms of the impacts on predicted ozone episodes.

Objective 3: Maps of ozone concentration and surface flux parameters for different agricultural crops and semi-natural species: 2007 and 2020

This Objective provided the Centre for Ecology and Hydrology (CEH) with UK maps for 24-hr mean ozone concentration outputs from the OSRM and ozone flux parameters for vegetation from the Surface Ozone Flux Model (SOFM) covering years 2007 and 2020. This work was reported in March 2012 and is not further mentioned in this report.

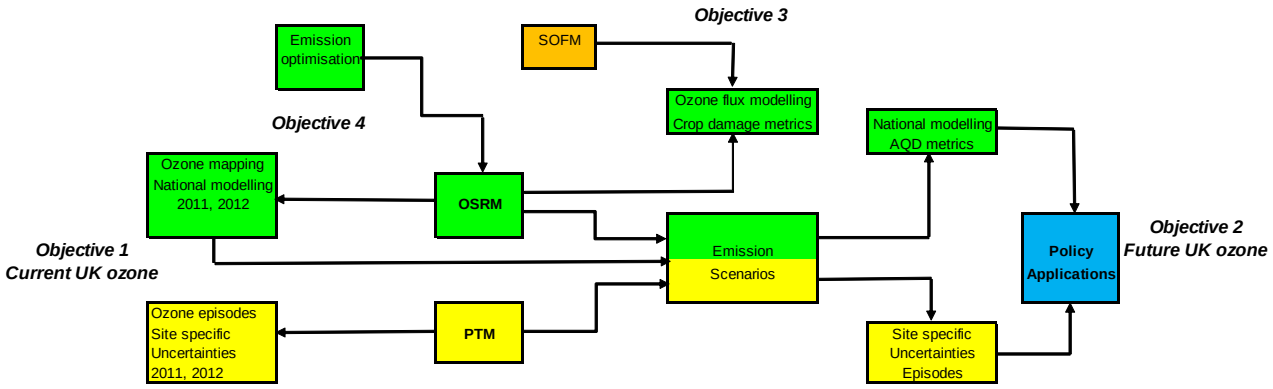
Objective 4: Optimising the use of emissions inventory information in the OSRM

This Objective aims to improve the efficiency and transparency of the OSRM by optimising the use of emissions inventory information particularly in terms of

- The emission scenario pre-processor, and
- The treatment of shipping emissions

Linkages between the four Objectives are shown in Figure 3.1.

Figure 3.1: Links between project objectives



Thus, the OSRM and PTM are used to model different aspects of the current ground level ozone climate using relevant emissions and meteorology data for the years 2011 and 2012 in Objective 1. Both models are used to predict future ozone concentrations for different emission scenarios. The OSRM predicts concentrations on a national scale, producing relevant health and vegetation-based metrics that can be compared with Air Quality Directive target values and inform policies on regional differences in exposure. The PTM is used to predict future episodes and probabilistic uncertainty analysis of future trends based on modelling at specific locations. The improvements developed in Objective 4 improve the transparency and efficiency of future OSRM runs for different emission scenarios in Objective 2. Objective 3 uses the OSRM for a specific task on ozone flux modelling, but further work of this nature will benefit from the maintenance of the OSRM carried out in Objectives 1 and 4.

The work on the OSRM across all objectives has been undertaken by **Ricardo-AEA** who is the lead contractor with overall project management responsibilities for the project. Work involving the PTM model in Objectives 1 and 2 has been undertaken **by Professor Dick Derwent (rdscientific).**

4 Modelling UK Ground-Level Ozone Concentrations in 2011

4.1 UK Scale Modelling Using the OSRM

The OSRM was used to model hourly ground-level ozone concentrations at 10 x 10 km resolution for 2011 in the same way as done for previous years. Meteorology data from the Met Office NAME model at 6-hourly intervals covering 23 boundary layer parameters over a domain 30ºW to 40ºE and 20º to 80ºN at 1 x 1º spatial resolution were used in conjunction with emissions inventory data for 2011. For UK emissions, the 1 x 1 km NAEI gridded data for 2010 were used,[5] projected to 2011 using the most up-to-date NAEI emission projections for each source sector based on DECC's UEP43 energy projections (Misra et al, 2012). For European emissions, the latest 50 x 50 km EMEP gridded data were used for 2010, also re-scaled to 2011 based on reported country totals for emissions for this year.

Ozone concentrations on each OSRM trajectory were initialised using daily concentration fields from the global tropospheric ozone model, STOCHEM, adjusted using monthly data for 2011 from measurements at Mace Head, Ireland, provided by Professor Derwent (rdscientific).

The OSRM was run to provide hourly ozone concentrations at mid-boundary layer height. The post-processor was then run to generate maps of ground-level ozone taking account of surface deposition and losses due to reaction with locally emitted NO_x. The post-processor generated concentrations for the two Long-Term Objective ozone metrics used in the EU Air Quality Directive reporting:

- Days greater than 120 $\mu g\ m^{-3}$ as a maximum daily running mean (DGT 120, the Long Term Objective for Human Health)
- AOT40 (Long Term Objective for Vegetation)

Concentrations for these metrics were also calculated at specific AURN monitoring sites for comparison with measurements.

4.1.1 Comparison of maps of OSRM and UKAAQA outputs for ozone metrics in 2011

Figures 4.1(a) and 4.2(a) show maps of AOT40 and DGT 120 calculated by the OSRM. Corresponding maps from the empirical modelling technique used in the UK Ambient Air Quality Assessments (UKAAQA) programme are also shown in Figures 4.1(b) and 4.2(b). These are developed at finer resolution on a 1 x 1 km grid.

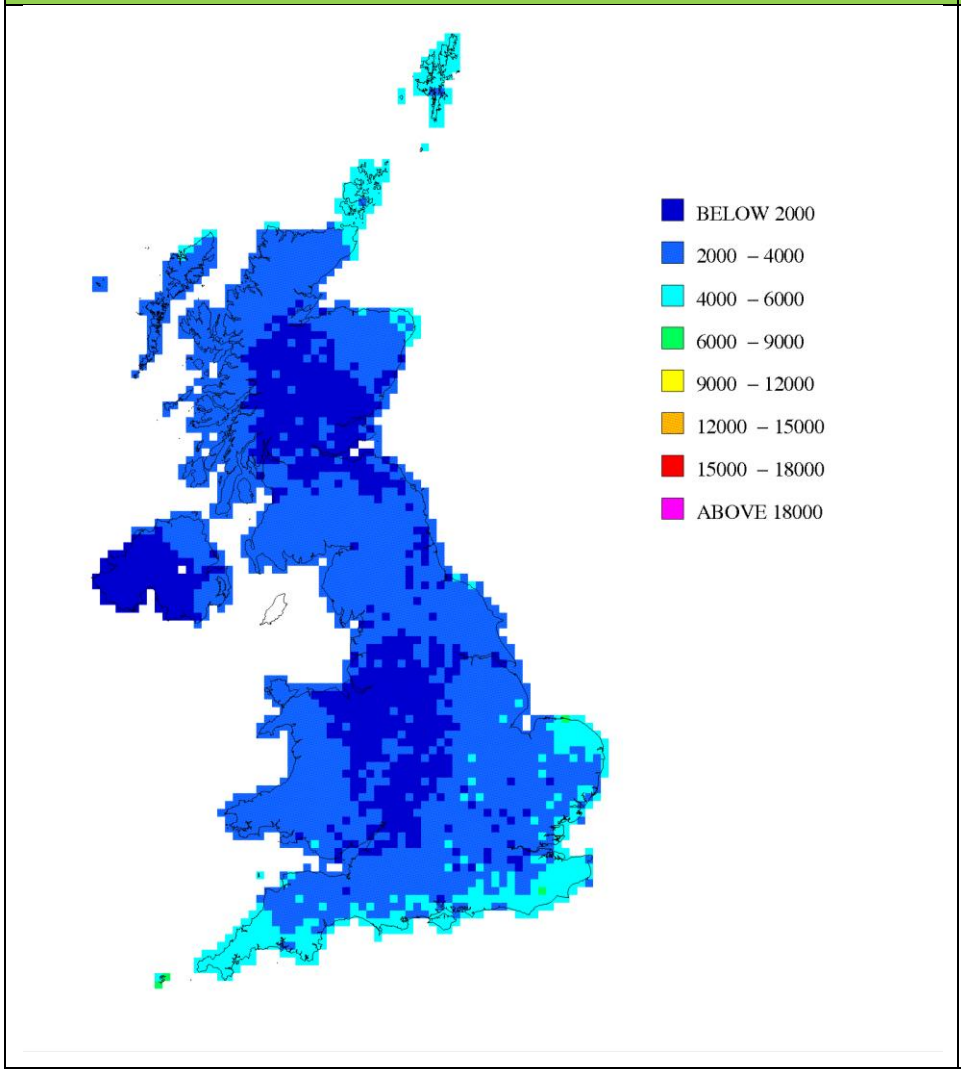
The OSRM maps show broadly similar patterns to the UKAAQA maps, with higher concentrations in East Anglia and in the south of England, but there are some specific spatial differences. The modelled metrics in North-East Scotland and South-West England are higher for the OSRM than the UKAAQA model, while the modelled metrics in East Anglia are higher for the UKAAQA than the OSRM.

An evaluation of OSRM and UKAAQA model performance has also been undertaken, comparing model results for 2011 with measured concentrations and against each other.

[5] http://naei.defra.gov.uk/data/map-uk-das

Figure 4.1: AOT40 maps of ozone for 2011 from the OSRM and UKAAQA models

(a) Map of AOT40 for 2011 (μgm^{-3}.hours) (OSRM)



(b) Map of AOT40 for 2011 (μgm^{-3}.hours) (UKAAQA)

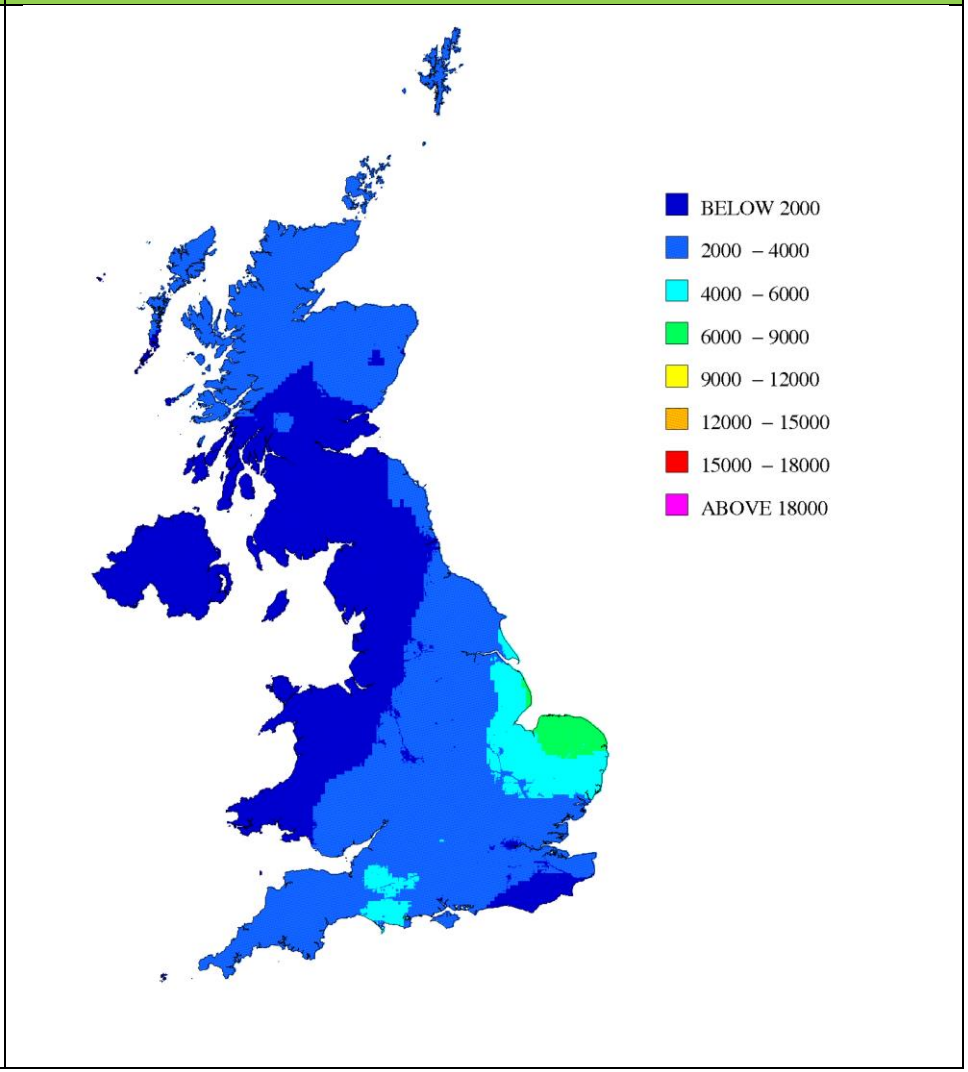


Figure 4.2: Number of days exceeding 120 µgm^{-3} maps for 2011 from the OSRM and UKAAQA models

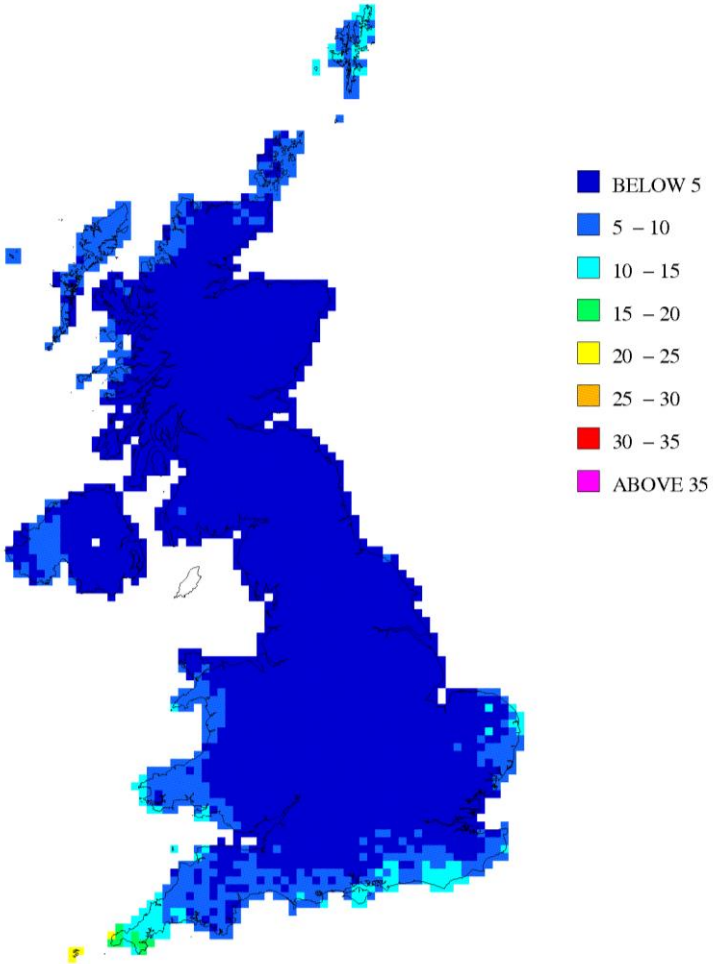
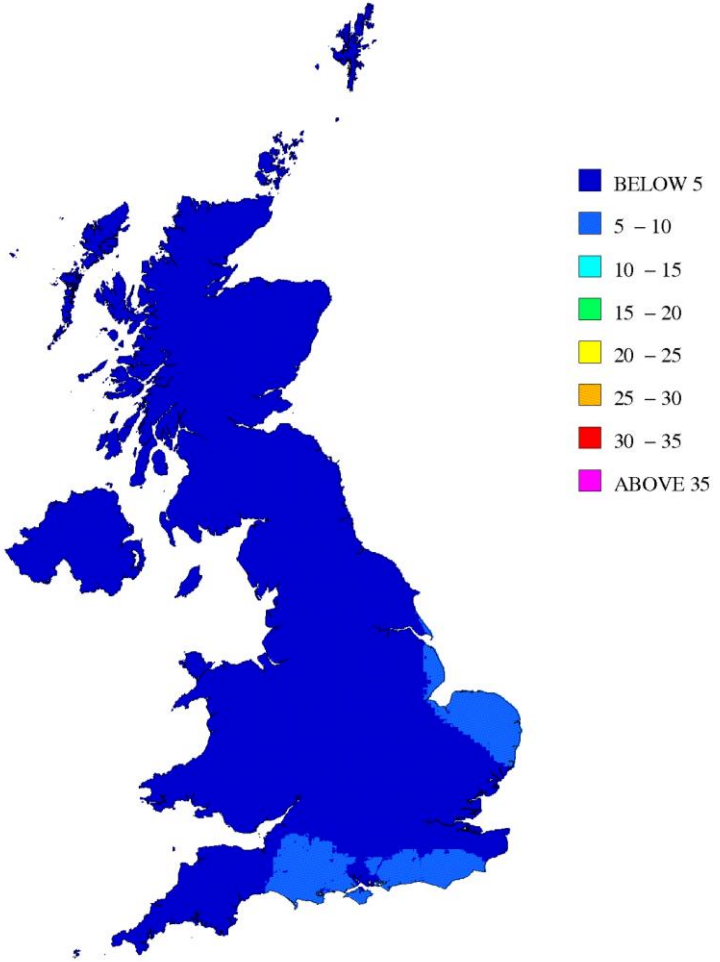
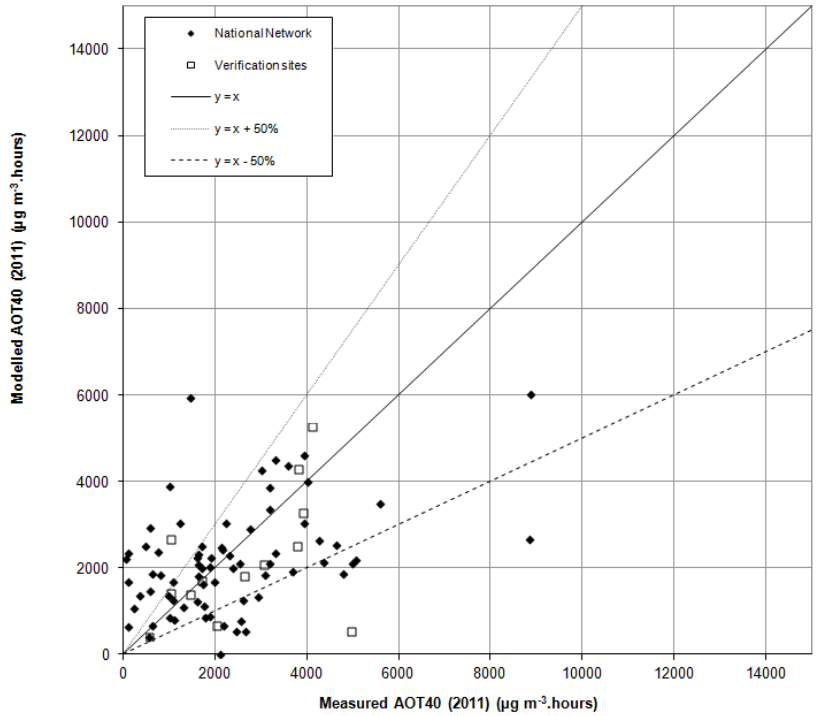
(a) Map of number of days exceeding 120 µgm^{-3} for 2011 (OSRM)	(b) Map of number of days exceeding 120 µgm^{-3} for 2011 (UKAAQA)
	

Figure 4.3: AOT40 verification plots for 2011 for the OSRM and UKAAQA models

(a) OSRM - AOT40 verification plot for 2011 (µgm^{-3}.hours)



(b) UKAAQA - AOT40 verification plot for 2011 (µgm^{-3}.hours)

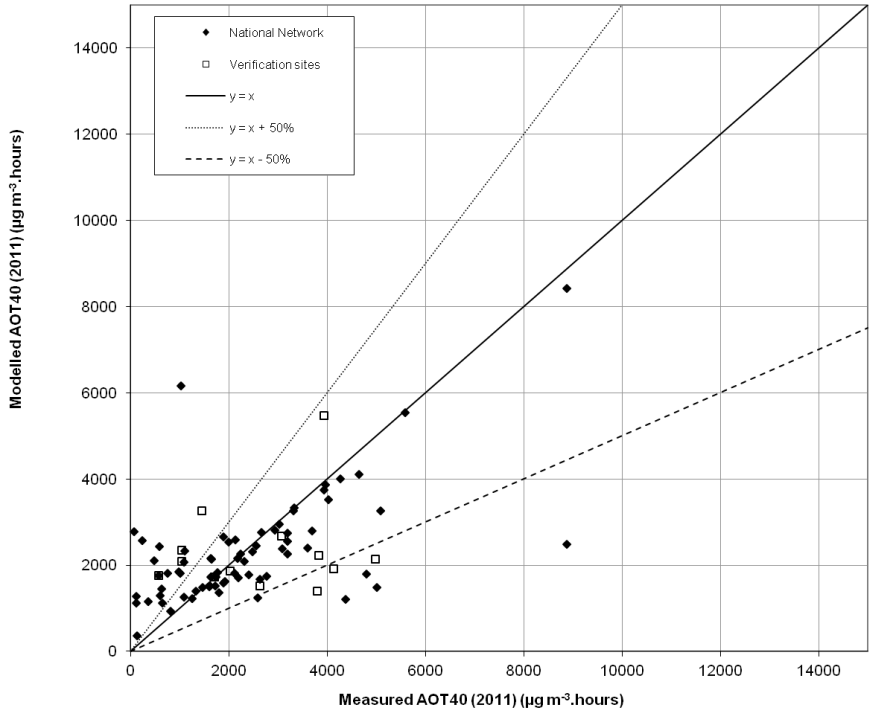
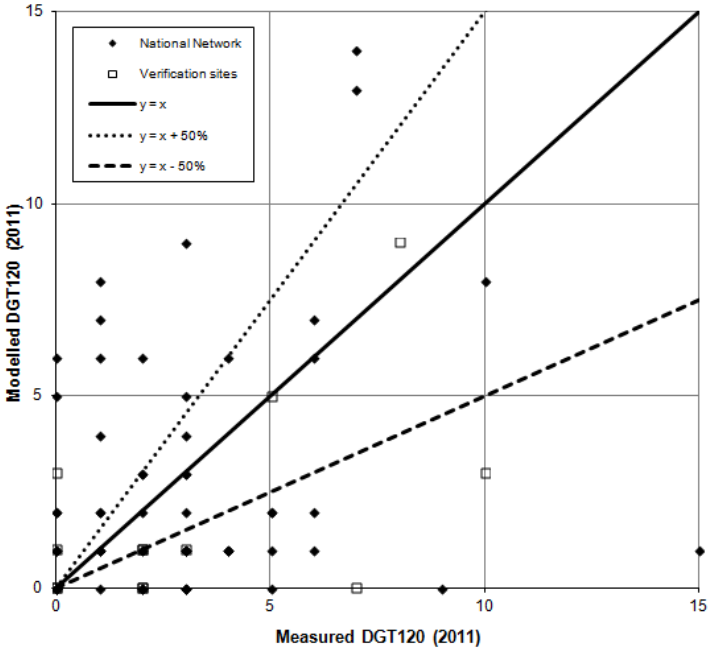
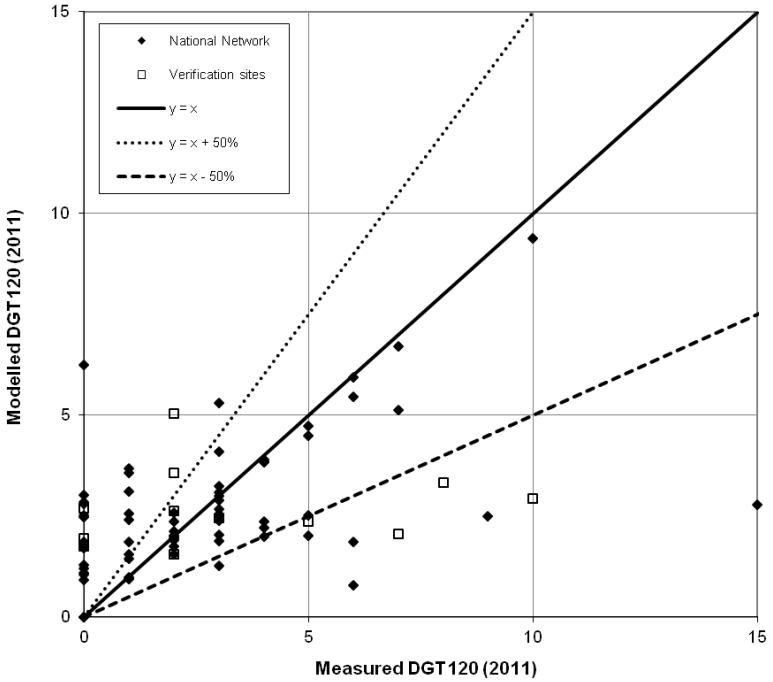


Figure 4.4: Number of days exceeding 120 µgm^{-3} verification plots for 2011 for the OSRM and UKAAQA models

(a) OSRM - number of days exceeding 120 µgm^{-3} verification plot for 2011	(b) UKAAQA - number of days exceeding 120 µgm^{-3} verification plot for 2011
	

Verification plots for the AOT40 and number of days exceeding 120 μgm^{-3} metrics are shown in Figures 4.3 and 4.4, respectively. Each plot shows data for the OSRM and UKAAQA. The 1:1 line and data quality objective (+/-50%) lines are also shown. Two groups of sites are presented in the verification charts: national network (AURN) monitoring sites and verification sites. The AURN sites were used as a direct input to the UKAAQA model and therefore provide a useful check during the verification process, but are not able to provide a completely independent representation of model performance. For this reason there is a separate group of sites labelled 'verification sites' that are completely independent from the UKAAQA model. These typically come from local authorities, research institutions and ad-hoc monitoring campaigns for which Ricardo-AEA holds and ratifies the data. These monitoring data are ratified to the same standard as the AURN. Both groups of sites provide an independent verification of the OSRM because this is a process based model which does not use monitoring data as an input or a calibration method. A data capture threshold of 75% has been applied to the monitoring data prior to analysis.

Table 4.1 summarises the average of the measured AOT40 concentrations, the average of the modelled AOT40 estimates, the R^2 of the fit line (Figure 4.3), the number of monitoring sites used and the percentage of these monitoring sites that fall outside the Data Quality Objective (DQO) for both the OSRM and UKAAQA results. Table 4.2 shows the equivalent data for the number of days exceeding 120 μgm^{-3} metric.

Table 4.1: AOT40 metric verification summary for the OSRM and UKAAQA model results for 2011

	Mean of measurements (μgm^{-3}.hours)	Mean of model estimates (μgm^{-3}.hours)	R^2	% outside DQO	No. sites used in assessment
National network (UKAAQA)	2333	2294	0.35	32%	72
Verification sites (UKAAQA)	2627	2339	0.03	54%	13
National network (OSRM)	2333	2171	0.20	42%	72
Verification sites (OSRM)	2627	2158	0.21	23%	13

Table 4.2: Number of days exceeding 120 μgm^{-3} metric verification summary for the OSRM and UKAAQA model results for 2011

	Mean of measurements (days)	Mean of model estimates (days)	R^2	% outside DQO	No. sites used in assessment
National network (UKAAQA)	2.6	2.6	0.24	42%	73
Verification sites (UKAAQA)	3.2	2.6	0.04	77%	13
National network (OSRM)	2.6	2.4	0.09	62%	73
Verification sites (OSRM)	3.2	1.8	0.30	54%	13

The measured mean values in the tables above for AOT40 and number of days exceeding 120 μgm^{-3} indicate that 2011 was a relatively low ozone year. This was evident from the maps when compared with maps developed for previous years. It can also be seen in the tables above that on average the OSRM under-predicts the metrics for 2011. This is unusual because previously the OSRM has tended to over-predict the metrics in a 'low' ozone year.

Past analysis (Hayman et al, 2006b, Murrells et al, 2009, Murrells et al, 2012) has shown that the OSRM slightly under-predicts measured concentrations in some cases and slightly over-predicts measured concentrations in others. In general, it has under-predicted ozone metrics in high ozone years (e.g. 2003 and 2006) and slightly over-predicted ozone metrics in low

ozone years (2004, 2005, 2007, 2009 and 2010). In 2008, which was considered a moderate ozone year, the OSRM generally under-predicted AOT40 concentrations.

Tables 4.3 and 4.4 below present the average measured and average OSRM modelled results for the years 2004-2011. These show the model performance in each year for both metrics, including during high (e.g. 2006) and low (e.g. 2004, 2005, 2007, 2009, 2010) ozone years. The OSRM results for later years are not directly comparable with earlier years because of model improvements to emissions and boundary conditions and changes in the meteorology data format. Nevertheless, the OSRM results seem to be consistent with the measurements for 2011 indicating it being a relatively low ozone year in terms of the mean of these metrics. The consistency also appears better in the simulations for 2011 than those for 2009 and 2010.

Table 4.3: OSRM results for AOT40 (µgm^{-3}.hours) for the years 2004 - 2011

		National network		Verification sites	
Year Modelled	**NAEI Year**	**Mean of measured**	**Mean of modelled**	**Mean of measured**	**Mean of modelled**
2004	2003	2888	2056	3681	2256
2005	2004	3650	4165	3810	3088
2006	2005	10497	5043	5061	6574
2007	2006	2281	4503	3061	5211
2008	2007	6025	4444	4913	4559
2009	2008	3182	4274	2738	3818
2010	2009	2244	4404	2518	4150
2011	2010	2333	2171	2627	2158

Table 4.4: OSRM results for number of days exceeding 120 µgm^{-3} for the years 2004 - 2011

		National network		Verification sites	
Year Modelled	**NAEI Year**	**Mean of measured**	**Mean of modelled**	**Mean of measured**	**Mean of modelled**
2004	2003	13	12	7	6
2005	2004	3	6	4	5
2006	2005	13	8	8	8
2007	2006	2	4	2	6
2008	2007	5	6	5	7
2009	2008	1	4	1	4
2010	2009	1	8	2	9
2011	2010	3	2	3	2

4.2 Analysis of Episodes of Photochemical Ozone in the United Kingdom during 2011 Using the PTM

4.2.1 Overview of ozone episodes in 2011

The 2011 ozone season began early with long dry periods of hazy sunshine across much of England during the second half of March. April 2011 was the warmest April in the entire 353-year long Central England Temperature (CET) record. It was exceptionally dry over much of eastern, central and southern England and sunshine duration was 50 – 75% above average in East Anglia and south east England. This fine, pleasant weather persisted into the first part of May but, after this, generally cyclonic weather systems prevailed throughout the remainder of May and for the remainder of the summer months (Eden, 2011). Anticyclonic conditions returned occasionally throughout the remainder of the 2011 ozone season which finished towards the end of October. As a result, the 2011 ozone season was unremarkable in terms of severity or number of its ozone episodes and its only memorable feature was the length of the season from early-April to early-October.

Air quality data were obtained from the UK air quality archive for the rural ozone monitoring stations and were benchmarked using the daily maximum 8-hour running mean ozone concentrations. Detailed focus has been given here to the rural Aston Hill monitoring station, a site near the mid-Wales-English border. This site reported 18 days with daily maximum 8-hour running mean levels in excess of 50 ppb, the WHO air quality guideline (WHO, 2006) during 2011. The highest daily 8-hour running mean of 66.5 ppb was recorded on the 21st April 2011.

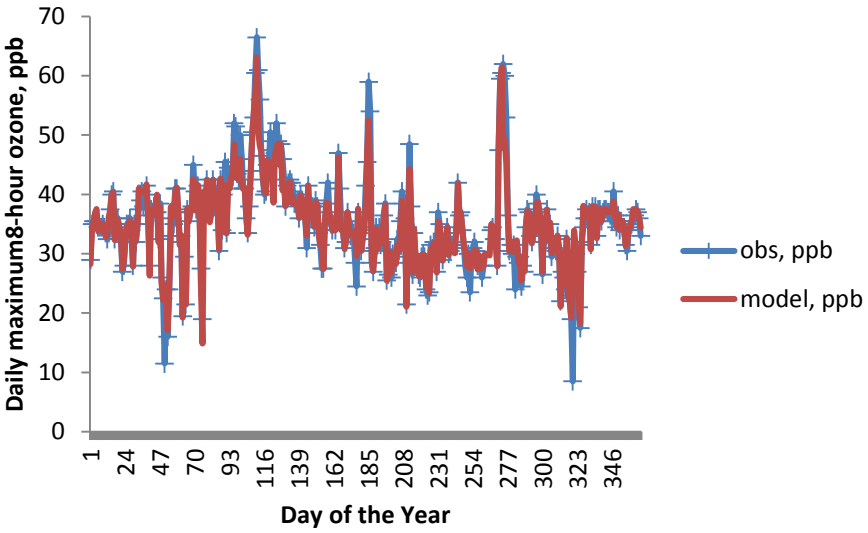
Back-track air mass trajectories from NILU (2012) were examined for the closest UK location at Harwell, Oxfordshire for each day of 2011. For 4 days out of the 18 potential episode days with daily maximum 8-hour running mean ozone levels in excess of 50 ppb, the air mass trajectories stretched backwards in a westerly direction and out over the North Atlantic Ocean. These days were classified as 'background' episodes and were given no further attention. This left 14 days that merited further attention with the PTM model as possible episodes of photochemical ozone.

4.2.2 Investigation of 2011 episodes of photochemical ozone at Aston Hill

The Photochemical Trajectory Model (PTM) was set up for 2011 conditions using appropriate input data where possible for a range of rural ozone monitoring stations. 96-hour back-track air mass trajectories were provided by the Met Office using the NAME model. 30 equally-probable, 96-hour 3-D back-track trajectories were made available for 15:00 to 15:15 hours on each day of 2011 for each site. Emissions data were obtained from EMEP by scaling national totals for SO_2, NO_x, VOCs, CO and NH_3. Initial and background concentrations for CH_4, CO, H_2 and O_3 were taken from the observed monthly mean baseline levels at Mace Head, Ireland.

The PTM model was run for each day of 2011 for Aston Hill using the CB-05 chemical mechanism and the results are shown in Figure 4.5, together with the observations. The observed average daily maximum 8-hour mean ozone level over 2011 was found to be 35.29 ppb compared with 34.05 ppb with the PTM model for the average mid-afternoon level. Over the year, the mean fractional bias was found to be 0.0068, indicating an excellent degree of model performance against observations. Over the episode days, model performance was somewhat poorer overall with a mean fractional bias of 0.099. The poorest model performance was found on 1st October, when the mid-afternoon predicted level from the PTM was 48.07 ppb compared to the observed daily maximum 8-hour running mean level of 62.0 ppb, a fractional bias of -0.22. This is just outside the target range for the fractional bias of -0.2 to +0.2 (Derwent et al., 2010c), representing acceptable model performance.

Figure 4.5. Observed daily maximum 8-hour running mean (+ signs) and model mid-afternoon (line) ozone levels for each day of 2011 for the rural Aston Hill monitoring site.



To assist in diagnosing the origins and understanding the nature of the 2011 episodes of photochemical ozone, four additional scenarios addressing man-made emissions were implemented in the PTM in addition to the base case scenario, as follows:

- S1: 30% reduction in NO_x emissions across the UK and the Rest of Europe (RoE),
- S2: 30% reduction in VOC emissions across the UK and the RoE,
- S3: 30% reduction in NO_x emissions across the UK,
- S4: 30% reduction in VOC emissions across the UK.

If the ozone response, base case – scenario case, to 30% emissions reductions was greater for S1 compared with S2, then that day was assigned as NO_x-sensitive and vice versa, VOC-sensitive. If the ozone response to 30% emissions reductions was greater for S4 compared with S2 – S4, then that day was assigned as UK-dominated and vice versa, RoE-dominated.

Table 4.5 presents the assignments as to NO_x- or VOC-sensitive and UK- or RoE-dominated for each of the 14 remaining ozone episode days, after the removal of the background days. A total of 10 out of the 14 days were assigned to the RoE-dominated category and 4 were UK-dominated. All of the days except one were assigned to the NOx-sensitive category. Generally speaking, ozone episodes have shown a tendency to fall into the VOC-sensitive, RoE-dominated category in the past. Examination of the NILU back-track air mass trajectories for the 14 episode days shows a greater than usual preponderance of southerly rather than easterly trajectories. Southerly trajectories are not generally associated with high ozone levels whereas easterly trajectories are most certainly. It is suggested that the preponderance of southerly trajectories for the 14 episode days was the reason that the majority were NOx-sensitive.

Table 4.5. NO_x- versus VOC-sensitivity and UK- versus RoE-dominated assignments for each ozone episode day during 2011 for Aston Hill, also showing the observed (obs) daily maximum 8-hour running mean ozone level.

Date	Obs, ppb	NO_x- or VOC-sensitivity	UK- or RoE-dominant
10/04/11	50.0	NO_x	UK
18/04/11	50.5	NO_x	RoE
19/04/11	53.0	NO_x	RoE
20/04/11	60.5	VOC	RoE
21/04/11	66.5	NO_x	RoE
22/04/11	61.0	NO_x	RoE
23/04/11	56.0	NO_x	UK
30/04/11	50.5	NO_x	UK
04/05/11	52.0	NO_x	RoE
04/07/11	59.0	VOC	UK
29/09/11	59.5	NO_x	UK
30/09/11	60.5	NO_x	RoE
01/10/11	62.0	NO_x	RoE
02/10/11	60.0	NO_x	RoE

5 Optimising Emissions Inventory Information in the OSRM

Objective 4 of the work programme involved improving the efficiency and transparency of outputs from the OSRM by optimising the use of emissions inventory information.

One of the main purposes of the OSRM is to simulate UK ozone concentrations for different precursor emission scenarios. This can require changes in emissions from sources in the UK or the rest of Europe, or both. Sometimes Defra require a whole batch of simulations to be run, with various permutations of emission changes for individual countries, groups of countries and sometimes for specific source sectors. The emission changes are applied as scaling factors to gridded datasets covering the UK (NAEI 1 x 1 km) and rest of Europe (EMEP 50 x 50 km).

With potentially large batches of runs required at any one time, it is important that the emission files in the OSRM can be set up efficiently using codes that are flexible enough to cope with an increasingly wide array of emission change scenarios. It is also important that emission pre-processing is sufficiently transparent for the model outputs to be traced back easily to the relevant input data files.

The OSRM has not kept pace with some of the changes that have been made to the emissions inventory information required for the type of model runs requested by Defra to support ozone policy development and analysis. For example, the OSRM needs to be able to treat different emission change factors for different countries. It is also necessary to use more detailed gridded datasets on emissions from shipping covering sea territories that overlap the areas covered by the NAEI and EMEP.

Work was therefore undertaken to improve the efficiency and flexibility of the emission scenario pre-processor and of the treatment of shipping emissions in the OSRM.

5.1 Treatment of Shipping Emissions

Although shipping emissions are not expected to have a significant impact on UK ozone, it is important that the OSRM has the capability of being able to change these emissions, just as it is for other source sectors when modelling any future emission scenario. This is particularly the case given that future levels of emissions from shipping are expected to change in response to regulations on ship engine emissions and fuels through the MARPOL Annex VI agreement and similar EU legislation, including the potential for NO_x emission control areas in the future.

There are also further refinements being made by the NAEI on how ship emissions are spatially resolved near major port areas. It will be necessary for the OSRM to be able to accommodate these improvements.

5.1.1 Improvements made

A new 1 x 1 km map of shipping emissions has been created, based on the 5 x 5 km map of shipping emissions around UK waters originally developed by Entec (2010)[6]. This new map extends out to cover almost the same area as the NAEI emissions maps for other sectors, including offshore oil and gas installations. Previously the NAEI shipping emissions maps

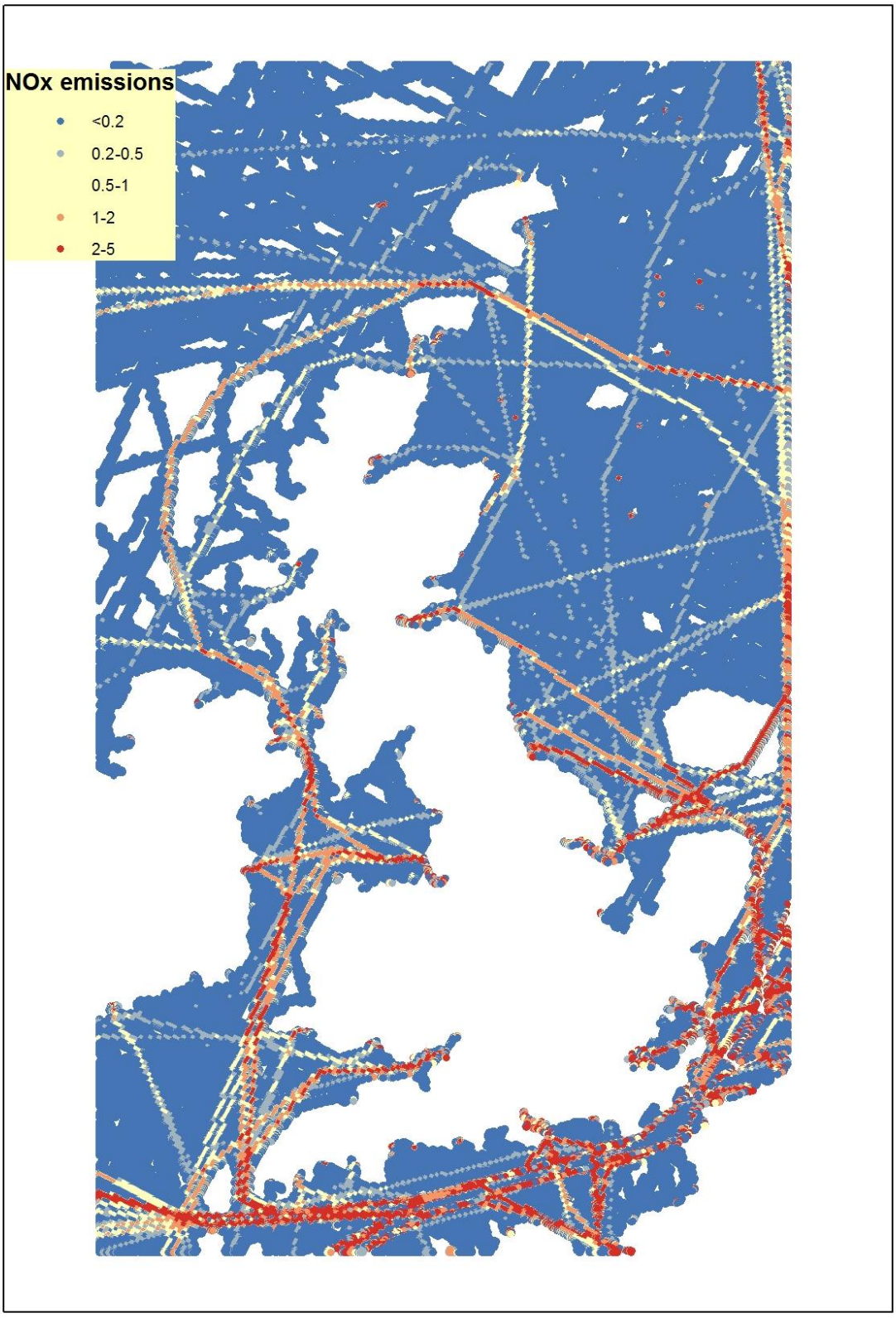
[6] The work was completed and reported on by Entec who now operate as AMEC, but the work is referred to as Entec (2010) throughout this report.

used in the OSRM only included shipping emissions within UK territorial waters, 12 nautical miles off the coastline.

The OSRM uses a mask to define the area where NAEI emissions, rather than EMEP emissions, are used as the input into the OSRM. Previously this mask covered UK land area and some of the surrounding sea. A new mask has been developed to use NAEI emissions over a larger area. This mask covers a much larger area than the previous one, with “holes” over southern Ireland and France where the EMEP emissions are used. This mask is needed in order for the OSRM to use the new shipping emissions map based on the Entec data developed for use in the OSRM rather than the EMEP data. The masked data are also being used in the CMAQ regional air quality model.

Figure 5.1 shows the new map of NO_x emissions from shipping developed from the NAEI and used for the OSRM. The shape of the land mass which makes up the UK can clearly be seen. The white areas which appear to distort the more familiar shape of the UK land mass include sea areas where there are no shipping emissions. However, the key shipping lanes can clearly be made out.

Figure 5.1: 1 x 1 km map of NO_x emissions from shipping used in the OSRM



As before, the OSRM pre-processor converts the 1 x 1 km grid of NAEI emissions to a 10 x 10 km grid for use in the model. A 1 x 1 km grid is used for post-processing the ozone results, taking account of reaction of ozone with the locally emitting NO_x.

5.1.2 Results from the OSRM using the new shipping emissions

The OSRM has been successfully run using the new shipping emissions data. Figures 5.2 and 5.3 show the UK maps for the AOT40 metric and the number of days exceeding 120 μgm^{-3} metric modelled using the new shipping emissions. These maps are shown alongside equivalent maps developed using the current version of the OSRM with old shipping emissions data, i.e. the same maps as shown earlier in Figures 4.1(a) and 4.2(a).

Figure 5.2: AOT40 maps of ozone for 2011 from the OSRM (using the old and new shipping emissions)

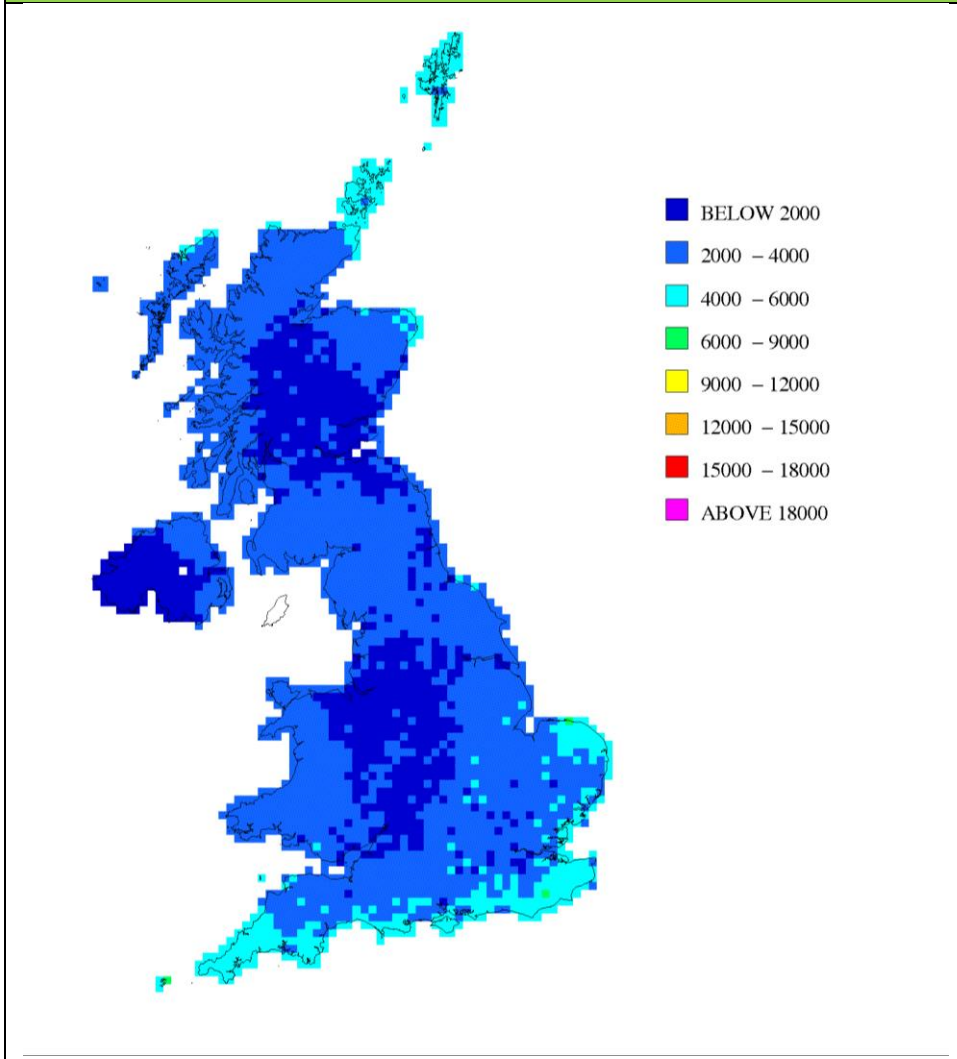
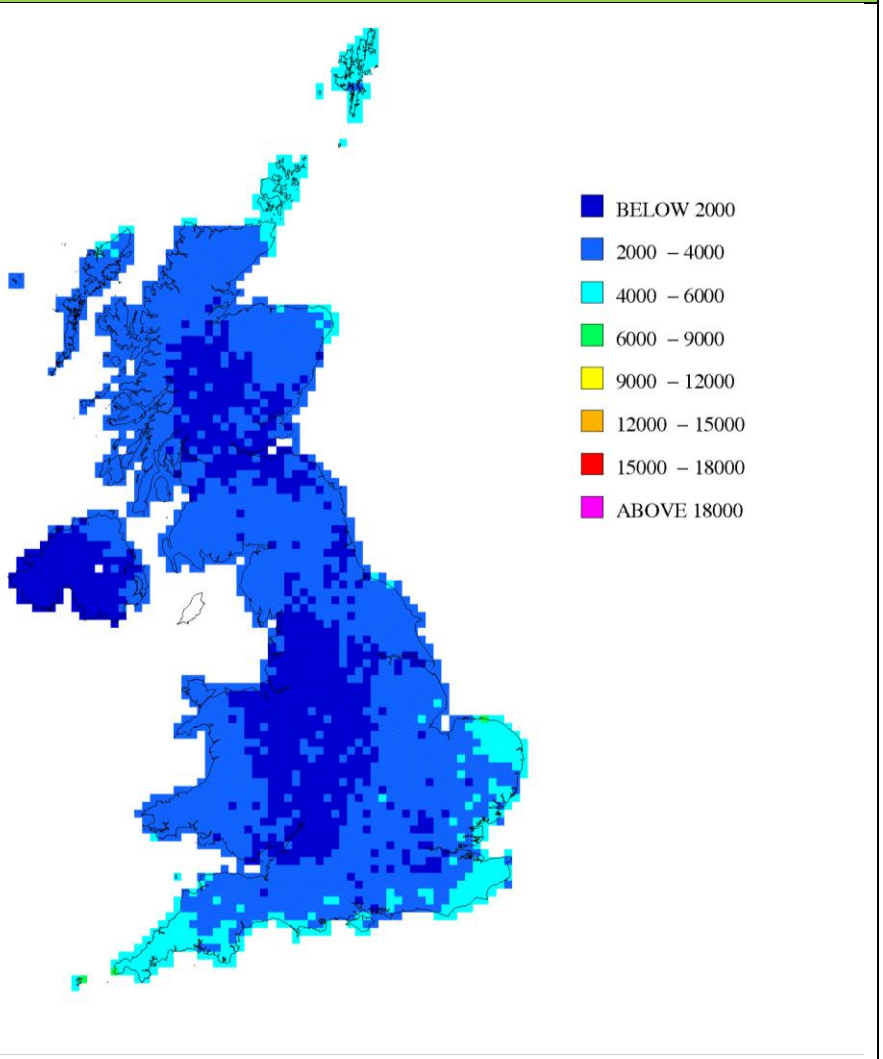
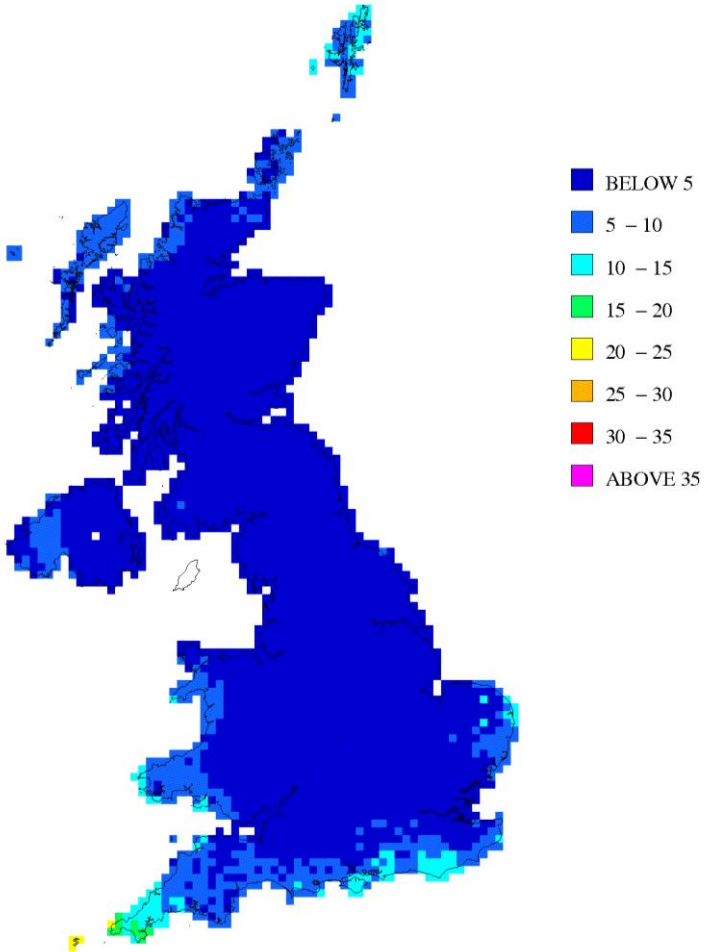
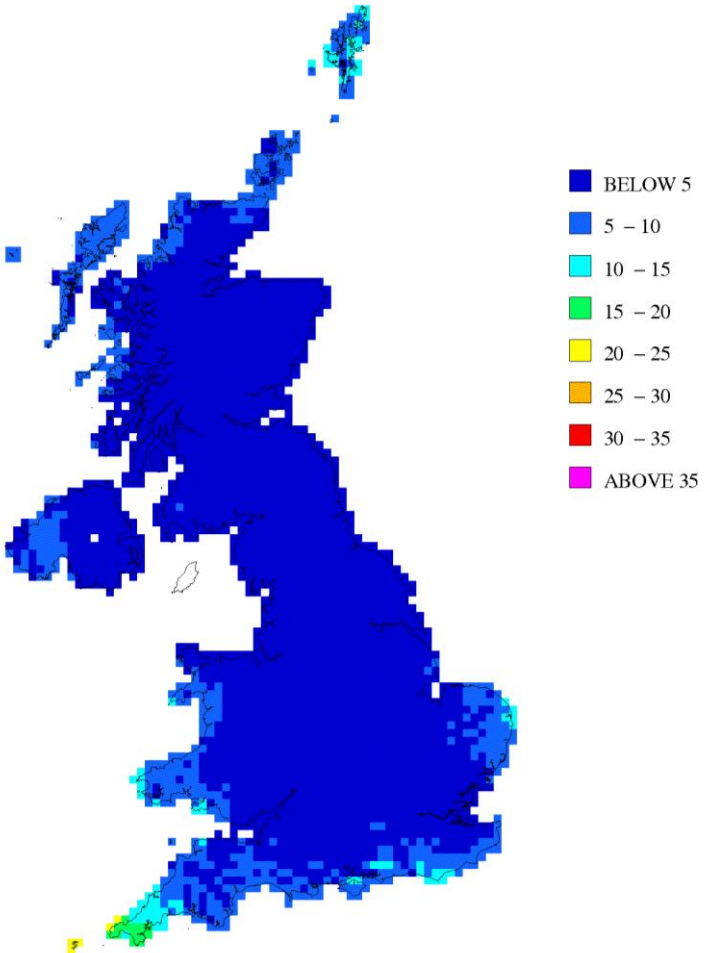
(a) Map of AOT40 for 2011 (μgm^{-3}.hours) (OSRM – old shipping)	(b) Map of AOT40 for 2011 (μgm^{-3}.hours) (OSRM – new shipping)
	

Figure 5.3: Number of days exceeding 120 µgm^{-3} maps for 2011 from the OSRM (using the old and new shipping emissions)

(a) Map of number of days exceeding 120 µgm^{-3} for 2011 (OSRM – old shipping)	(b) Map of number of days exceeding 120 µgm^{-3} for 2011 (OSRM – new shipping)
	

Tables 5.1 and 5.2 are summary tables showing the average of the measured concentrations, the average of the modelled estimates and the R^2 of the fit line in terms of the relationship between measured and modelled values for the OSRM results using both the old and new shipping emissions. The results for the AOT40 metric are shown in Table 5.1 and the results for the number of days exceeding 120 μgm^{-3} metric are shown in Table 5.2.

Table 5.1: AOT40 metric verification summary for the OSRM model results (using the old and new shipping emissions) for 2011

	Mean of measurements (μgm^{-3}.hours)	Mean of model estimates (μgm^{-3}.hours)	R^2	% outside DQO	No. sites used in assessment
National network (OSRM old shipping)	2333	2171	0.20	42%	72
Verification sites (OSRM old shipping)	2627	2158	0.21	23%	13
National network (OSRM new shipping)	2333	2034	0.18	43%	72
Verification sites (OSRM new shipping)	2627	2061	0.18	31%	13

Table 5.2: Number of days exceeding 120 μgm^{-3} metric verification summary for the OSRM model results (using the old and new shipping emissions) for 2011

	Mean of measurements (days)	Mean of model estimates (days)	R^2	% outside DQO	No. sites used in assessment
National network (OSRM old shipping)	2.6	2.4	0.09	62%	73
Verification sites (OSRM old shipping)	3.2	1.8	0.30	54%	13
National network (OSRM new shipping)	2.6	2.2	0.07	55%	73
Verification sites (OSRM new shipping)	3.2	1.3	0.20	38%	13

Overall, the results produced by the OSRM for 2011 using the new shipping emissions are similar to those produced using the old shipping emissions. The averages of the AOT40 metric at both national network and verification sites are slightly lower with new shipping emissions. This means that the average AOT40 values from the OSRM run using old shipping are slightly closer to the average measured values than the average values from the OSRM run using new shipping emissions. The R^2 and percentage outside DQO statistics are not as good for the new shipping as the old shipping, though the changes in these statistics are very slight. For the number of days exceeding 120 μgm^{-3} metric, the average values are slightly lower for the national network sites and verification sites when using the new shipping emissions. This means that the average values from the OSRM run using the new shipping are not as close to the average measured values as the average values from the OSRM run using old shipping emissions. The R^2 values are slightly worse when using the new shipping emissions, but the percentage outside DQO statistics are slightly better when the new shipping emissions are used. However, like the AOT40 results, the changes in these statistics are not large.

The improvements to the shipping emissions data used in the OSRM were not expected to make large differences in the output metrics. The use of the new shipping emissions data in the OSRM will increase the flexibility to run more complex shipping emission scenarios,

accommodating the greater detail in the shipping inventory that may be required in the future. These improvements also mean that the OSRM is using higher resolution input data that have been produced in a more robust and traceable way. Use of a larger mask means the OSRM is now able to capture the NAEI maps of other offshore emission sources at higher resolution than before when these sources would have been taken from the EMEP grid.

5.2 Improvements to the OSRM Emission Scenario Pre-processor

Work has been undertaken to improve the efficiency and flexibility of the emission scenario pre-processor in the OSRM. Essentially, a new OSRM pre-processor for NAEI emissions has been built. This new pre-processor has been built using VBA in a spreadsheet, rather than in Fortran code which has been used previously. This new pre-processor allows more individual sub-sectors to be scaled in an automated way for scenarios. Previously, only the 8 SNAP sectors could be individually scaled in an automated way and for more complex scenarios, the emissions for individual sub-sectors had to be calculated manually and re-entered into the model. The use of this new NAEI pre-processor improves the flexibility in which scenarios can be modelled without manually having to re-calculate the emissions totals.

As the new pre-processor calculates new emissions totals in a spreadsheet, scaling factors given to many decimal places can be used. Previously, even if the scaling factors were calculated manually to many decimal places only two decimal places could be used for scaling the emissions within the OSRM, limiting the accuracy by which scenarios could be represented and modelled.

The new pre-processor is simpler and more transparent and improves the traceability and audit trail of the emissions used in the OSRM. It also improves the efficiency of calculating the emissions needed for OSRM modelling.

The emission inputs to the OSRM that are produced by the new pre-processor are in the same format as produced by the old pre-processor, so no changes were needed to the OSRM model code.

Improvements have also been made to the EMEP emissions pre-processor for the OSRM. The code has been adapted so that csv format input files are used, rather than prn format files which were used previously. The prn files were very sensitive to the formatting and spacing within the files and it was time consuming to ensure that this was correct. Using csv files therefore improves the efficiency of the EMEP pre-processing.

Further development of the EMEP pre-processor has not yet been carried out. This is because the Webdab EMEP projections data previously used (http://www.ceip.at/webdab-emission-database/emissions-as-used-in-emep-models/) are not currently available. These data are not likely to be available in the near future, so thought will be needed on what to use as an alternative. This work will also be informed by the ongoing NAEI improvements programme which is investigating appropriate European emissions data for modellers.

The new pre-processors have been tested and the results indicate that the new pre-processors are functioning correctly and can now be used in the pre-processing of emissions for OSRM model runs.

6 Policy Applications of the OSRM and PTM

Objective 2 involves the use of the OSRM and PTM to model ozone concentrations in the UK for different emission scenarios or meteorological conditions specified by Defra. During 2012/13 the OSRM has been used to model five scenarios, all involving national scale annual runs producing maps of ozone concentrations and population- and area-weighted means of health- and ecosystem-based metrics. The PTM has been used to model episodic peak ozone at 12 rural sites in 2011 for precursor emission reductions defined by the revised Gothenburg Protocol for EU27 countries.

6.1 OSRM runs for 2008 for Comparison with Future Year Simulations Done for Different Emission Scenarios

The OSRM had previously been used in 2011/12 to simulate UK ozone in 2020-2030 for different UK and European emission scenarios. The model was run using 2006 and 2007 meteorology as years representing photochemically active years with high ozone episodes in the summer (2006) and less active years with few ozone episodes (2007). The UK emission scenarios were based on the DECC UEP43 energy projections. Details of the OSRM runs undertaken then were given in the '*Modelling of Tropospheric Ozone'* Annual Report 2011 (Murrells et al, 2012).

Defra requested ozone simulations for a recent historic emissions year under the same meteorological conditions for a comparison with the future year model results to gain a better understanding of the trends over time in ozone concentrations due to the changes in emissions.

The year 2008 was run for this analysis. The aim was to be as consistent as possible with the emissions data used for the UEP43 future year scenarios. As there was limited time available to carry out these runs, emissions that were already processed and ready to use had to be used. Therefore the 2008 NAEI emissions totals and maps were used for the UK. The 2008 EMEP emissions maps and national totals were used for other European countries.

The OSRM was run twice using these emissions, once with 2006 meteorology and once with 2007 meteorology.

For these runs the Mace Head correction normally made for the initialisation conditions on each trajectory run for current year simulations was not used. This was to be as consistent as possible with the future year runs where the Mace Head correction was not used.

6.1.1 Results for 2008 runs

The results for the 2008 runs described above are presented in Tables 6.1 to 6.3. The tables show area-weighted means of the AOT40 metric, population-weighted means of the number of days exceeding 120 μgm^{-3} metric and area-weighted means of the annual mean metric respectively for different regions of the UK and the UK as a whole. In addition to the 2008 results, the results for 2020, 2025 and 2030 based on UK emissions from the UEP 43 central case (CCC) energy scenario are included in the tables for comparison. The future year results had been presented in Section 6.1 of the 2011 project report (Murrells et al, 2012).

Table 6.1: Area-weighted AOT40 (µgm^{-3}.hours)

Scenario	Scotland	Wales	Northern Ireland	Inner London	Outer London	Rest of England	All UK
2008 Base (Met 2006)	5771	6861	5983	4922	5015	6777	6391
2020 UEP43 CCC UK (Met 2006)	8855	10814	8912	12967	12414	12217	10789
2025 UEP43 CCC UK (Met 2006)	9809	11887	9850	15031	14338	13668	12016
2030 UEP43 CCC UK (Met 2006)	10958	13192	10957	16603	15870	15201	13377
2008 Base (Met 2007)	6773	5927	5227	3347	3620	5777	6075
2020 UEP43 CCC UK (Met 2007)	9893	9755	7623	10195	9788	10238	9929
2025 UEP43 CCC UK (Met 2007)	10835	11033	8407	12197	11578	11561	11093
2030 UEP43 CCC UK (Met 2007)	11942	12350	9406	13715	13044	12942	12356

Table 6.2: Population-weighted number of days exceeding 120 µgm^{-3}

Scenario	Scotland	Wales	Northern Ireland	Inner London	Outer London	Rest of England	All UK
2008 Base (Met 2006)	4.0	10.3	6.7	6.1	6.6	7.8	7.4
2020 UEP43 CCC UK (Met 2006)	7.0	15.6	11.5	14.3	13.7	13.8	13.3
2025 UEP43 CCC UK (Met 2006)	8.8	17.9	13.4	16.1	15.7	15.8	15.2
2030 UEP43 CCC UK (Met 2006)	11.7	20.9	15.8	17.3	18.7	18.3	17.8
2008 Base (Met 2007)	5.5	2.8	1.2	1.0	3.0	4.1	3.8
2020 UEP43 CCC UK (Met 2007)	10.4	8.1	2.6	11.6	11.0	8.5	8.8
2025 UEP43 CCC UK (Met 2007)	11.9	11.1	3.9	13.9	13.3	10.7	11.0
2030 UEP43 CCC UK (Met 2007)	13.5	14.8	5.4	15.8	15.6	13.2	13.4

Table 6.3: Area-weighted annual mean ozone (μgm^{-3})

Scenario	Scotland	Wales	Northern Ireland	Inner London	Outer London	Rest of England	All UK
2008 Base (Met 2006)	63.1	64.5	63.7	48.0	48.3	56.6	59.8
2020 UEP43 CCC UK (Met 2006)	68.9	71.5	69.4	63.2	62.5	66.2	67.7
2025 UEP43 CCC UK (Met 2006)	70.0	72.9	70.5	65.5	64.6	67.9	69.1
2030 UEP43 CCC UK (Met 2006)	71.0	74.0	71.3	66.5	65.7	69.0	70.2
2008 Base (Met 2007)	66.6	64.9	65.7	46.6	47.2	57.0	61.3
2020 UEP43 CCC UK (Met 2007)	72.1	72.3	71.8	62.6	62.1	66.7	69.3
2025 UEP43 CCC UK (Met 2007)	73.2	73.8	72.9	65.1	64.4	68.5	70.7
2030 UEP43 CCC UK (Met 2007)	74.1	75.0	73.8	66.2	65.6	69.6	71.8

Figures 6.1 and 6.2 show maps produced by the OSRM for 2008 and the 2020 UEP 43 central case (CCC) emission scenario (modelled using 2006 meteorology) for the AOT40 metric and the number of days exceeding 120 μgm^{-3} metric, respectively. Figures 6.3 and 6.4 show maps for the same metrics modelled by the OSRM using 2007 meteorology. The maps for the 2025 central case (CCC) scenario and the 2030 central case (CCC) scenario (and the other 2020 UEP 43 emission scenarios) are not reproduced here, but can be found in the project Annual Report 2011 (Murrells et al 2012).

The 2008 emission results from the OSRM runs using 2006 meteorology are lower for all metrics shown in the tables above than any of the results for the future emission years (with 2006 meteorology). The results from the runs using 2007 meteorology are also lower for all metrics shown in the tables above for 2008 emissions than for the future years. This is especially the case for London for the AOT40 and annual mean metrics.

The maps also show lower ozone concentration metrics across the country, but particularly for the south and east of England for the 2008 emission scenario relative to the 2020 case.

6.1.2 Interpretation of UK ozone trends predicted from 2008 to 2020

This is a feature previously observed from OSRM runs whereby ozone concentrations tend to be higher in future years when emissions are reduced and all other conditions are maintained the same. The increase has been attributed to the reductions in NO_x emissions. Although NO_x is a precursor to ozone production, local emissions of NO_x (e.g. from traffic) react with ozone creating a local ozone decrement. Thus, while reductions in NO_x across Europe will reduce ozone production, the reductions in NO_x across the UK will reduce the ozone decrement, in some areas leading to a net increase in ozone concentrations.

This feature has been observed in other modelling studies. For example, in a multi-model assessment of projected exposure to ozone using an ensemble of Chemical Transport Models (including EMEP), Collete et al (2012) noted that annual means of daily ozone predicted for 2030 increase over the Benelux/UK/Germany/Northern France area as a result of a less efficient titration by NO_x, which shows that the area was still saturated in NO_x. The

Figure 6.1: AOT40 maps for 2008 and 2020 UEP43 CCC emission scenario (with 2006 meteorology) modelled by OSRM

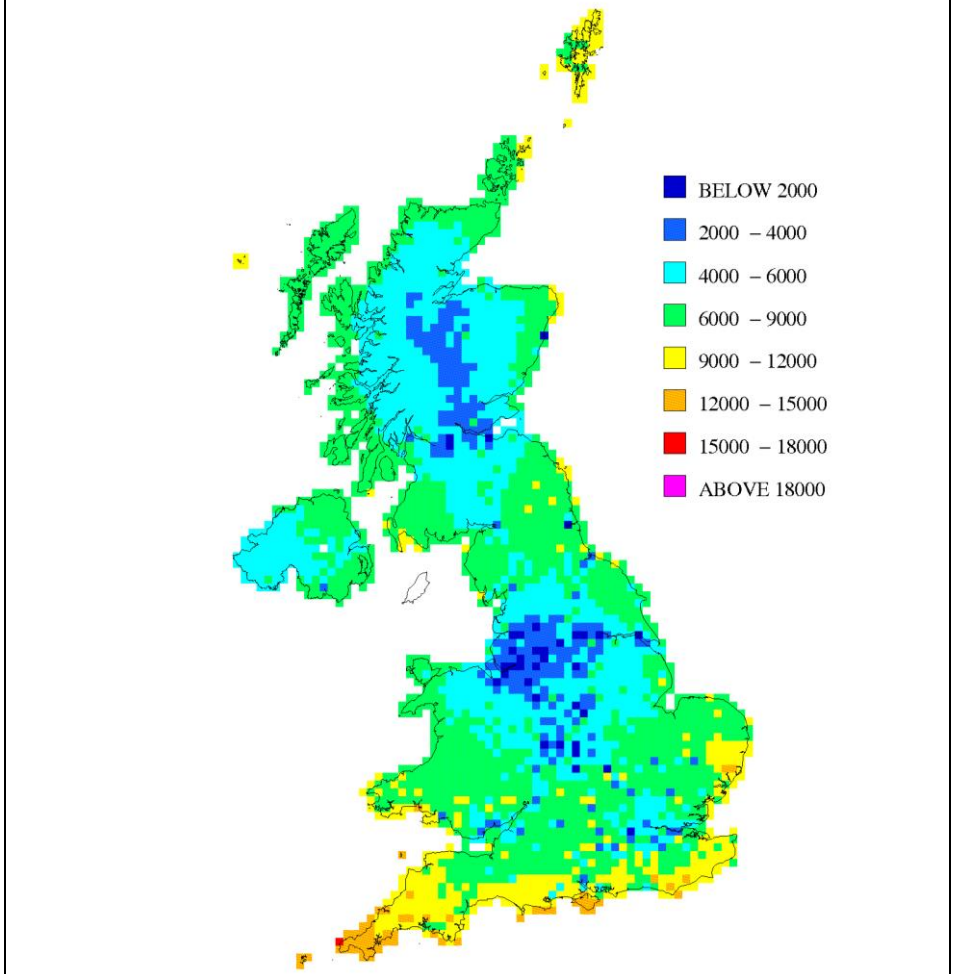
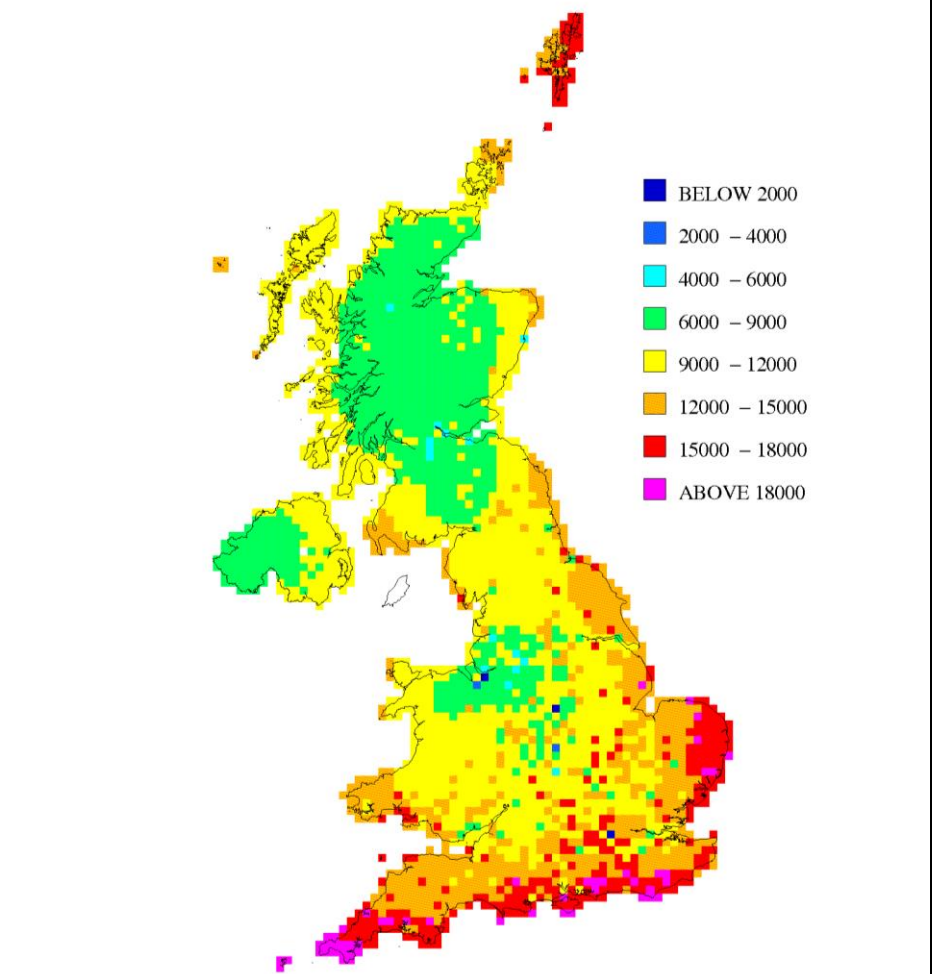
Map of AOT40 for 2008 (µgm^{-3}.hours) (2006 meteorology)	Map of AOT40 for 2020 CCC emission scenario (µgm^{-3}.hours) (2006 meteorology)
	

Figure 6.2: Number of days exceeding 120 µgm^{-3} maps for 2008 and 2020 CCC scenario (with 2006 meteorology) modelled by OSRM

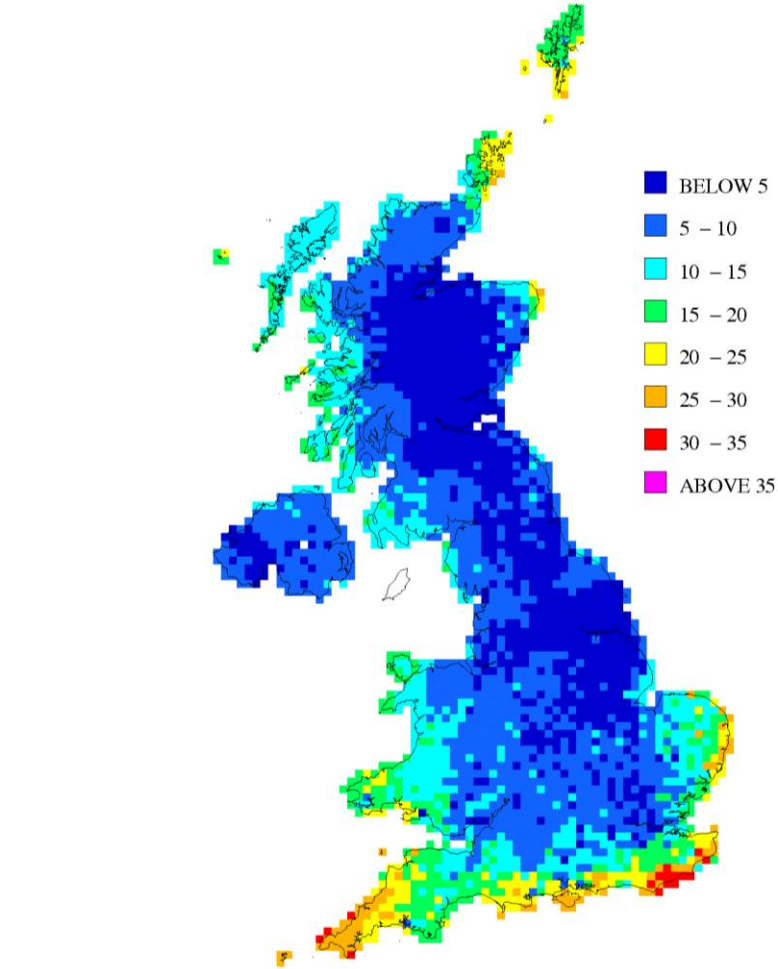
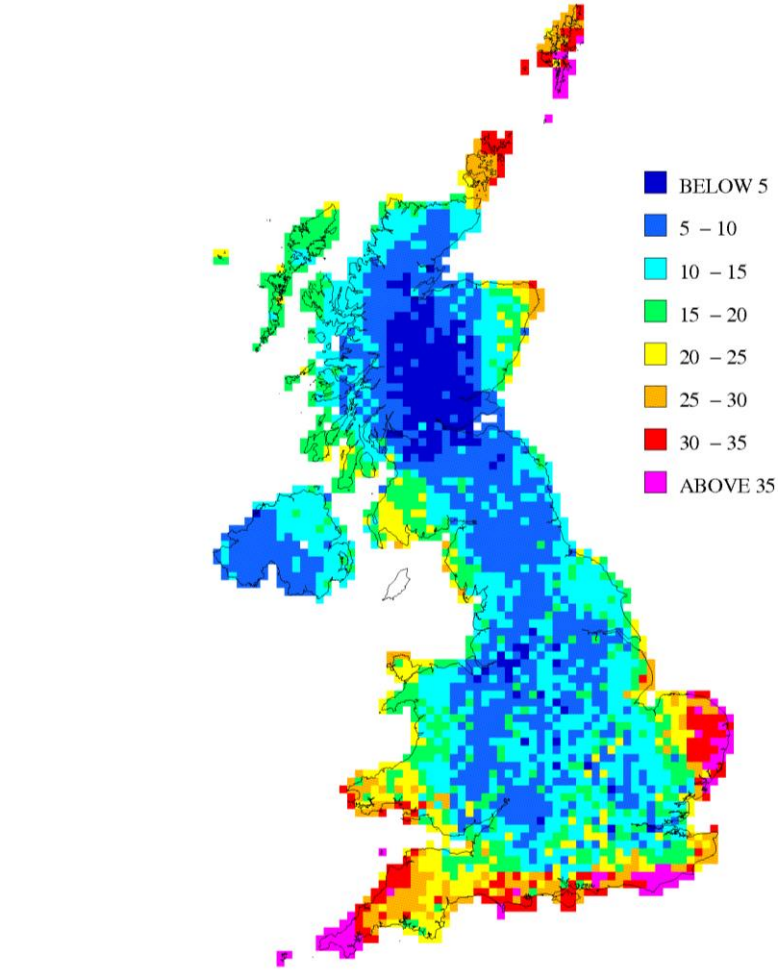
Map of number of days exceeding 120 µgm^{-3} for 2008 (2006 meteorology)	Map of number of days exceeding 120 µgm^{-3} for 2020 CCC emission scenario (2006 meteorology)
	

Figure 6.3: AOT40 maps for 2008 and 2020 UEP43 CCC emission scenario (with 2007 meteorology) modelled by OSRM

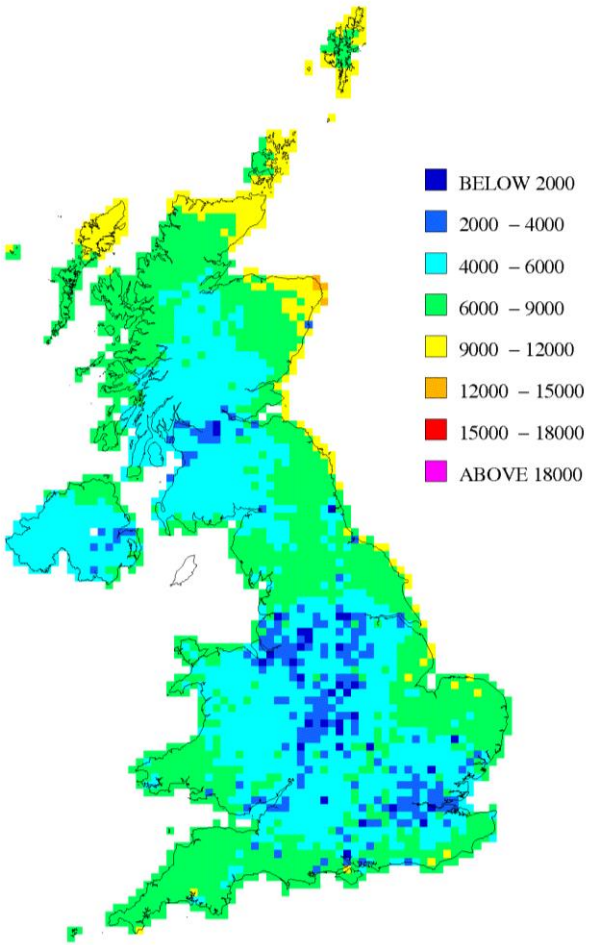
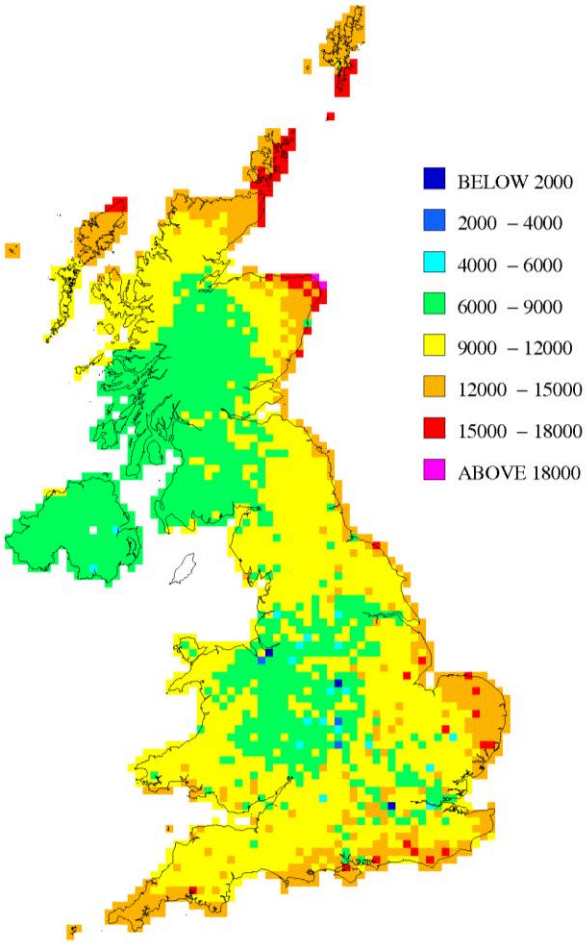
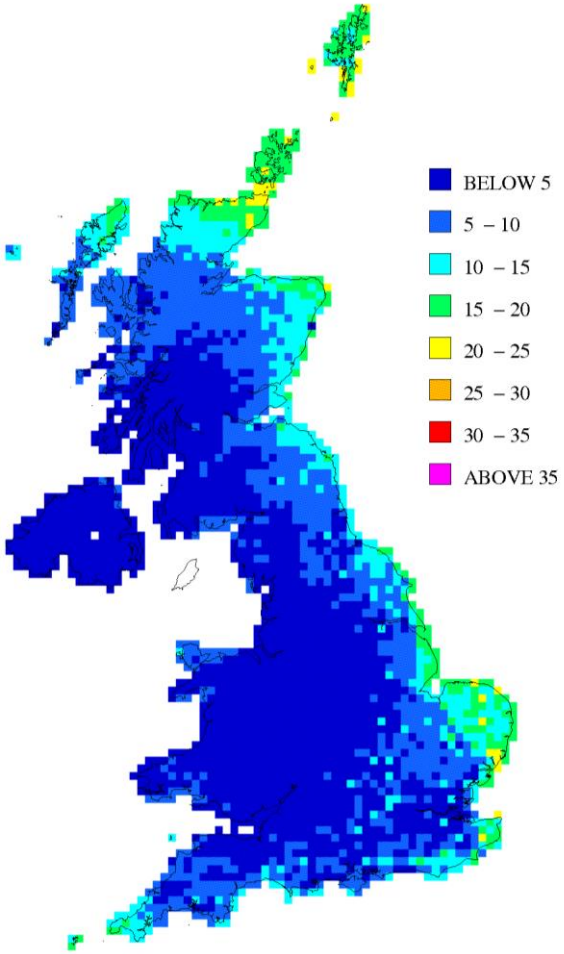
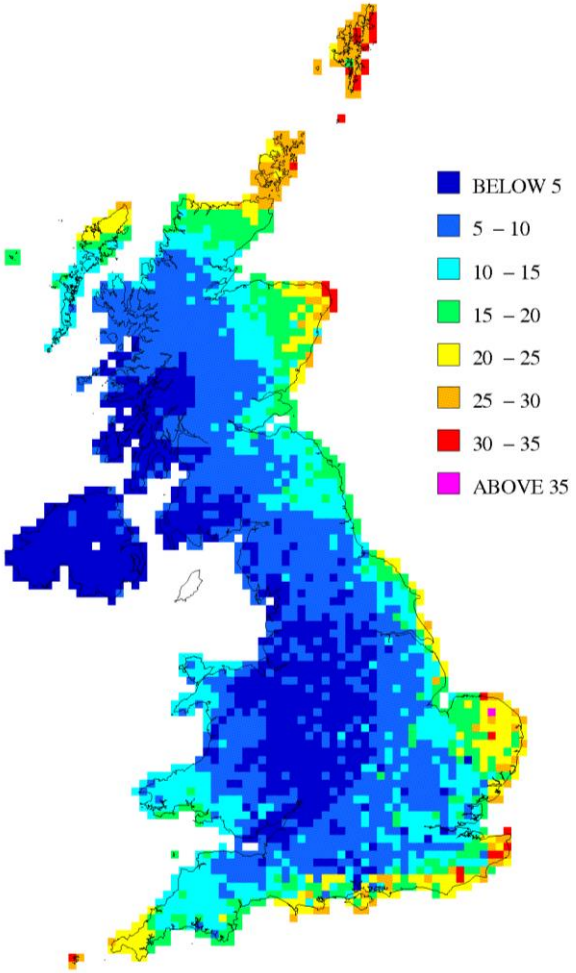
Map of AOT40 for 2008 (µgm^{-3}.hours) (2007 meteorology)	Map of AOT40 for 2020 CCC scenario (µgm^{-3}.hours) (2007 meteorology)
	

Figure 6.4: Number of days exceeding 120 µgm^{-3} maps for 2008 and 2020 CCC scenario (with 2007 meteorology) modelled by OSRM

Map of number of days exceeding 120 µgm^{-3} for 2008 (2007 meteorology)	Map of number of days exceeding 120 µgm^{-3} for 2020 CCC scenario (2007 meteorology)
	

effect is more significant in urban and suburban areas. In other parts of Europe, the projections to 2030 showed decreases in ozone concentrations. The situation is quite complex though as Colette et al (2012) point out that average ozone is sensitive to the NO_x titration effect that influences mainly ***low*** O_3 levels. The higher quantiles of the O_3 distribution will respond in a quite different manner and decreases of ozone peaks in conjunction with an increase of the ozone mean have been predicted.

The AQEG (2009) report on ozone also makes a similar point concerning trends in future UK ozone based on analysis of the condition of the 5-week TORCH campaign in 2003 projected to 2020 using the PTM and the CRI chemical reaction mechanism and projections in future emissions. The study showed trends in ozone distribution statistics for the August 2003 episode at Writtle in Essex as a function of emissions from 1990-2020. The report shows how the maximum ozone concentrations are predicted to initially decline with future reductions in emissions between 1990 and 2010, before then levelling off over the period from 2010-2020 whilst the mean remains relatively static throughout the time series.

The modelling study for the TORCH campaign also showed the relative variation in the calculated number of hours with ozone ≥ 180 μgm^{-3} over the period 1990-2020. This is similar to the Days Greater than 120 μgm^{-3} statistic. A similar picture to that predicted by the OSRM is apparent, whereby the simulated ozone statistic for the episode condition at Writtle falls throughout the 1990s until around 2005, but then shows no further decline with future emission trends and a slight increase between 2010 and 2020. AQEG suggested this was owing to the increasing trend in the emitted VOC/NO_x ratio, in part due to the uncontrolled biogenic component of the VOCs, and the fact that ozone formation tended to be VOC limited over NW Europe for the conditions of the campaign.

6.2 Modelling for Updating Ozone Damage Costs Relationships with Emissions

The UKAAQA has previously been used to establish relationships between various pollution concentration metrics and incremental changes in UK emissions in order for Defra to update health-, crop- and materials damage cost functions. Previous relationships for ozone had used the OSRM to provide outputs on the changes in various ozone concentration metrics for decreases in precursor NO_x and VOC emissions in the UK.

Defra required these relationships between ozone metrics and emission changes to be updated. The approach was to model ozone from a 2010 basecase (meteorology and emissions) for an arbitrary 10% reduction in UK NO_x and VOC emissions separately.

The year and emissions data used were chosen to be as consistent as possible with the assessments for other pollutants (PM_{10} and $PM_{2.5}$) made using the Pollution Climate Mapping (PCM) model in the UKAAQA programme. The PCM damage costs runs for 2010 used the 2009 NAEI emissions scaled forward to 2010 for all sectors except road transport, which used updated emission factors for NO_x and new fleet data informed by Automatic Number Plate Recognition data from DfT (see Passant et al, 2012). These updated road transport data were used in the more recent 2010 version of the NAEI. An exactly equivalent OSRM base case run (2010 modelled using the 2009 NAEI with updated road transport data) was not available, so a new base case for 2010 run using the 2010 NAEI was needed.

Three simulations were therefore carried out for Defra to provide the necessary information:

- Base case 2010 using 2010 meteorology and NAEI emissions and 2010 EMEP emissions
- 2010 as above with 10% reduction in UK NO_x emissions
- 2010 as above with 10% reduction in UK VOC emissions

Various ozone metrics were calculated from the OSRM output. These metrics were specified by Defra and are used to calculate human health impacts, materials damage and crops/vegetation damage.

For calculating the human health impacts the population-weighted mean (PWM) of three metrics were calculated:

- sum of the daily maximum of the running 8-hour mean ozone, calculated with a cut off at 0 μgm^{-3} (SOMO0),
- sum of the daily maximum of the running 8-hour mean ozone, calculated with a cut off at 20 μgm^{-3} (SOMO10)
- sum of the daily maximum of the running 8-hour mean ozone, calculated with a cut off at 70 μgm^{-3} (SOMO35) and
- sum of the daily maximum of the running 8-hour mean ozone, calculated with a cut off at 100 μgm^{-3} (SOMO50).

For calculating materials damage the population-weighted annual mean concentration was calculated.

For calculating crops/vegetation damage the area-weighted mean of the AOT40 metric was calculated.

The metrics provided include SOMO10, which is an additional health-based metric suggested at a WHO workshop. This is similar to the other SOMO metrics, but with a 20 μgm^{-3} cut off. This is a new metric, not previously set up as an output option in the OSRM post-processor. This metric has now been added as an output option, so will be easily available in the future.

6.2.1 Results for Damage Cost Runs

The results for the new 2010 basecase and the two emission reduction scenarios are shown for each ozone metric in Tables 6.4 to 6.9. Results are shown for each country in the UK and for London as well as the UK as a whole. Each of these refers to emission changes from a 2010 base using 2010 meteorology.

Table 6.4: Population-weighted SOMO0 (μgm^{-3}.days)

	Scotland	Wales	Northern Ireland	Inner London	Outer London	Rest of England	All UK
Base 2010 (2010 NAEI)	22277	23815	24154	20902	21049	21728	21858
10% reduction in VOC emissions 2010	22121	23659	24057	20718	20874	21551	21686
10% reduction in NOx emissions 2010	22990	24675	24662	22576	22617	22905	23013

Table 6.5: Population-weighted SOMO10 (μgm^{-3}.days)

	Scotland	Wales	Northern Ireland	Inner London	Outer London	Rest of England	All UK
Base 2010 (2010 NAEI)	15207	16826	17042	14150	14285	14873	14982
10% reduction in VOC emissions 2010	15055	16674	16947	13969	14112	14699	14813
10% reduction in NOx emissions 2010	15877	17627	17519	15756	15786	15980	16071

Table 6.6: Population-weighted SOMO35 (μgm^{-3}.days)

	Scotland	Wales	Northern Ireland	Inner London	Outer London	Rest of England	All UK
Base 2010 (2010 NAEI)	1783	2664	2503	1835	1900	2039	2041
10% reduction in VOC emissions 2010	1741	2597	2477	1755	1825	1969	1974
10% reduction in NOx emissions 2010	1961	2957	2652	2492	2503	2424	2426

Table 6.7: Population-weighted SOMO50 (μgm^{-3}.days)

	Scotland	Wales	Northern Ireland	Inner London	Outer London	Rest of England	All UK
Base 2010 (2010 NAEI)	122	319	252	149	150	243	225
10% reduction in VOC emissions 2010	120	306	251	136	136	234	216
10% reduction in NOx emissions 2010	133	361	268	251	234	295	277

Table 6.8: Population-weighted annual mean ozone (μgm^{-3})

	Scotland	Wales	Northern Ireland	Inner London	Outer London	Rest of England	All UK
Base 2010 (2010 NAEI)	47.2	49.9	53.3	41.5	41.9	44.0	44.6
10% reduction in VOC emissions 2010	46.8	49.6	53.1	41.0	41.5	43.6	44.1
10% reduction in NOx emissions 2010	49.3	52.5	55.0	46.1	46.3	47.4	47.9

Table 6.9: Area-weighted AOT40 (μgm^{-3}.hours)

	Scotland	Wales	Northern Ireland	Inner London	Outer London	Rest of England	All UK
Base 2010 (2010 NAEI)	1669	2125	1394	2494	2602	3010	2392
10% reduction in VOC emissions 2010	1577	1944	1347	2319	2433	2801	2234
10% reduction in NOx emissions 2010	1716	2258	1397	3721	3601	3548	2705

The results for all the metrics show a consistent trend whereby the 10% reduction in VOC emissions leads to a small decrease in all the metrics calculated. The decrease is apparent in all the different regions. On the other hand the 10% reduction in NO_x emission leads to an increase in all the metrics calculated and the increase is larger than the decrease apparent from the same percentage reduction in VOC emissions.

These results will be used by Defra in their damage cost calculations.

6.3 The Impact of the Revised Gothenburg Protocol on Episodic Peak Ozone

After five years of negotiation, a revised Gothenburg Protocol was agreed on 4th May 2012 at a meeting in Geneva of the Parties to the UN ECE Convention on Long-range Transboundary Air Pollution (CLRTAP). Most EU member states have emission reductions for 2020 that were less ambitious or, at best, were in line with business-as-usual, that is, reductions that would be achieved by implementing existing legislation. The revised Gothenburg Protocol was negotiated by the EU on behalf of the EU-27 so the EU member states commitments are expressed jointly relative to the respective emissions in 2005[7]. Across the EU-27, SO_2 emissions are to be cut by 59%, NO_x by 42%, NH_3 by 6%, VOCs by 28% and PM by 22% from 2005 to 2020 (Agren, 2012).

The revised Gothenburg Protocol was implemented in the Photochemical Trajectory Model (PTM) by reducing the SO_2, NO_x, NH_3, VOC emissions across-the-board by the above percentages. A reduction in CO emissions was also implemented by assuming a 28% cut in emissions in line with the VOC emissions. Meteorology was set to 2011 using NAME model back-track air mass trajectories and the PTM was run to 12 UK rural ozone monitoring stations. The impact of the revised Gothenburg Protocol was estimated on episodic peak ozone as indicated by the exceedances of the World Health Organization (WHO, 2006) running 8-hour average guideline set at 50 ppb.

Tables 6.10 – 6.21 present the impact of the revised Gothenburg Protocol on the 50 ppb-episode days across the 12 chosen UK rural ozone monitoring stations. Over the 12 sites there were 211 50 ppb-episode days observed during 2011.

6.3.1 April 17th – 26th Episode

This was the most intense episode of the 2011 season and was undoubtedly photochemical in nature. It began in the south west at Yarner Wood on April 17th, then spread eastwards to Aston Hill and Narberth on April 18th, to Glazebury, High Muffles and Strath Vaich on April 19th and to Eskdalemuir, Lullington Heath and Sibton on April 20th. Maximum 8-hour levels above 50 ppb were monitored at all sites on April 21st and 22nd and at all but Glazebury and Narberth on April 23rd. The episode continued in the south and east until April 25th at Harwell, Lullington Heath, Sibton and Yarner Wood. The episode finished in the east at Sibton on April 26th.

The highest 8-hour running mean level of 83 ppb was observed at High Muffles on April 22nd, second highest of 75 ppb at Strath Vaich also on April 22nd and third highest of 74 ppb at Eskdalemuir on April 22nd and Yarner Wood on April 21st.

The PTM model accurately predicted the episode maxima at only 5 out of the 12 sites, that is, at Aston Hill, Bush Estate, Glazebury, Lullington Heath and Narberth. At these well-predicted sites, the PTM model indicated that the revised Gothenburg Protocol would lead to reductions in episodic peak levels of between -1.1 and 11.2 ppb, averaging 4.4 ppb over the 5 sites. Although this represented a substantial reduction in episodic peak levels, it was not sufficient to bring all the sites below the 50 ppb WHO guideline.

6.3.2 September 28th – October 3rd Episode

This episode was also photochemical in nature and occurred unusually late in the 2011 season. The episode began on September 28th at Bush estate and spread to Aston Hill, Harwell and Lullington Heath on September 29th. All sites recorded the episode on September 30th and all but Eskdalemuir and Rochester on October 1st. The episode continued in the south and east at Aston Hill, Harwell, Lullington Heath, Rochester and Sibton on October 2nd and 3rd.

[7] http://www.unece.org/fileadmin/DAM/env/lrtap/full%20text/Informal_document_no_17_No23_Consolidated_text_checked_DB_10Dec2012_-_YT_-_10.12.2012.pdf

The highest 8-hour running mean level of 63 ppb was observed at Lullington Heath on October 1st, the second highest of 62.5 ppb at Rochester on October 2nd and the third highest of 62 ppb at Aston Hill on October 1st.

The PTM model accurately predicted the episode maxima at 6 out of the 12 sites, that is, at Bush estate, Eskdalemuir, Glazebury, High Muffles, Narberth and Rochester. At these well-predicted sites, the PTM model indicated that the revised Gothenburg Protocol would lead to reductions in episodic peak levels of between -2.3 and 9.7 ppb, averaging 1.4 ppb over the 6 sites. Although this represented an overall reduction in episodic peak levels, it was necessarily a fine balance between some sites where ozone increased and those where it decreased. The impact of the revised Gothenburg Protocol on this photochemical episode was therefore considered to be only a marginal improvement. The sites where ozone increased on some of days were Bush Estate, Eskdalemuir and High Muffles.

6.3.3 Other Episodes

The Bush Estate alone recorded an exceedance of the 50 ppb WHO 8-hour running mean guideline on 4th February which is very early in the 2011 year. This is almost certainly a 'background' episode and was not predicted by the PTM model.

Aston Hill, Strath Vaich and Yarner Wood recorded a number of exceedances of the WHO guideline between April 2nd and April 15th. Again, these were not predicted by the PTM model and they were almost certainly 'background' episodes.

All the sites recorded exceedances of the WHO guideline between April 27th and May 10th. These exceedances were recorded earliest at Eskdalemuir, Harwell, Narberth and Yarner Wood and latest at Bush Estate, Harwell, High Muffles, Yarner Wood and Sibton (particularly). The character of the exceedances appeared to change during the episode from largely 'background' episodes to 'photochemical' in origin. Maximum 8-hour running mean levels were observed to reach 64 ppb at Sibton on May 6th. The later episodes were well predicted at High Muffles, Lullington Heath, Narberth and Yarner Wood. At these well-predicted sites, the PTM model indicated that the revised Gothenburg Protocol would lead to reductions in episodic peak levels of between -8.5 and 6.9 ppb, averaging 2.6 ppb over the 4 sites. Although this represented a substantial reduction in episodic peak levels, it was not sufficient to bring all the sites below the 50 ppb WHO guideline.

All sites recorded exceedances of the WHO guideline between July 3rd and July 5th. Maximum 8-hour running mean levels were observed to reach 62 ppb at Rochester on July 3rd. The highest observed levels at each site during these episodes were only well predicted by the PTM at Lullington Heath and Narberth where the impact of the revised Gothenburg Protocol was found to be reductions of 2.2 ppb and 2.6 ppb, respectively.

The sites in the south and east recorded exceedances of the WHO guideline between July 28th and August 4th. The highest observed level was found to be 72 ppb at Rochester on August 3rd. These episodes were all photochemical in origin and were generally well predicted by the PTM model. The impact of the revised Gothenburg Protocol was found to produce a 4.7 ppb reduction in episodic peak levels when the predictions for High Muffles, Lullington Heath and Rochester were averaged. Again, although this represented a substantial reduction in episodic peak levels, it was not sufficient to bring all the sites below the 50 ppb WHO guideline.

The Sibton site alone recorded an exceedance of the WHO guideline on September 3rd which was well predicted by the PTM model. The impact of the revised Gothenburg Protocol was found to be a reduction of 3.4 ppb which would on this occasion be enough to reduce the episodic peak levels below the WHO guideline.

6.3.4 Summary

The impact of the revised Gothenburg Protocol on episodic peak ozone levels was found to be substantial, bringing about reductions in episodic peak levels of between -8.4 ppb

(indicating an increase in ozone) and 11.2 ppb and averaging 3.0 ± 4.7 ppb. However, the impact was not sufficient to bring all the episodes, at all the sites during 2011, below the 50 ppb WHO guideline.

Table 6.10. Impact of the revised Gothenburg Protocol (GP) on the 50 ppb episode days in 2011 at Aston Hill. A negative difference indicates an increase in ozone for the GP on ozone.

Date	Observed maximum 8-hour average, ppb	Base case model ppb	Revised GP model ppb	Difference ppb
6th April	52	48.452	44.823	3.629
7th April	51.5	42.931	42.931	0
10th April	50	45.959	45.300	0.659
18th April	50.5	50.787	48.26	2.527
19th April	53	53.676	47.581	6.095
20th April	60.5	56.363	55.021	1.615
21st April	66.5	63.059	62.469	0.590
22nd April	61	50.884	47.518	3.366
23rd April	56	48.191	44.435	3.756
30th April	50.5	44.540	41.379	3.161
4th May	52	47.850	46.083	1.767
4th July	59	52.465	51.524	0.941
5th July	54	42.628	39.734	2.894
29th September	59.5	58.702	44.194	14.508
30th September	60.5	61.438	54.612	6.826
1st October	62	48.073	42.44	5.633
2nd October	60	49.375	43.913	5.462
3rd October	53	45.335	40.482	4.853
Average	**56.2**	**50.60**	**46.82**	**3.793**

Table 6.11. Impact of the revised Gothenburg Protocol (GP) on the 50 ppb episode days in 2011 at Bush Estate. A negative difference indicates an increase in ozone for the GP on ozone.

Date	Observed maximum 8-hour average, ppb	Base case model ppb	Revised GP model ppb	Difference ppb
4th February	52	43.195	43.176	0.019
21st April	54	49.616	49.693	-0.077
22nd April	54	54.586	52.558	2.028
23rd April	52.5	44.447	44.447	0
30th April	52.5	46.497	46.497	0
1st May	50.5	40.056	40.056	0
4th May	50.5	45.935	45.466	0.469
8th May	51	48.395	48.445	-0.050
5th July	51	43.826	43.966	-0.140
28th September	50.5	49.919	41.3	8.619
30th September	59	57.041	58.874	-1.833
1st October	51.5	47.529	44.530	2.999
Average	**52.4**	**47.59**	**46.58**	**1.003**

Table 6.12. Impact of the revised Gothenburg Protocol (GP) on the 50 ppb episode days in 2011 at Eskdalemuir. A negative difference indicates an increase in ozone for the GP on ozone.

Date	Observed maximum 8-hour average, ppb	Base case model ppb	Revised GP model ppb	Difference ppb
20th April	58.5	53.140	52.0	1.140
21st April	64.5	52.282	51.329	0.953
22nd April	74	67.231	64.356	2.965
23rd April	54	42.794	42.609	0.185
28th April	51.5	48.979	48.979	0
30th April	51	46.428	46.428	0
1st May	54.5	41.462	39.886	1.576
2nd May	50.5	38.617	38.594	0.023
4th May	57.5	47.078	45.403	1.675
5th May	50	46.562	43.102	3.460
30th September	56.5	56.336	57.699	-1.363
Average	**56.6**	**49.17**	**48.20**	**0.965**

Notes:

a. Data capture at this site was below that typically expected for rural ozone monitoring stations during 2011.

Table 6.13. Impact of the revised Gothenburg Protocol (GP) on the 50 ppb episode days in 2011 at Glazebury. A negative difference indicates an increase in ozone for the GP on ozone.

Date	Observed maximum 8-hour average, ppb	Base case model ppb	Revised GP model ppb	Difference ppb
19th April	55.5	55.284	49.182	6.102
20th April	58.5	56.937	55.121	1.816
21st April	67	64.412	58.253	6.159
22nd April	72	75.904	75.914	-0.010
30th April	54	46.180	44.402	1.778
1st May	52	41.292	37.569	3.723
4th July	60.5	48.739	46.591	2.148
30th September	50	49.247	48.248	0.999
1st October	52.5	53.764	52.313	1.451
Average	**58.0**	**54.64**	**51.95**	**2.685**

Table 6.14. Impact of the revised Gothenburg Protocol (GP) on the 50 ppb episode days in 2011 at Harwell. A negative difference indicates an increase in ozone for the GP on ozone.

Date	Observed maximum 8-hour average, ppb	Base case model ppb	Revised GP model ppb	Difference ppb
21st April	57	57.056	53.164	3.892
22nd April	65	49.62	46.666	2.954
23rd April	69	51.901	51.317	0.584
24th April	52	47.989	48.444	-0.455
25th April	51.5	47.137	47.121	0.016
28th April	52.5	49.469	49.408	0.061
30th April	59.5	47.586	44.033	3.553
1st May	57.5	43.467	43.366	0.101
2nd May	52.5	38.913	37.206	1.707
5th May	54.5	43.126	38.757	4.369
6th May	62.5	41.601	37.87	3.731
7th May	57.5	37.443	29.973	7.47
8th May	50	37.821	36.074	1.747
3rd July	51	38.602	36.365	2.237
4th July	60	49.509	46.404	3.105
28th July	52.5	42.012	40.78	1.232
31st July	57.5	45.448	44.58	0.868
1st August	60	37.306	34.985	2.321
3rd August	53.5	36.265	34.951	1.314
29th September	54.5	56.621	47.013	9.608
30th September	56	52.457	44.955	7.502
1st October	58.5	44.08	41.268	2.812
2nd October	57.5	40.608	36.628	3.98
3rd October	52	45.606	36.048	9.558
Average	**56.4**	**45.07**	**41.97**	**3.094**

Table 6.15. Impact of the revised Gothenburg Protocol (GP) on the 50 ppb episode days in 2011 at High Muffles. A negative difference indicates an increase in ozone for the GP on ozone.

Date	Observed maximum 8-hour average, ppb	Base case model ppb	Revised GP model ppb	Difference ppb
19th April	62.5	56.924	48.963	7.961
20th April	69	65.761	54.881	10.88
21st April	81.5	70.571	73.586	-3.015
22nd April	83	67.776	69.897	-2.121
23rd April	69	57.275	53.143	4.132
29th April	52	44.238	42.354	1.884
30th April	53.5	44.908	44.464	0.444
1st May	53	41.663	37.36	4.303
2nd May	50.5	39.317	37.524	1.793
3rd May	50	44.461	43.193	1.268
4th May	52	46.633	46.289	0.344
7th May	54	52.804	47.226	5.578
8th May	51.5	51.008	47.63	3.378
27th June	58	58.499	54.986	3.513
3rd July	52	37.023	35.037	1.986
4th July	55.5	43.237	42.875	0.362
5th July	56	34.62	34.23	0.39
31st July	56	51.254	49.059	2.195
3rd August	50.5	53.928	52.148	1.78
4th August	50.5	40.722	38.605	2.117
30th September	51	50.863	53.137	-2.274
1st October	50	52.175	48.113	4.062
Average	**57.3**	**50.26**	**47.94**	**2.316**

NOTES: a. the data capture for this site was not as high as is typically seen at rural ozone monitoring network sites.

Table 6.16. Impact of the revised Gothenburg Protocol (GP) on the 50 ppb episode days in 2011 at Lullington Heath. A negative difference indicates an increase in ozone for the GP on ozone.

Date	Observed maximum 8-hour average, ppb	Base case model ppb	Revised GP model ppb	Difference ppb
20th April	51	50.694	42.236	8.458
21st April	61	62.992	53.789	9.203
22nd April	59.5	56.524	52.177	4.347
23rd April	62.5	56.164	49.267	6.897
24th April	66	65.35	54.164	11.186
25th April	65	63.318	58.744	4.574
5th May	54	53.883	47.689	6.194
6th May	56	58.342	66.799	-8.457
3rd July	52.5	47.374	45.17	2.204
1st August	54	51.29	50.27	1.02
2nd August	53.5	51.494	47.463	4.031
3rd August	52.5	52.321	53.201	-0.88
29th September	53.5	55.147	46.755	8.392
30th September	62.5	63.248	54.528	8.72
1st October	63	47.54	42.905	4.635
2nd October	62	54.284	50.282	4.002
3rd October	52	55.561	49.98	5.581
Average	**57.7**	**55.62**	**50.91**	**4.712**

Table 6.17. Impact of the revised Gothenburg Protocol (GP) on the 50 ppb episode days in 2011 at Narberth. A negative difference indicates an increase in ozone for the GP on ozone.

Date	Observed maximum 8-hour average, ppb	Base case model ppb	Revised GP model ppb	Difference ppb
18th April	51.5	52.36	51.926	0.434
19th April	59.5	56.219	55.987	0.232
20th April	59.5	57.963	49.967	7.996
21st April	66.5	67.308	58.914	8.394
22nd April	56.5	49.907	51.015	-1.108
28th April	50	47.706	47.695	0.011
29th April	50.5	47.851	43.926	3.925
30th April	58	57.517	50.612	6.905
1st May	52.5	49.956	49.494	0.462
2nd May	52.5	44.954	43.766	1.188
4th May	51	46.286	41.704	4.582
3rd July	53.5	51.169	49.762	1.407
4th July	57	52.64	50.089	2.551
5th July	51.5	30.458	29.851	0.607
30th September	53.5	48.444	43.557	4.887
1st October	54	54.017	48.856	5.161
28th November	51	42.955	41.128	1.827
29th November	52	39.982	38.824	1.158
1st December	50.5	45.521	45.995	-0.474
Average	**54.3**	**49.64**	**47.00**	**2.639**

Table 6.18. Impact of the revised Gothenburg Protocol (GP) on the 50 ppb episode days in 2011 at Rochester. A negative difference indicates an increase in ozone for the GP on ozone.

Date	Observed maximum 8-hour average, ppb	Base case model ppb	Revised GP model ppb	Difference ppb
27th June	55	50.974	43.846	7.128
3rd July	62	49.694	49.889	-0.195
5th July	57	42.463	39.153	3.31
15th July	51	47.076	45.681	1.395
28th July	52	51.347	49.787	1.56
31st July	51.5	46.978	46.484	0.494
1st August	51.5	44.683	42.268	2.415
2nd August	51	52.321	46.791	5.53
3rd August	72	71.658	60.666	10.992
30th September	58.5	54.175	44.478	9.697
2nd October	62.5	58.268	51.031	7.237
3rd October	51.5	38.761	35.104	3.657
Average	**56.3**	**50.70**	**46.26**	**4.435**

NOTES:

a. the data capture for this site was not as high as is typically seen at rural ozone monitoring network sites.

Table 6.19. Impact of the revised Gothenburg Protocol (GP) on the 50 ppb episode days in 2011 at Sibton. A negative difference indicates an increase in ozone for the GP on ozone.

Date	Observed maximum 8-hour average, ppb	Base case model ppb	Revised GP model ppb	Difference ppb
20th April	59.5	57.45	75.192	-17.742
21st April	66	67.796	65.038	2.758
22nd April	52.5	52.491	46.591	5.9
23rd April	66.5	59.311	56.542	2.769
24th April	62.5	62.143	57.534	4.609
25th April	54.5	50.695	50.221	0.474
26th April	52.5	47.392	44.346	3.046
30th April	51	50.382	46.002	4.38
1st May	54	43.681	40.498	3.183
2nd May	52	42.396	40.739	1.657
3rd May	52.5	45.743	45.743	0
4th May	51.5	41.314	41.314	0
6th May	64	56.82	54.18	2.64
7th May	58	62.364	62.626	-0.252
9th May	50.5	50.857	46.95	3.907
10th May	52.5	47.816	47.379	0.437
21st May	52	47.277	46.267	1.01
27th June	60	58.383	50.627	7.756
3rd July	51.5	47.037	44.967	2.07
5th July	55	45.462	43.375	2.087
15th July	53	46.064	42.968	3.096
28th July	50	48.942	44.61	4.332
1st August	51	52.343	50.844	1.499
2nd August	54.5	56.171	61.245	-5.074
3rd August	69	54.93	46.39	8.535
3rd September	50.5	48.338	44.941	3.397
30th September	53	50.202	44.615	5.587
1st October	59	60.399	54.767	5.632
2nd October	60.5	50.918	44.226	6.692
3rd October	53	48.804	45.633	3.171
Average	**55.7**	**51.81**	**49.55**	**2.252**

Table 6.20. Impact of the revised Gothenburg Protocol (GP) on the 50 ppb episode days in 2011 at Strath Vaich. A negative difference indicates an increase in ozone for the GP on ozone.

Date	Observed maximum 8-hour average, ppb	Base case model ppb	Revised GP model ppb	Difference ppb
3rd April	51	43.331	43.313	0.018
4th April	51	43.935	43.931	0.004
5th April	50.5	43.648	43.641	0.007
7th April	51.5	42.585	42.58	0.005
14th April	51	46.843	46.843	0
15th April	53	44.926	44.924	0.002
19th April	50	44.387	43.279	1.108
20th April	60	47.54	43.542	3.998
21st April	54	45.933	45.933	0
22nd April	75	60.298	56.699	3.599
23rd April	63.5	42.666	42.512	0.154
26th April	50	45.324	45.320	0.004
27th April	52	48.531	48.531	0
29th April	54	47.389	45.03	2.359
30th April	55.5	46.088	46.085	0.003
20th May	50.5	40.229	40.011	0.218
Average	**54.5**	**45.85**	**45.14**	**0.717**

Table 6.21. Impact of the revised Gothenburg Protocol (GP) on the 50 ppb episode days in 2011 at Yarner Wood. A negative difference indicates an increase in ozone for the GP on ozone.

Date	Observed maximum 8-hour average, ppb	Base case model ppb	Revised GP model ppb	Difference ppb
2nd April	51.5	47.746	45.428	2.318
6th April	50	45.419	41.726	3.693
8th April	50.5	46.092	44.636	1.456
17th April	60	54.658	50.566	4.092
18th April	54	51.456	46.601	4.855
19th April	61.5	55.109	49.003	6.106
20th April	67	46.653	42.691	3.962
21st April	74	64.553	62.7	1.853
22nd April	62	44	40.904	3.096
23rd April	59.5	48.835	44.351	4.484
25th April	54	54.971	51.165	3.806
27th April	52	48.464	42.261	6.203
28th April	51	51.311	51.265	0.046
30th April	52.5	51.104	48.053	3.051
1st May	54.5	50.355	44.043	6.312
2nd May	52	46.196	45.183	0.383
3rd May	52	49.887	48.29	1.597
8th May	50.5	36.669	36.14	0.529
2nd July	53.5	44.999	43.906	1.093
3rd July	52.5	57.945	54.767	3.178
4th July	54.5	42.236	39.848	2.388
Average	**55.7**	**49.46**	**46.39**	**3.071**

NOTES:

a. the data capture for this site was not as high as is typically seen at rural ozone monitoring network sites.

7 Model Intercomparison Activities

Both the OSRM and PTM have taken part in the Defra Model Intercomparison Exercise (MIE). For Phase 2 of the exercise, results had been submitted for four different emissions scenarios:

1. Reduce total anthropogenic NOx and VOC by 30% across the UK + Europe
2. Reduce total anthropogenic NOx and VOC by 30% across the UK only
3. Reduce anthropogenic NOx by 30% across UK + Europe
4. Reduce anthropogenic VOC by 30% across UK + Europe

The base year was 2006 (emissions and meteorology). Hourly ground-level concentrations were provided for ozone and other indicator species, namely NO, NO_2, NO_y, HNO_3 and H_2O_2.

The results for each scenario were sent to David Carslaw (King's College London) for statistical analysis and comparison with results from other regional scale models.

These activities continued during 2012, mainly through the supply of additional information required on emissions data and the temporal variability assumed in the models and on boundary layer height. The project partners completed further questionnaires on the OSRM and PTM and took part at a meeting covering the regional group of the MIE in May 2012.

The MIE is now coming to an end and partners will attend a final meeting of the regional group in April 2013.

7.1 Comparison of Model Sensitivity to Ozone Precursor Emissions During Episode Conditions

Whilst the final conclusions of the regional group of the MIE are still being prepared, the ozone results from the various modelling groups that took part have been further analysed in this project to compare and understand differences in the sensitivity of the model results to changes in ozone precursor emissions during episode conditions.

In effect, the ability of these models to respond to typical questions on emission changes of the sort that is relevant to Defra's policy on ozone air quality is addressed. We look at the range of possible answers to typical policy-relevant questions from a range of different models and we try to find explanations of the wide variations in the model scenario predictions. For simplicity, we focus on two policy-relevant questions: is it better to reduce NO_x or VOC emissions and is it better for any action to be taken concertedly across Europe or unilaterally within the United Kingdom?

The models employed in this study have all been employed to describe photochemical ozone formation across north west Europe and across the UK. Full details of the models themselves are given in the Supplementary Information provided by the different modelling groups to the Defra MIE and covered in the report on Phase 1 of the MIE by the Defra Air Quality Modelling Review Steering Group (Williams et al, 2011). The detailed analysis of the MIE Phase 2 results from the various models involved in the regional group is given in the draft report by Carslaw (2012).

The models involved in the regional group which focused on ozone include 3-dimensional Eulerian grid models, Lagrangian dispersion models and trajectory models and employ a wide range of chemical mechanisms to describe photochemical ozone formation from volatile

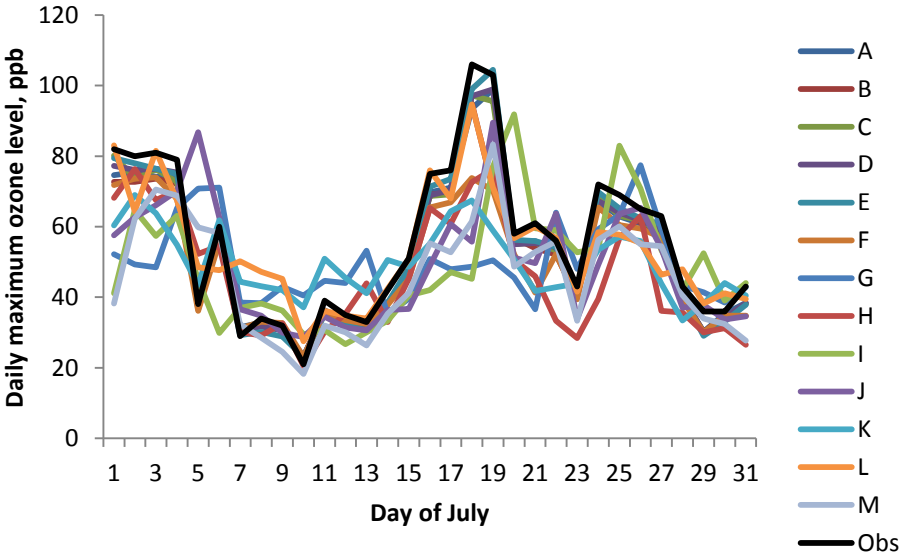
organic compound (VOC) and nitrogen oxide (NO_x) emissions. A brief summary is given below:

1. Community Multi-scale Air Quality (CMAQ) model, (3 independent versions),
2. Air Quality Unified Model (AQUM),
3. European Monitoring and Evaluation Programme for the UK (EMEP4UK) model,
4. Numerical Atmospheric Dispersion Model Environment (NAME) model,
5. Ozone Source Receptor Model (OSRM),
6. Photochemical Trajectory Model (PTM), (using 6 independent chemical mechanisms).

To reduce the scope and complexity of the study to a level which was tractable, detailed attention was given to the behaviour of ground-level ozone during July 2006 at a rural EMEP site, Harwell in Oxfordshire, United Kingdom. This site is situated about 80 km due west of London and is surrounded by agricultural fields and a large campus of research establishments.

Each of the 13 models was set up with base case conditions for July 2006 and run in their standard configurations as described in the Supplementary Information. The highest hourly maximum ozone level modelled each day was recorded from each set of model outputs and these levels, together with the observations are plotted in Figure 7.1. All of the models were able to account satisfactorily for the observed day-to-day variations in daily peak ozone levels in that they exhibited peak levels during the periods July 1st – 3rd, 15th – 20th and 23rd – 27th, with relatively lower, background levels between Jul 7th – 14th and 28th – 31st. Normalised mean biases NMBs were calculated for daily ozone maxima for July 2006 for each model and were found to lie in the range from -0.18 to -0.04. In view of the simple evaluation metrics based on NMBs being in the range -0.2 < NMB < 0.2 as proposed by Derwent et al., (2010c), model performance was considered entirely satisfactory for all 13 models.

Figure 7.1. Daily maximum ozone levels for all 13 models and observations for each day of July 2006 at the rural Harwell, Oxfordshire site.



7.1.1 NO_x- versus VOC-sensitivity

An important issue for policy-makers involved in developing strategies for ground-level O_3 has been whether to reduce NO_x emissions or VOC emissions or both. To address this issue, attention has been focussed on the impact of some simple policy-relevant emission scenarios:

- S1: 30% reductions in NO_x and VOC emissions carried out across Europe,
- S2: 30% reductions in NO_x and VOC emissions carried out across the United Kingdom,
- S3: 30% reductions in NO_x emissions carried out across Europe,
- S4: 30% reductions in VOC emissions carried out across Europe.

The choice of 30% is arbitrary and has no direct policy relevance. It has been chosen because it is neither too small nor too large and to be consistent with a large literature on photochemical ozone model sensitivity to VOC and NO_x emissions, see, for example, Sillman (1999) and Sillman and He (2002). To assess the impact of 30% across-the-board reductions in man-made NO_x and VOC emissions relative to the 2006 base case, each model ran the S3 and S4 emission scenario cases. The maximum hourly ozone levels predicted for the base case and the two scenario cases for each day of July 2006 were determined for each model. If the difference in the maximum hourly ozone levels, calculated as base case – scenario case, was greater for the 30% reduction in VOC emissions than for NO_x emissions, then that day was assigned as VOC-sensitive and vice versa.

The impact of the 30% reductions in NO_x emissions carried out across the UK and the Rest of Europe (RoE) on the July mean daily maximum ozone levels varied considerably between the 13 models. O_3 responses (base case – scenario case) covered the range from -2.0 to 3.8 ppb, with 2 models producing an increase (-ve response) and 11 models producing a decrease (+ve response). Figure 7.2 presents a 'box and whisker' plot of the 13 model responses. The interquartile range, shown as a shaded box, confirms that although the responses showed a degree of uncertainty, the mean model response of 0.9 ppb was statistically significantly different from zero. However, there was a 15% chance that the response would be negative and July mean daily maximum ozone levels would increase in response to a 30% reduction in NO_x emissions.

In contrast, the impact of 30% reductions in VOC emissions shown in Figure 7.2 covered the range from 0.4 to 3.4 ppb with a mean model response of 1.7 ppb. All models produced a positive response demonstrating an improvement in ozone air quality as indexed by their July mean daily maximum ozone levels. The interquartile range of the responses to 30% VOC emission reductions confirmed that the mean model response was statistically significantly different from zero. VOC reductions always produced an improvement in air quality, in contrast to NO_x reductions, using the July mean daily maximum ozone index.

Table 7.1 shows the NO_x- versus VOC-sensitivity assignments for each model and for each day of July 2006. There was complete agreement as to NO_x- or VOC-sensitivity on only four days and some disagreement between models on the remaining 27 days. It was apparent that assignment of the sensitivity to ozone precursors, whether NO_x or VOCs, varied from day-to-day.

To understand why there is so much conflict in the NO_x- versus VOC-sensitivity assignments, the analysis will need to move on from categoric assignments, either NO_x- or VOC-sensitive. This is because each model is inherently uncertain and policy advice should necessarily be probabilistic rather than categoric. Figure 7.3 presents a scatter plot of the O_3 responses to 30% NO_x reduction plotted out against the O_3 responses to 30% VOC reduction for the 13 models and 15 O_3 episode days. Also shown is the 1:1 correspondence line representing the locus of the points with equal responses to 30% NO_x reduction and 30% VOC reductions.

Figure 7.2. Box and Whiskers plots of the responses of the July mean daily maximum ozone levels to 30% NO_x emission reductions, 30% VOC emission reductions and 30% NO_x and VOC emission reductions carried out across the UK and the Rest of Europe, together with those to 30% NO_x and VOC emission reductions carried out across the UK.

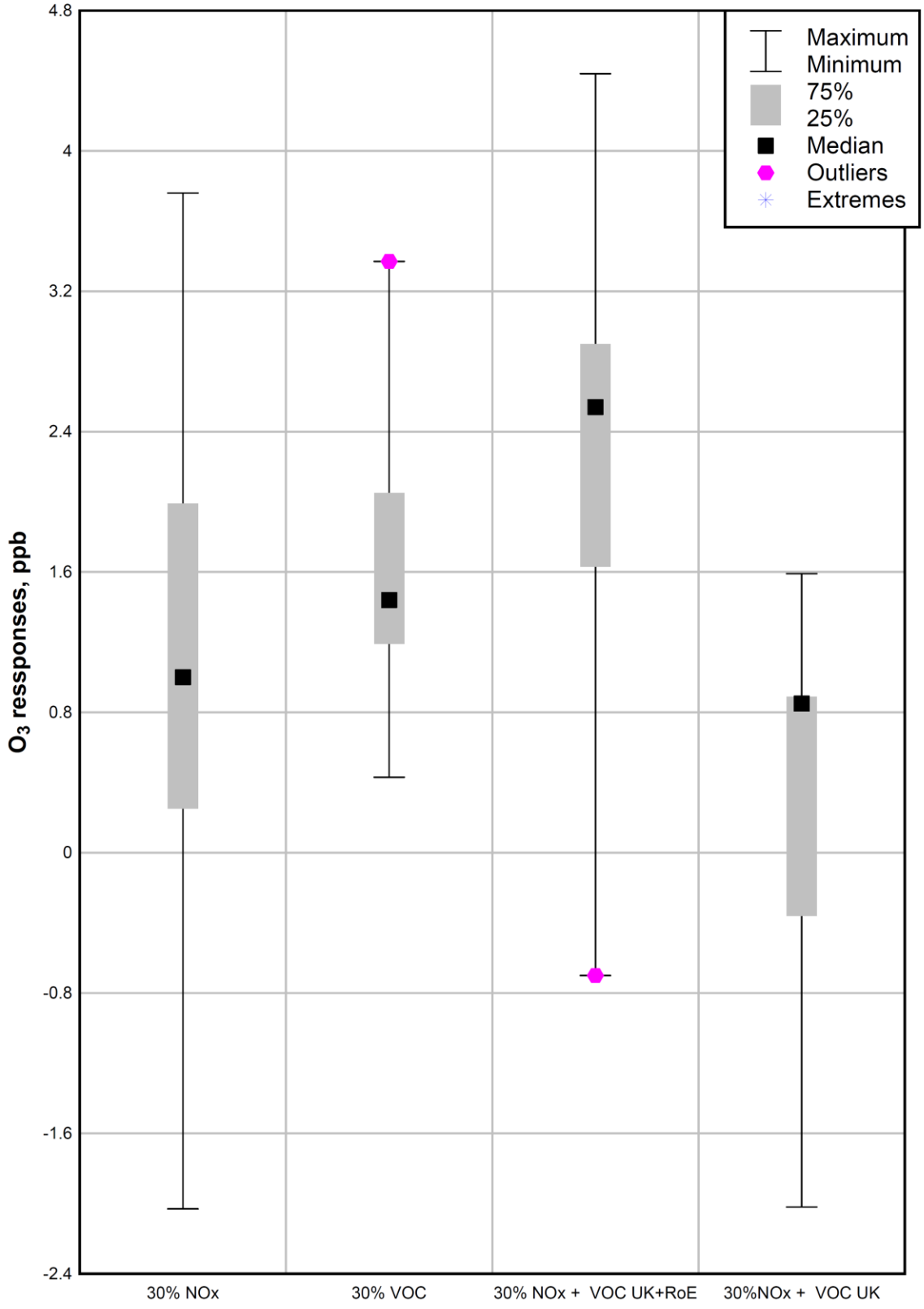
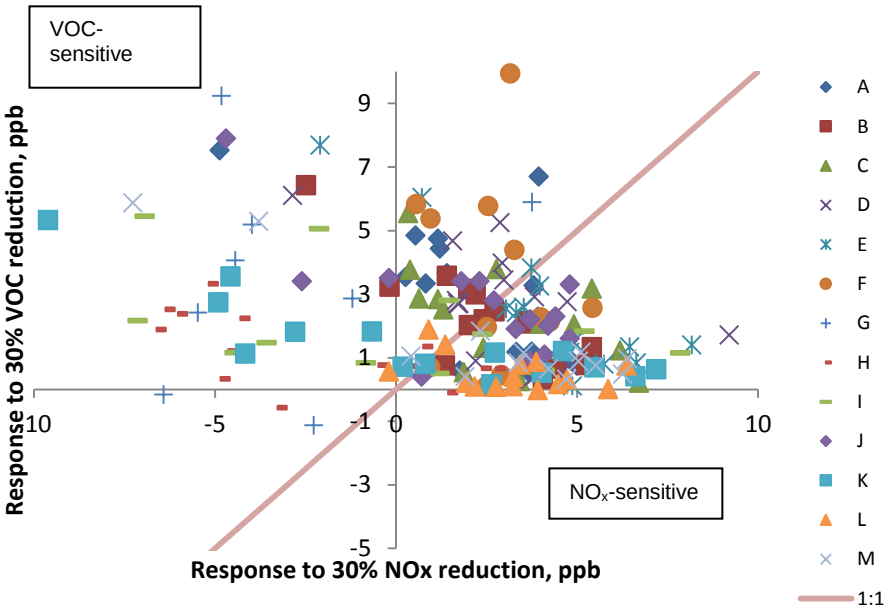


Table 7.1. Assignments as to NO_x- versus VOC-sensitivity for each model and for each day of July 2006.

Model	A	B	C	D	E	F	G	H	I	J	K	L	M
1st	VOC	NOx	NOx	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC
2nd	NOx	NOx	NOx	NOx	NOx	VOC	NOx	NOx	NOx	NOx	NOx	NOx	NOx
3rd	NOx	NOx	NOx	NOx	NOx	NOx	VOC	VOC	NOx	VOC	VOC	NOx	NOx
4th	NOx	VOC	NOx	NOx	NOx	NOx	VOC	VOC	VOC	VOC	NOx	NOx	VOC
5th	NOx	NOx	NOx	NOx	NOx	NOx	VOC	VOC	VOC	NOx	NOx	NOx	VOC
6th	VOC	VOC	VOC	VOC	NOx	VOC	VOC	NOx	VOC	NOx	NOx	NOx	NOx
7th	NOx	VOC	NOx	NOx	NOx	NOx	VOC	VOC	VOC	VOC	NOx	NOx	VOC
8th	NOx	NOx	NOx	NOx	NOx	NOx	VOC	NOx	NOx	NOx	NOx	NOx	NOx
9th	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	VOC
10th	NOx	NOx	NOx	NOx	NOx	NOx	VOC	VOC	NOx	NOx	VOC	VOC	VOC
11th	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	VOC	NOx	NOx	NOx	NOx
12th	NOx	NOx	NOx	NOx	NOx	NOx	VOC	VOC	VOC	VOC	VOC	VOC	VOC
13th	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC
14th	NOx	NOx	NOx	NOx	NOx	NOx	VOC	VOC	NOx	NOx	VOC	NOx	NOx
15th	NOx	NOx	NOx	NOx	NOx	NOx	VOC	VOC	VOC	NOx	VOC	VOC	NOx
16th	VOC	VOC	VOC	VOC	NOx	NOx	VOC	VOC	VOC	VOC	VOC	VOC	NOx
17th	VOC	NOx	NOx	VOC	NOx	VOC	VOC	VOC	VOC	VOC	VOC	NOx	VOC
18th	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC	NOx	NOx	VOC
19th	NOx	NOx	NOx	NOx	NOx	NOx	VOC	VOC	NOx	VOC	NOx	VOC	NOx
20th	NOx	NOx	NOx	NOx	NOx	NOx	VOC	VOC	NOx	NOx	NOx	NOx	NOx
21st	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx
22nd	VOC	VOC	VOC	VOC	NOx	VOC	VOC	VOC	NOx	NOx	VOC	NOx	NOx
23rd	VOC	VOC	VOC	NOx	NOx	VOC	VOC	VOC	NOx	VOC	NOx	NOx	VOC
24th	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC	NOx	VOC
25th	NOx	NOx	NOx	NOx	NOx	NOx	VOC	VOC	VOC	NOx	NOx	NOx	NOx
26th	VOC	NOx	VOC	VOC	NOx	VOC	VOC	VOC	VOC	NOx	NOx	NOx	NOx
27th	VOC	VOC	VOC	VOC	VOC	VOC	VOC	VOC	NOx	NOx	VOC	NOx	NOx
28th	NOx	NOx	NOx	NOx	NOx	NOx	VOC	VOC	NOx	VOC	VOC	NOx	NOx
29th	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx
30th	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx	NOx
31st	NOx	NOx	NOx	NOx	NOx	NOx	VOC	VOC	NOx	NOx	NOx	NOx	NOx

Points above the 1:1 line have responses to 30% VOC reduction that are greater than to 30% NO_x reduction and so have been assigned as VOC-sensitive. Points below the 1:1 correspondence line have been assigned as NO_x-sensitive. It appears that the vast majority of the points are located above the x-axis showing that almost all of the points show positive responses to 30% VOC reductions and that O_3 air quality almost always improves. In contrast, there are a small but significant number of points to the left of the y-axis showing that some model results show negative responses to 30% NO_x reductions and that O_3 air quality may deteriorate.

Figure 7.3. Scatter plot of the 13 model O_3 responses to 30% NOx reductions versus 30% VOC reductions for the episode days of July 2006. Also shown is the 1:1 correspondence line above which points are VOC-sensitive and below which they are NO_x-sensitive.



The majority of the points in the scatter plot in Figure 7.3 form a 'wedge-shaped' pattern. This pattern starts at the right-hand side of the plot, at high NO_x-response – low VOC-response and widens towards the left-hand side of the plot. There is a tendency for VOC-responses to be smallest when NO_x-responses are greatest and VOC-responses to be greatest when NO_x-responses are negative. This characteristic tendency has its origins in the theory underpinning NO_x- and VOC-sensitivity as demonstrated by Sillman (1999) and Sillman and He (2002). Superimposed on this characteristic tendency is the impact of model uncertainty which is manifest in terms of the relative scatter between the sets of model points. The axis of this 'wedge-shaped' pattern is almost at right angles to the 1:1 correspondence line. As a result, the characteristic tendency and the model uncertainty strongly impact on the location of the points relative to the 1:1 correspondence line and hence the NO_x- versus VOC-sensitivity assignments. Categoric conclusions about NO_x- versus VOC-sensitivity on episode days do not appear to be robust. What can be said is that there are 90 points out of 195 that are VOC-sensitive and 105 points that are NO_x-sensitive. On this basis, the balance of probabilities is that episode days are slightly more likely to be NO_x-sensitive than VOC-sensitive. The number of episode days assigned to be NO_x-sensitive by the PTM-like models A – F was no different to that assigned NO_x-sensitive by the other models G – M, both averaging about 7 days out of 15 days, though the assignments on any given episode day were not robust within the two groups of models.

7.1.2 UK- versus Rest of Europe dominance

A further important issue for policy-makers has been whether the balance of the effort in terms of O_3 precursor emission reductions should be made at home on UK emissions or abroad on the emissions from the Rest of Europe (RoE). To assess this issue, attention has been directed to the simple policy-relevant emission scenarios, S1 and S2, which focus on the influence of O_3 precursor sources in the UK versus those in the RoE. Since the UK emissions were included in the European emissions, an estimate of the impact of the RoE emissions could be obtained by subtraction of the UK impacts from the European (=UK+RoE) impacts. Therefore if, for that day and that model, the impact of the 30% reduction in both VOC and NO_x emissions carried out in the UK was greater than the

differences in impacts between across Europe and across the UK, then that day was assigned as UK-dominant. If however, the impact of the reductions carried in the UK was less than the differences in impacts between across Europe and across the UK, then that day was assigned as RoE-dominant. This subtraction assumes that O_3 responses are linear and additive, a reasonable working assumption for these small percentage reductions in O_3 precursor emissions.

Table 7.2 shows the UK- versus RoE-dominant assignments for each model and for each day. There was complete agreement as to UK- or RoE-dominance on only three days and some disagreement on the remaining 28 days. Again, it was apparent that assignment of the major source region of ozone precursors, whether in the UK or in the RoE, varied from day-to-day and back again. The question remains: which of the model assignments is correct and for each day.

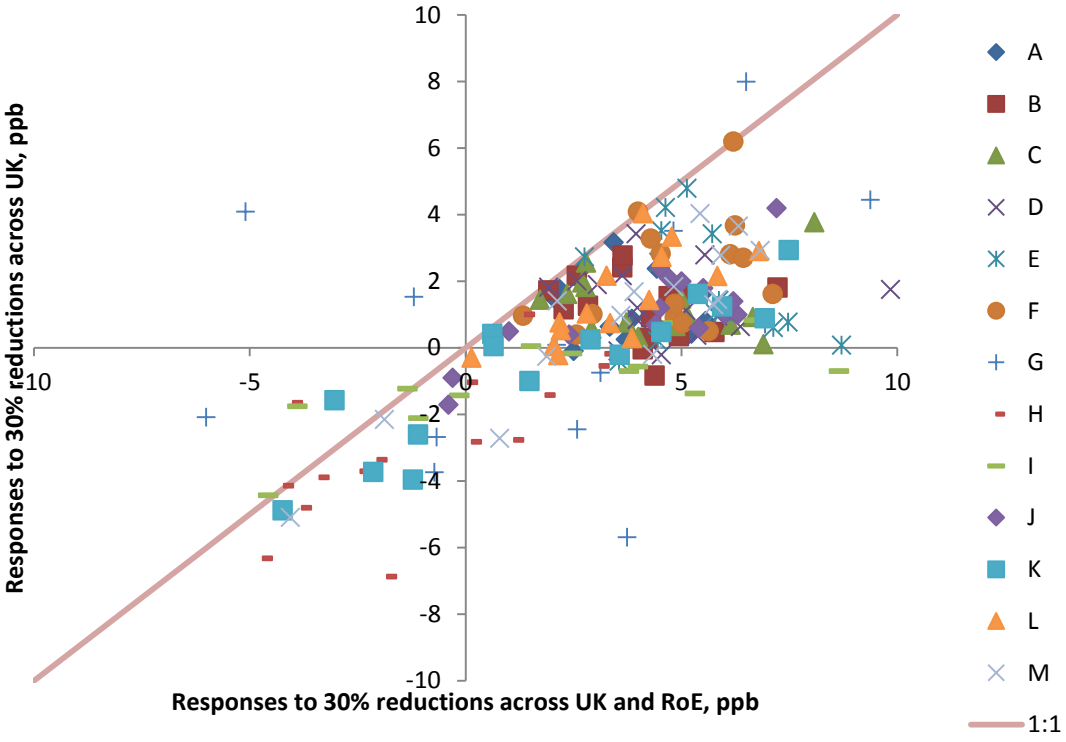
Table 7.2: Assignments as to UK- versus Rest of Europe-dominance for each model and for each day of July 2006.

Model	A	B	C	D	E	F	G	H	I	J	K	L	M
1st	RoE	RoE	RoE	UK	RoE	UK	UK	RoE	UK	RoE	RoE	RoE	RoE
2nd	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE
3rd	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE
4th	RoE	RoE	RoE	RoE	RoE	RoE	UK	RoE	RoE	RoE	RoE	RoE	RoE
5th	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	UK	RoE	RoE	RoE
6th	UK	UK	UK	UK	UK	UK	RoE	RoE	RoE	UK	RoE	RoE	RoE
7th	UK	RoE	UK	UK	UK	UK	RoE	RoE	RoE	RoE	UK	UK	RoE
8th	RoE	RoE	RoE	UK	UK	RoE	RoE	RoE	RoE	RoE	RoE	RoE	UK
9th	UK	RoE	RoE	UK	UK	RoE	RoE	RoE	RoE	RoE	UK	RoE	RoE
10th	UK	RoE	RoE	RoE	RoE	RoE	UK	RoE	RoE	RoE	UK	RoE	RoE
11th	UK	UK	UK	UK	UK	UK	UK	UK	RoE	UK	UK	UK	UK
12th	UK	UK	UK	UK	UK	UK	RoE	RoE	RoE	RoE	RoE	RoE	RoE
13th	RoE	RoE	RoE	RoE	UK	UK	RoE	RoE	RoE	RoE	RoE	RoE	RoE
14th	UK	UK	UK	UK	UK	UK	RoE	RoE	RoE	UK	RoE	UK	UK
15th	UK	UK	UK	UK	UK	UK	UK	RoE	RoE	UK	RoE	RoE	UK
16th	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE
17th	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	UK	RoE	RoE	RoE
18th	RoE	UK	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE
19th	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	UK
20th	RoE	RoE	RoE	RoE	RoE	RoE	RoE	UK	RoE	RoE	RoE	RoE	RoE
21st	RoE	RoE	RoE	RoE	UK	RoE	RoE	UK	RoE	RoE	RoE	UK	UK
22nd	UK	UK	UK	UK	UK	UK	UK	RoE	RoE	RoE	RoE	RoE	RoE
23rd	UK	UK	UK	UK	UK	UK	RoE	RoE	UK	RoE	UK	UK	RoE
24th	UK	UK	UK	UK	UK	UK	UK	RoE	RoE	RoE	UK	UK	RoE
25th	RoE	RoE	RoE	RoE	RoE	RoE	UK	RoE	RoE	RoE	RoE	RoE	RoE
26th	RoE	RoE	RoE	RoE	RoE	RoE	RoE	RoE	UK	RoE	RoE	UK	RoE
27th	UK	UK	UK	UK	UK	UK	RoE	RoE	RoE	RoE	RoE	UK	UK
28th	UK	UK	UK	UK	UK	UK	RoE	RoE	RoE	RoE	RoE	UK	UK
29th	RoE	RoE	RoE	RoE	RoE	RoE	UK	RoE	RoE	RoE	RoE	RoE	RoE
30th	UK	UK	UK	UK	UK	UK	RoE	UK	UK	RoE	UK	UK	UK
31st	RoE	UK	RoE	RoE	UK	UK	RoE	RoE	RoE	RoE	RoE	UK	RoE

Following on from the examination of the NO_x- versus VOC-sensitivity assignments, to make progress the analysis of UK- versus RoE-dominance will need to move on from categoric assignments. However, although the presence of an underlying relationship between NO_x- and VOC-sensitivity could be anticipated from theory, there is no such relationship between UK- and RoE-dominance. Figure 7.4 presents a scatter plot of the O_3 responses to 30% reductions in both NO_x and VOC emissions carried out across the UK and RoE plotted out

against the responses to 30% reductions carried out across the UK only, for all models and all episode days. Also shown is the 1:1 correspondence line which represents the locus of points where the responses to emission reductions carried out across the UK and the RoE are equal to those carried out across the UK. If the model responses were linear and additive, then there would be no points above the 1:1 correspondence line since that would imply that the response to the emission reductions carried out across the RoE, by difference, would be negative. Figure 7.4 shows that only a very small fraction of the points lie above the line and that the vast majority of the points lie below the line.

Figure 7.4. Scatter plot of the O_3 responses to 30% reductions in NO_x and VOC emissions carried out across the UK and the RoE versus the O_3 responses to 30% reductions in NO_x and VOC emissions carried out across the UK for episode days and all models.



Subtracting the O_3 response to the emission reductions carried out across the UK only from that from the UK+RoE yields an estimate for that carried out across the RoE only. The greater the response to emission reductions carried out across the RoE, the further the points move below the 1:1 correspondence line in Figure 7.4. RoE only responses are thus seen to be relatively large compared with UK only responses on all episode days and with all models. Nevertheless, there is considerable amount of scatter apparent in this figure. So much so, that it is difficult to draw robust conclusions about UK- or RoE-dominance on specific days using specific models.

Over all the episode days and all the models, the average O_3 response to 30% reductions in both NO_x and VOC emissions carried out across the UK was 0.6 ± 1.5 ppb, whereas that to reductions carried across the RoE was considerably greater at 2.6 ± 0.7 ppb. Episode days are highly likely to be RoE-dominant, on this basis, and this conclusion is robust to choice of model group, whether PTM-based or non-PTM based.

7.1.3 Conclusions

With considerable help from the Defra modelling teams, it has been possible to examine assignments as to NO_x- versus VOC-sensitivity and UK- versus RoE-dominance within the framework of the Defra model intercomparison exercise. The conclusion is that these assignments are not robust between the different days of July 2006 and between the different models. Robustness only appears when the assignments are averaged over a number of episode days or over a number of models.

This work will be written up for publication during the next project year.

8 Summary and Conclusions

The OSRM and PTM have been used to model and interpret the UK ground level ozone concentrations for 2011 and a range of different emission reduction scenarios to support Defra's ozone air quality policies.

As far as the EU Air Quality Directive compliance metrics are concerned, the measurements showed that 2011 was a year with relatively low ozone concentrations and this was supported by model runs using the OSRM. The OSRM tended to under-predict these ozone metrics for 2011, but was overall fairly consistent with the measurements. This is unusual because previously the OSRM has tended to over-predict the metrics in a 'low' ozone year.

The ozone season in 2011 was unremarkable in terms of severity or number of ozone episodes. However, there were photochemical ozone episodes during 2011 including periods in April and in late September/early October. In fact, the only memorable feature of 2011 was the length of the ozone season from early-April to early-October.

The PTM was used to diagnose and understand the nature of the 2011 episodes by modelling the responses of peak ozone concentrations at the Aston Hill site to hypothetical changes in NO_x and VOC emissions in the UK and rest of Europe. From the results, it was concluded that most of the episodes at this site were dominated by emissions from the rest of Europe and all but one of the episode days was NO_x-sensitive rather than VOC-sensitive. The latter finding was quite unusual because ozone episodes in the UK have a tendency to be in the VOC-sensitive category. There was a greater than usual preponderance for southerly trajectories during episode days in 2011. These are not normally associated with high ozone levels and it is suggested that the ozone episodes were predominantly NO_x sensitive in 2011 because there was this greater than usual preponderance for southerly trajectories.

Work has been undertaken to improve the efficiency and flexibility of the emission scenario pre-processor and of the treatment of shipping emissions in the OSRM. A new 1 x 1 km map of shipping emissions has been extended out to cover a much larger sea area, increasing the flexibility of the OSRM to run more complex scenarios accounting for future reductions in emissions from shipping. A new emission pre-processor has been developed for the OSRM that allows individual emission sub-sectors to be scaled in a more automated, flexible and transparent way for scenarios than is currently possible.

The OSRM and PTM have been used to model UK ozone for a range of different emission reduction scenarios of relevance to Defra's ozone air quality policy. The OSRM was used to model UK ozone for emissions in 2008 for direct comparison with simulations previously run for Defra based on projected emissions for 2020-2030 assuming the same meteorological conditions. The simulations consistently show how ozone concentrations are predicted to be higher in future years when emissions are reduced and all other conditions are maintained the same. The increase has been attributed to less efficient titration of ozone by NO_x in urban and suburban areas, in an area of NW Europe still saturated in NO_x. Moreover, there is an increasing trend in the emitted VOC/NO_x ratio in these future scenarios in an area of NW Europe which is VOC-limited.

The OSRM was also used to model various ozone metrics for an arbitrary 10% reduction in UK NO_x and VOC emissions separately. The results will be used by Defra to update health-, crop- and materials damage cost functions in relation to ozone to combine with those updated for other air pollutants. Under the conditions modelled, all metrics again show a small decrease for the 10% VOC reduction scenario and a larger increase for the 10% NO_x reduction scenario.

The PTM was used to model the impact that the 2020 emission reductions associated with the revised Gothenburg Protocol would have on UK ozone episodes experienced during 2011. The impact was found to be substantial, bringing about both increases and decreases in ozone at the different locations modelled under the 2011 episode conditions. However the overall impact of the Gothenburg Protocol emission reductions, averaging a net decrease in peak ozone of around 3.0 ± 4.7 ppb, was not sufficient to bring all the episodes, at all the sites during 2011, below the 50 ppb WHO guideline.

Finally, the results from the Defra Model Intercomparison Exercise were further analysed to compare and understand differences in the sensitivity of the model results to changes in ozone precursor emissions during episode conditions. With considerable help from the Defra modelling teams, it has been possible to examine assignments of ozone episodes in July 2006 to NO_x- versus VOC-sensitivity and UK- versus rest of Europe-dominance. The conclusion is that these assignments are not robust between the different days of July 2006 and between the different models. Robustness only appears when the assignments are averaged over a number of episode days or over a number of models.

9 Acknowledgements

We acknowledge the support provided by the Department for Environment, Food and Rural Affairs (Defra) and the Devolved Administrations (the Welsh Assembly Government, the Scottish Executive and the Department of the Environment for Northern Ireland) under contract AQ0722. We especially acknowledge the help and support of Peter Coleman and Sarah Honour of Atmosphere and Local Environment Programme (ALE) of Defra in facilitating the direction of the project in 2012/13.

10 References

Abdalmogith, S.S., Harrison, R.M., Derwent, R.G. (2006). *Particulate sulphate and nitrate in southern England and Northern Ireland during 2002/3 and its formation in a photochemical trajectory model.* Science of the Total Environment 368, 769–780.

Agren, C. (2012). *New Gothenburg Protocol adopted*. Acid News No.2, June.

AQEG (2009). *Ozone in the United Kingdom*. Report of the UK Air Quality Expert Group, AQEG. Prepared for the Department for Environment Food and Rural Affairs, the Scottish Executive, the Welsh Assembly and the Department of the Environment in Northern Ireland. Defra publications, London, 2009. ISBN 978-0-85521-184-4. http://archive.defra.gov.uk/environment/quality/air/airquality/publications/ozone/documents/aqeg-ozone-report.pdf

Bush T, I Tsagatakis, N Passant, A Griffin and B Pearson (2010). *UK Emission Mapping Methodology 2007*. Report for The Department for Environment, Food and Rural Affairs, Welsh Assembly Government, the Scottish Executive and the Department of the Environment for Northern Ireland. AEA Report AEAT/ENV/R/2863 (September 2010). http://www.airquality.co.uk/reports/cat07/1010011332_UKMappingMethodReport2007.pdf

Carslaw D (2012). *Defra Phase 2 regional model evaluation*. Draft working document for Defra, May 2012.

Colette A, C. Granier, Ø. Hodnebrog, H. Jakobs, A. Maurizi8, A. Nyiri, S. Rao, M. Amann, B. Bessagnet, A. D'Angiola, M. Gauss, C. Heyes, Z. Klimont, F. Meleux, M. Memmesheimer, A. Mieville, L. Rou¨ıl, F. Russo, S. Schucht, D. Simpson, F. Stordal, F. Tampieri, and M. Vrac (2012). "*Future air quality in Europe: a multi-model assessment of projected exposure to ozone*". Atmos. Chem. Phys., 12, 10613–10630 , 2012. http://www.atmos-chem-phys-discuss.net/12/14771/2012/acpd-12-14771-2012.html

Derwent, R.G., Jenkin, M.E., Saunders, S.M. (1996). *Photochemical ozone creation potentials for a large number of reactive hydrocarbons under European conditions*. Atmospheric Environment 30, 181–199.

Derwent, R.G., Jenkin, M.E., Saunders, S.M., Pilling, M.J. (1998). *Photochemical ozone creation potentials for organic compounds in northwest Europe calculated with a Master Chemical Mechanism*. Atmospheric Environment 32, 2429–2441.

Derwent, R., Witham, C., Redington, A., Jenkin, M., Stedman, J., Yardley, R., Hayman, G., (2009). *Particulate matter at a rural location in southern England during 2006: model sensitivities to precursor emissions*. Atmospheric Environment 43, 689–696

Derwent, RG, M E. Jenkin, S R. Utembe, D E. Shallcross, T P. Murrells, N R. Passant (2010a). *Secondary organic aerosol formation from a large number of reactive man-made organic compounds*. Science of the Total Environment 408 (2010) 3374–3381

Derwent, RG, C.S. Witham, S. R. Utembe, M. E. Jenkin and N. R. Passant (2010b). *Ozone in Central England: the impact of 20 years of precursor emission controls in Europe*. Environmental Science & Policy 13 (2010) 195-204

Derwent RG, A Fraser, J Abbott, M Jenkin, P Willis and T Murrells (2010c). *Evaluating the Performance of Air Quality Models*. Report published by for The Department for Environment, Food and Rural Affairs, Welsh Assembly Government, the Scottish Executive and the Department of the Environment for Northern Ireland, June 2010. http://www.airquality.co.uk/reports/cat05/1006241607_100608_MIP_Final_Version.pdf

Eden, P. (2011). Weather Log. April 2011. Weather 66 (6), i-iv.

Entec (2010). UK Ship Emissions Inventory. Final Report to Defra, November 2010. http://uk-air.defra.gov.uk/reports/cat15/1012131459_21897_Final_Report_291110.pdf

Hayman, G.D., J. Abbott, C. Thomson, T. Bush, A. Kent, RG Derwent, ME Jenkin, MJ Pilling, A. Rickard and L. Whitehead, (2006a) "*Modelling of Tropospheric Ozone*". Final Report (AEAT/ENV/R/2100 Issue 1) produced for the Department for Environment, Food and Rural Affairs and the Devolved Administrations on Contract EPG 1/3/200

Hayman, G.D., Y Xu, J. Abbott, T. Bush, (2006b) "*Modelling of Tropospheric Ozone"*. Report on the Contract Extension produced for the Department for Environment, Food and Rural Affairs and the Devolved Administrations on Contract EPG 1/3/200, AEA Report AEAT/ENV/R/2321 Issue 1, October 2006.

Hayman, G.D., J. Abbott, T.J. Davies, C.L. Thomson, M.E. Jenkin, R. Thetford and P. Fitzgerald (2010). *The ozone source–receptor model – A tool for UK ozone policy*. Atmospheric Environment, 44(34), 4283-4297.

Johnson, D., Utembe, S.R., Jenkin, M.E., Derwent, R.G., Hayman, G.D, Alfarra, M.R., Coe, H., McFiggans, G., 2006. *Simulating regional scale secondary organic aerosol formation during the TORCH 2003 campaign in the southern UK.* Atmospheric Chemistry and Physics 6, 403-418

Misra, A, N R Passant, T P Murrells, G Thistlethwaite, Y Pang, J Norris, C Walker, R A Stewart, J MacCarthy, M Pierce (2012). *UK Emission Projections of Air Quality Pollutants to 2030*. Report of the National Atmospheric Emissions Inventory. AEA Report AEA/ENV/R/3337. March 2012. http://uk-air.defra.gov.uk/reports/cat07/1211071420_UEP43_(2009)_Projections_Final.pdf

Monks P., R.S. Blake and P Borrell (2007). *Review of tools for modelling tropospheric ozone formation and assessing impacts on human health & ecosystems*. Report to Defra, November 2007

Murrells, T.P., Cooke, S., Kent, A., Grice, S., Derwent, R.G., Jenkin, M., Pilling, M.J., Rickard, A. and Redington, A (2008). "*Modelling of Tropospheric Ozone. First Annual Report"* produced for The Department for Environment, Food and Rural Affairs, Welsh Assembly Government, the Scottish Executive and the Department of the Environment for Northern Ireland under Contract AQ03508, AEA Report AEAT/ENV/R/2567, January 2008

Murrells, TP, S Cooke, A Kent, S Grice, A Fraser, C Allen, RG Derwent, M Jenkin, A Rickard, M Pilling, M Holland (2009). *Modelling of Tropospheric Ozone: Annual Report 2008*. Report for The Department for Environment, Food and Rural Affairs, Welsh Assembly Government, the Scottish Executive and the Department of the Environment for Northern Ireland. AEA Report AEAT/ENV/R/2748, February 2009.

Murrells, TP, S Cooke, J Abbott, A Fraser, RG Derwent, M Jenkin, A Rickard and J Young (2011). *Modelling of Tropospheric Ozone: Annual Report 2010*. Report for The Department for Environment, Food and Rural Affairs, Welsh Assembly Government, the Scottish Executive and the Department of the Environment for Northern Ireland. AEA Report AEAT/ENV/R/3134, February 2011

Murrells, TP, S Cooke, J Abbott, A Fraser, RG Derwent and M Jenkin (2012). *Modelling of Tropospheric Ozone: Annual Report 2011*. Report for The Department for Environment, Food and Rural Affairs, Welsh Assembly Government, the Scottish Executive and the Department of the Environment for Northern Ireland. AEA Report AEAT/ENV/R/3271, March 2012

NILU (2012). *FLEXTRA – air mass trajectories*. Norwegian Institute for Air Research. http://www.nilu.no/projects/ccc/trajectories/.

Passant, N.R. (2002) *Speciation of UK emissions of non-methane volatile organic compounds*. AEA Technology Report ENV-0545. Culham, Abingdon, United Kingdom, 2002

Passant NR, TP Murrells, A Misra, Y Pang, HL Walker, R Whiting, C Walker, NCJ Webb, J MacCarthy, 2012. UK Informative Inventory Report (1970 to 2010). '*Annual Report for submission under the UNECE Convention on Long-Range Transboundary Air Pollution*.' AEAT Report, March 2012. http://uk-air.defra.gov.uk/reports/cat07/1203221052_UK_IIR_2012_final.pdf

Sillman, S (1999). *The relation between ozone, NO_x and hydrocarbons in urban and polluted rural environments.* Atmospheric Environment 33, 1821-1845.

Sillman, S. and He, D. (2002). *Some theoretical results concerning O_3-NO_x-VOC chemistry and NO_x-VOC indicators*. Journal of Geophysical Research 107, D22, 4659, doi10.1029/2001JD001123.

WHO (2006). *Air quality guidelines. Global update 2005.* World Health Organisation Regional Office for Europe. Copenhagen

Williams, M, R Barrowcliffe, D Laxen and P Monks (2011). *Review of Air Quality Modelling in Defra. A report by the Air Quality Modelling Review Steering Group*. April 2011. http://uk-air.defra.gov.uk/reports/cat20/1106290858_DefraModellingReviewFinalReport.pdf

RICARDO-AEA

The Gemini Building
Fermi Avenue
Harwell
Didcot
Oxfordshire
OX11 0QR

Tel: 01235 75 3000

www.ricardo-aea.com