CMAQ Development for UK National Modelling

Phase 2: CMAQ-UK Final Report

Report for Department for Environment, Food and Rural Affairs

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Executive summary

The Community Multi-Scale Air Quality model (CMAQ) is an open-source model developed by the USEPA able to produce outputs for a range of air pollutants and processes simultaneously for research and regulatory purposes.

The Department for Environment, Food and Rural Affairs (Defra) commissioned a review involving a collaboration of three of the groups in the UK using CMAQ for national scale policy, the overall aim of which was to investigate and demonstrate how CMAQ might meet Defra's needs with respect to the national modelling and assessment of UK air quality policies and to develop a configuration optimised for those needs. The three groups participating in the project were King's College London (KCL), University of Hertfordshire (UH) and Ricardo-AEA, with further input from rdscientific (Professor Dick Derwent).

The project has involved the development and evaluation of the CMAQ model optimised in a way that will best meet Defra's future modelling and assessment needs. The overall work programme has been delivered in two phases. The initial evaluation of CMAQ and development and testing of a provisional optimised configuration of the model (CMAQ-UK) was carried out in Phase 1. The second phase of the project commenced in 2013 and has involved further more targeted development and evaluation of CMAQ-UK and an extensive demonstration of an application of CMAQ-UK to assess compliance with EU air quality objectives for $PM_{2.5}$ in 2020, a key date for achieving the $PM_{2.5}$ exposure reduction target and limit values required under EU Directive 2008/50/EC.

In its current configuration, the overall performance of CMAQ-UK for predicting concentrations of different air pollutants in the years 2009-2011 has been shown to be good, particularly on a regional scale. From the version developed in Phase 1, some improvements were seen in the predictions of NO_x and O_3, but $PM_{2.5}$ and PM_{10} remain under predicted by around 25%.

Based on the assumptions made concerning the meteorological conditions in 2020 and the latest UK and European emission projections, and taking into account the tendency for CMAQ-UK to under predict ambient $PM_{2.5}$, the results showed that the Average Exposure Indicator value for the 2019-2021 period will be 45% below the measured value for 2009-2011 indicating the exposure reduction target of 15% will be met.

The results demonstrate that CMAQ-UK can be used to produce some (but not all) of the required outputs for Air Quality Directive compliance reporting but this work has not considered developing and testing the model for compliance assessment purposes. The Model has been demonstrated to be more usefully used for the type of policy assessments currently performed by other models for Defra. It has been constructed with an efficient and transparent emission processing tool designed for testing a range of emission scenarios relevant to Defra policy applications.

CMAQ-UK has the advantage of providing a single modelling framework for Defra's policy applications and will benefit from future developments made by the wider WRF/CMAQ community and from developments and future availability of boundary conditions and biogenic emissions data arising from international projects.

Our main recommendations are:

- CMAQ-UK can be used for Defra's policy applications covering key pollutants at a regional scale. Further work is required for urban modelling and to ensure consistent behaviour between PM mass and PM components.

- The current configuration is well suited for regional-scale applications and coupling with other models would be required for improved finer-scale modelling and this

would require further development.

- Further work is required to develop the PM emissions speciation for individual source sectors and the quantification from sources such as dust resuspension.

- CMAQ-UK will need to be maintained to accommodate improvements in meteorology, boundary conditions, chemistry schemes and those parameters affecting $PM_{2.5}$ concentrations, including the description of secondary organic aerosol (SOA) formation.

- The 2019-2021 demonstration runs for $PM_{2.5}$ should be repeated with 2009-2011 meteorology so that the changes in $PM_{2.5}$ concentrations due to changes in emissions can be seen in isolation from the effects of changes in meteorology.

- Some further limited demonstration runs exploring the response of CMAQ-UK predictions to changes in emissions should be carried out. These could include runs to separate the effect of changes in UK vs European emissions as a typical policy application.

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1 Introduction

The Community Multi-Scale Air Quality model (CMAQ) is an open-source model developed by the USEPA able to produce outputs for a range of air pollutants and processes simultaneously for research and regulatory purposes.

The Department for Environment and Rural Affairs (Defra) commissioned a review of CMAQ involving a collaboration of three of the groups in the UK using CMAQ for national scale policy. The overall aim was to investigate and demonstrate how CMAQ might meet Defra's needs with respect to the national modelling and assessment of UK air quality policies and to develop a configuration optimised for those needs. The three groups participating in the project are King's College London (KCL), University of Hertfordshire (UH) and Ricardo-AEA, with further input from Professor Dick Derwent, rdscientific.

The project involves the development and evaluation of the CMAQ model optimised in a way that will best meet Defra's future modelling and assessment needs. The overall work programme has been delivered in two phases.

Phase 1 started in October 2011 and consisted of two main strands:

- Demonstration of CMAQ for Defra's Evidence Needs

- Development and evaluation of a provisional CMAQ-UK Configuration

The work was completed in July 2012 with a tested provisional version of CMAQ-UK optimised for Defra applications. The results were described in three detailed technical reports and one summary report at the end of Phase 1.

Following stakeholder meetings with CMAQ developers from the USEPA and with UK air quality modelling experts in June 2012, a targeted programme of further development and evaluation of CMAQ-UK was proposed for Phase 2, to be concluded with a more extensive demonstration due for completion in August 2014.

The main objectives for Phase 2 were to:

- Further optimise the configurations in the Weather Research and Forecast (WRF) meteorology model used in conjunction with CMAQ-UK, the boundary conditions and emissions data and provide further evaluation of the CMAQ-UK model performance.

- Conduct a set of simulations to demonstrate the applications of CMAQ-UK to a real policy issue, regarding the assessment of compliance with EU regulatory air quality obligations for $PM_{2.5}$ in 2020, a key date for achieving the $PM_{2.5}$ exposure reduction target and limit values required under EU Directive 2008/50/EC.

The demonstration tasks were to include CMAQ-UK runs for the three year periods 2009-2011 and 2019-2021. In parallel with this, further work was undertaken to specifically understand the treatment of secondary organic aerosol formation in CMAQ and to examine the benefit of the decision support tools available in the latest version of CMAQ.

This report summarises the main findings of the Phase 2 work, explaining the further optimisations made and the results and analysis of the demonstration runs. Further details of the model optimisation and sensitivity testing undertaken will be given in supplementary reports.

The report concludes with a summary of the report and key recommendations.

2 Optimisation of CMAQ-UK

Phase 2 had the initial objective to further optimise CMAQ-UK with development of methods and tools to enable optimisation and subsequent evaluation. The specific optimisation tasks were based on feedback from stakeholder meetings with CMAQ developers from the USEPA and with the UK air quality modelling community in June 2012. These included:

- **Weather Research and Forecast (WRF) meteorological model optimisation and sensitivity tests.** At the end of Phase 1, questions were raised about the role of the WRF meteorological model in the over prediction of observed NO_x (and an under prediction of O_3). Additional sensitivity tests were performed to answer these questions.

- **Boundary conditions development**. In Phase 1 the GEOS-Chem boundary conditions were used. This requires the GEOS-Chem model to be run. It was recommended in Phase 1 that the MACC global boundary conditions should undergo further investigation as a more viable alternative for CMAQ-UK modelling.

- **Emission processor development and evaluation of emissions**. Phase 2 requires new spatially-resolved emission data sets for 2009-2011 and 2019-2021 and a new emissions model process have been created to address this requirement. Emissions for 2006 using the new methodology were developed for testing against the CMAQ-UK results for 2006 derived in Phase 1.

This Section summarises the data and configuration options used for the optimisations. This configuration was subsequently used for the $PM_{2.5}$ demonstration runs. A comparison in the performance of CMAQ-UK developed in Phase 1 and this Phase 2 is given in Appendix A of this report.

To assess Defra's requirements on the compliance with regulatory air quality obligations for 2020, the WRF-CMAQ modelling system was used to simulate the present (2009-2011) and future (2019-2021) regional air quality over the UK. This required emissions inventory, meteorology and boundary conditions data developed specifically for these years.

The meteorological conditions for CMAQ version 5.0.1 air quality simulations have been prepared using WRF version 3.4.1 with horizontal grid resolutions at 50x50 km (covering the whole of Europe) and at 10 x 10 km (for the UK). The meteorological boundary conditions for the WRF simulation are from NCEP-FNL analysis and HadGEM-ES2 for present and future years respectively. The MACC reanalysis (present) and HadGEM-ES2 (future) are used as chemical boundary conditions for CMAQ simulations. The emission inventory for the CMAQ simulations is based on various sources which include the TNO-MACC emission inventory for Europe, the National Atmospheric Emissions Inventory (NAEI) area and point source emissions for the UK and ENTEC's gridded shipping emissions for surrounding sea territories.

The methodology and the options used for the WRF-CMAQ simulations are described in the sections below.

2.1 WRF Meteorology Data

Based on the evaluation of performance of WRF and CMAQ, the optimum configuration for WRF-CMAQ simulations for present years (2009-2011) and the future years (2019-2021) is shown in the Table 1. Table 1 describes the recommended surface layer, land surface and PBL schemes, grid nudging settings and vertical layer structure for WRF simulations.

Although the use of ACM2 scheme improved the performance of WRF and CMAQ predictions, there was no clear cut "best" model for all conditions and in all locations. In summary the recommended scheme tended to work best in urban areas which is important for $PM_{2.5}$ exposure evaluation and less well in rural locations. The ACM2 WRF-CMAQ model improved the prediction of NO_X, NO_2 and O_3 during the evening and overnight periods, although there is scope for improvement. It proved difficult to interpret the new NO_X results using the model's turbulence and surface meteorological performance alone. As a consequence, an additional set of diagnostic analysis was undertaken, using vertical mixing intensity (Kz) and ethane observations.

Table 1 Options used in WRF simulations

Parameter	Assumption
WRF version	3.4.1
Grid resolution	50km (Europe) ,10km (UK)
Spatial projection	ETRS89-LCC
IC/BC	NCEP-FNL reanalysis
Land use	USGS
Vertical layers	23
Nudging	Grid (U,V,T,Q)
Radiation (SW/LW)	Dudhia/RRTM
Cumulus	Kain-Fritsch
Microphysics	WSM 6 – class graupel
PBL	ACM2
Land surface	RUC
Surface layer	Pleim-Xiu

The use of Kz and ethane proved to be beneficial in interpreting the modelled concentrations and in resolving the relative role of emissions and dispersion in the performance of the model. The new diagnostic analysis suggested that NO_X emissions played an important role in the model's performance in January and that dispersion was important in July. It is therefore recommended that the use of these two diagnostics be incorporated into any further model evaluation. The analysis of cloud and precipitation observations showed that all model configurations under predict cloud cover by 50-60% and under predict precipitation by a factor of two in winter and at night time in summer. To improve the model's performance, further sensitivity analysis would be required to investigate the performance of different microphysics and cumulus schemes within the CMAQ-UK model.

The grid nudging analysis indicated that the Phase 1 CMAQ-UK provisional configuration, i.e. nudging all model layers every 6h, with a nudging coefficient of 3×10^{-4} s^{-1} for u and v wind component, temperature (T) and water vapour mixing ratio (Q), is most suitable for retrospective modelling. Sensitivity tests of the model using different layer structures showed that only the 35 layer scheme improved the model performance, but at the same time increased run times by a factor of two compared with the Phase 1 recommended scheme. With such small improvements it is recommended that the Phase 1 layer settings be retained.

2.2 Boundary Conditions

2.2.1 Meteorological Boundary Conditions

The NCEP-FNL reanalysis and HadGEM2-ES are used for present years (2009-2011) and future years (2019-2021) meteorological boundary conditions for WRF simulations, respectively.

NCEP FNL

The NCEP-FNL (Final) Operational Global Analysis data are on 1°x 1° grids prepared operationally every six hours. The data sets are publically available from July 1999 to present day from http://rda.ucar.edu/datasets/ds083.2. NCEP FNL is a part of the Global Data Assimilation System (GDAS), which continuously collects observational data from the Global Telecommunications System (GTS), and other sources. The analyses are available on the surface, at 26 pressure levels from 1000 mb to 10 mb, in the surface boundary layer and at some sigma layers, the tropopause and a few others. Parameters include surface pressure, sea level pressure, geo-potential height, temperature, sea surface temperature, soil values, ice cover, relative humidity, u- and v- winds, vertical motion and vorticity. The WRF pre-processing System (WPS)[1] prepares the initial and boundary meteorological conditions for the WRF[2] main module.

HadGEM2-ES

HadGEM2[3] is a global climate model run by the UK Meteorological Office and used for future climate predictions. The HadGEM2 family includes a coupled atmosphere-ocean configuration, with or without a vertical extension in the atmosphere to include a well-resolved stratosphere, and an Earth-System configuration which includes dynamic vegetation, ocean biology and atmospheric chemistry.

The standard atmospheric component has 38 levels extending to ~40km height, with a horizontal resolution of 1.25 degrees of latitude by 1.875 degrees of longitude, which produces a global grid of 192 x 145 grid cells. An interface, UM-WRF has been developed in order to force WRF with HadGEM2-ES data. The interface reads the HadGEM files (pp format), vertical interpolates to convert hybrid height into sigma pressure levels and writes to the WRF intermediate format. Parameters include sea surface temperature, temperature at 2m, relative humidity at 2m, sea-level pressure, surface pressure, wind-speed U at 10m, wind-speed V at 10m, water equivalent of accumulated snow depth, land-sea mask, surface geopotential heights, four layers soil moisture and soil temperature and 3D temperature, pressure, relative humidity, wind speed U and V and geopotential heights. These have been processed using the UM-WRF interface. Simulations for 2019-2021 used WPS and WRF to downscale global climate output into 50km and 10km for the EU and UK domain, respectively.

2.2.2 Chemical boundary conditions

The chemical boundary conditions used for CMAQ-UK simulations are MACC reanalysis for present years (2009-2011) and HadGEM2-ES for future years (2019-2021).

MACC

The MACC I/II (Modelling Atmospheric Composition and Climate[4]) is a research project funded by the European Union under the FP7 programme. The main aim of the project is to

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[1] http://www2.mmm.ucar.edu/wrf/users/download/get_sources.html#WPS
[2] http://www2.mmm.ucar.edu/wrf/users/download/get_sources.html#WRF-ARW
[3] Hadley Centre Global Environment Model version 2
[4] , www.gmes-atmosphere.eu

establish a global and regional atmospheric environmental service that covers European air quality, global atmospheric composition, climate and UV and solar energy delivered as a component of Europe's GMES (Global Monitoring for Environment and Security) initiative. The global model and data assimilation system used in MACC are based on the European Centre for Medium-Range Weather Forecasts' (ECMWF) Integrated Forecast System (IFS). Inness et al, (2012)[5] described the main modelling components as well as the assimilation of satellite data in the global model to generate the MACC reanalysis atmospheric composition data sets. The global MACC reanalysis service provides a reanalysis for the years 2003-2012 of trace gas and aerosol concentrations.

The experience gained by generating the reanalysis of atmospheric composition data as part of the GEMS project assisted with the development of MACC reanalysis data for this project. The latest version of MACC is used and benefited from the assimilation of more reprocessed satellite data compared with the one used in the GEMS project. This MACC modelling system has produced reanalysis of atmospheric composition data for the period 2003 to recent years, by assimilating satellite data to constrain O_3, CO, NO_2, CO_2, CH_4, and aerosol optical depth. There are 13 gas phase species that can be mapped from MACC directly onto the CB05 chemical scheme in CMAQ. These are: NO_2, NO, O_3, HNO_3, H_2O_2, CH_2O, CH_3CHO, CO, PAN, C_5H_8, SO_2, OH and C_2H_6. Particulate species available from MACC are primary organics, elemental carbon, sodium, chloride, SO_4 and desert dust. Desert dust is not treated explicitly in CMAQ and is therefore added to the non-speciated PM mass carried by the model.

HadGEM2-ES

The HadGEM2 Earth system model (HadGEM2-ES) is derived from the HadGEM1 with improvements in its components. The atmospheric component uses a horizontal resolution of 1.25 x 1.875 in latitude and longitude with 38 layers in the vertical layers extending over 39 km in height. In HadGEM2, the tropospheric chemistry was provided through climatological distribution. The main improvement of HadGEM2-ES is the inclusion of the UKCA chemistry scheme for modelling tropospheric chemistry and this will allow changes in air pollutants in the troposphere to be defined according to variations in the climate. Collins et al. (2011)[6] have provided a detailed description on the HadGEM2-ES model components, aerosol and tropospheric chemistry and the feedback mechanisms. There are 24 gas phase species that can be mapped from HadGEM2-ES directly onto CB05. These are: NO_2, NO, O_3, NO_3, N_2O_5, HNO_3, HONO, HNO_4, H_2O_2, HCHO, CH_3CHO, C_2H_5CHO, CO, CH_4OOH, CH_3OH, HCOOH, PAN, CH_3COOOH, CH_3COOH, C_3H_6, C_2H_4, MGLY, C_5H_8, and C_2H_6. Particulate species available from HadGEM2 are sulphate, elemental carbon and biomass-burning.

Methodology

The methodology to prepare boundary conditions for CMAQ simulations from MACC and HadGEM2-ES global models was developed as the part of the AQMEII project Phase 1[7] with the help of the US Environmental Protection Agency (EPA).

The main tools to prepare CMAQ boundary conditions from MACC/HadGEM2 global model data include:

- NCF2IOAPI – this tool reads the gas species and aerosol components from the original MACC/HadGEM2-ES reanalysis data and converts them into ioapi format for further use.

[5] Inness, A., Baier, F., Benedetti, A., Bouarar, I., Chabrillat, S., Clark, H., Clerbaux, C., Coheur, P., Engelen, R. J., Errera, Q., J. Flemming, M. George, C. Granier, J. Hadji-Lazaro, V. Huijnen, D. Hurtmans, L. Jones, J. W. Kaiser, J. Kapsomenakis, K. Lefever, J. Leitão, M. Razinger, A. Richter, M. G. Schultz, A. J. Simmons, M. Suttie, O. Stein, J.-N. Thépaut, V. Thouret, M. Vrekoussis, C. Zerefos, and the MACC team (2012). The macc reanalysis: an 8-yr data set of atmospheric composition. Atmospheric Chemistry & Physics Discussions, 12:31247–31347
[6] Collins, W. J., Bellouin, N., Doutriaux-Boucher, M., Gedney, N.,Halloran, P., Hinton, T., Hughes, J., Jones, C. D., Joshi, M., Lid-dicoat, S., Martin, G., O'Connor, F., Rae, J., Senior, C., Sitch,S., Totterdell, I., Wiltshire, A., and Woodward, S.: Development and evaluation of an Earth-system model HadGEM2, Geosci. Model Dev. Discuss., 4, 997–1062, doi:10.5194/gmdd-4-997-2011 , 2011.
[7] http://aqmeii.jrc.ec.europa.eu/

- MACC/HadGEM2-ES 2 CMAQ – this tool maps the species from the MACC/HadGEM2-ES to CMAQ with the CB05 chemical mechanisms. In order to put boundary condition data on to the CMAQ grid, the MACC/HadGEM2-ES 2 CMAQ tool uses MCIP grid description files as well as meteorological conditions.

2.3 Emissions Data

Emissions inventory data are collated primarily in formats for compliance with international inventory reporting commitments (UNECE and EU).

These inventory data sets, maps and projections are expressed as annual rates of emissions and are used to create the hourly emissions required by CMAQ. Creating emissions for CMAQ modelling involves identifying suitable emissions inventories and applying factors to convert the annual emission rates to hourly emission rates for the chemical species required for the chemical transformation schemes in the CMAQ model.

The National Atmospheric Emissions Inventory (NAEI) programme has recently undertaken a number of tasks to prepare emissions data suitable for all of Defra's air quality modelling activities. These studies focused on developing a range of supplementary data for the air pollution modelling community including profiles defining the temporal (hourly, daily and monthly) variation in emissions for different source sectors and pollutant species, recommendations for methods to calculate biogenic emissions, specific data to characterise point source emissions and recommended sources of inventory data to convert emission maps for the UK and rest of Europe for current years back to 2005 and forward to 2020 on a consistent basis.

More specifically, these include:

- Recommendations on European Emission Inventories for air pollution modelling in the UK
- Updated gridded emissions data for 2005 (used for 2006 modelling) based on the 2010 inventory methodology and source-specific scaling factors for other years
- Temporal emission profiles for source sectors at the SNAP 1 level.
- More detailed information on the temporal variation in emissions from road transport
- Monthly profiles for ammonia emissions from agriculture

In Phase 1, the emissions were created using 2006 emission maps generated in 2008, but the European and UK maps have since been updated using methodologies consistent with more recent versions of the inventory. In Phase 2 the emissions have been recreated using these latest NAEI updates and recommendations.

2.3.1 Anthropogenic emissions

Table 2 summarises the sources of anthropogenic emissions used to prepare CMAQ-UK data. The base inventory maps are developed periodically e.g. the NAEI has recently redeveloped the 2005 NAEI maps using the same methodology as for 2010 and for the years between 2005 and 2010 scaling factors are applied to generate maps for the required year based on trends in the national totals for each sector**.**

Table 2 Summary of inventory data used for the European (EU50) and UK (UK10) anthropogenic emissions grids

	European 50km grid	UK 10km grid
Europe – area	TNO-MACC (2006) (including UK)	TNO-MACC (2006) (excluding UK and shipping)
Europe - point	TNO-MACC (2006) (including UK)	TNO-MACC (2006) (excluding UK)
UK - area		NAEI (2005 scaled to 2006) (2011 scale to 2009, 2010, 2019, 2020, 2021)
UK - point		NAEI (from the 2011 database)
Shipping	TNO-MACC (2006)	Entec (for 2007) scaled by the NAEI to other years consistent with shipping emission regulations TNO-MACC outside the Entec area.

The anthropogenic inventories used are:

- TNO-MACC: the MACC II[8] emissions inventory for Europe for area and point sources (based on 2006) is widely used in Europe, including the MACC II regional air quality forecast and the international model intercomparison initiative, AQMEII, coordinated by the JRC in the EU and the US EPA. Based on the completeness of the data coverage, the spatial representation and the ability to manipulate the data to represent hourly emissions and policy scenarios the TNO-MACC maps are recommended for CMAQ modelling[9]. However this will be reviewed as new maps become available from EMEP, MACC and TNO. The 2006 TNO-MACC data have been scaled to 2009 using country- and sector-specific trends data provided by TNO. This has been cross-checked against 2009 data available from EMEP and found to be consistent at country level. To develop consistent European gridded emissions data for 2010 and 2011, the TNO-MACC data have been scaled to these years using country-specific data reported to EMEP[10]. In line with other Defra projects the 2019-2021 country scaling factors used to prepare future emissions are based on the TSAP_Mar13_CLE ("Current Legislation") scenario available through the GAINS Europe modelling system.[11] Examples of the factors used are in Appendix E .

- NAEI area source emissions: for the UK, the NAEI[12] is used. An updated 1x1km gridded dataset for UK emissions in 2005 developed using factors and methodologies consistent with the latest NAEI emission maps developed for 2010. These maps are accompanied by a set of sector- and pollutant-specific scaling factors to enable a consistent set of gridded emissions for 2006 to be processed from the 2005 map. The most recent maps for 2011 are used as the base maps for 2009-2011 and 2019-2021. These are scaled using source- and pollutant-specific factors recommended by the NAEI.

- NAEI point emissions: the point source emissions data for the UK are stored along with stack information in a database. This is updated each year to take account of new and more accurate information. These are then applied to historic data and

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[8] Pouliot, G., Pierce, T, Denier van der Gon, H., Schaap, M., Nopmongcol, U., 2012. Comparing Emissions Inventories and Model-Ready Emissions Datasets between Europe and North America for the AQMEII Project. Atmospheric Environment (AQMEII issue) 53, 4–14
[9] NAEI report "Recommendations on European Emission Inventories for Air Pollution Modelling in the UK" available from http://naei.defra.gov.uk/about-the-website
[10] EMEP/CEIP (2014): Present state of emissions as used in EMEP models; http://www.ceip.at/ms/ceip_home1/ceip_home/webdab_emepdatabase/reported_emissiondata/
[11] Amann M, Bertok I, Borken-Kleefeld J, et al. (2013) Policy scenarios for the revision of the thematic strategy on air pollution. TSAP Report #10. International Institute for Applied Systems Analysis, Laxenburg, Ausrtia. www.iiasa.ac.at/web/home/research/researchPrograms/MitigationofAirPollutionandGreenhousegases/TSAP-review.en.html report #10
[12] http://naei.defra.gov.uk/data/mapping

projected forward to provide a consistent time series. The points data used for all years in this study are from the 2011 database.

- <u>Entec</u>: a gridded shipping emissions inventory was developed by Amec (formerly Entec) under contract to Defra[13] in sea territories around the UK based on vessel movements in 2007. During the modelled period, the revised MARPOL Annex VI Regulations[14] came into force affecting SO_2 and PM emissions in different sea areas around the UK. UK grid maps have been created for 2006, 2009-2011 and 2020 using a new method to take into account up-to-date and projected estimates of the impacts of MARPOL and European Sulphur Content of Marine Fuels Directive on PM and SO_2 emissions from ships operating within the Emission Control Area[15] .

Further information on the emission projections used for the 2019-2021 simulations are given in Section 3.2.4.

2.3.2 Natural emissions

Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.0.4 was used for the preparation of the biogenic emissions. Twenty biogenic emission species including isoprene and various terpenes were simulated with the MEGAN based on the meteorology[16]. The simulated biogenic emissions at the model's lowest layer were merged with the anthropogenic emissions data generated. The sea salt contribution to particulate matter is calculated within CMAQ based on the meteorology.

2.3.3 Factors

Factors are used to convert the basic annual inventory emission rates into gridded hourly emissions required by CMAQ. As described above scaling factors are used to generate emission maps for the years where maps are not available. Temporal and speciation factors have been developed based on UK activity patterns to convert the annual rate of emissions data into the hourly data required by CMAQ. Vertical profiles are used to introduce the point source emissions into the correct layer of the model.

2.3.3.1 Temporal emission factors

The NAEI made recommendations for temporal profiles for each source sector following a review of UK activity data and profiles developed by TNO[17]. The UK temporal profiles are applied across the European area, taking into account time zones.

Recommendations have been made for the following main SNAP sectors:

- **Power generation emissions –** new factors are based on 2010 and 2011 emissions data from primarily coal fired power stations in England and Wales.
- **Domestic and commercial and institutional combustion –** this sector is a complex mixture of emissions sources including domestic homes and institutions such as schools, hospitals, prisons, council offices. For 2006, SNAP 2 emission maps were not separated into sub SNAP sectors. To address this, weekly and hourly profiles were developed that took account of the relative contribution of residential and non-residential sources on a monthly basis. New monthly profiles based on daily temperature were developed from a degree-days concept.
- **Road transport –** the road transport profiles in Phase 1 were based on the London profiles developed by King's College and consisted of daily profiles for weekdays and weekends. New profiles for average UK traffic have been developed for passenger cars

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[13] http://uk-air.defra.gov.uk/reports/cat15/1012131459_21897_Final_Report_291110.pdf
[14] MARPOL (2010): Revised MARPOL Annex VI, Prevention of Air pollution from shipping.
http://www.imo.org/OurWork/Environment/PollutionPrevention/AirPollution/Pages/Air-Pollution.aspx
[15] Tsagataskis, I., Brace, S., Passant, N., Cooke, S. (2013): UK Emission Mapping Methodology 2011
http://uk-air.defra.gov.uk/reports/cat07/1403100909_UK_Emission_Mapping_Methodology_2011-Issue_1.pdf
[16] http://acd.ucar.edu/~guenther/MEGAN/MEGAN.htm
[17] Temporal Emission Profiles – report of the NAEI (January 2014)

and commercial vehicles on different road types in Phase 2[18] based on Dft traffic statistics[19].

- **Ammonia emissions from agriculture –** Monthly estimates of ammonia emissions were estimated[20] using monthly activity data for different sources of emissions. This includes estimates for cattle, pigs, poultry and sheep taking into account the different husbandry methods, emission from manure stores and application of nitrogen fertiliser. It is more difficult to allocate emission by hour of the day; many of the emission are continuous over 24hrs whilst others have a diurnal profile based on temperature or husbandry methods. In Phase 1 a constant profile was used, in Phase 2 a diurnal profile has been introduced.

2.3.3.2 Speciation factors

Speciation factors are used to disaggregate total emissions of VOCs and PM into their chemical components. There has been no change in the VOC and PM speciation between Phase 1 and Phase 2.

- **VOC speciation**. - VOC speciation to forms relevant to different chemistry schemes is calculated using the spreadsheet developed by Ricardo-AEA and is based on the NAEI's existing NMVOC speciation profile[21].
- **PM Speciation** - $PM_{2.5}$ speciation is based on the AQMEII recommendations. The new species required for CMAQv5 have been introduced with zero values.
- **NOx Speciation** – Based on the NAEI factors referring to the fraction of NO_x emitted directly as NO_2 (f-NO_2) [22] [23], the road traffic profile is year specific. For 2006, the average f-NO_2 for all traffic is 12.7 % increasing to 19.9%, 20.4% and 21.5% for 2008, 2010 and 2011 respectively and 22.1%, 21.5% and 21.0% for 2019, 2020 and 2021.

2.3.3.3 Point Source emissions

The UK points database has over 5000 records. A number of approaches have been considered for applying this to CMAQ-UK. In Phase 2 a simple method has been used based on the vertical profiles of distribution of emissions as outlined in Bieser[24] (2011). There are 73 profiles provided in the paper. Taking this and the vertical structure of CMAQ-UK into account, three vertical profiles are used for autumn/winter, spring and summer to release the emissions into layers 5, 6 and 7. There are approximately 200 point sources above the 95th percentile of emissions for CO, NO_x, SO_2 or PM_{10} where stack details are available. These can be released directly into CMAQ using in-line processing (CMAQv5) if required. For Phase 2 all point sources were treated in the same way.

The same vertical factors are used for the European point source emissions as no stack details are available with the TNO-MACC data.

2.3.4 Emissions Processor

A system for processing the inventory data into CMAQ-ready data was developed. A new suite of programs was developed based on the R programming language to create the CMAQ emissions data. This will be described in a supplementary document, but includes a series of models including:

- MEGAN biogenic model

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[18] Temporal variation in emissions from road transport - report of the NAEI (January 2014).
[19] https://www.gov.uk/government/statistical-data-sets/tra03-motor-vehicle-flow Table TRA0307
[20] Temporal Profiles for Ammonia Emissions from Agriculture - report of the NAEI (March 2013)
[21] Passant, N (2002). Speciation of UK emissions of non-methane volatile organic compounds. Report of the NAEI AEAT/ENV/R/0545 (February 2002). http://uk-air.defra.gov.uk/assets/documents/reports/empire/AEAT_ENV_0545_final_v2.pdf
[22] Murells et al., (2010) An Emissions Inventory for Primary NO_2 and Projections for Road Transport: 2008 NAEI: NAEI reference 48954007 http://naei.defra.gov.uk/datachunk.php?f_datachunk_id=324
[23] Murrells, T., MacCarthy, J., Passant, N. An Emissions Inventory for Primary NO2 and Projections for Road Transport: 2008 NAEI. Report 21 March 2010, NAEI Ref 48954007. Not published
[24] J. Bieser, A. Aulinger, V. Matthias, M. Quante, H.A.C. Denier van der Gon, Vertical emission profiles for Europe based on plume rise calculations , Environmental Pollution 159 (2011)

- Anthropogenic Emissions Toolkit (AET). The process steps are written as a series of R functions; these are generic and can be applied to any modelling grid on which the base emissions have been created.
- The final stage combines the anthropogenic and biogenic emissions and writes the data into the CMAQ file format.

This is a flexible system, separating the preparation of the base emissions inventory data (annual emission rates) from the application of the scaling, temporal, speciation and scenario factors so that the choice of emission inventory or changes to the factors can be made independently without the need to reprocess all the data. The factors can be developed independently based on pollutant, emission sector, time and country to create CMAQ-ready emissions data for complex scenarios that may be required by Defra. It is only the final stage of the process that is CMAQ specific and this can be adapted for other models.

2.4 CMAQ version

The CMAQ-UK consortium has recommended to use CMAQ version 5.0.1 for the present (2009-2011) and future (2019-2021) year scenario model simulations.[25] The decision was made on the basis of a comparison of CMAQ simulation results from v4.7.1 (used in Phase 1) and the more recent version v5.0.1 (see Appendix A) where the model results are compared with the observations for various air pollutants in the UK.

To summarise, Table 3 shows the main options used in the CMAQv5.0.1 for Phase 2.

Table 3 Options used in CMAQ-UK simulations

Parameter	Assumption
CMAQ version	5.0.1
Grid resolution	50km (Europe) to 10km (UK)
Spatial projection	ETRS89-LCC
Vertical layers	23 (7 below 1km)
IC/BC	MACC2/HadGEM2-ES
Chemical Scheme	CB05tucl_ae6_aq
Aerosol Scheme	Aero 6
Emissions processor	EMST (MEGAN and AET)
Area anthropogenic emissions	TNO-MACC/NAEI
Temporal emissions profiles	NAEI (Ricardo-AEA)
Point anthropogenic emissions	NAEI
Natural emissions	MEGAN

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[25] http://www.airqualitymodeling.org/cmaqwiki/index.php?title=CMAQ_version_5.0_%28February_2012_release%29_Technical_Documentation

3 CMAQ-UK Model Results

This section discusses the evaluation of CMAQ-UK air quality simulations for the year 2009 – 2011 and 2019 - 2020 periods, using methods from Defra's model inter-comparison exercise (MIE). The input emissions, boundary conditions and meteorology vary across years, with detailed setup of these inputs described in Section 2. This section also discusses the influence that the yearly variation of boundary conditions has on WRF and CMAQ-UK performance.

Further details of the evaluation are given in the following sections, but the main findings of this work are summarised as follows:

- The CMAQ-UK model predictions of annual mean NO_2 compare reasonably well with observations. The number of days when $NO_2 > 200$ µg m^{-3} predicted by the model is below the EU limit value, which is typical of the background concentrations.
- The CMAQ-UK model predictions of the ozone daily maximum 8-h running mean are in good agreement with observations although slightly less so for the number of days that daily maximum 8h running mean > 40 µg m^{-3} and AOT40 metrics.
- The bias in O_3 predictions at rural sites is partly attributed to model boundary conditions.
- The CMAQ-UK model predicts PM_{10} and $PM_{2.5}$ daily and annual averages considerably better than hourly averages. In common with other chemistry transport models it under predicts PM.
- PM_{10} predictions at rural sites tend to agree better with observations than at urban sites.
- The measurements show that the urban increment of PM_{10} can be as high as 9 µg m^{-3} while the modelled urban increment is only ~3 µg m^{-3} in 2011. A similar conclusion can be drawn for $PM_{2.5}$.
- The bias in NO_2, O_3, PM_{10} and $PM_{2.5}$ predictions can be improved by improving the spatial distribution of the precursor emissions.
- The model predicts most PM components well although there is a positive bias for SO_4^{2-} and a negative bias for carbonaceous species.
- There remains a need for further development of CMAQ-UK's prediction of organic aerosols.
- The model captures the inter-annual variability of meteorology well and has performed consistently in predicting pollutant concentrations.
- The model is successfully applied to predicting $PM_{2.5}$ and exposure reduction target in 2020.
- The model suggests a large reduction in future $PM_{2.5}$ concentrations, as well as reductions in PM_{10} and NO_2 towards 2020. The reduction in O_3 is half of the reductions shown by $PM_{2.5}$, PM_{10} and NO_2. For $PM_{2.5}$, this would suggest that the EU exposure reduction target would be met in the UK, although this is based on the meteorology used for the 2019-2021 period.
- The reduction of $PM_{2.5}$ in 2020 is driven by a reduction of its precursor emissions, particularly NOx (44%) and SO_2 (40%), and may be partly attributed to changes in meteorological conditions over the UK, such as increasing rainfall rates by ~50%, or boundary conditions.
- To ensure that the CMAQ-UK's future predictions of $PM_{2.5}$ are robust, a comparison of model and measured trends in PM components should be undertaken.

3.1 Analysis of 2009-2011 Simulations

3.1.1 NOx and NO2

The 2009-2011 modelled surface concentrations of NO_2, NO_x, O_3, PM_{10} and $PM_{2.5}$ have been compared against 23 monitoring sites used in Defra's Modelling Intercomparison Exercise[26] (see dashboard in Appendix B and Figure1). The statistical measures of model performance show that the model bias is within +/- 25% for both NO_2 and NO_x with NO_2 in year 2009 having the largest bias. On average, over 50% of predicted NO_x and NO_2 are within a factor of two of the measurements with a RMSE of 51 and 19 µg m^{-3},respectively. For NO_x, the r value is low at ~0.4 and COE is ~0.24 and for NO_2, the r value is ~0.6 and COE is ~0.27, meaning that the model does not describe well the hourly variance in observations.

Figure 1: Statistical performance of CMAQ-UK in predicting concentrations of NO2, NOx, O3, PM10 and PM2.5 for the 2009-2011period

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[26] http://uk-air.defra.gov.uk/research/air-quality-modelling?view=intercomparison

For policy metrics, NO_2 annual mean predictions are compared with observations at rural, suburban and urban sites (Figure 2). The predictions of annual mean NO_2 are mostly within a factor of two of the measurement especially at urban sites, although a relationship between model bias and the magnitude of NO_2 is evident with a negative model bias for the highest concentrations (> 40 µg m^{-3}) and a positive model bias for the remaining sites.

Note, the number of days when NO_2 > 200 µg m^{-3} is below the EU limit value, which is typical of the background concentrations predicted by the model so the performance of CMAQ in predicting this metric is not discussed further.

Figure 2: Scatter plot of annual means NO_2 concentrations for 2009, 2010 and 2011

2009   2010   2011
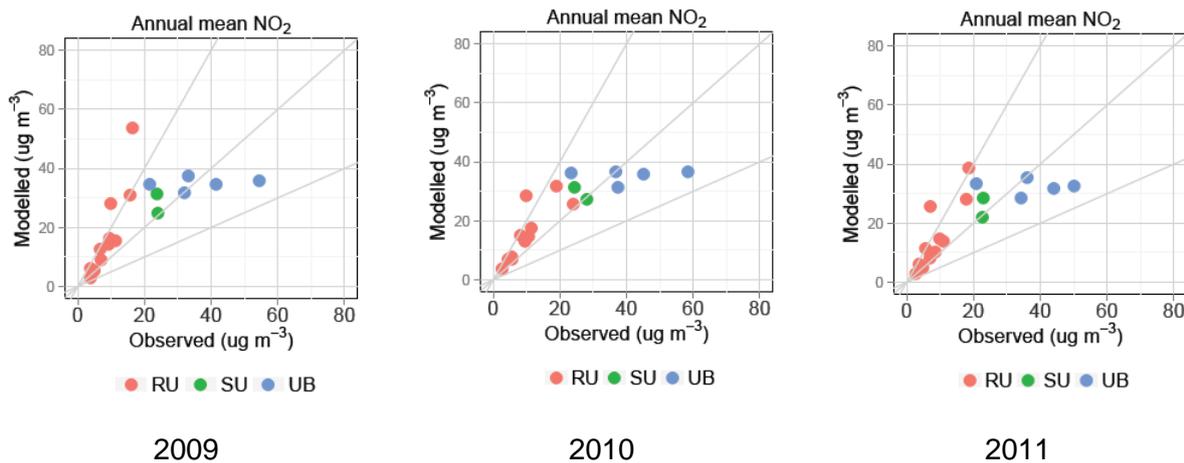
3.1.2 Ozone

The model evaluation statistics indicate that of all the pollutants, CMAQ is most skilful in predicting O_3. Over 75% of modelled results are within a factor of two of the measurements. The MB and NMB values are very small ~1% and with an RMSE of ~20 µg m^{-3}, r of ~0.7 and COE ~ 0.25, CMAQ has good performance overall.

The model performance in predicting daily maximum 8-h running mean (8hDx), number of days that daily maximum 8-h running mean > 100 µg m^{-3} (8hDx100) and AOT40 for 2009, 2010 and 2011 are shown in the O_3 dashboard (Appendix C). The modelled 8hDx is consistently in good agreement with the measurements across all three years. More than 80% of the 8hDx predictions are within factor of two of the observations. The MB and NMB values at both rural and urban sites are within +/- 20%, although slightly over 20% at suburban sites in 2009. The NMB values at the rural sites are small (≤3%). The r and COE values at the urban sites tend to be larger than the rural sites.

The predictions of 8hDx100 and AOT40 reflect the absolute concentrations of O_3 above the threshold values. The statistics indicate a larger bias in predicting the 8hDx100 and AOT40 than the 8hDx or short-term averages. However, over 50% of predicted 8hDx100 and AOT40 are in a factor of two of the measurements. There is a positive bias for 8hDx100 and AOT40 values at urban sites with the opposite true at rural sites.

3.1.3 PM$_{10}$

Averaged across the years 2009 to 2011, the model under predicts PM$_{10}$ by 24%. Over 60% of the model results are within factor of two of the measurements and with an r value of ~ 0.6 the model is reasonably able at describing the observed PM$_{10}$ variance. The RMSE is ~11 µg m^{-3}, and COE is ~ 0.11. Lack of PM species measurements makes it difficult to identify the precise parameters responsible for the bias in PM$_{10}$ predictions, however the analysis at Harwell and North Kensington in 2009 suggest that improvement of coarse mode PM would improve total PM$_{10}$ predictions.

CMAQ-UK's performance in predicting PM_{10} policy metrics are shown in the PM_{10} model dashboard (Appendix C), and shows that PM_{10} predictions are well within a factor of two of the measurements, particularly for annual mean concentrations. The model predicts long-term averages considerably better than short-term averages and this is reflected in improvements in all evaluation statistics. The predictions at rural sites tend to have better agreement with observations than at urban sites. The measurements in 2011 show that the urban increment on PM_{10} can be as high as 9 µg m^{-3} while the modelled urban increment is only ~3 µg m^{-3}. This indicates that the urban PM emissions may be underestimated. Through underestimating PM_{10} the model predicts a very small number of days where the daily maximum PM_{10} concentration > 50 µg m^{-3}.

3.1.4 $PM_{2.5}$

Over 60% of the $PM_{2.5}$ predictions are within factor of two of the measurements, although the model underestimates the measurements by ~25%. The RMSE is ~10 µg m^{-3}, r is ~0.6 and COE is ~0.13.

Similar to PM_{10}, the predicted long-term average concentrations are better than the short-term average metrics. The statistics such as FAC2, RMSE and r show that daily mean $PM_{2.5}$ concentrations are better than hourly mean values.

Apart from 2009, the negative biases at rural sites are smaller than urban sites, suggesting that the predictions of urban increment are smaller than the measurements. Further investigation on spatial distribution of PM emissions would have potential benefits in model performance.

3.1.5 PM_{10} and $PM_{2.5}$ components

CMAQ-UK's performance in predicting PM chemical species is analysed against the only available measured datasets for the UK, including: SO_4^{2-}, NO_3^-, Cl^-, OC and EC (all are of PM_{10}) at Harwell and North Kensington in 2009 and SO_4^{2-}, NO_3^-, NH_4^+ (all are of $PM_{2.5}$) at Auchencorth Moss in 2010 and 2011 (see an example of PM components dashboard in the Appendix C).

The model captures the seasonal trends of all PM components well, although it over predicts PM_{10} NO_3^- in spring 2009. The model over predicts PM_{10} NO_3^- by 1 µg m^{-3} (~42%). The $PM_{2.5}$ NO_3^- and $PM_{2.5}$ NH_4^+ are in good agreement with the measurements. The model predicts small positive bias for PM_{10} Cl^- (~0.14 µg m^{-3} or 11%).

The model has a tendency to under predict OC by ~1.7 µg m^{-3} (~60%) and EC by 0.4 µg m^{-3} (~ 36%) and over predicts PM_{10} SO_4^{2-} by 1.3 µg m^{-3} (~85%) and $PM_{2.5}$ SO_4^{2-} by 0.5 µg m^{-3} (~36%). SOA is grossly underestimated and this contributes to the underestimation of $PM_{2.5}$ and PM_{10}.

The analysis at Harwell and North Kensington indicates that the model under predicts coarse mode PM, while it agrees well with the magnitude of coarse PM at the Auchencorth Moss. This indicates that the model may benefit from further development in the spatial distribution of coarse mode PM emissions.

3.1.6 Meteorological evaluation

The performance of WRF in predicting relative humidity (rh %), ambient temperature (ta ^{0}C), and wind speed (ws m/s), from 2009 to 2011, is quantified at 11 Defra MIE meteorological sites (see Appendix D for WRF model performance dashboard). The model predicts the inter-annual variability of the meteorology well. Overall, the statistics (Table 4) indicate that over 80% of the predictions of rh, ta and ws are within a factor of two of the observations. The model captures the monthly and diurnal trends of these three variables well although a small positive bias (5%) is observed in rh predictions in spring and between afternoon and evening. On average the model over predicts rh by only 3% with RMSE of 10.9%, r of 0.7 and COE of 0.27. The model is able to capture the magnitude, size and temporal distribution

of temperature and wind speed well. The median values of ta and ws are observed to align with the perfect model area. Small negative biases of ta (5%) and ws (4%) are observed. For ta, RMSE is 1.6°, r is 0.95 and COE is 0.71. The ws has a RMSE of 2.1 m/s, r of 0.74 and COE of 0.34.

Table 4 Statistical performance of WRF3.4.1 in predicting relative humidity (rh, %), ambient temperature (ta, ^{O}C), and wind speed (ws, m/s) for 2009 - 2011.

Param.	# data	Observed mean	Modelled mean	FAC2	MB	MGE	NMB	NMGE	RMSE	r	COE
rh	287006	81.7	84.0	1.0	2.36	8.3	0.029	0.10	10.9	0.67	0.27
ta	271122	10.6	10.4	0.95	-0.28	1.2	-0.026	0.12	1.6	0.95	0.71
ws	287881	4.8	4.7	0.84	-0.17	1.5	-0.035	0.32	2.1	0.74	0.34

3.1.7 Boundary conditions

In Phase 1 we have investigated the impact of chemical boundary conditions from different global chemical models (STOCHEM, GEMS, GEOS-Chem and MACC) on the concentrations of gaseous and aerosol species simulated in the CMAQ-UK modelling system for the year 2006[27]. CMAQ-UK has been used to simulate January 2006 and July 2006 as representations of winter and summer seasons. Comparison of spatial distribution of ozone and PM concentration from different CMAQ simulations suggested that the use of boundary conditions from different global models has different influences on the overall chemical mechanism and transport of air pollutants (horizontally as well as vertically) in the regional models.

The CMAQ simulation with MACC boundary conditions simulated high ozone and particulate matter concentrations as compared to CMAQ simulation with STOCHEM, GEMS, and GEOS-Chem boundary conditions. The comparison of modelled and observed vertical distribution of ozone at Lerwick ozonesonde station in the UK showed that all the model simulations reproduced the vertical structure of ozone fairly, but with some under estimation. CMAQ simulations with MACC boundary conditions reproduced the vertical structure better compared with other CMAQ simulations. Overall the hourly time series comparison and the statistical measures revealed that CMAQ simulation with MACC boundary conditions performed better than CMAQ simulations with STOCHEM, GEMS, and GEOS-Chem boundary conditions for winter and summer months of the year 2006.

To further examine the sensitivity of MACC boundary conditions on the prediction of tropospheric ozone concentration, CMAQ-UK modelling simulations have been performed for the years 2009, 2010 and 2011. The evaluation of the performance of CMAQ-UK on the concentration of ozone is done by comparing the results with ozone concentrations observed at the Mace Head station. The Mace Head station is one of the most important background stations located on the west coast of Ireland. It is a unique site which offers the opportunity to study atmospheric composition under Northern Hemispheric background conditions as well as European continental emissions when the winds favour transport from that region. It also offers westerly exposure to the North Atlantic Ocean.

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[27] Beevers S, Kitwiroon N, Beddows A, Carslaw, D, Good N, Chemel C, Xavier Francis, Sokhi R, Derwent D, Fraser A, Murrells T, and Venfield H. (2012). CMAQ Development for UK National Modelling - Development of a provisional CMAQ-UK Configuration. Report to Defra Version: 31st August 2012"

Figure 3: Variation of bias of daily ozone concentration at Mace Head station from MACC reanalysis and CMAQ for the 50km European (left hand panel) and 10km UK (right hand panel) simulations.
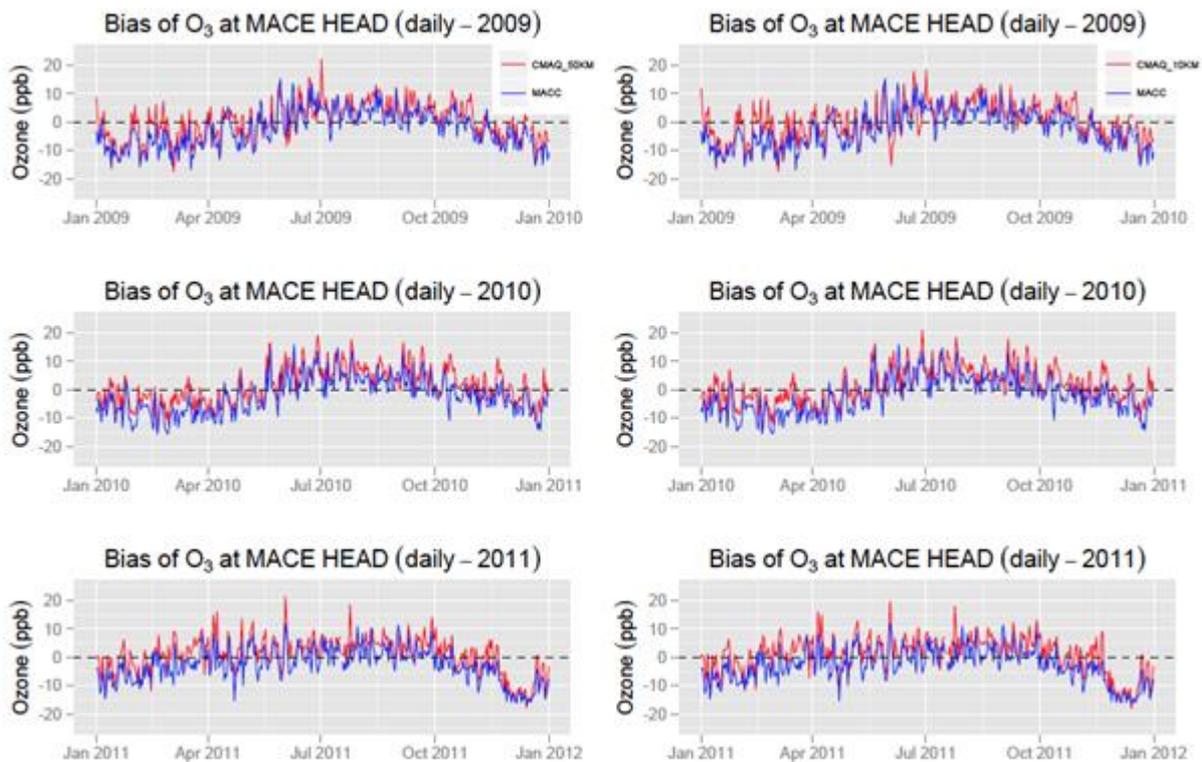
Figure 3 shows the variation of bias of daily ozone concentration at Mace Head station simulated in the CMAQ and MACC reanalysis for the years 2009, 2010 and 2011. The left hand panel shows the bias of concentration of ozone from MACC reanalysis and CMAQ simulation with 50 km horizontal resolution. The right hand panel shows the bias of ozone concentration from MACC reanalysis and CMAQ simulation with 10 km horizontal resolution. A similar behaviour in terms of variability of ozone concentration bias from both CMAQ simulations is observed. The ozone concentration bias for the years 2009 and 2010 shows that an underestimation of ozone concentration lasts from January to May and the largest underestimation (negative bias, 10-15 ppb) is from the end of February to beginning of March. During June-August both MACC reanalysis and CMAQ simulations over-estimated the ozone concentration (positive bias, 2-15 ppb). In 2011, both model simulations and MACC reanalysis show less bias as compared to model simulations and MACC reanalysis of years 2009 and 2010.

Figure 4: Variation of bias of daily maximum ozone concentration at Mace Head station from MACC reanalysis and CMAQ for the 50km European (left hand panel) and 10km UK (right hand panel) simulations.
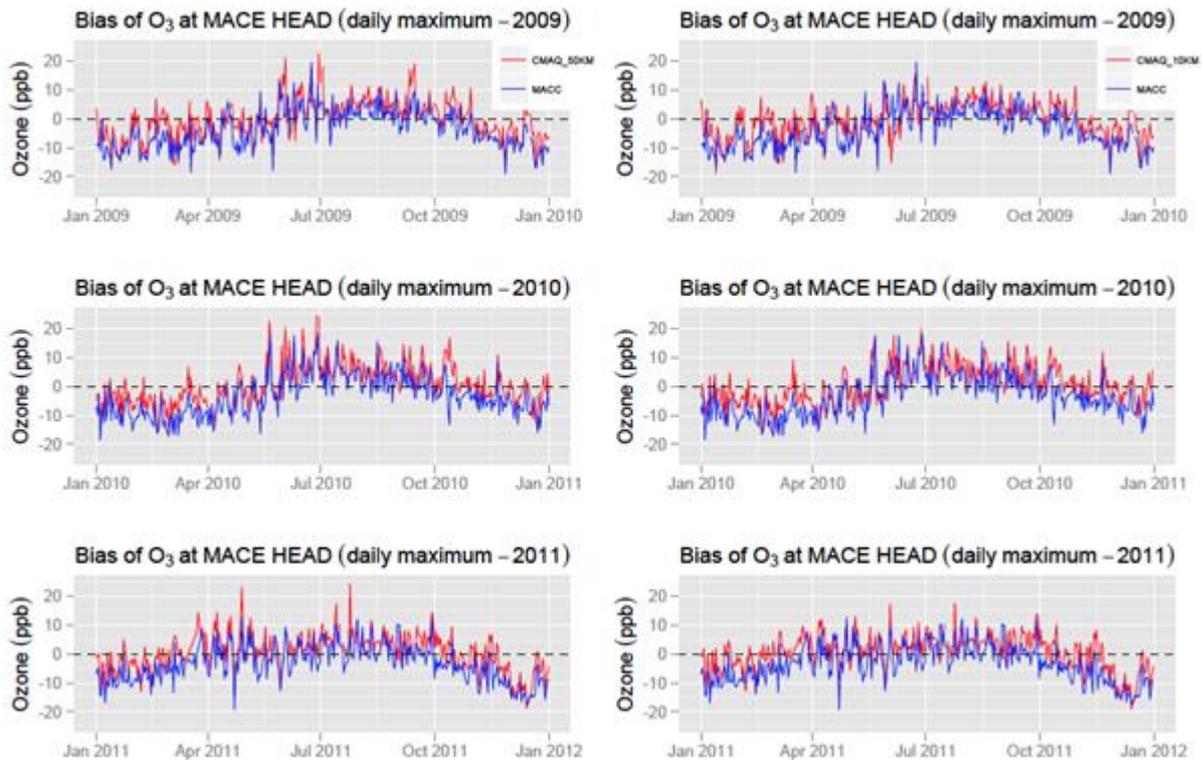
Figure 4 shows the variation of **daily maximum ozone** concentration bias at Mace Head station simulated in CMAQ simulations and MACC reanalysis for the years 2009, 2010 and 2011. The figure shows that the variation of bias is very similar to results found in the Figure 3. Figure 3 and Figure 4 show that the bias variation in CMAQ simulations always follows the bias variation in MACC reanalysis. This shows that any variations in the boundary conditions will directly affect the simulation of ozone concentrations in CMAQ at Mace Head station.

Figure 5 depicts the monthly averaged ozone concentration at Mace Head from observation, MACC reanalysis and CMAQ simulations for years 2009, 2010 and 2011. The variation of monthly averaged ozone concentration shows no significant difference in the CMAQ simulations with 50 km and 10 km horizontal resolution at Mace Head station. The figures show that MACC boundary conditions underestimate ozone by close to 10 ppb in winter and overestimate by 5 ppb in summer 2009 and 2010 at Mace Head. In 2011, as depicted in Figures 3 and 4, MACC reanalysis show less bias during winter (negative bias) and summer (positive bias) seasons.

Figure 5: Variation of monthly ozone concentration at Mace Head station from observation, MACC reanalysis and CMAQ simulations.
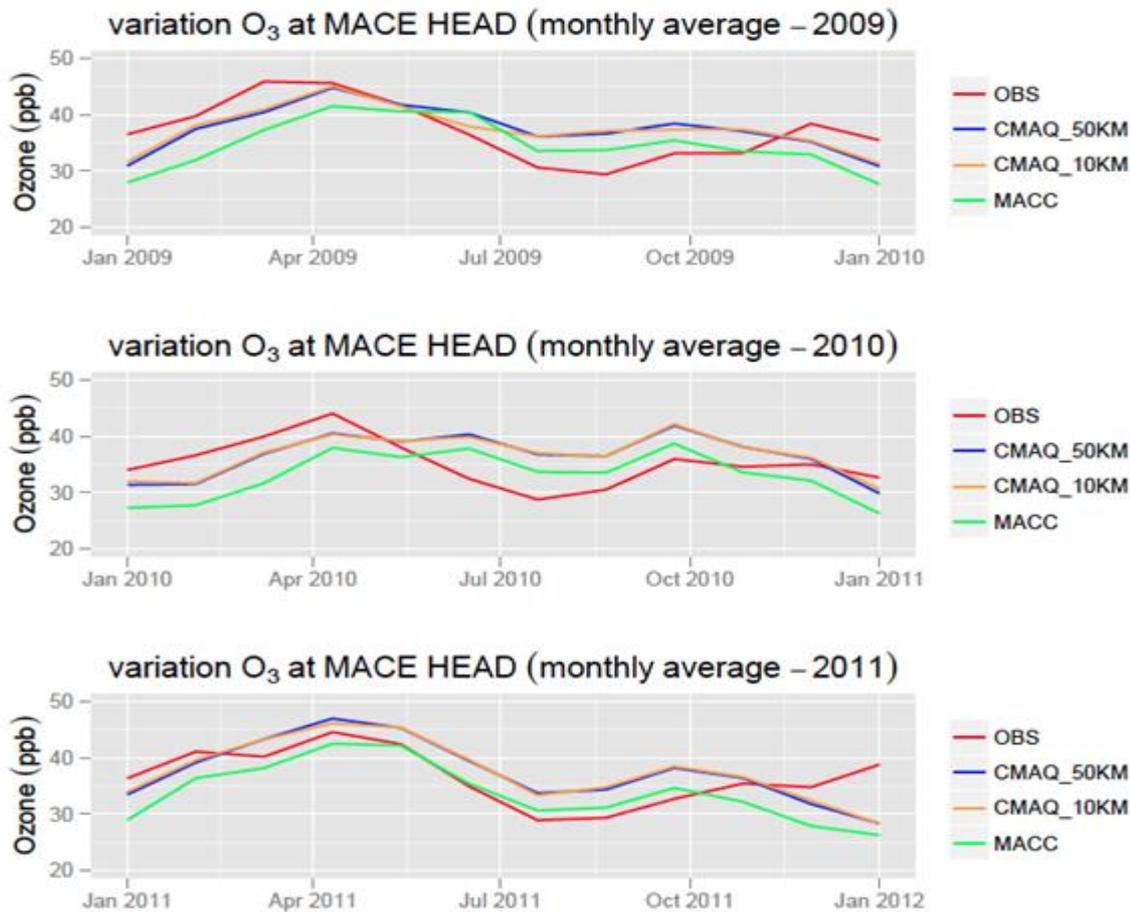
Overall the results show that any variations in boundary conditions will directly affect the ozone concentrations in CMAQ simulations and follows the variations generated in MACC reanalysis. This is clearly evidenced in CMAQ ozone concentration with underestimation during winter months and over-estimation during summer months, similar to variation of ozone concentration in the MACC reanalysis. Also, the MACC results appear to have a difference in the seasonal cycle compared with observations for Mace Head. Cross-over months occur in May (2009, 2010, 2011), November (2009, 2010) and October (2011) for ozone concentrations.

3.2 Analysis of 2019-2021 Simulations

CMAQ-UK simulations for 2019-2021 were performed assuming the same meteorology and boundary conditions based on HadGEM2-ES climatology for 2020, as described in Section 2.2. Emissions specific to each of the three different years were used in the simulations, although the differences in emissions were small.

3.2.1 CMAQ-UK Results for 2019-2021

The predicted concentrations of NO_2, O_3, PM_{10} and $PM_{2.5}$ in 2019-2021 show similar spatial distributions across the three future years and a reduction in concentration compared to 2010 (see Figure 6). Across the UK mainland, the model forecasts reductions of up to 40% for NO_2, $PM_{2.5}$ and PM_{10}. The concentrations of O_3 are also reduced by up to 30% outside the city areas while the opposite change over the cities is insignificant (mostly less than 5%).

The decrease in NO_2 concentrations in 2020 is driven by the reduction in NO_x emissions of ~44%. The reduction in O_3 concentrations as NO_x and VOC emissions decrease (VOCs by 7%), suggests that by 2020 ozone may be NO_x sensitive. In urban areas the O_3 levels are stable or may increase by up to ~5%. The influence of rising temperature in 2020 which would promote the production of O_3 is negligible. The analysis of boundary conditions (BCs) in section 3.2.3 shows that the reduction of O_3 in 2020 may partly be attributed to a reduction of O_3 BCs of ~10-20 % in the boundary layer and $\geq$30 % in the upper layers.

Figure 6: Percentage differences of NO_2, O_3, $PM_{2.5}$ and PM_{10} concentrations between the 2009-2010 and 2019-2021 periods; 100*(2010 period – 2020 period)/ 2010 period
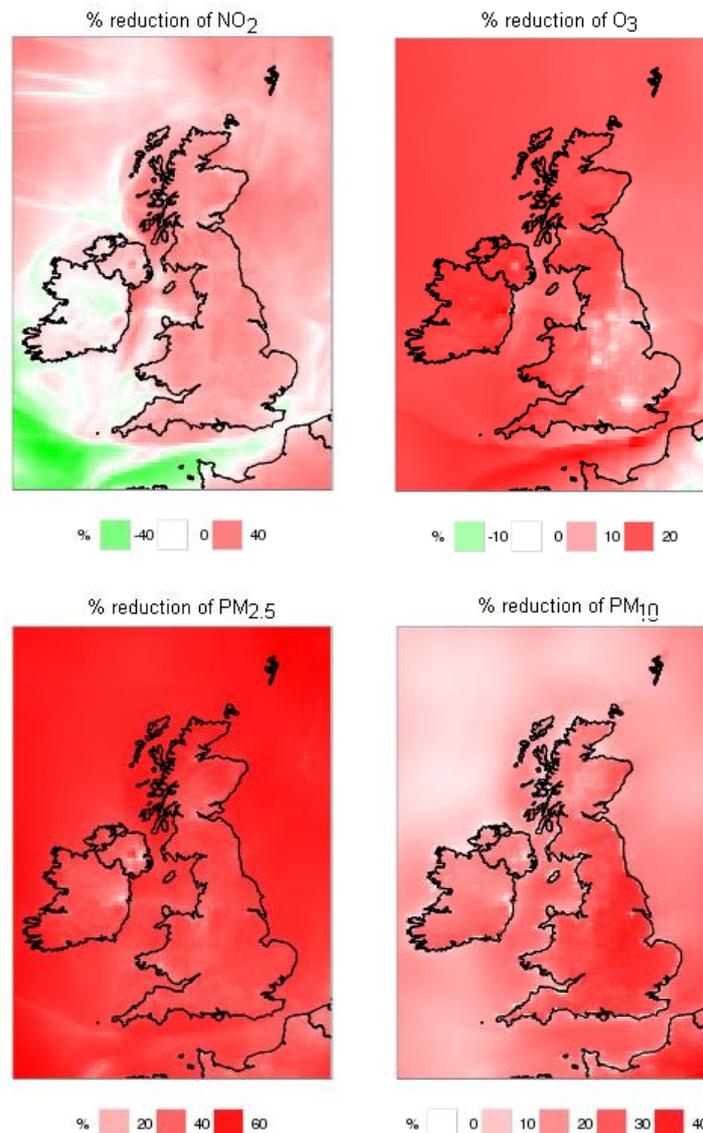
For PM_{10} and $PM_{2.5}$, the concentrations are driven by the reduction of their precursor emissions (i.e., 44% NO_x, 40% SO_2, 7% VOCs and 3% primary PM_{10} and $PM_{2.5}$), an increase of rainfall rate of ~50% over England (see Section 2.2) and/or a decrease of boundary conditions of the key components such as the reduction of SO_4^{2-} by ~30-50% (see section 3.2.3).

The analysis of fractions of $PM_{2.5}$ components (Figure 7) shows that the 45% reduction of $PM_{2.5}$ is attributed to the reductions of 30% in SO_4^{2-}, 20% NO_3^- and 18% NH_4^+, 17% $PM_{2.5}$

others, 6% POA and 4% EC and <1% anthropogenic Secondary Organic Aerosol (SOAA) and biogenic Secondary Organic Aerosol (SOAB) components.

Figure 7: Annual mean of PM$_{2.5}$ and its components for the period of 2009-2011 and 2019-2021

so4=sulphate, nh4=ammonium, no3=nitrate, NaCl=sodium chloride, ec=elemental carbon, poa=primary organic aerosol, soaa=secondary anthropogenic aerosol, soab=secondary biogenic aerosol, others=unclassified components
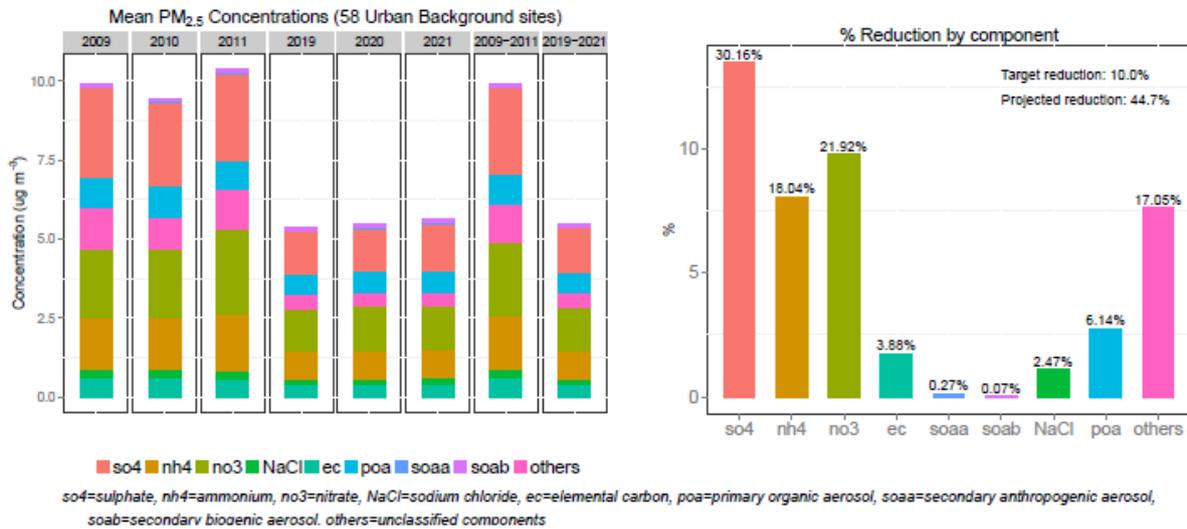
The small reduction of SOAB in 2020 suggests that the influence of rising temperature on PM$_{10}$ and PM$_{2.5}$ is insignificant. The influence of rainfall is determined through the analysis of wet deposition of PM$_{10}$ components including SO$_4^{2-}$, NO$_3^-$ and NH$_4^+$. Figure 8 shows the 3 year annual mean of wet deposition of PM$_{10}$ SO$_4^{2-}$, PM$_{10}$ NO$_3^-$ and PM$_{10}$ NH$_4^+$ for the 2010 and 2020 3-year periods, including the percentage reduction of these components in the 2020 period relative to the 2010 period. The Figure suggests ~50% reduction of wet deposition of these components over the UK mainland. This suggests a complex picture with an increase in wet deposition of between 50% and 100% of these components over the North Sea where the predicted increase in precipitation rate is as high as 200%. This suggests that the influence of precipitation on aerosol concentrations is significant for that region and may, along with other changes in meteorology predicted for 2019-21, require further investigation.

Figure 8: 3 year annual mean and percentage differences between 2010 period and 2020 periods of wet deposition of PM$_{10}$ sulphate, PM$_{10}$ nitrate and PM$_{10}$ ammonium
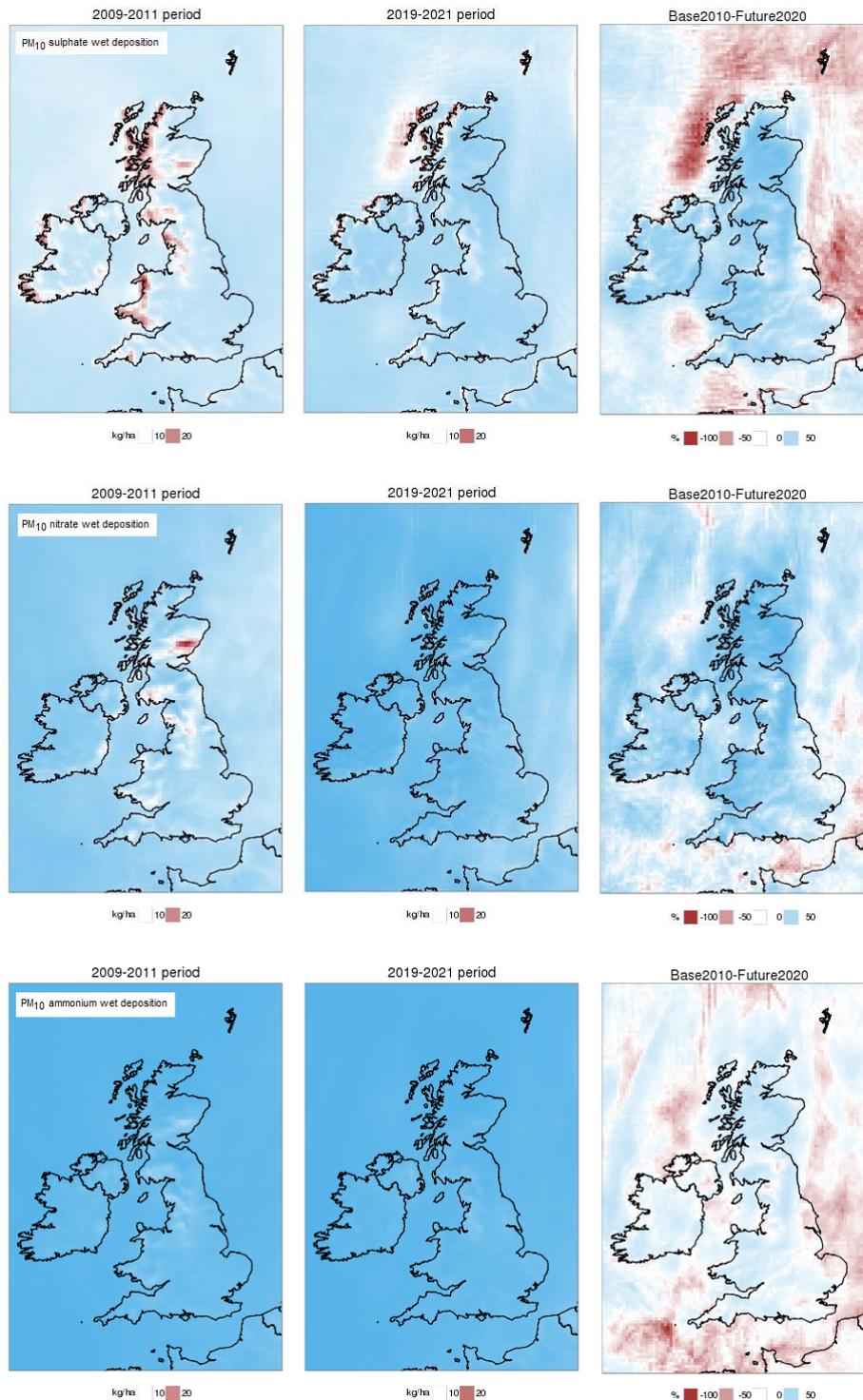
3.2.2 2020 Meteorology

The spatial distributions of 3 year annual mean values of ambient temperature and rainfall rate, including the absolute differences in temperature and percentage differences in precipitation between 2010 and 2020 are shown in Figure 9.

The temperature in 2020 is predicted to be warmer, especially in the central and southern part of the UK where the temperature is predicted to increase by ~1^0 C. In some southern parts of the model domain, the temperature rises by up to 2^0 C in 2020. In 2020, the precipitation over most part of the UK increases by ~ 50% except over Scotland where the opposite trend is predicted. The precipitation significantly increases by up to ~200% over the North Sea.

Figure 9: Three year annual mean of temperature and precipitation and the percentage differences between 2010 and 2020 periods
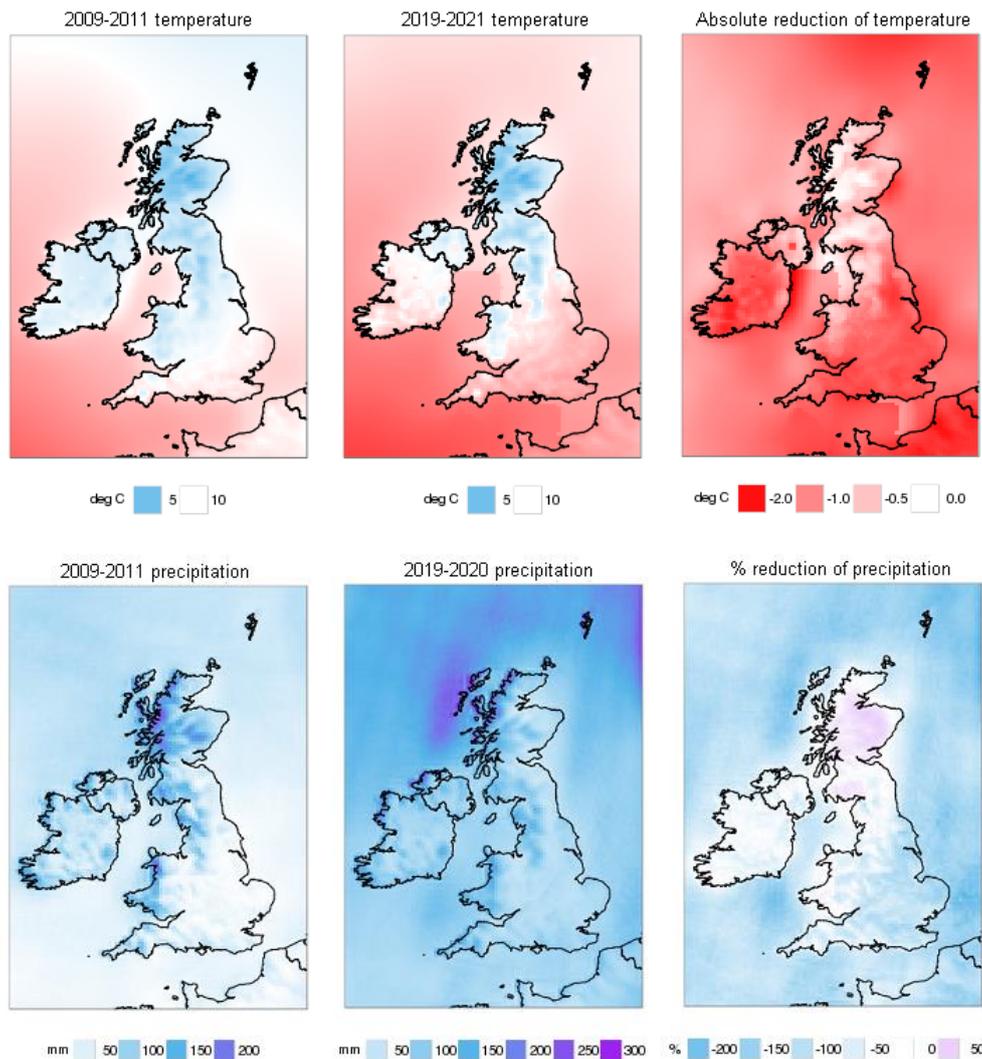
3.2.3 Boundary Conditions

Figure 10 shows the vertical distribution of absolute and percentage difference of ozone (O_3) and NO_x concentrations used at four boundaries (South, East, North and West) of the model domain between the future (ensemble average of 2019, 2020 and 2021) and present (ensemble average of 2009, 2010 and 2011) year simulations. Figure 10 shows a reduction (5 -10 ppb) in the ozone concentration at all four of the boundaries, but at a very small area of the south boundary of the domain there is an increase (0 – 5 ppb) at the ground surface in the future years.

Figure 10: Vertical distribution of absolute and percentage difference of Ozone and NOx concentrations used at four boundaries (South, East, North and West) of the model domain between the future (ensemble average of 2019, 2020, and 2021) and present (ensemble average of 2009, 2010, and 2011) year simulations
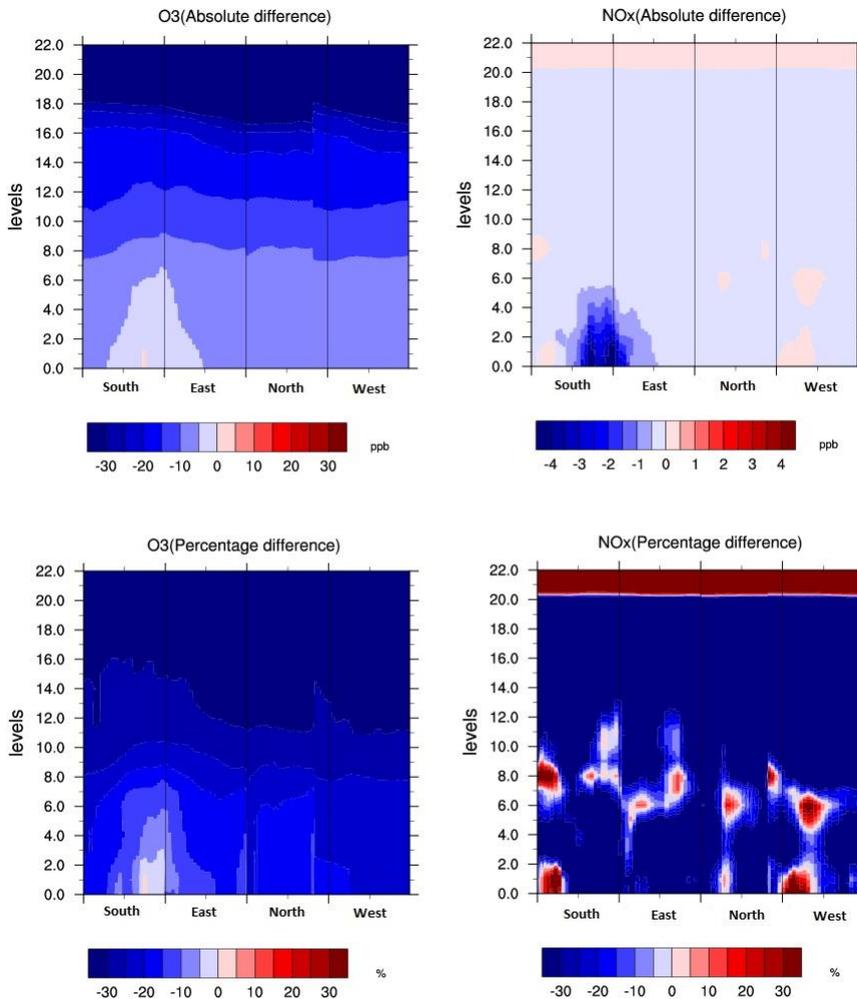
The percentage change of annual average ozone concentrations shows a decrease of 10-20% in the first 8 layers and the reduction of ozone concentration is more than 30% in the upper layers. At the ground level, NOx concentrations reduced in the range of 0-1 ppb at three of the boundaries but the highest reduction is at south boundary at more than 4 ppb. Also, at the west boundary there is an increase of NOx concentration in the range of 0-1 ppb (20-30%). The percentage difference for NOx concentration shows patches of increase (0-30%) of NOx concentrations in between the levels 5 and 8 for all boundaries for the future years.

Figure 11: Vertical distribution of absolute and percentage difference of sulphate and elemental carbon concentrations used at four boundaries (South, East, North and West) of the model domain between the future (ensemble average of 2019, 2020, and 2021) and present (ensemble average of 2009, 2010, and 2011) year simulations.
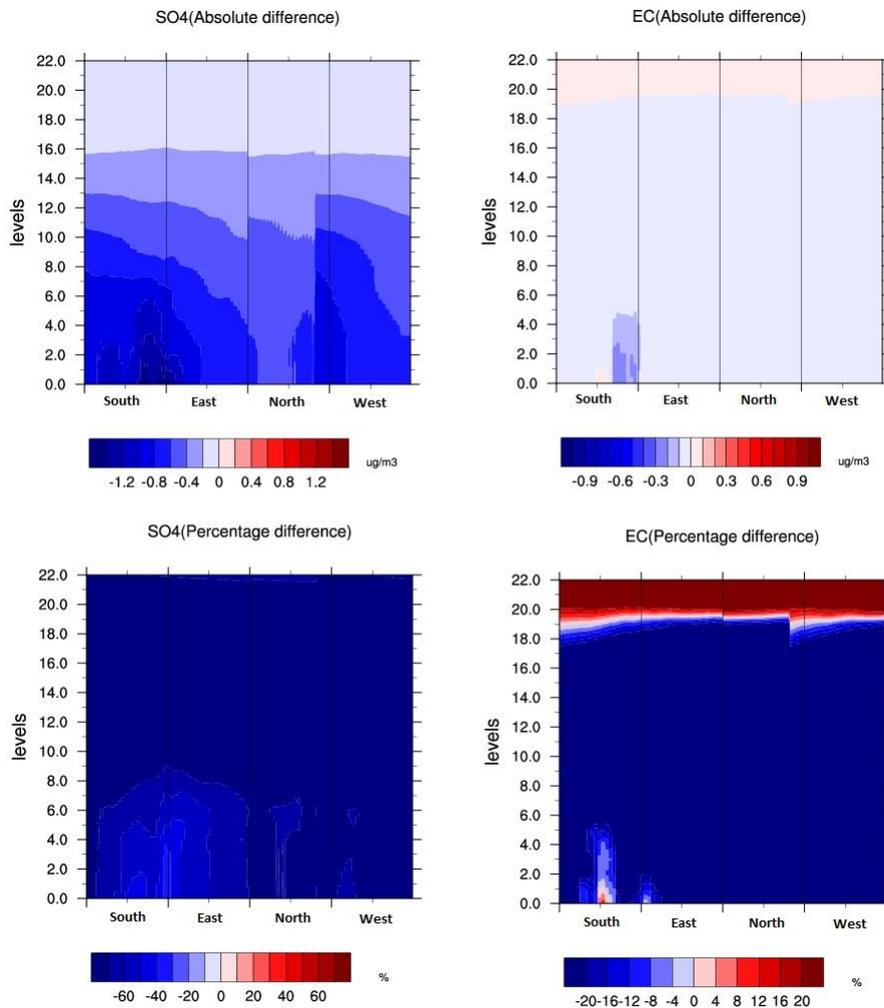
Figure 11 shows the vertical distribution of absolute and percentage difference of sulphate (SO_4^{2-}) and elemental carbon (EC) concentrations used at four boundaries (South, East, North and West) of the model domain between the future (ensemble average of 2019, 2020 and 2021) and present (ensemble average of 2009, 2010 and 2011) year simulations. Figure 11 shows a reduction (0.2 – 1.2 µg m^{-3}) in the concentration of sulphate at the ground level of four model boundaries. At ground level, the highest reduction in the absolute concentration of sulphate is at south and east boundaries amounting to a 30-50% reduction. The vertical distribution of absolute difference of EC concentration shows a reduction of 20% concentration of elemental carbon in the future years at the ground surface with the exception at south boundary where there is a patch of increase of elemental carbon concentration.

Overall the results show a reduction in the concentration of ozone (10-20 %) and sulphate (30-50 %) at all model boundaries for the future years. NO$_x$ concentration reduced more than 30% at the ground levels for the future years with exception of increase of NO$_x$ concentrations (20-30 %) at south and west boundaries. The vertical distribution of

percentage difference of elemental carbon showed a reduction (more than 20%) of concentration at the ground level in the future years.

3.2.4 Emissions for 2020

In line with other Defra projects the 2019-2021 scaling factors used to prepare future emissions for European countries other than the UK are based on the TSAP_Mar13_CLE ("Current Legislation") scenario available through the GAINS Europe modelling system[28]. Table 5 summarises the scaling factors used to convert the 2009 TNO-MACC emission maps to the 2020 emissions for a selection of countries in Europe. These are based on the ratios of national emission totals between these years.

Table 5 Country scaling factors used to scale 2009 European maps to 2020

	CO	NH3	NOx	PM10	PM2.5	SO2	VOC
Austria	0.76	1.07	0.53	0.90	0.81	0.82	0.83
Belgium	1.43	0.91	0.70	0.87	0.77	0.77	0.84
Denmark	0.44	0.82	0.61	0.70	0.60	0.68	0.66
Finland	0.71	0.94	0.64	0.82	0.77	0.76	0.72
France	0.75	0.99	0.55	0.89	0.78	0.47	0.74
Germany	0.80	0.99	0.60	0.85	0.77	0.74	0.83
Greece	0.80	0.89	0.65	0.69	0.70	0.35	0.65
Ireland	1.12	0.97	0.82	1.00	0.95	0.75	0.90
Italy	0.67	1.01	0.60	0.82	0.77	0.65	0.69
Netherlands	0.76	0.87	0.62	0.89	0.78	0.89	0.85
Norway	1.12	1.04	0.83	0.95	0.92	0.98	0.65
Portugal	0.69	1.02	0.62	0.87	0.83	0.78	0.81
Spain	0.92	1.05	0.61	0.96	0.92	0.44	0.81
Sweden	0.69	0.92	0.56	0.93	0.87	0.87	0.80
Switzerland	0.63	1.00	0.63	0.84	0.74	0.76	0.81

For the UK, the scaling factors are taken from the NAEI's UK sector-specific projections. Figure 12 shows the mapped difference in emissions between the first Monday in July of 2010 and 2020 (using the first Monday of the month eliminates variation due to day of the week). The maps on the left show differences in emissions in absolute terms the maps on the right in fractional terms. For all species except ammonia there is a decrease in emissions. The absolute reduction in emissions is centred on the urban areas for NO, VOC and CO, but in fractional terms are similar across the UK. PM and SO_2 emissions show larger differences over the areas of sea and clear demarcation of the emission control areas. The scaling factors applied for Ireland and France in the UK grid are based on the GAINS factors. These are at a country level and hence show no spatial variation.

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[28] Amann M, Bertok I, Borken-Kleefeld J, et al. (2013) Policy scenarios for the revision of the thematic strategy on air pollution. TSAP Report #10. International Institute for Applied Systems Analysis, Laxenburg, Ausrtia.
www.iiasa.ac.at/web/home/research/researchPrograms/MitigationofAirPollutionandGreenhousegases/TSAP-review.en.html report #10

Figure 12: Maps of the difference in UK emissions for first Monday in July of 2010 and 2020 as a) moles s-1 except for PM nitrate (PM_NO3) which are g s-1 (left panel), and b) as factors (right panel)

PM_NO3 1st Mon in July — max 0.01 ; min -1.22

PM_NO3 1st Mon in July fractional

NH3 1st Mon in July — max 0.44 ; min -0.34

NH3 1st Mon in July fractional

CO 1st Mon in July — max 2.64 ; min -13.83

CO 1st Mon in July fractional
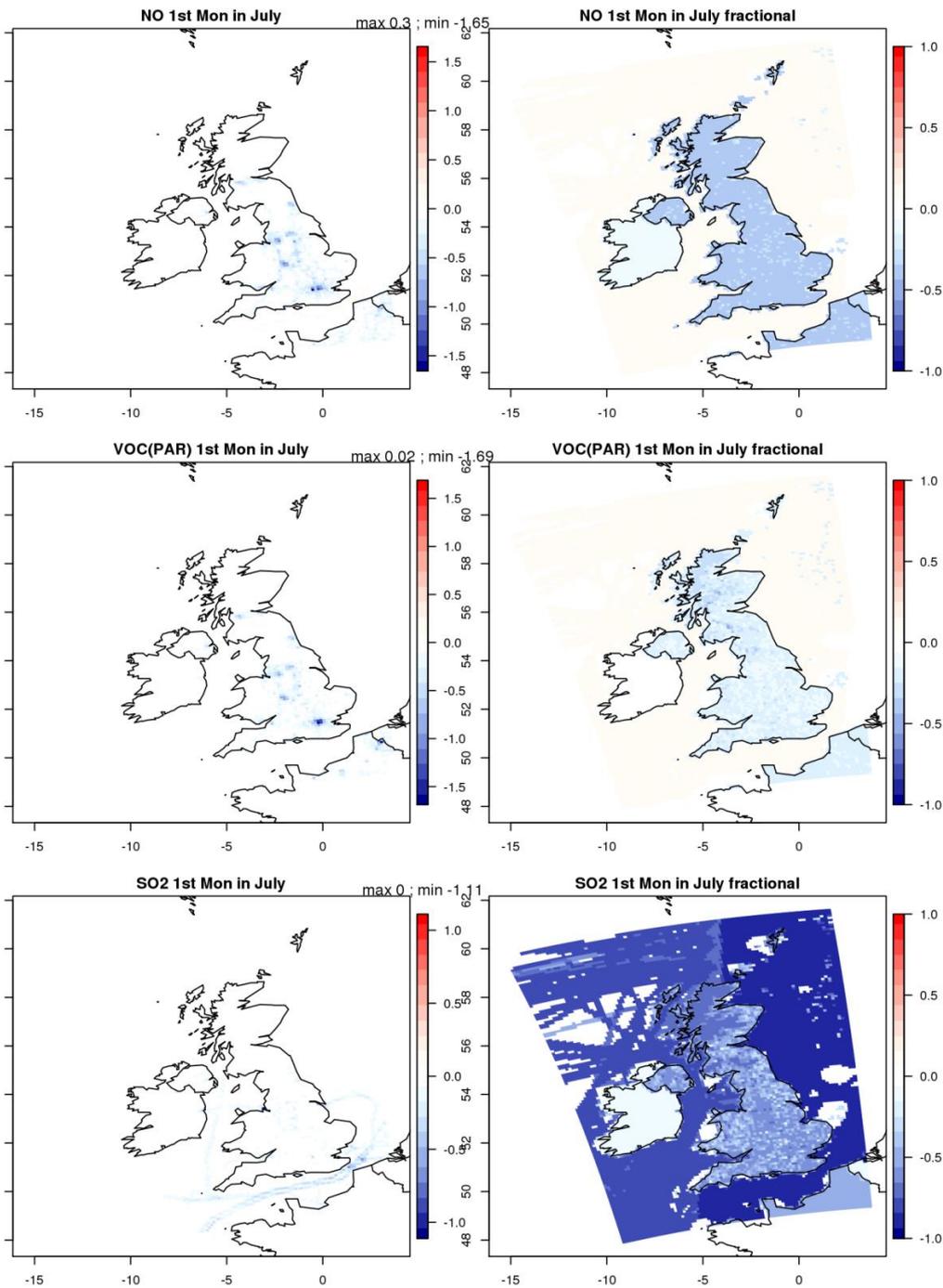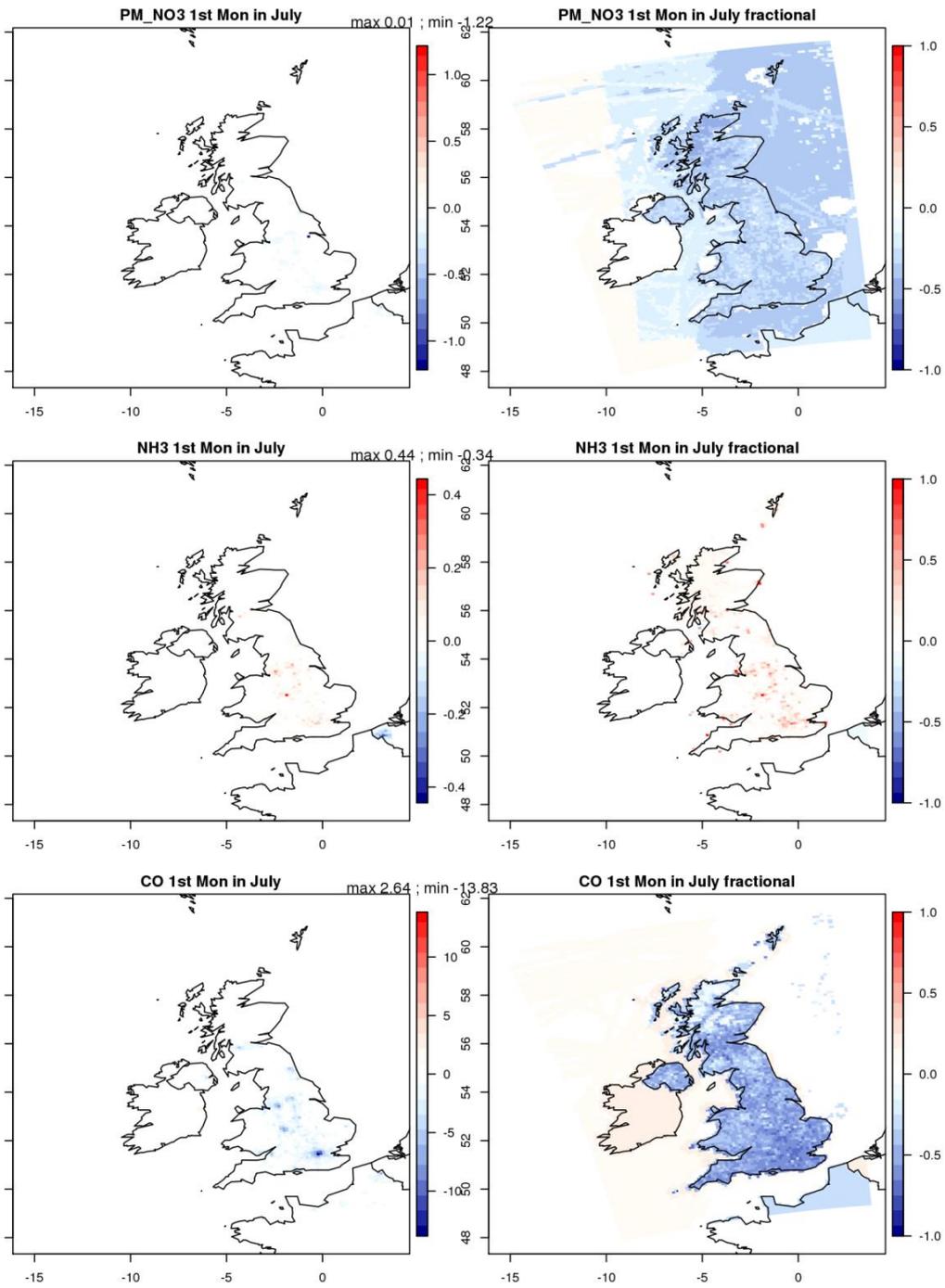
3.3 Potential Improvements for SOA Chemistry

Further investigation has been made in Phase 2 to understand the under prediction of SOA formation in CMAQ. This is a highly uncertain component of PM and formation is not well represented in the chemical reaction schemes within CMAQ. However, simplified parameterisations can be made to correct for these under predictions derived from the Master Chemical Mechanism implemented in the Photochemical Trajectory Model (PTM).

This section summarises the analysis undertaken, but the main findings are that CMAQ v4.7.1 and v5.0.1 model predictions for daily mean total organic carbon PM mass concentrations for Harwell for 2009 grossly under predicted the available observations. If the SOA formation from toluene and terpene photo-oxidation had been scaled as described in our earlier work, then total SOA would have been many times higher and total organic carbon PM mass would have been overestimated. The CMAQ model results thus, with and without scaling, bracket the available observations for total organic carbon PM.

3.3.1 Background

Carbon-containing compounds make up an important component of ambient particulate matter (PM). Policy actions to control and reduce PM levels need to take into account the presence of this carbonaceous component and its responses to emission reductions. There are a number of different carbon-containing PM components including elemental or black carbon, carbonates and organic PM. Organic PM is the main concern of in this phase of the project.

Organic PM may be directly emitted into the atmosphere in the form of organic compounds absorbed into or onto the surface of solid particles or dissolved in droplets or other liquid particles. This material is known as primary organic PM. Some organic PM is formed in the atmosphere by chemical reactions in which volatile organic compounds are oxidised to form low volatility species which are subsequently absorbed onto pre-existing particles. This material is known as secondary organic PM or secondary organic aerosol (SOA). The absorption process may be reversible so that the organic compounds can desorb from the particles and pass back into the gas phase. There they can be again absorbed by pre-existing particles. Alternatively they may be further oxidised in the gas phase to carbon dioxide and water or to species with even lower volatility which may then be taken up again by pre-existing particles. The low volatility gas phase organic compounds thus act as SOA precursors. SOA precursors may be directly emitted into the atmosphere or they may be present there by desorption from primary organic PM or secondary organic PM.

The relationship between primary and secondary organic PM is therefore highly dynamic and remarkably complex. Many aspects of this dynamic relationship between VOCs or primary organic PM emissions and SOA precursors, and between SOA precursors and SOA are still poorly understood. Progress is currently hampered by a lack of adequate observations of SOA and its precursors and by a lack of adequate emissions data for primary organic PM and SOA precursors.

3.3.2 Previous SOA Studies using CMAQ

Previous work on this contract under Phase 1 and 2 has concentrated on CMAQ modelling for the year 2006 using CMAQ v4.7.1 and the chemical mechanism CB05cl_ae5_aq. Initial results showed that the CMAQ predictions for primary organic carbon for Harwell for 2006 were reasonably within the bounds of expectations, based on the Defra NPL organic carbon observations, albeit for 2009. However, it was considered likely that the CMAQ predictions for secondary organic aerosol grossly underestimated the real world situation. On this basis, the CMAQ implementation underestimated organic PM by factors of 2.5 – 4.0.

In order to make progress despite the dearth of PM observations, a detailed set of comparisons was made between the CMAQ v4.7.1 predictions for Harwell and the results from the PTM model. These comparisons led to the identification of scaling factors which

would be required to scale the production of anthropogenic and biogenic SOA to reduce the magnitude of the gross under prediction found with CMAQ v4.7.1. It was found necessary to scale SOA formation from toluene photo-oxidation by a factor of 25 and that from terpene photo-oxidation by a factor of 20 to reproduce the PTM results for SOA with CMAQ-UK. When these scaling factors were applied, the CMAQ-UK SOA results lined up well with those from the PTM for Harwell in 2006.

3.3.3 Recent Progress with the CMAQ-UK Modelling for 2009

As part of the demonstration phase for the national CMAQ-UK modelling, an evaluation has been performed on the organic PM predictions for 2009 from the CMAQ v4.7.1 and v5.0.1 implementations. This evaluation has focussed on the CMAQ predictions for Harwell because of the availability of daily elemental carbon and organic carbon observations carried out as part of the Defra Particle Number and Composition Network.

The observed annual mean organic PM concentration for Harwell for 2009 was 2.13 µg m^{-3} compared with 0.82 and 1.01 µg m^{-3} predicted by CMAQ v4.7.1 and v5.0.1, respectively. This points to model underestimations of 62% and 53%. The corresponding observed annual mean elemental carbon PM concentration was 0.44 µg m^{-3} which compared well with 0.41 and 0.55 µg m^{-3} in CMAQ v4.7.1 and v5.0.1, respectively.

Figures 13 and 14 present scatter plots of the observed daily mean organic PM concentrations versus the CMAQ v4.7.1 and v5.0.1 model predictions, respectively. The correlation coefficients (R^2 = 0.19 and 0.14) indicated that there was no statistically significant correlation between the observations and the model predictions. Only 28% (CMAQ v4.7.1) and 38% (CMAQ v.5.0.1) of the model predicted daily mean values were within ± a factor of two of the observations. Altogether, CMAQ model performance against observations for Harwell in 2009 was less than satisfactory, irrespective of model version.

Figure 13: Scatter plot of CMAQ v4.7.1 model predicted organic PM concentrations against observations for Harwell for 2009
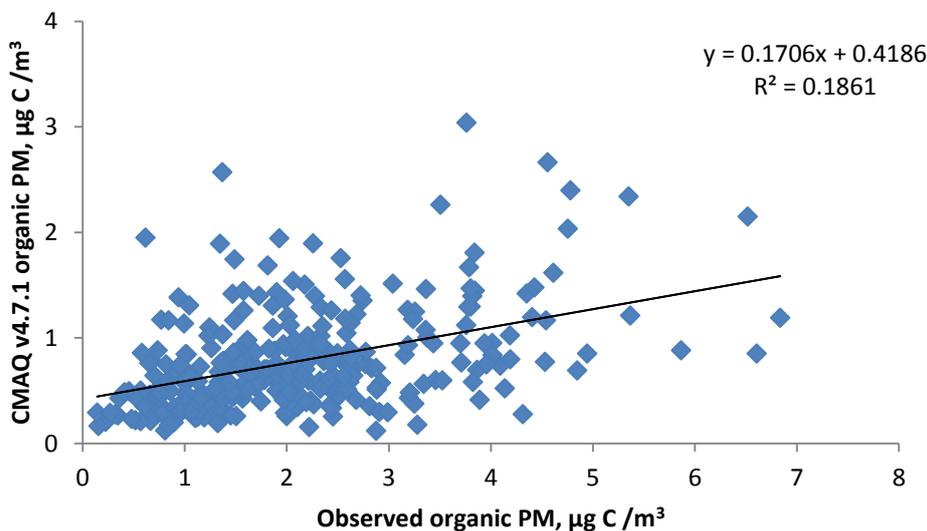
Figure 14: Scatter plot of CMAQ v5.0.1 model predicted organic PM concentrations against observations for Harwell for 2009.
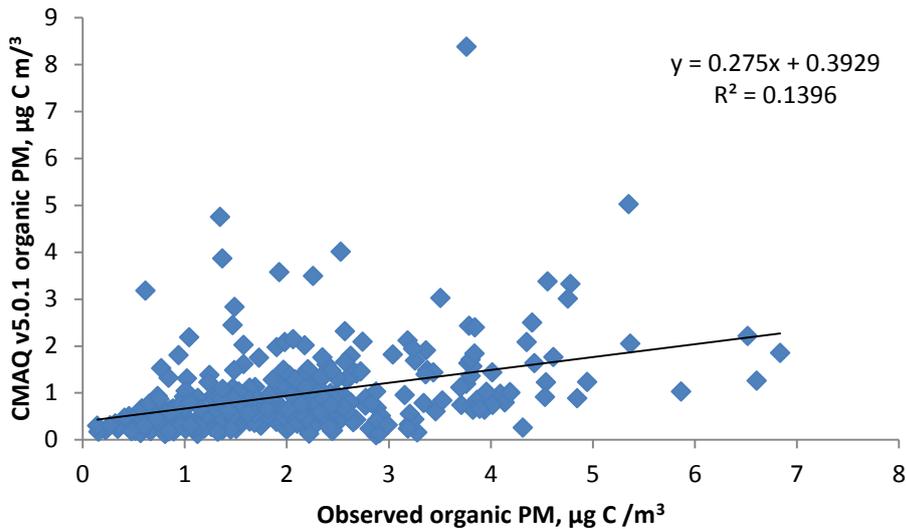
Table 6 presents a detailed comparison of the CMAQ model predictions of the components of the carbonaceous PM with the observations at Harwell for 2009. Primary organic carbon was found to be the largest predicted component of the CMAQ total organic carbon PM, with negligible contributions predicted for SOA. Consequently, the CMAQ model predictions grossly underestimated the observed total organic carbon PM mass concentration. If the SOA formation from toluene and terpene photo-oxidation had been scaled as described in our earlier work, then the results would have been as presented in the 'scaled CMAQ' column, see Table 6. In this column, total SOA was many times higher at about 2.55 µg C /m^3, increasing total organic carbon PM to 3.40 µg C /m^3, significantly higher than the observed value of 2.13 µg C /m^3. The CMAQ model results thus, with and without scaling, would bracket the available observations for total organic carbon PM.

Table 6 A detailed comparison of the observed and CMAQ model predicted components of the carbonaceous PM mass concentration in µg C /m^3 for Harwell in 2009.

Component in µg C /m^3	Observations	CMAQ v4.7.1	CMAQ v5.0.1	Scaled CMAQ
Primary organic carbon		0.667	0.856	0.856
Anthropogenic SOA		0.032	0.034	0.563
Biogenic SOA		0.118	0.109	1.986
Total SOA		0.150	0.153	2.549
Total organic carbon PM	2.130	0.817	1.009	3.405
Elemental carbon PM	0.437	0.405	0.547	0.547
Total carbonaceous PM	2.568	1.222	1.556	3.952

4 Demonstration of CMAQ-UK for Policy Evaluation

This section aims to show how CMAQ-UK can be used to provide the additional types of model outputs required to inform Defra's air quality policy and for compliance reporting to Air Quality Directive (AQD) requirements. The first section is a demonstration to show the use of the CMAQ-UK results described In Section 3 in assessing whether the UK will comply with the Air Quality Directive exposure reduction targets for PM$_{2.5}$. The following section compares concentration results from CMAQ-UK expressed in AQD metrics with results from other models used to provide these health- and ecosystem-based metrics for Defra, namely the Pollution Climate Mapping model (PCM) and Ozone Source Receptor Model (OSRM). The PCM outputs currently provide the supplementary data required for AQD compliance reporting. The final section examines the potential application of one of CMAQ's Decision Support Tools, namely the Decoupled Direct Method (DDM), as an efficient means of developing emission sensitivity coefficients that could be used to inform Defra's air quality policy analysis and development.

4.1 Achieving the 2020 Exposure Reduction Target for PM$_{2.5}$

The European Directive 2008/50/EC sets out exposure reduction targets relative to an Average Exposure Indicator (AEI) value in 2010, and these are summarised in Table 7. The AEI, expressed in μg/m^3, is based upon measurements in urban background locations in zones and agglomerations throughout the UK. It is assessed as a three-calendar year running annual mean concentration averaged over all sampling points. In addition there is an obligation to reach an AEI concentration of PM$_{2.5}$ of 20μgm^{-3} by 2025.

We have carried out an analysis of the capability of the UK to meet the national exposure reduction targets in Table 7 using the CMAQ-UK model output data from the demonstration runs performed in Phase 2 of this project. According to the Directive, the AEI for the reference year 2010 is the mean concentration of the years 2008, 2009 and 2010. As the CMAQ-UK model runs were for the years 2009, 2010 and 2011 we have instead extracted the concentration of PM$_{2.5}$ measured at the 41 urban background locations for these three years to perform this evaluation. Where data is not available for 2008, the Directive allows member states to use concentrations for these three years.

Table 7 shows the measured and modelled AEI results for 2009-2011 including a breakdown of the number of sites that fall within each initial concentration bin. Results for the individual sites are in Appendix F . In all cases, the modelled 3-year average PM$_{2.5}$ concentration for 2009-2011 is lower than the AEI derived from measurements. Figure 15 presents a scatterplot showing the correlation between the model and the measurements. The results are reasonably well correlated, but a linear fit to the data indicates that the model underestimates the measurements by a factor of ~1.31.

The AEI for the year 2020, which is used for the examination of whether the national exposure reduction target is met, should be the three-year running mean concentration averaged over all those sampling points for the years 2018, 2019 and 2020. As the CMAQ-UK project has considered the years 2019 to 2021 we have instead calculated the AEI for these three years. Table 7 also shows the modelled 3-year average PM$_{2.5}$ concentration for the years 2019 to 2021.

The AEI for 2009-2011 derived from measurements is 13.2µgm^{-3} which requires an exposure reduction target of 15% to be met. The equivalent AEI derived from the CMAQ model results is 9.9 µgm^{-3} for this time period, lower than the value based on measurements as noted above. The modelled AEI value for 2019-2021 is 5.4 µgm^{-3}, but if we assume that a correction factor of 1.35 can be applied to every year of model results to correct for the model underestimation bias then the AEI for 2019-2021 increases to 7.3 µgm^{-3}, providing a better comparison with the AEI for 2009-2011. This AEI value for 2019-2021 is 45% below the measured value for 2009-2011 indicating the exposure reduction target of 15% will be met.

Table 7 AEI National exposure reduction target for PM$_{2.5}$

Average Exposure Indicator		Measured	Modelled		
		2009-2011	2009-2011	2019-2021	2019-2021 Scaled (1.31)
		13.2 µgm^{-3}	9.9 µgm^{-3}	5.4 µgm^{-3}	7.3 µgm^{-3}
Exposure reduction target relative to the AEI in 2010 to be reached by 2020[29]		Number of sites with and average PM$_{2.5}$ concentrations falling in the exposure reduction target bands (41 urban background sites)			
Initial concentration µg/m3	Reduction target %				
< 8,5 = 8,5	0 %	1	8	41	35
> 8,5 — < 13	10 %	15	33	0	6
= 13 — < 18	15 %	25	0	0	0
= 18 — < 22	20 %	0	0	0	0
≥ 22	All appropriate measures to achieve 18 µg/m3	0	0	0	0

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[29] Where the AEI in the reference year is 8,5 µg/m^3 or less the exposure reduction target shall be zero. The reduction target shall be zero also in cases where the AEI reaches the level of 8,5 µg/m3 at any point of time during the period from 2010 to 2020 and is maintained at or below that level.

Figure 15: Average PM$_{2.5}$ concentration at urban background sites in 2009 to 2011. The AEI value is presented in red and the site specific 3-year average concentrations are blue.
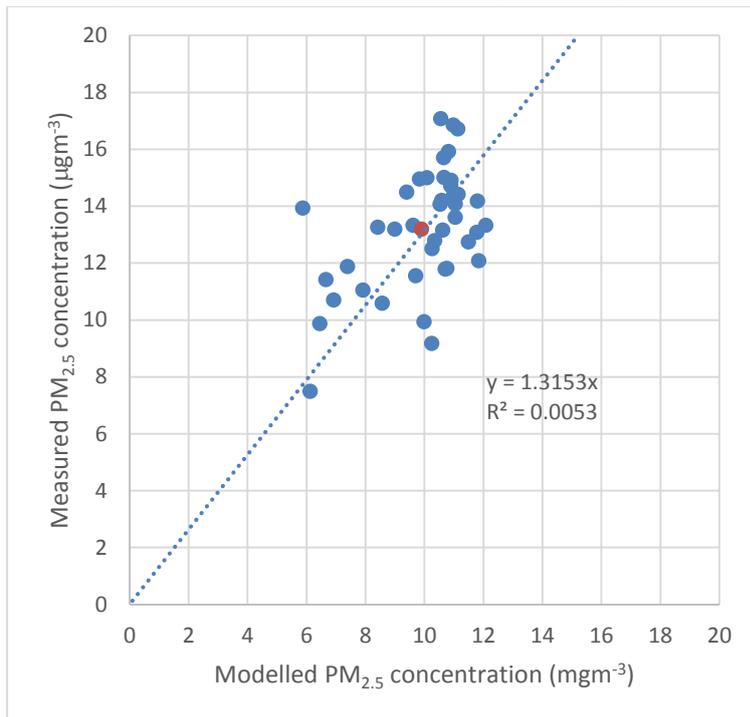
4.2 Comparison with Results from Other Models Used by Defra

This section aims to compare the results from CMAQ-UK with outputs from current models used by Defra and the Devolved Administrations to assess a range of pollutants and provide evidence for a number of policy drivers. Model results that were accessible for comparison are results from the Ozone Source Receptor Model (OSRM) and the Pollution Climate Mapping (PCM) model.

OSRM is a Lagrangian trajectory model developed to describe photochemical ozone production in the UK. It produces hourly concentrations of ozone at named receptors or on a 10 x 10 km UK grid. It is used by Defra to advise on the effects of planned or proposed policies affecting precursor emissions on ozone concentrations.

The PCM model is a collection of models designed to fulfil part of the UK's EU directive (2008/50/EC) requirements to report annually on the concentrations of certain pollutants in the atmosphere. There is one model for each modelled pollutant PM$_{10}$, PM$_{2.5}$, O$_3$, SO$_2$, NO$_X$, NO$_2$, benzene, benzo(a)pyrene (BaP), arsenic, nickel, lead and cadmium. The PCM provides outputs on a 1x1 km grid of background conditions, plus around 9000 representative roadside values. PCM is also used for scenario assessment and population exposure calculations to assist policy developments and provides model runs to support the writing of Time Extension Notification (TEN) applications for PM$_{10}$ and NO$_2$.

CMAQ-UK provides an output on a 10 x 10 km grid and is suitable for comparison with the OSRM model and the background PCM model, but not the roadside PCM model. In the following sections the output of the CMAQ-UK model runs for 2009, 2010 and 2011 are evaluated relative to relevant existing model results and measurements. The CMAQ-UK ozone outputs are compared to OSRM ozone outputs and PCM background outputs and the NO$_x$, NO$_2$, PM$_{10}$, PM$_{2.5}$ and SO$_2$ outputs are compared to the background PCM model outputs. In addition, secondary inorganic aerosol and sea salt components of PM are compared to background outputs from the PCM model. In the current configuration metals,

benzene and BaP concentrations are not output by the CMAQ-UK model, although modelling of these species by CMAQ would be possible.

To evaluate the performance of CMAQ-UK for compliance reporting and policy assessment purposes for each section the relevant limit values and target values stated in the EU air quality directive are summarised and, where appropriate, CMAQ-UK is evaluated with respect to the relevant compliance metrics. Spatial distributions of the pollutant concentrations or metrics are plotted and compared to the spatial distributions output by the alternative models. In addition concentrations at UK national monitoring and verification monitoring sites have been extracted and used to evaluate CMAQ-UK model concentrations. A similar comparison is presented between the alternative model and measurement values. Such evaluations have been presented previously in the annual reports on projects from the PCM and OSRM models. Supplementary maps are in Appendix G and Tables of comparative statistics are in Appendix H - Table 23 to Table 42.

4.2.1 Ozone

Two target values (TV) for ambient ozone concentrations are set out in the Air quality directive (AQD) and are specified for the protection of human health and vegetation. These are:

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

AOT40 is defined as the sum of the difference between hourly concentrations greater than 80 µgm^{-3} (equivalent to 40 ppb) over a given period using the one-hour values measured between 8.00 and 20.00 each day. The TV's apply from 01/01/2010

Two long term objectives (LTO) for ambient concentrations ae set out in the AQD, these are:

- a maximum daily 8-hour mean concentration of 120 µgm^{-3} within a calendar year
- AOT40 of 6000 µg m^{-3} h May to July

These LTOs have been specified for the protection of human health and the protection of vegetation, respectively. The date for compliance with the LTO's has not been defined.

Concentrations for these metrics have been calculated from the hourly CMAQ ozone concentrations and the results are plotted in Table 8 and Table 9. In addition, concentrations values for these metrics have also been calculated at specific AURN monitoring sites for comparison with these measurements. Table 23-Table 28 summarise comparison to measurements.

The spatial distribution of AOT40 for CMAQ-UK is similar to the PCM with both showing high levels of ozone in the south-east corner of England and lower ozone levels in northern England and much of Scotland. There are also high levels of AOT40 off the far north east of Scotland, around the Shetland Islands, a feature also shown by the OSRM. Compared to the spatial distribution of AOT40 from OSRM, CMAQ tends to show lower AOT40 in coastal areas, northern England and southern Scotland. Higher levels of AOT40 in and around London are observed by CMAQ-UK than in either of the other models, this is particularly apparent in the 2009 model results.

The model verification summaries in Appendix H Table 23-Table 25 indicate that CMAQ-UK tends to output high AOT40 levels at national and verification measurement site locations. This is consistent with the results in section 3.1 which show that CMAQ-UK tends to over predict 8 hour maximum ozone concentration between the May to July period for which the AOT metric is calculated for. This analysis also indicated that CMAQ-UK over predicts ozone concentrations more at urban sites than rural sites. At urban sites local emissions have a greater impact on ozone concentrations.

The days exceeding 120 µg m^{-3} (DGT120, see Appendix HTables 26-28) predicted by CMAQ is low, with almost all locations predicted to have less than 5 days exceeding this metric for all three years. The PCM model similarly predicts a small number of days exceeding this metric with areas in eastern, southern and south-western England predicted to have up to 10 days exceeding this metric for one or more year and other areas are predicted to have less than five days exceeding. OSRM tends to predict a higher number days exceeding this metric than the other models and the spatial distribution plots indicate that southern and coastal areas tend to show more days exceeding this metric.

Table 8: AOT 40 metric plots from CMAQ-UK, OSRM and PCM compared.
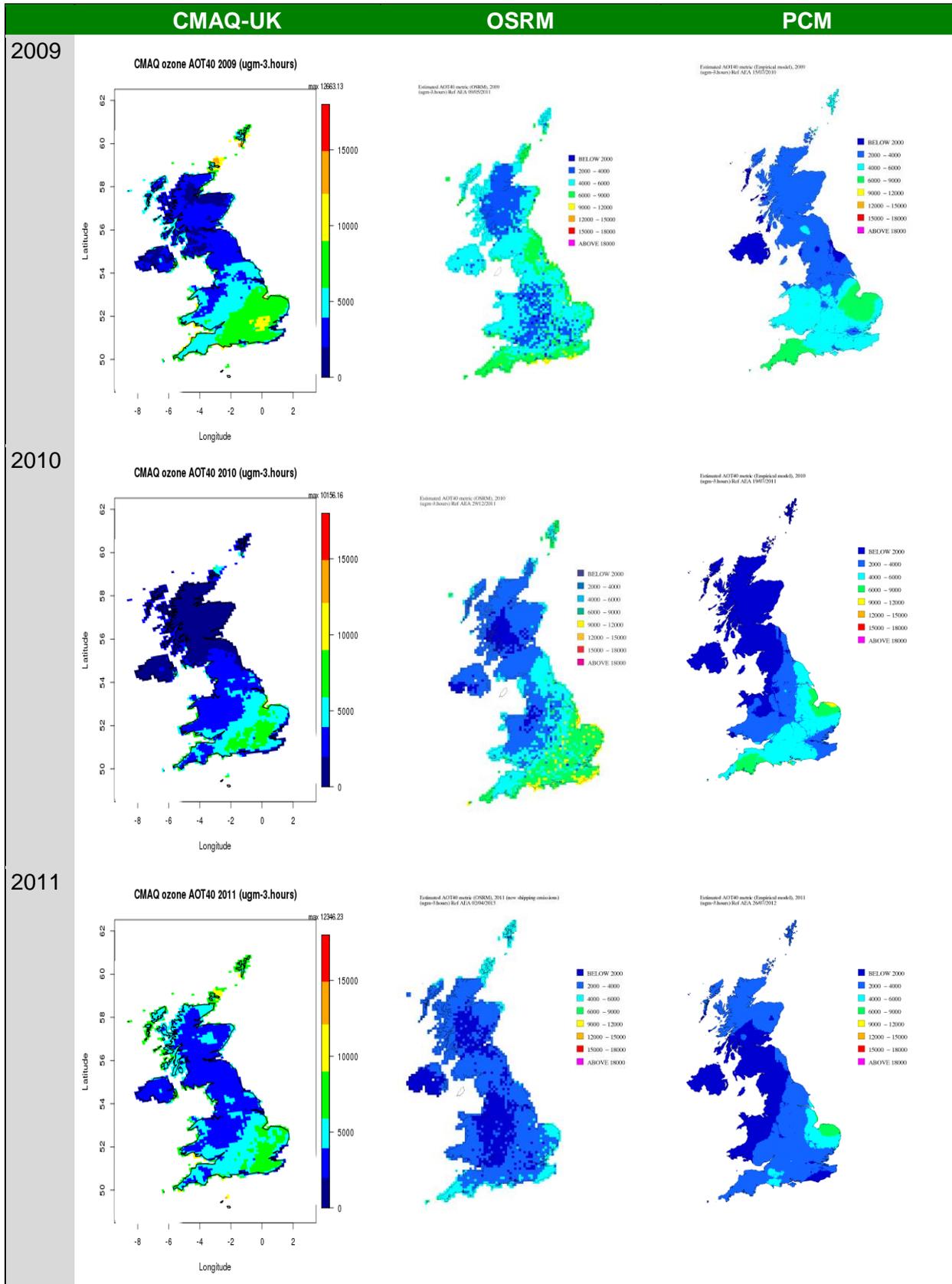
Table 9: Maps of DGT120 metric from CMAQ-UK, OSRM and PCM compared.

	CMAQ-UK	OSRM	PCM
2009			
2010			
2011			
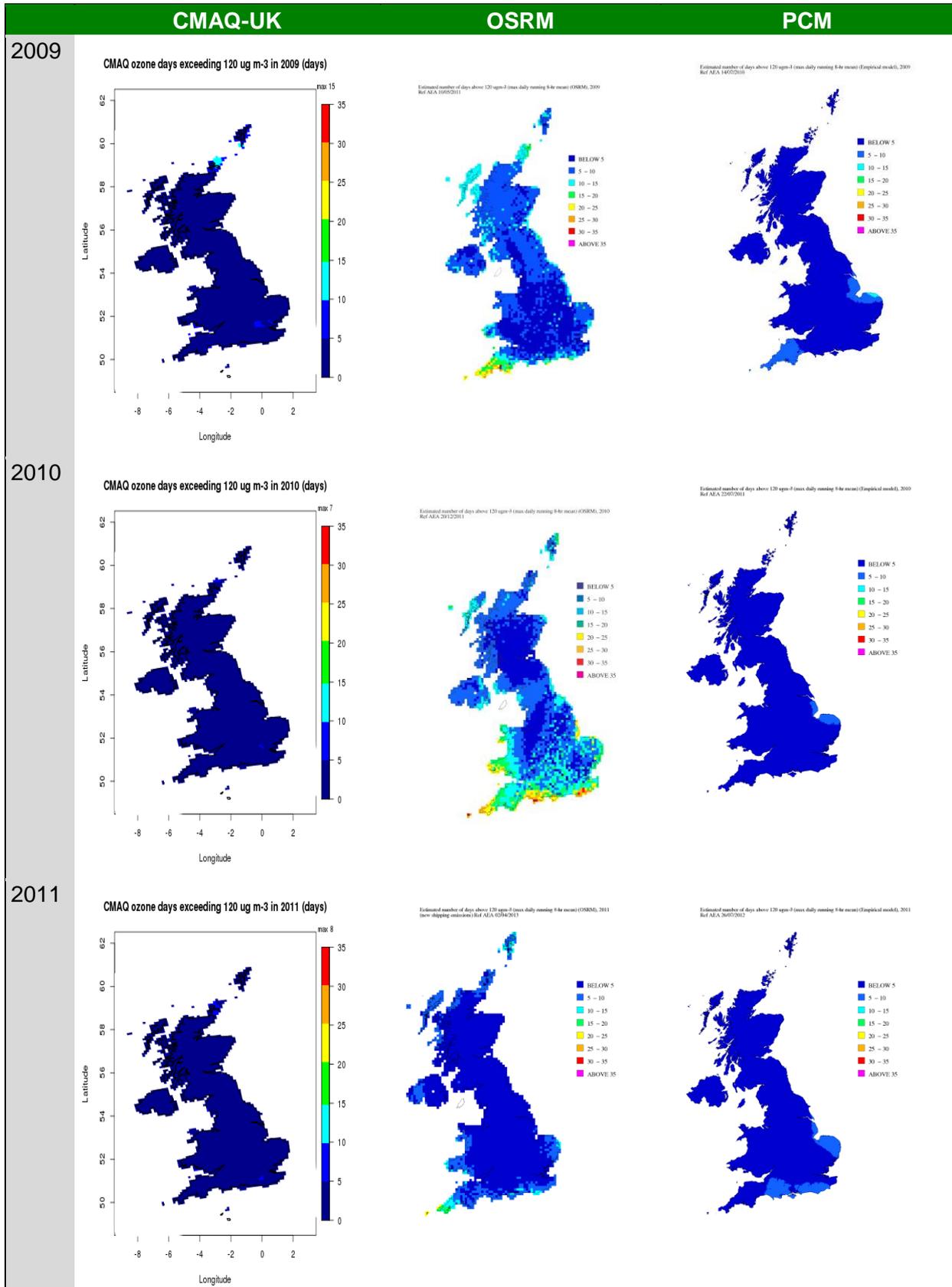
4.2.2 PM$_{10}$

Two limit values for ambient PM$_{10}$ concentrations are set out in the AQD. These have been specified for the protection of human health. These limit values are:

- An annual mean concentration of 40 µg m^{-3}.
- A 24-hour mean concentration of 50 µg m^{-3}, with 35 exceedances permitted each year (no comparison has been made here because the PCM has no mapped output).

In this report the focus is on comparison of the CMAQ-UK model results for annual mean PM$_{10}$ concentration with the annual mean concentration results from the PCM model which were reported for compliance assessments in 2009, 2010 and 2011. Annual mean concentrations of PM$_{10}$ from CMAQ-UK have been calculated from the hourly CMAQ results. The results are plotted in Table 10 where they are also compared to the PCM model results. Annual mean concentrations of PM$_{10}$ have been extracted at specific AURN and verification monitoring site locations for comparison with measurements. Summary statistics for the comparison between modelled and measured PM$_{10}$ for CMAQ-UK and PCM are presented in Appendix H Table 29 and

Table 30.

The verification summary tables indicate that CMAQ generally under predicts the annual mean concentration of PM$_{10}$ compared to the measurements by an average of ~4 to 6 µgm^{-3}. PCM also tends to slightly under predict annual mean PM concentrations but by a smaller amount. The spatial distribution of PM$_{10}$ from CMAQ-UK is similar to the spatial distribution from PCM with higher concentrations in south eastern England and lower concentrations in northern Scotland. CMAQ-UK tends to show a relatively lower concentration of PM$_{10}$ in south eastern England than the PCM model.

4.2.3 PM$_{2.5}$

The Air Quality Directive (AQD) includes a target value (TV) for annual mean PM$_{2.5}$ which came into force from 01/01/2010. This target value is:

- An annual mean concentration of 25 µg m^{-3}.

Two limit values have also been set for ambient PM$_{2.5}$ concentrations in the AQD. These limit values are:

- Stage 1 limit value – An annual mean concentration of 25 µg m^{-3}.
- Stage 2 indicative limit value – An annual mean concentration of 20 µg m^{-3}

The Stage 1 limit value is due to come into force on 01/01/2015, the Stage 2 limit value is due to come into force 01/01/2020.

Annual mean concentrations of PM$_{2.5}$ from CMAQ-UK have been calculated from the hourly CMAQ results. The results are plotted in Table 11 where they are also compared to the PCM model results. Annual mean concentrations of PM$_{2.5}$ have been extracted at specific AURN and verification monitoring site locations for comparison with measurements. Summary statistics for the comparison between modelled and measured PM$_{2.5}$ for CMAQ-UK and PCM are presented in Appendix HTable 31 and Table 32.

The verification summary tables indicate that CMAQ generally under predicts the annual mean concentration of PM$_{2.5}$ compared to the measurements by approximately 3 µgm^{-3}. PCM predicts annual mean PM$_{2.5}$ concentrations well. The spatial distribution of PM$_{2.5}$ from CMAQ-UK is similar to the spatial distribution from the PCM with higher concentrations in southern and eastern England and low concentrations in northern Scotland.

Table 10: Maps of annual mean PM₁₀ concentration from CMAQ-UK and PCM compared.
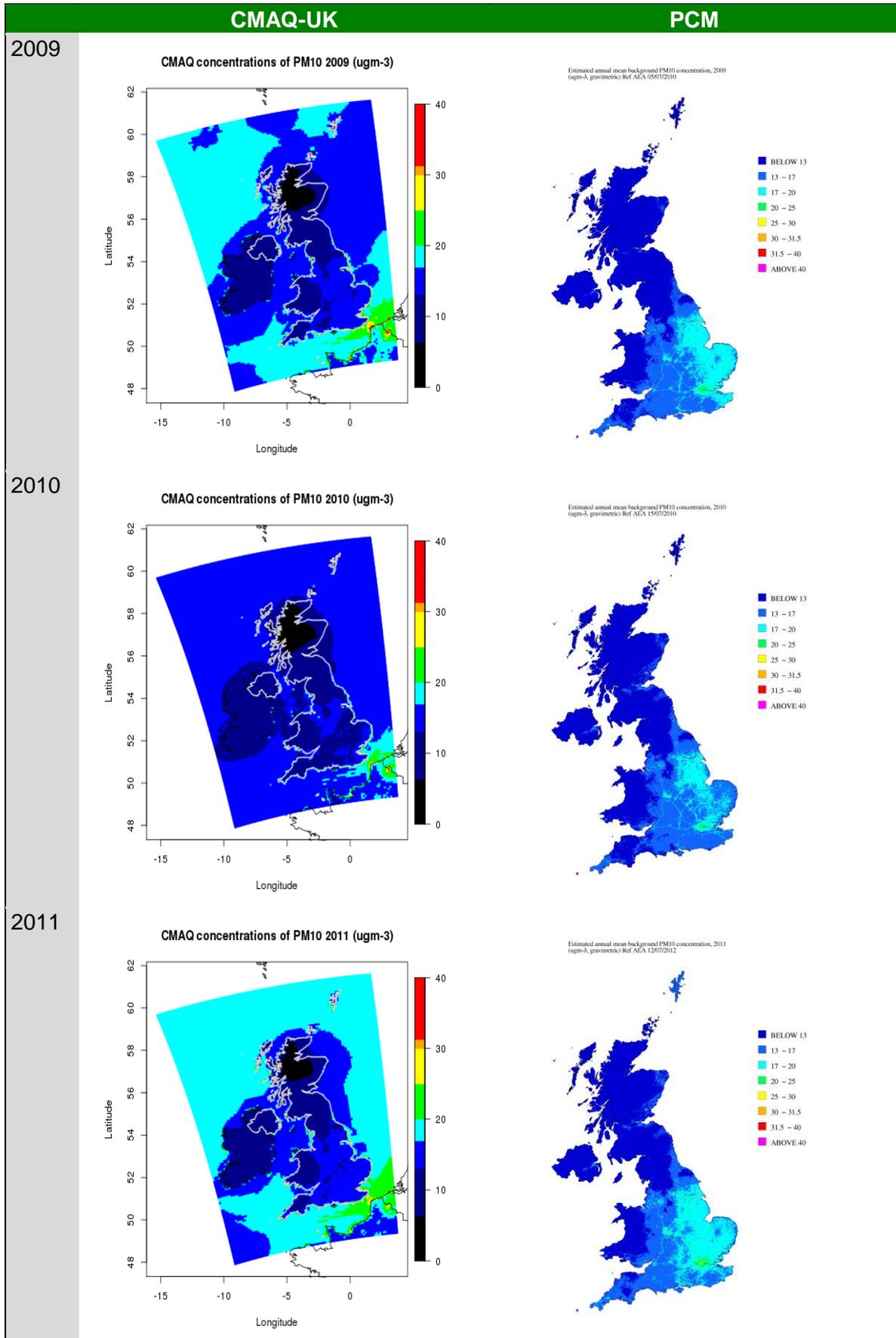
Table 11: Maps of annual mean PM2.5 concentration from CMAQ-UK and PCM compared.
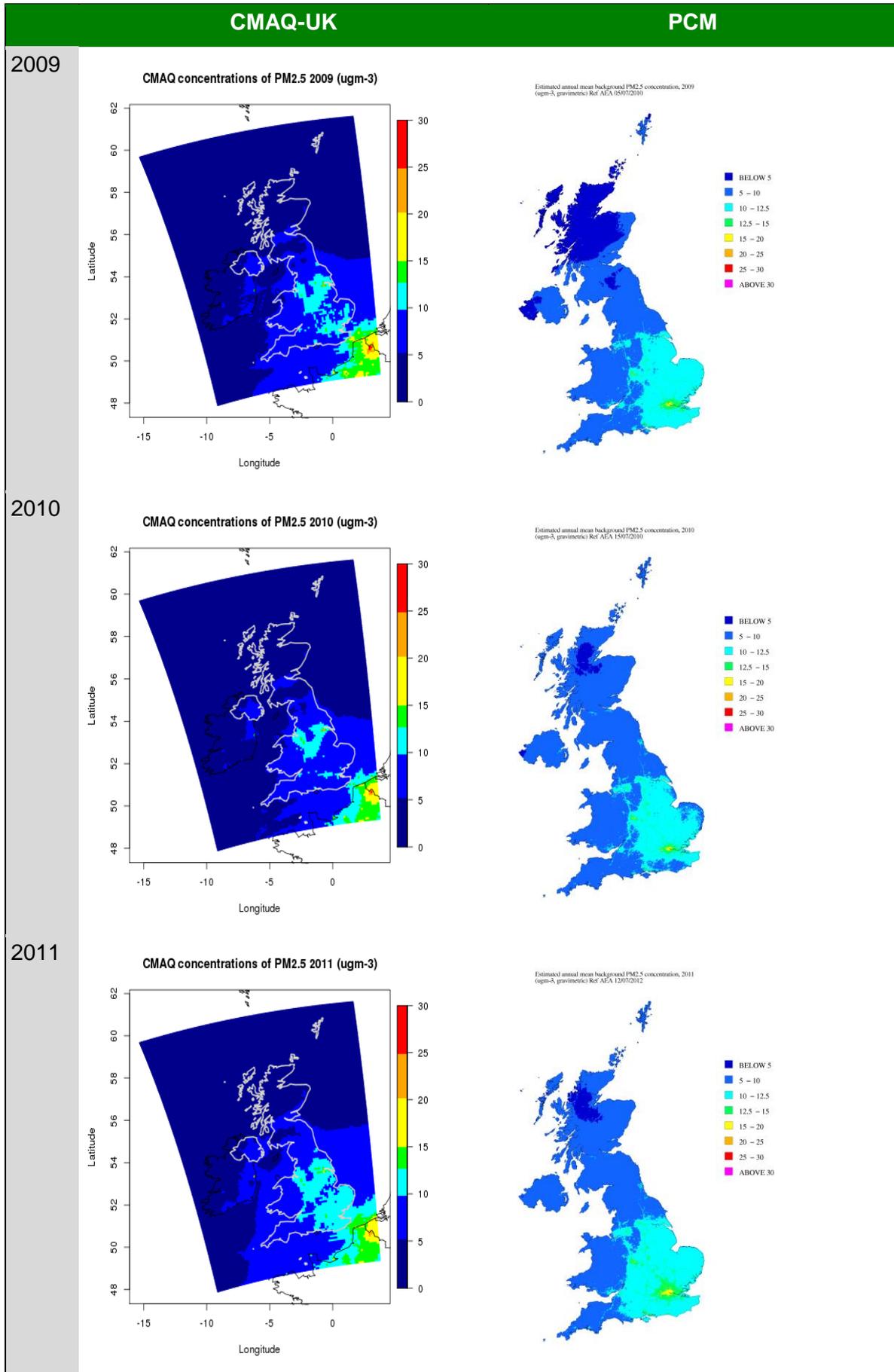
4.2.4 Secondary inorganic components of PM

In addition to the comparison between annual mean PM_{10} and $PM_{2.5}$ concentrations, an analysis of the secondary inorganic components of PM (nitrate, sulphate and ammonium) has been carried out. These grids are output from the CMAQ model. In the PCM model maps of secondary inorganic aerosol (SIA) concentrations across the UK are calculated from rural measurements of sulphate, nitrate and ammonium concentrations by interpolation. Monthly measurements are available for 28 rural monitoring sites within the UKEAP AGAnet[30]. Concentration surfaces on a 5 km x 5 km grid were calculated from the measurement data using Krigging techniques.

Comparison between the maps of sulphate concentration show that CMAQ-UK over predicts the sulphate SIA component by a factor of two or greater in most areas compared to the empirical PCM maps. The spatial distribution is broadly similar with higher sulphate concentrations in the south than the north, however CMAQ-UK shows elevated sulphate concentrations around Birmingham, Manchester and Sheffield which are not seen in the empirical PCM maps.

The maps of ammonium concentration show that CMAQ-UK tends to over predict ammonium concentrations compared to the empirical PCM maps. The spatial distribution predicted by CMAQ-UK is similar to the spatial distribution predicted for sulphate concentration. The spatial distribution is broadly similar to the empirical PCM maps which tend to show increasing ammonium concentrations from the north west to the south east. However, CMAQ-UK predicts high concentrations around Birmingham, Manchester and Sheffield which are not seen in the empirical PCM maps.

The maps of nitrate concentration show that CMAQ-UK tends to over predict nitrate concentrations by around 1 μgm^{-3} compared to the empirical PCM maps. The spatial distribution predicted by CMAQ-UK is similar to the empirical PCM maps with concentration increasing from the north west to the south east.

A Comparison of CMAQ-UK sulphate, ammonium and nitrate grids to the empirical PCM SIA grids are shown in Appendix G, Supplementary maps and tables of comparative statistics between CMAQ, PCM and OSRM Table 17, Table 18 and Table 19 respectively.

4.2.5 Sea salt

The AQD requires member states to discount exceedances of limit values due to natural sources when reporting the results of air quality assessments. The definition of natural sources in this directive includes sea spray. The PCM model currently derives the contribution to ambient PM from sea salt directly from measurements of particulate chloride[31]. Data from 28 rural sites were interpolated by Krigging methods onto a 5 km x 5 km grid. It is assumed that sodium chloride is the only marker for sea salt and a mass scaling factor of 1.648 was applied to convert elemental chloride mass to sodium chloride mass. Full details of the PCM sea salt subtraction method are reported in Brookes et al. 2012[32].

The CMAQ-UK outputs the sea salt component of PM directly. This grid includes calculated natural sources of sodium chloride and sulphate. Table 20 (Appendix G) presents the PCM empirical chloride grids which are directly proportional to the calculated sea salt grids. These are compared to the concentrations of sea salt from CMAQ-UK which are divided by a scaling factor of 1.648 to provide a direct comparison to the empirical PCM maps. The CMAQ-UK sea salt concentrations are of similar magnitude to PCM concentrations and the spatial distributions are broadly similar. However, the CMAQ-UK concentrations show a sharper increase towards the coast line, with a relatively constant concentration of sea salt

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[30] CEH Edinburgh, Centre for Ecology and Hydrology, Natural Environment Research Council. http://pollutantdeposition.defra.gov.uk/aganet
[31] CEH Edinburgh, Centre for Ecology and Hydrology, Natural Environment Research Council. http://pollutantdeposition.defra.gov.uk/aganet
[32] http://uk-air.defra.gov.uk/assets/documents/reports/cat09/1312231525_AQD_DD4_2012mapsrepv0.pdf

inland. In general, elevated sea salt concentrations extend further inland from the west and south coast of the UK than the east coast. The PCM empirical tend to show higher sea salt concentrations the south and west coast of the UK, but show no significant increase in sea salt concentrations at the east coast. These differences may arise due to the limited number of chloride measurements interpolated to create the PCM empirical maps.

4.2.6 SO$_2$

Two limit values for ambient SO$_2$ concentrations are set out in the AQD for the protection of human health. These limit values are specified as follows:

- An hourly concentration of 350 µg m^{-3}, with 24 exceedances permitted each year
- A 24-hour mean concentration of 125 µg m^{-3}, with 3 exceedances permitted each year.

A critical level for SO$_2$ for the protection of vegetation has also been specified in the AQD:

- An annual mean and winter mean concentration of 20 µg m^{-3}.

The critical level is designed to protect vegetation so it only applies in vegetation areas as defined in the Directive.

Annual mean SO$_2$ concentrations have been calculated from the CMAQ-UK hourly model output. In addition, maps of 99.73 percentile of hourly mean and 99.18 percentile of 24-hour mean SO$_2$ concentrations have been calculated from the CMAQ-UK model output. These percentile concentrations correspond to the number of allowed exceedances of the 1-hour and 24-hour limit values for SO$_2$ described above. Maps of annual mean SO$_2$ concentration from CMAQ-UK are presented in Table 21 (Appendix G) and are compared to PCM maps. The PCM maps have had an ecosystems and vegetation mask applied which removes areas close to agglomerations, built-up areas, industrial locations and motorways, as defined by the AQD (1999/30/EC Annex VI para 1(b)). The 24-hour mean and hourly mean percentile maps from CMAQ-UK and PCM models are presented in Table 12 and Table 22 (Appendix G). Summary statistics for the comparison between modelled and measured SO$_2$ annual, 24-hr and hourly metrics for CMAQ-UK and PCM are presented in Appendix HTable 33-Table 38.

The spatial distributions of the SO$_2$ annual mean, 24-hr and 1-hour from CMAQ-UK are similar to the spatial distributions from the PCM model, although the maps indicate higher concentrations of SO$_2$ are predicted by CMAQ-UK in urban areas in the midlands. The tables of summary statistics indicate that CMAQ-UK tends to over predict SO$_2$ annual mean and 1hr SO$_2$ metrics compared to measurements at national and verification sites but gives a reasonably good prediction of the SO$_2$ 24-hr mean metric. R-squared values relating the correlation of the CMAQ-UK model results to the measured values are generally comparable or better (higher) than the corresponding R-squared values for the PCM model indicating reasonable spatial agreement with the measurements.

Table 12: Maps of 99.73 percentile of hourly SO2 concentrations from CMAQ-UK and PCM.
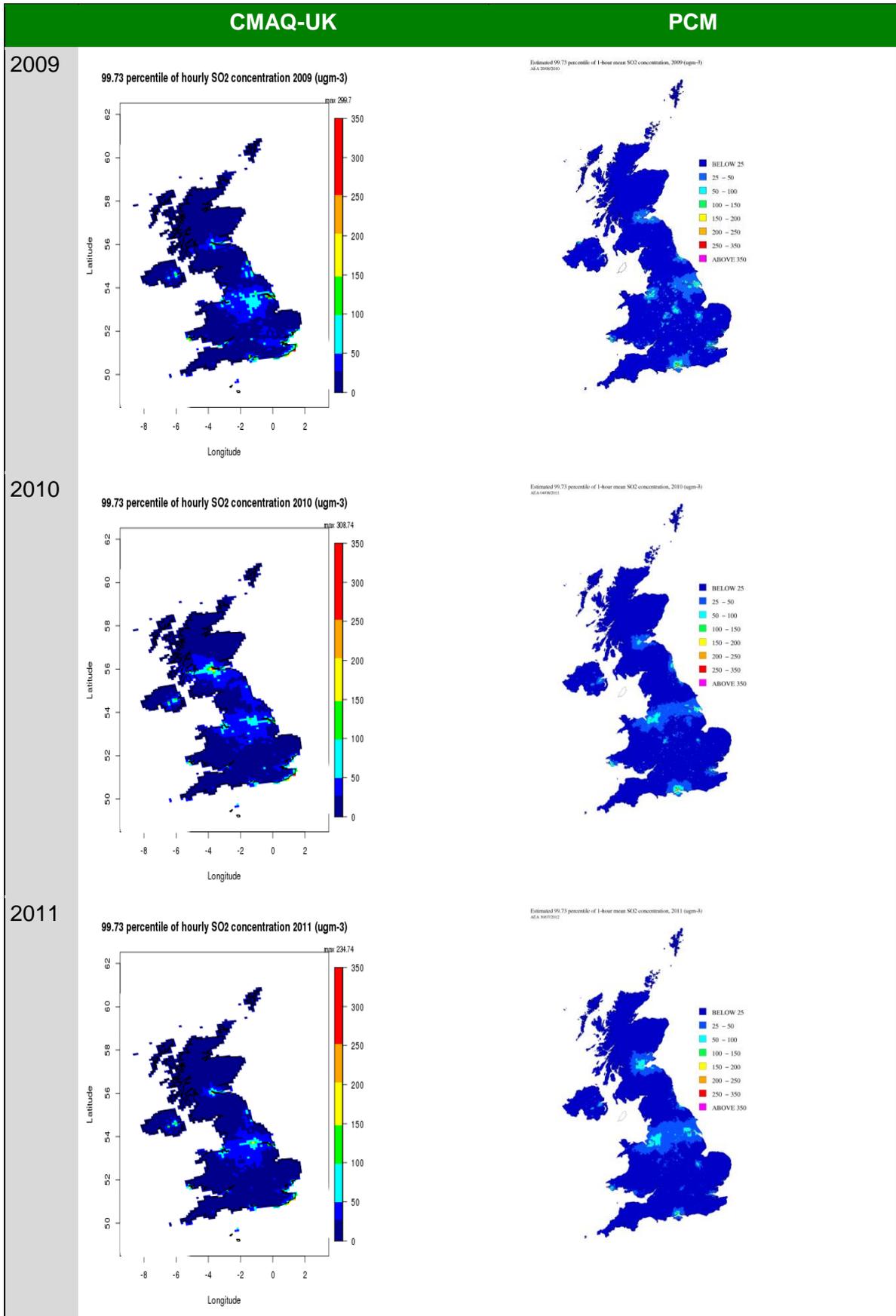
4.2.7 NOx/NO$_2$

Two limit values for ambient NO$_2$ concentrations are set out in the Air Quality Directive (AQD). These have been specified for the protection of human health and came into force from 01/01/2010. These limit values are:

- An annual mean concentration of 40 µg m^{-3}.
- An hourly concentration of 200 µg m^{-3}, with 18 exceedances permitted each year

A critical level for NO$_X$ for the protection of vegetation has also been specified in the Directive:

- An annual mean concentration 30 µg m^{-3} (NO$_X$ as NO$_2$).

Because this critical level is designed to protect vegetation, it only applies in vegetation areas as defined in the Directive.

In this document the focus is on the annual mean limit value. However, as CMAQ model outputs hourly concentration data it has the potential to be used to assess the hourly concentration limit values. Annual mean concentrations of NOx and NO$_2$ have been calculated from the hourly CMAQ results. The results are plotted in Table 13 and Table 14 where they are also compared to the PCM model results. Annual mean concentrations of NO$_x$ and NO$_2$ have been extracted at specific AURN and verification monitoring site locations for comparison with measurements. Summary statistics for the comparison between modelled and measured NO$_x$ and NO$_2$ for CMAQ-UK and PCM are presented in Appendix HTable 39, Table 40, Table 41 and Table 42, respectively.

The spatial distributions are similar between the PCM and CMAQ NOx and NO$_2$ grids. In the CMAQ maps elevated concentrations around urban areas and major roads tend to extend further into the rural areas than indicated by the PCM grids which may be a result of the lower resolution of the CMAQ-UK grid (10x10 km compared to the 1x1 km PCM resolution).

The verification statistics indicate the mean of the CMAQ-UK concentrations of both NOx and NO$_2$ at measurements at national and verification monitoring sites is in good agreement with the mean of the measurements. However the R-squared values for the CMAQ-UK analysis are significantly poorer than the R-squared values derived from the comparison of PCM concentrations with measurements indicating that the CMAQ-UK NOx and NO$_2$ concentrations are less well correlated to measurements.

Table 13: Maps annual mean NOx concentrations from CMAQ-UK and PCM.
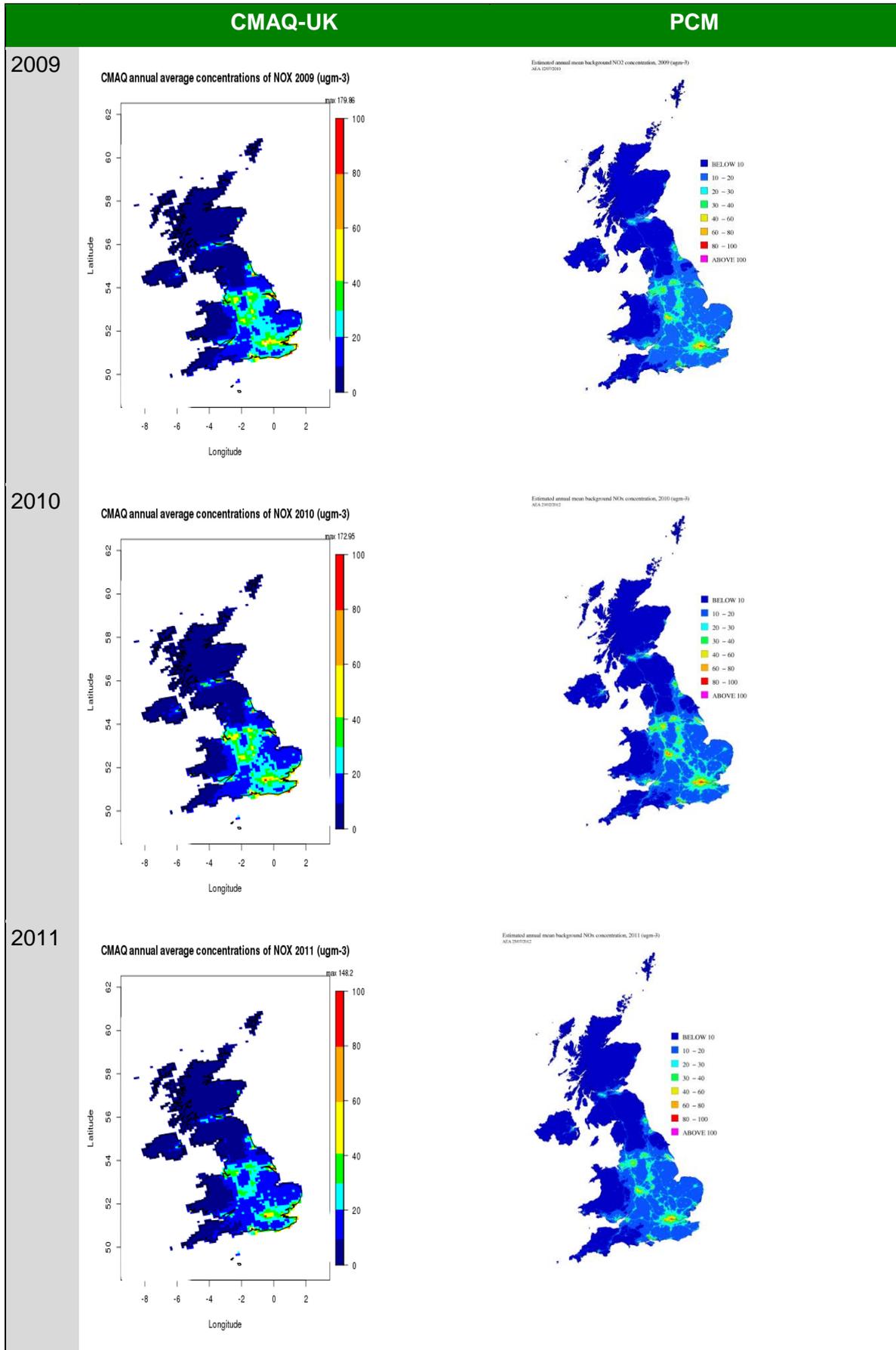
Table 14: Maps annual mean NO2 concentrations from CMAQ-UK and PCM.

CMAQ-UK	PCM
2009	CMAQ annual average concentrations of NO2 2009 (ugm-3)	Estimated annual mean background NO2 concentration, 2009 (ugm-3)
2010	CMAQ annual average concentrations of NO2 2010 (ugm-3)	Estimated annual mean background NO2 concentration, 2010 (ugm-3)
2011	CMAQ annual average concentrations of NO2 2011 (ugm-3)	Estimated annual mean background NO2 concentration, 2011 (ugm-3)
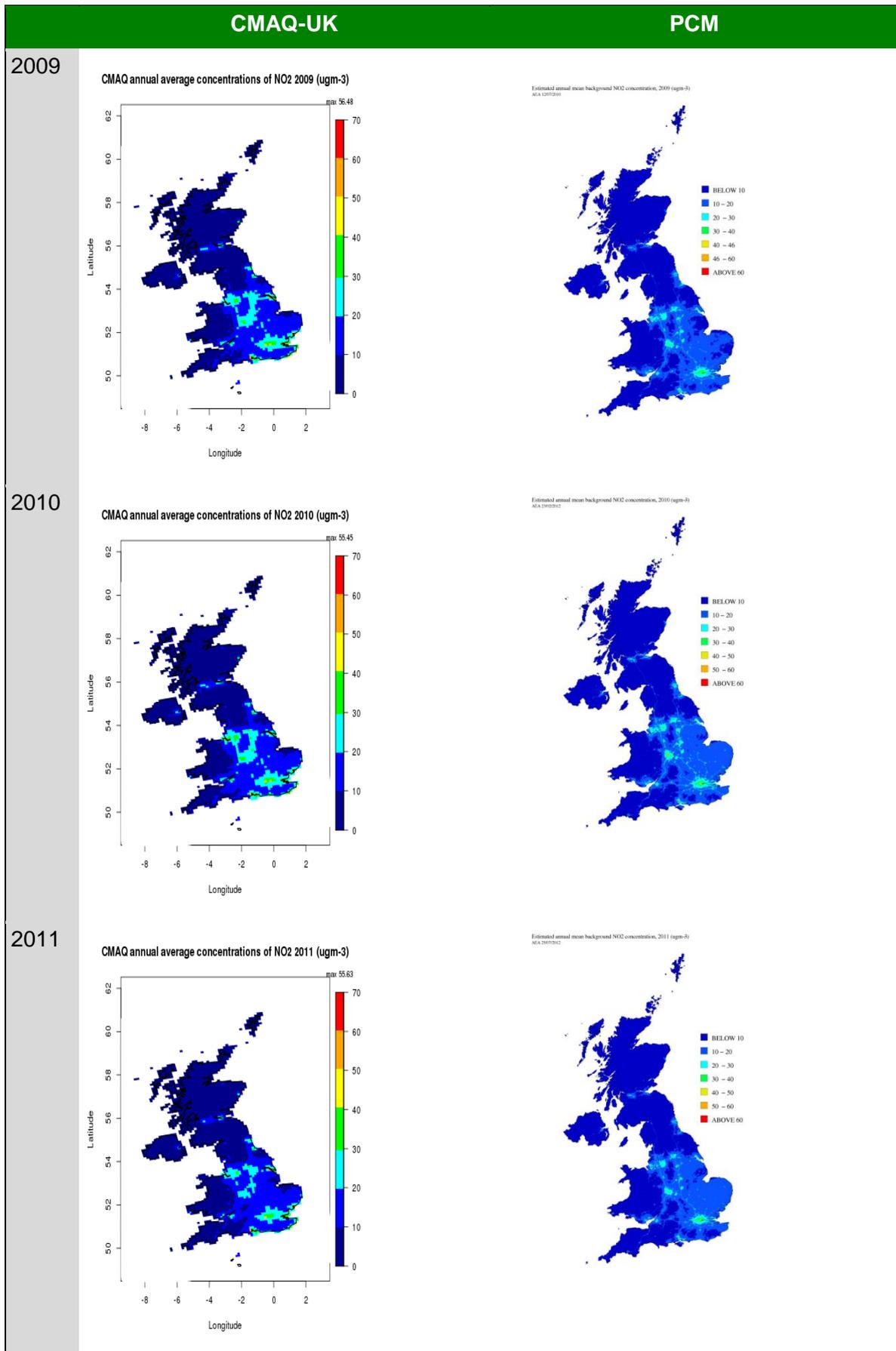
4.3 Evaluation of CMAQ Decision Support Tools – CMAQ-DDM

The 2014 release of CMAQ v5.0.2 included instrumented versions of CMAQ. These include additional modelling routines to aid the application of CMAQ for air quality policy development. The instrumented versions are:

- **CMAQ Decoupled Direct Method in 3 Dimensions (CMAQ- DDM)** – a model allowing simultaneous computation of sensitivity coefficients while air pollutant concentrations are being computed. The sensitivity coefficients represent the change in concentration, of any modeled species, associated with a change in a model input (e.g., an initial condition, boundary condition or emission rate) or a model parameter (e.g., a reaction rate) .

- **CMAQ Integrated Source Apportionment Method (CMAQ-ISM)**– a model that uses an emission-based approach generally provides a physical ground for source tracking results, as opposed to track emissions contributions from source groups and/or regions to ambient levels and deposited amounts of pollutants.

- **CMAQ-Sulphur Tracking Model (CMAQ-STM)**, - a model configuration that provides detailed information on the modeled sulphur budget. This model version tracks sulfate production from gas- and aqueous-phase chemical reactions, as well as contributions from emissions and initial and boundary conditions.

CMAQ-DDM was identified as an instrumented version that may provide benefits for Defra policy applications. It will allow a number of sensitivity tests to be run simultaneously. This may include: sensitivity to the uncertainty range in the emissions, sensitivity to emissions from a sector e.g. road transport or sensitivity of the UK to emission from Europe. Being a 'one atmosphere' model the impact of the sensitivity to changes in emissions can be analysed in all pollutants at the same time allowing analysis of unintended impacts to be taken into account.

A series of sensitivity tests focussing on emissions were run to establish:

- The flexibility of the sensitivity definitions
- The additional processing time
- An understanding of the model sensitivity coefficients

In order to maximise our understanding the tests were focused on a series of days in April 2011 when ozone and PM concentrations were above normal. This allows the sensitivity to be analysed during a period when conditions have most impact on regulatory limits.

The number and complexity of sensitivity tests included in each model run was varied. CMAQ-DDM takes longer to run than a single CMAQ simulation. The number of sensitivity tests had an effect on run times although the complexity of the sensitivity test had little effect. As the number of sensitivity tests are increased the incremental increase in run time decreases. In the limited tests undertaken in Phase 2, CMAQ-DDM performs better than separate CMAQ simulation when 4 or more sensitivity tests are included. The US EPA reports that CMAQ-DDM is computationally more efficient for 3 or more sensitivity tests. In a recent example which required CMAQ input to develop a reduced form air quality model required either 213 'brute-force' simulations or CMAQ-DDM run with 90 sensitivities calculated in a single run. The CMAQ-DDM ran ~9x faster than the brute force method (Foley 2014)[33].

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[33] Foley, K. M., et al. (2014). "Two reduced form air quality modeling techniques for rapidly calculating pollutant mitigation potential across many sources, locations and precursor emission types." Atmospheric Environment 98(0): 283-289

The DDM computes sensitivity coefficients, $S_{ij}^{(1)}$, which represent the change in concentration of a modelled species to a perturbation in a model input or parameter. The calculated sensitivity is a local, first order measure of sensitivity, therefore its accuracy in characterising the impact of input perturbations depends on the size of the perturbation and the nonlinearity of the response. Ozone and some PM species are secondary pollutants formed by complex, nonlinear chemical processes and the response of these species to perturbations in model inputs and parameters therefore may not be well represented simply by the first order sensitivity coefficient. For these species, the DDM can additionally calculated a higher (second) order sensitivity coefficient, $S_{ij}^{(2)}$, which represents the second derivative or local curvature of the species-input or species-parameter relationship and accounts partially for nonlinearities in response. The sensitivity coefficients output by the DDM are semi-normalised to the size of the unperturbed model input or parameter and therefore have the units of concentration. Combining the first and second order sensitivities in a Taylor series approximation (Equation 1) allows the calculation of the change in concentration of a species ΔC_i resulting from a fractional change in a model input or parameter $\Delta \varepsilon_j$.

$$\Delta C_i = \Delta \varepsilon_j S_{ij}^{(1)} + \tfrac{1}{2}\Delta \varepsilon_j^2 S_{ij}^{(2)} \qquad\qquad\qquad (1)$$

Previous studies by US-EPA have shown that ozone response calculated using DDM compared well to brute-force emissions for changes up to approximately a 50% reduction in emissions.[33,34,35] The PM response to emissions calculated using the DDM compare well spatially and temporally with the traditional brute-force approach, particularly for species responses to emissions of their "parent" precursor (e.g., sulfate to SO_2 emissions).[36] The DDM approach is less prone to numerical noise when compared to the brute force method, particularly for small perturbations.

Some demonstration sensitivity tests have been run for a range of emission scenarios relevant to Defra policy questions. The DDM version of CMAQ was not released until near the end of Phase 2 of this project and the analysis was limited. To gain the maximum experience of how the DDM works most of the sensitivity tests were restricted to 5-6 days in April 2011. The boundary conditions sensitivity test was run for 30 days in April 2011 which allows a 20 day average to be calculated after removing a spin-up period.

Sensitivity tests for ozone, NOx, VOC and PM were tested. CMAQ has a lot of VOC and PM components and this requires a lot of species to be followed within the sensitivity test. This slows the process and results is a large amount of data being generated. Within the DDM sensitivity definition file all the CMAQ species and not just the emissions species need to be defined, and each species needs to be included in the output file.

Demonstration 1: Sensitivity of O_3, NO_2 and $PM_{2.5}$ to the boundary conditions for O_3 and NO_X.

Phase 2 of this project has highlighted influence of the boundary conditions on the under prediction in winter and over prediction in summer ozone. In response to this one of the demonstrations has focused on how the DDM can be used to investigate the influence of the boundary conditions. This test focused on the sensitivity of ozone, NO_2 and $PM_{2.5}$ to ozone from the boundary conditions but did not include sensitivity of individual component PM species. First order sensitivity coefficients were calculated in each case, but not higher order sensitivity coefficients.

This demonstration is done for April 2011, a period when there is no significant under or over prediction. The sensitivity is run for April 2011 with the first 4 days discarded to allow a spin-

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[34] Cohan et al., (2005). "Nonlinear Response of Ozone to Emissions: Source Apportionment and Sensitivity Analysis." Environmental Science and Technology 39 (17), 6739–6748
[35] Baker et al., (2012), "A direct sensitivity approach to predict hourly ozone resulting from compliance with the National Ambient Air Quality Standard", Environmental Science and Technology. 47 (5), 2304-2313.
[36] Napelenok et al., Decoupled direct 3D sensitivity analysis for particulate matter (DDM-3D/PM), Atmospheric Environment 40 (2006) 6112-6121.

up of the boundary conditions. Data have been averaged for 20 days to give an indication of the monthly average. The version of the DDM tested did not allow for the sensitivity to be nested although this is available in a research version. This means the sensitivity of the UK10 grid to the global boundary conditions cannot be evaluated directly.

The sensitivity to boundary conditions are demonstrated at two different scales:

1. European sensitivity - the EU50 grid demonstrates the sensitivity to the global boundary conditions. These represent the importance of the contribution of long range transport of pollutants on the UK and the importance of getting the boundary conditions right for modelling Europe.
2. UK sensitivity - the UK10 grid demonstrates the sensitivity to the CMAQ-UK EU50 grid and the effect of medium range transport of pollutants from outside UK and Ireland on the UK.

European sensitivity

Figure 16 shows the first order coefficients for the sensitivity of ozone to ozone from the boundary conditions and nitrogen dioxide to nitrogen dioxide from the boundary conditions. The sensitivities are presented as a percentage of the modelled pollutant concentration and represent the extent of the influence of the boundary conditions on the concentrations modelled within the European EU50 grid if it is assumed that nonlinearities in the sensitivity responses are small. During the period 5-25th April 2011 ozone imported from the boundary conditions has an influence over the whole modelled area where as for NO_2 the effect of the boundary conditions are confined to the boundary of the grid, with a greater influence in the north-west corner as a response to the prevailing wind.

Figure 16: First order sensitivities of a) ozone and b) NO_2 concentrations to global boundary ozone and NOx, respectively that can be attributed to the contribution of global boundary ozone and NOx respectively averaged over the period 5-25th April 2011.

a)   b)
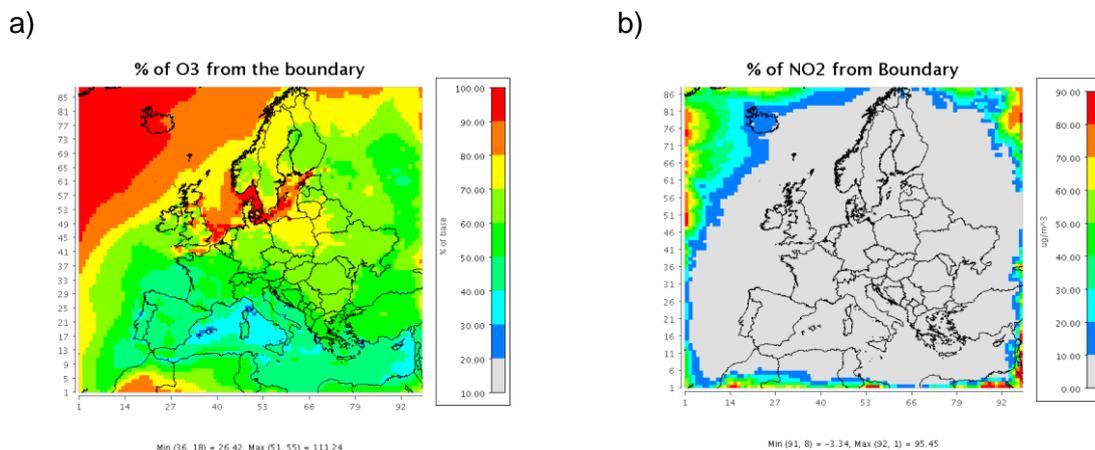
Figure 17 presents the sensitivity of modelled ozone, NO_2 and $PM_{2.5}$ concentrations to ozone from the global boundary conditions. In this figure semi-normalised values of the sensitivity coefficients are presented. As noted above, the sensitivity of ozone to ozone from the boundary conditions is extensive, but the figure also demonstrates the influence of ozone from the boundary condition on NO_2 and $PM_{2.5}$ concentrations. The interaction of ozone, nitrogen species and $PM_{2.5}$ is complex and a more complete process analysis would be required to explain the interactions.

Figure 17: Sensitivities of O₃, NO₂ and PM₂.₅ to O₃ concentrations from Global boundary conditions on the EU50 model domain (µgm⁻³) averaged over the period 5th-25th April 2011.
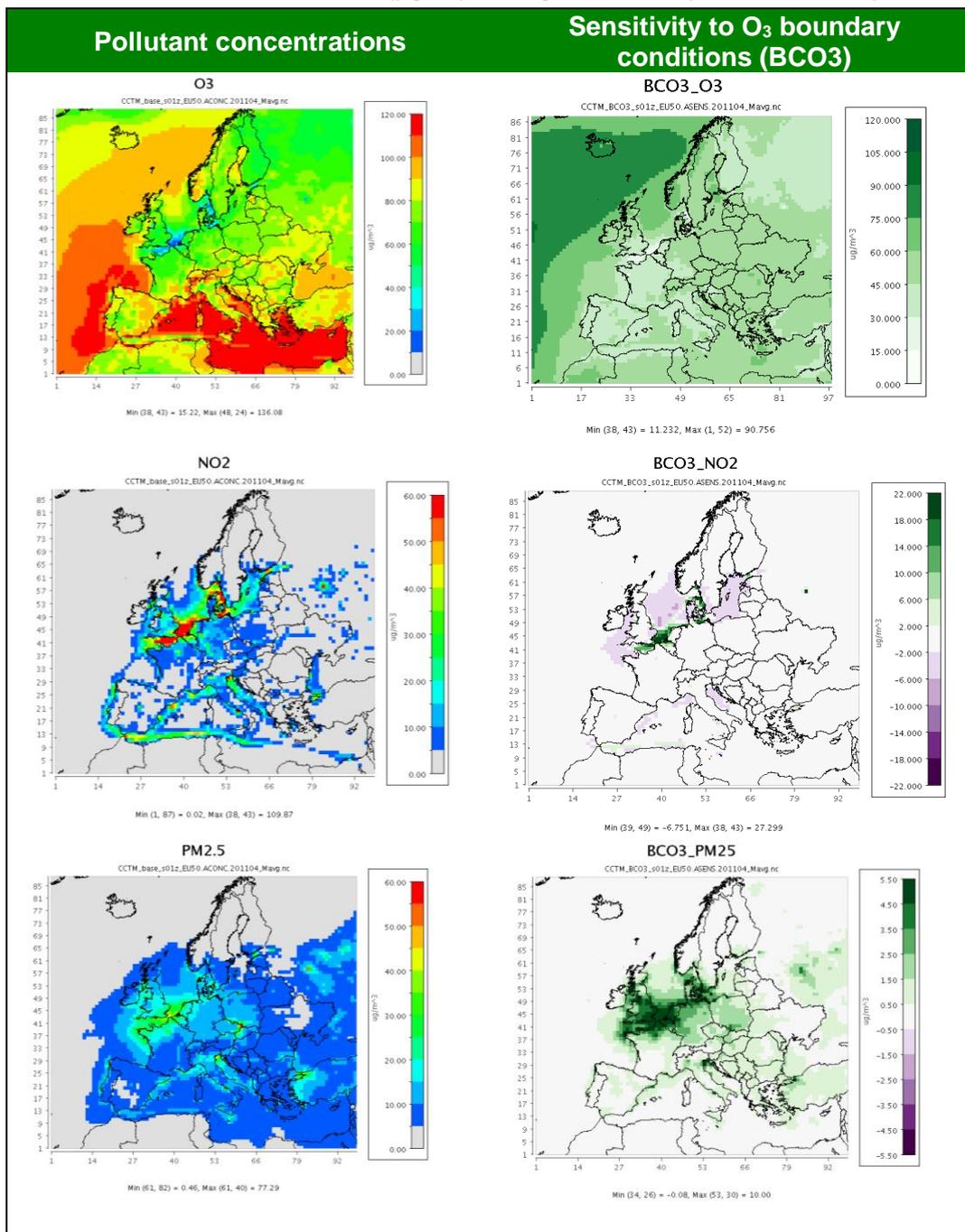
To demonstrate the influence of ozone from the boundary conditions to modelled ozone concentrations, a comparison of ozone measurements and modelled concentrations along with the sensitivity of ozone to ozone imported from the global boundary is show in Figure 18. This includes results for 3 sites:

1. Mace Head - Remote site west coast of Ireland
2. Auchencorth Moss – Rural site South of Edinburgh
3. Harwell – Rural site South of Oxford

At the Mace Head site the magnitude of the sensitivity of ozone to ozone from the global boundary conditions is approximately the same as the modelled concentration of ozone for most days of the modelled period. Mace Head is used as an indicator of background conditions for the UK, and these results are consistent with a high proportion of model ozone

being derived from the global boundary conditions at this site. At Harwell and Auchencorth Moss the sensitivity coefficients of ozone to ozone from the boundary conditions tends to be lower than the modelled concentration of ozone but remain a significant fraction of the ozone concentration. This suggests that the contribution of ozone imported from the boundary conditions remains a significant contributor to total ozone concentration at these sites. This short demonstration therefore shows the importance of high quality, accurate boundary conditions for ozone.

Figure 18: Hourly sensitivities of ozone to ozone from the Global boundary conditions contributions at Mace Head, Auchencorth Moss and Harwell sites on the EU50 model domain. Hourly modelled and measured concentrations, EU50 O3 and Obs O3 respectively, are compared to the hourly sensitivities, EU50 BCO3_O3.
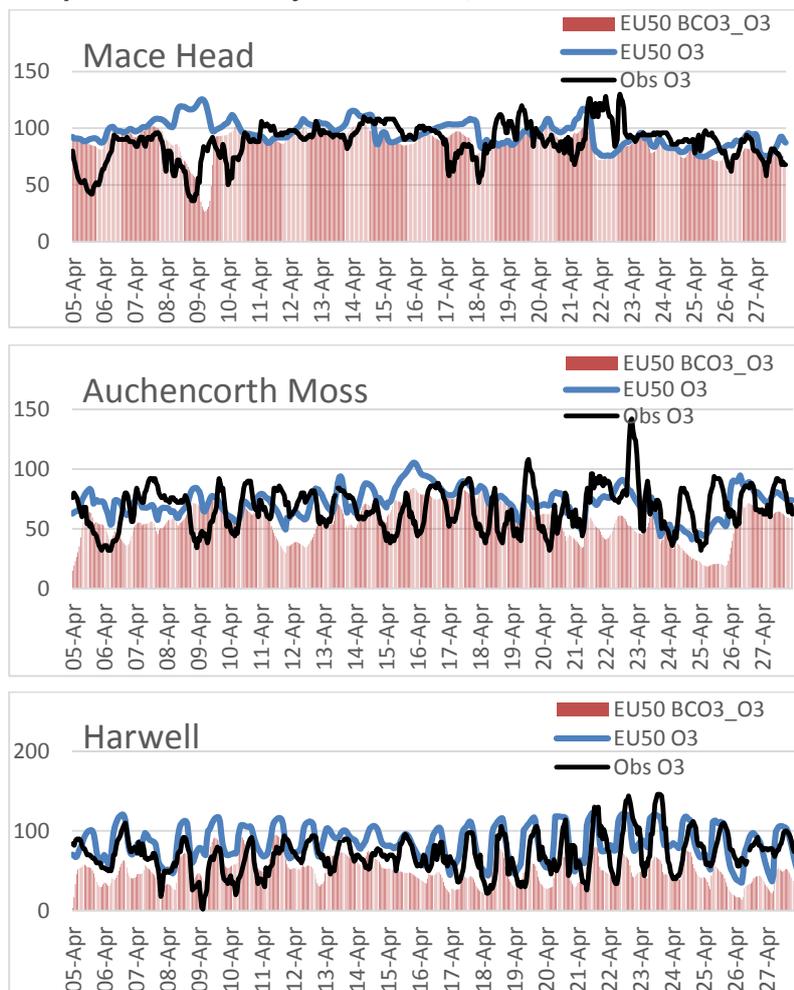
UK sensitivity

The spatial pattern in sensitivity of modelled ozone, nitrogen dioxide and PM$_{2.5}$ concentrations to ozone and NO$_X$ imported from the European nest is similar to the sensitivities to the global boundary conditions on the European domain (Figures 16 and 17). However, there is a greater influence of the boundary conditions from north-west to south-east (Figure 19). The effect on PM$_{2.5}$ is greater and is spatially associated with the areas of higher NOx and PM emissions and imported pollution from the south-east. Unlike the European sensitivity test, the UK ozone is sensitive to NOx boundary conditions. The NOx imported from the eastern boundary reduces the ozone in East Anglia, this is a small effects

relative to the influence from the ozone boundary conditions. The NOx imported from the boundary had little influence on NO_2 and $PM_{2.5}$.

Figure 19: Sensitivities of O_3, NO_2, and $PM_{2.5}$ to O_3 and NO_X from European boundary conditions (μm^{-3}) in the UK10 domain averaged over the period 5th-25th April 2011
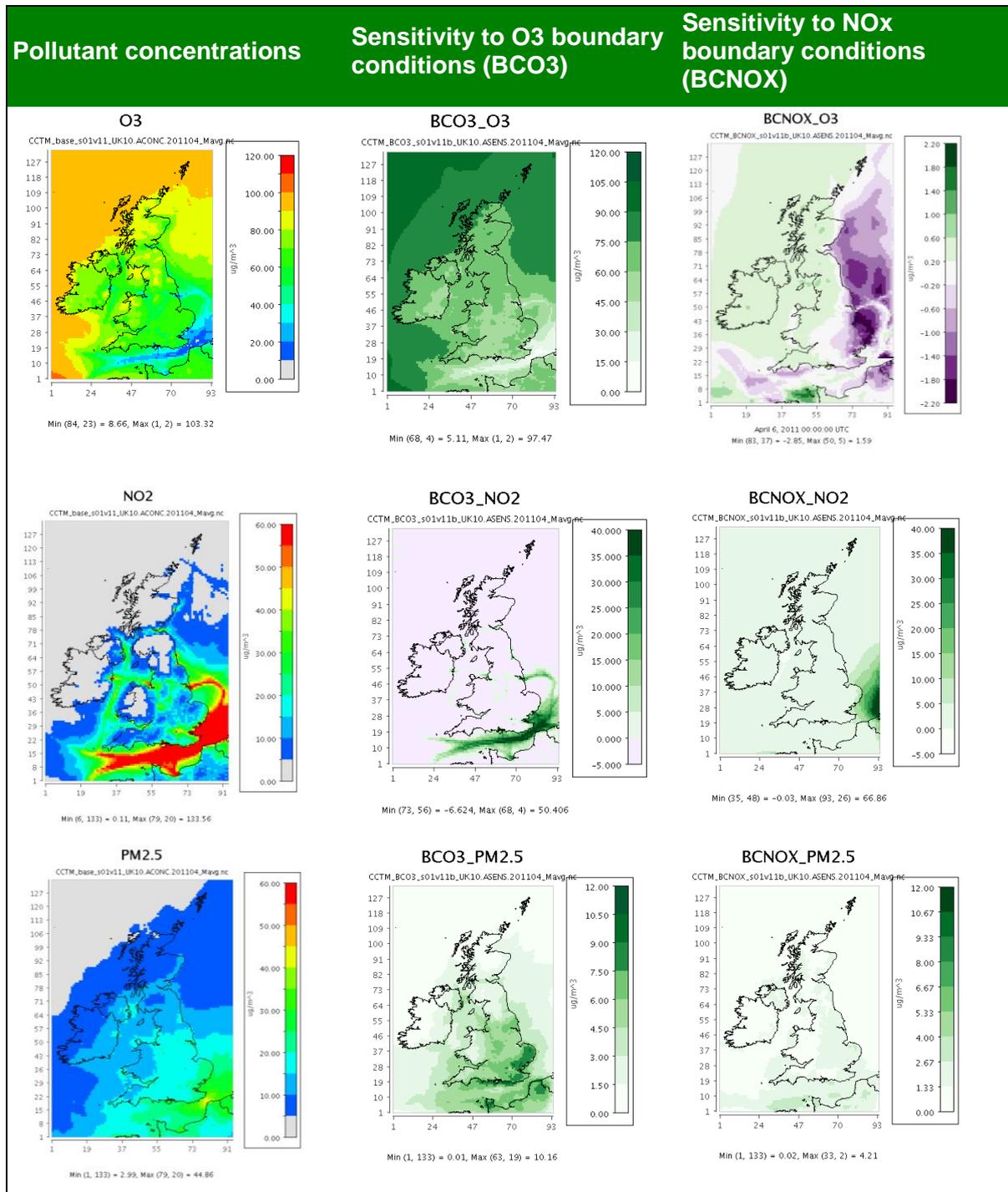
Figure 20 shows the influence of ozone from the EU50 boundary conditions to ozone concentrations modelled for the UK10 domain at the same 3 sites as presented in Figure 18. At the Mace Head site, sensitivity coefficients for the sensitivity of ozone to ozone from the European domain tend to be close to 100% of the total modelled ozone concentrations and the concentrations are stable for most of the period. During the periods where the ozone concentration is more variable, the sensitivity of ozone to ozone from the European nest tends to be a smaller fraction of the total modelled concentration. This suggests that local

conditions from within the UK10 model are more significant in these areas. Modelled ozone concentrations at Auchencorth Moss and Harwell can also be attributed predominantly to ozone transported in from the European model domain for most of the period under investigation.

This is a simple demonstration and a full process analysis would be required to explain the different contributing processes.

Figure 20: Sensitivity of ozone to ozone from the EU50 nested boundary conditions at Mace Head, Auchencorth Moss and Harwell on the UK 10 model domain. Hourly modelled and measured concentrations, UK10 O3 and Obs O3 respectively, are compared to the hourly sensitivities, UK10 BCO3_O3.
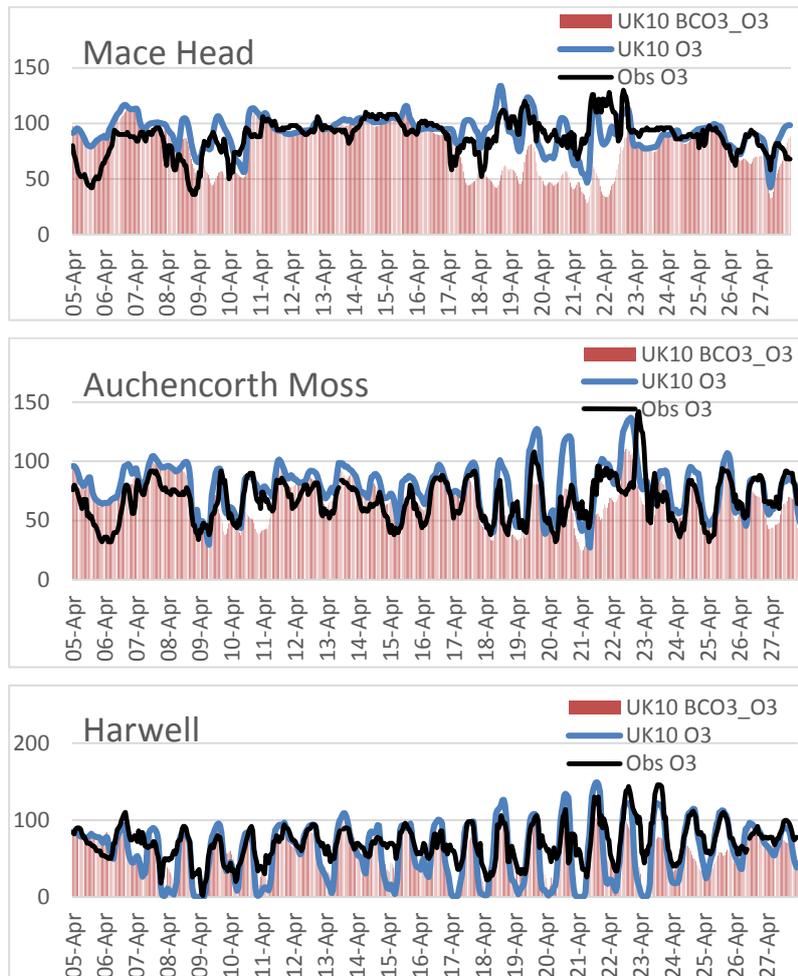
A similar sensitivity test can be done to investigate the PM precursors and component species.

Demonstration 2: Sensitivity of ozone to nitrogen oxide and VOC emissions from road transport

Policies targeting emissions controls are one of the key options for governments to manage pollutant concentrations. Policies may target emissions of specific pollutants from certain sectors, but may impact upon concentrations of many pollutants due to complex chemical process in the atmosphere. This demonstration exemplifies the use of the DDM to investigate the influence of the change in emissions for one pollutant on the concentration of another pollutant. This demonstration focuses on the influence of changes in emissions of nitrogen

oxides (NO_X=NO_2+NO) and VOCs from road transport on modelled concentrations of ozone. CMAQ-DDM was run for a five day period 20-24th April 2011 and hourly first and second order sensitivities of ozone concentration to nitrogen oxide and VOC emissions from road transport were calculated. The results presented below focus on a single day in this time period, 21st April 2011, and hourly results have been aggregated to provide daily average mapped results.

Figure 21: Base case daily average ozone concentration on 21st April 2011 (µgm[-3]).
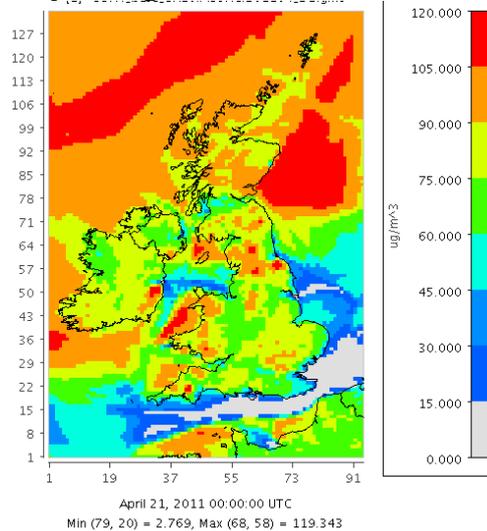
Figure 21 presents the daily average concentration of ozone calculated on 21st April 2011 without perturbation of emissions from road transport (i.e. the base case concentrations). First and second order sensitivity coefficients of ozone to NO_X and VOC are presented in Figure 22. The first order sensitives of ozone to NO_X emissions from road transport are negative showing that concentrations of ozone tend to increase as emissions of NO_X from road transport decrease. This is a result of the depression of ozone concentrations by NOx titration. Spatially, the magnitude of the first order sensitivity of ozone to NO_X is greatest at the locations of major cities and motorways. Additionally, the effect of wind direction can be seen in the spatial distribution of the sensitivity coefficients as sensitivities are seen to be high off the west coast of the UK which was downwind of NO_X emissions during this time period. The first order sensitivities of ozone to VOC emissions from road transport are positive showing that concentrations of ozone tend to decrease as VOC emissions from road transport decrease. In contrast to sensitivities to NOx emissions from road transport the spatial distribution of the sensitives to VOC indicate the sensitivity is greatest in rural and offshore locations downwind of transport emissions. These observations are consistent with NO_X limited ozone formation in urban areas and around major roads, while ozone formation is both NO_X and VOC dependent in rural and offshore location.

Figure 22: Daily average first and second order sensitivities of O₃ to NOₓ and VOC emissions from road transport on 21st April 2011.
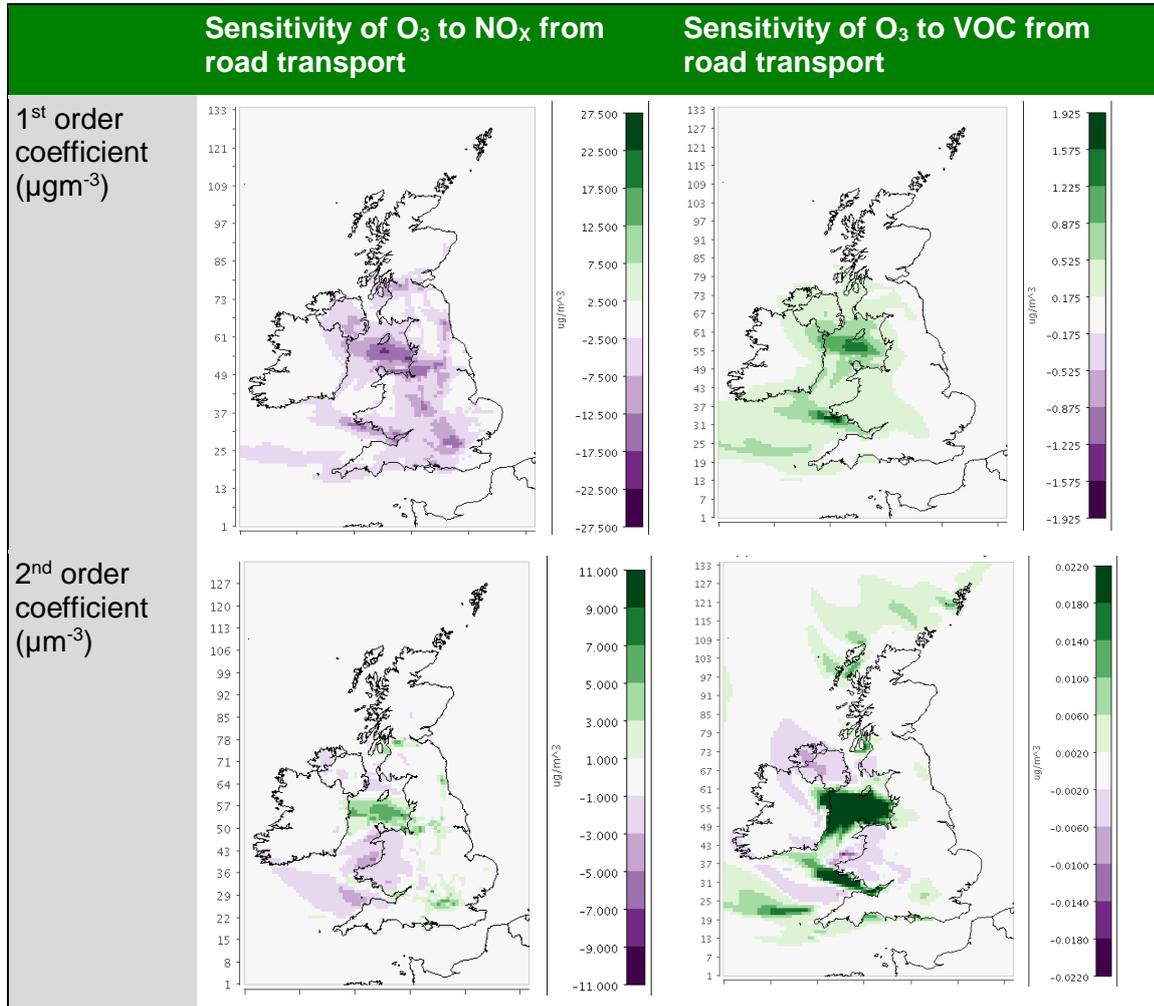
Second order sensitivities of ozone to NO_X and to VOC emissions from road transport are significantly smaller in magnitude that the corresponding first order sensitivity coefficients. Second order sensitivities to NO_X emissions from road transport and to VOC emissions from road transport tend to be positive in major urban areas and around major roads, but are negative in downwind rural locations reflecting local conditions.

Figure 23 demonstrates how the first and second order sensitivity coefficients calculated using the DDM can be combined using a Taylor series expansion (Equation 1) to calculate the spatial distribution of changes in ozone concentrations for a specific reductions in NOx and VOC emissions for road transport. The examples presented are for a 30% reduction in NOx emissions from road transport and a 30% reduction in VOC emissions from road transport. The plots demonstrate that on 21st April 2011 a 30% reduction in NO_X emissions from road transport would increase ozone concentration by up to approximately 6 μgm^{-3} in urban and roadside locations in the UK, while a 30% reduction in VOC emissions from road transport would lead to a small decrease in ozone concentrations (~0.1 μm^{-3}) in downwind rural locations.

Figure 23: Change in daily average surface ozone concentration (µgm^{-3}) modelled with a 30% reduction in NOx emissions (LHS) and a 30%in VOC emissions (RHS) from road transport sources, calculated from first and second order sensitivity coefficients output by the DDM. Results are presented for 21st April 2011.
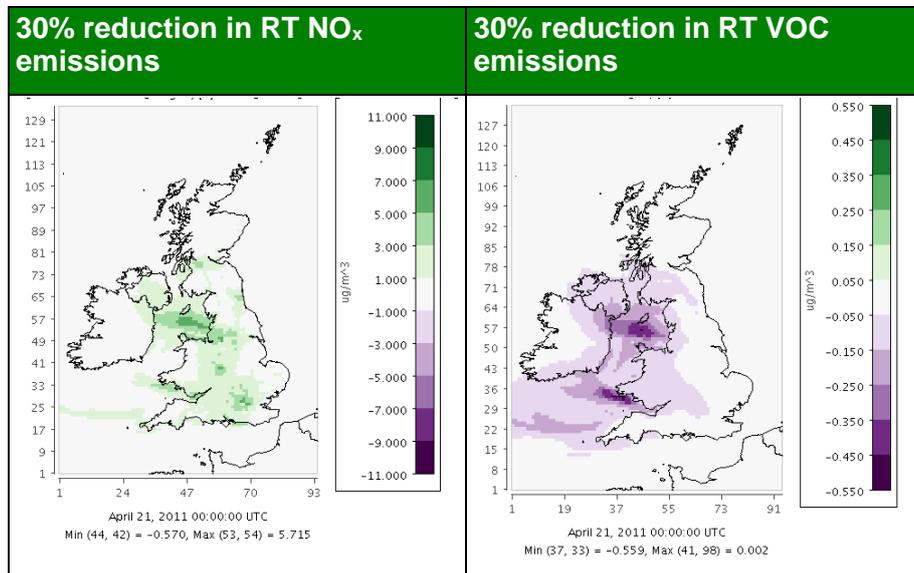
Summary recommendations

The primary focus of this short study was to learn how the CMAQ-DDM can be used and to understand the types of outputs it can generate. The study has demonstrated that DDM is a powerful and flexible tool to generate model sensitivity data. It provides a more efficient and potentially faster way of exploring the sensitivity of modelled concentrations to a range of different model input parameters which could be beneficial for policy applications, e.g. on the sensitivity to emission changes or uncertainties in emission inventories. However, the design of the sensitivity tests needs to be thought out carefully to ensure the relevant data are generated and the results also need careful interpretation. By designing the sensitivity tests the data can be used to investigate model performance or to develop sensitivity factors to be used to answer policy questions or include in other models.

The DDM is developed to look at gas, PM and deposition species, and includes first order sensitivity to emissions, boundary conditions, reaction rates and second order sensitivity of ozone precursors. Sensitivities may be calculated across the domain or for user specified regions. The DDM will continue to expand as more applications are identified.

The US EPA have used CMAQ-DDM (Foley 2014)[37] to develop a reduced form air quality model for rapidly calculating pollutant mitigation potential across many sources, locations and precursor emission types.

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[37] Foley, K. M., et al. (2014). "Two reduced form air quality modeling techniques for rapidly calculating pollutant mitigation potential across many sources, locations and precursor emission types." Atmospheric Environment 98(0): 283-289

5 Summary and Recommendations

After further optimisation of the WRF meteorology settings, the boundary conditions and development of the emissions data using the most up-to-date inventory information, an updated version of CMAQ-UK has been developed in Phase 2 which has been tested and evaluated against monitoring data. There was an overall improvement in the prediction of concentrations of NO_x and O_3 with a reduction in the bias in the concentrations predicted from the version of CMAQ-UK developed in Phase 1. The switch to CMAQ v5.0.1 which was released during Phase 1 saw no significant change in the predictions of O_3, PM_{10} and $PM_{2.5}$ concentrations from the version 4.7.1 used in Phase 1 when all other inputs were left unchanged.

A robust and traceable system has been developed for preparing all the key inputs necessary for a typical Defra policy application, including an efficient and transparent emission processing tool designed for testing a range of emission scenarios and a dashboard system for evaluating the CMAQ results. The emission processing tool can also be adapted for other Defra air quality models.

The updated version of CMAQ-UK has shown good overall performance for modelling different air pollutant concentrations over three consecutive years: 2009-2011. However, the updated version continues to under predict PM_{10} and $PM_{2.5}$ concentrations when compared with measurement data by around 25%. Over 60% of the $PM_{2.5}$ predictions are within a factor of two of the measurements and the long-term average concentrations are generally predicted better than the short-term average metrics. Apart from in 2009, the negative biases at rural sites are smaller than urban sites.

We recommend further work is required to develop the PM emissions speciation for individual source sectors and the quantification from sources such as dust resuspension. Secondary organic aerosols, a component of the PM mass, are severely under predicted in CMAQ-UK. Simplified parameterisations can be made to correct for these under predictions derived from the Master Chemical Mechanism implemented in the Photochemical Trajectory Model (PTM), by scaling up the SOA formation from toluene and terpene photo-oxidation. However, further work is required to determine the extent of scaling necessary for different years.

CMAQ-UK performs consistently well for ozone. Some seasonal over predictions are likely to be due to the influence of boundary conditions.

An application of CMAQ-UK has been demonstrated for a real policy issue, regarding the assessment of compliance with EU regulatory air quality obligations for $PM_{2.5}$ in 2020, a key date for achieving the $PM_{2.5}$ exposure reduction target. Based on the assumptions made concerning the meteorological conditions in 2020 and the latest UK and European emission projections, and taking into account the tendency for CMAQ-UK to under predict ambient $PM_{2.5}$, the results showed that the Average Exposure Indicator value for the 2019-2021 period will be 45% below the measured value for 2009-2011 indicating the exposure reduction target of 15% will be met.

These reductions are partly due to the reductions in precursor emissions and partly due to the meteorological and boundary conditions assumed for 2020. The latter are taken from the Met Office climate model, HadGEM2. To separate out the effects of differences in emissions, meteorology and boundary conditions between the 2009-2011 and 2019-2021 runs, we recommend repeating the 2019-2021 runs with 2009-2011 meteorology so that the changes in $PM_{2.5}$ concentrations due to changes in emissions can be seen in isolation.

Further demonstrations of CMAQ-UK have compared the outputs for a range of pollutants expressed in various concentration metrics defined in the EU Air Quality Directive with outputs from other models used for Defra policy, including the PCM model used for AQD compliance reporting. PCM is a semi-empirical model, linked to monitoring data, but the results demonstrate CMAQ can be used to produce the required outputs for these purposes. For regional applications in particular, the performance of CMAQ-UK is comparable to the PCM. CMAQ-UK is a chemical transport process model, so is better able to take into effect the effects of changes in precursor emissions and meteorology on secondary air pollutant formation. However, in the current configuration, the concentrations of metals and air toxics such as benzene and benzo(a)pyrene are not output although modelling of these species would be possible.

CMAQ-UK has the advantage of providing a single modelling framework for Defra's policy applications covering a range of pollutants. It has been constructed with an emissions processor that can readily feed in updates to UK and European emissions inventories. It will also benefit from future developments made by the wider WRF/CMAQ community, including chemical reaction schemes and meteorology, and from developments and future availability of boundary conditions and biogenic emissions data from international projects. This, and the open-source nature of WRF/CMAQ, is a key strength of the CMAQ-UK system. Initial analysis has also shown the potential benefits of CMAQ's decision support tools such as the DDM for assessing the effects of the uncertainties in key input data such as the emissions.

The current version of CMAQ-UK is optimised for regional scale modelling. The model will need to be coupled with another system for roadside and fine-scale modelling. Further work is also required to implement and test the source apportionment features of CMAQ.

To summarise the main recommendations:

- CMAQ-UK can be used for Defra's policy applications covering key pollutants at a regional scale. Further work is required for urban modelling and to ensure consistent behaviour between PM mass and PM components.

- The current configuration is well suited for regional-scale applications and coupling with other models would be required for improved finer-scale modelling.

- Further work is required to develop the PM emissions speciation for individual source sectors and the quantification from sources such as dust resuspension.

- CMAQ-UK will need to be maintained to accommodate improvements in meteorology, boundary conditions, chemistry schemes and those parameters affecting $PM_{2.5}$ concentrations, including the description of SOA formation.

- The 2019-2021 demonstration runs for $PM_{2.5}$ should be repeated with 2009-2011 meteorology so that the changes in $PM_{2.5}$ concentrations due to changes in emissions can be seen.

- Some further limited demonstration runs exploring the response of CMAQ-UK predictions to changes in emissions should be carried out. These could include runs to separate the effect of changes in UK vs European emissions as a typical policy application.

Appendix

A. Phase 1 to Phase 2 comparison and progress

Phase 1 analysis of the CMAQ-UK project showed that NO_x concentrations were overestimated from the late afternoon and evening, with opposite results observed for O_3. One possible factor responsible for this behaviour has been identified as the meteorological model WRF, which showed negative temperature and wind speed bias during the same afternoon and evening period. As part of Phase 2 of the project a comprehensive sensitivity analysis has been undertaken on WRF, with recommendations provided for its use within CMAQ-UK. In addition, a new set of emissions estimates and boundary conditions have together resulted in a considerable number of changes between Phase 1 and 2 of the CMAQ-UK model. These are summarised in Table 15.

Table 15 Phase 1 and Phase 2 CMAQ-UK configuration

Parameter	Phase 1	Phase 2
WRF version	3.3.1	3.4
CMAQ	4.7.1	v4.7.1 -> v5.0.1
Grid projection	50km (Europe) to 10km (UK)	No change
Spatial projection	Lambert conformal	No change
IC/BC	ECMWF/GFS	No change
Land use	MODIS (or USGS)	No change
Vertical layers	23 (7 layers below 1km)	No change
Nudging	Grid (U,V,T,Q)	No change
Radiation (SW/LW)	RRTM/Dudhia	No change
Cumulus	Kain-Fritsch	No change
Microphysics	WSM 6	No change
PBL	MYNN	ACM2
Land surface	NOAH	RUC
Surface layer	M-O	P-X
IC/BC	GEOS-Chem (interim)	MACC
Chemical Scheme	CB-05	CB05_AERO5 -> CB05_AERO6
Point source details	Included at a fixed height	change based on profiles from Bieser[38]
Emissions processor	AEA python based process	Ricardo-AEA NAEI emissions processor
Area anthropogenic emissions	EMEP/NAEI	TNO-MACC/NAEI (based on new maps)
Temporal emissions profiles	AEA, plus -King's College London for Traffic.	Ricardo-AEA (NAEI) profiles
Point anthropogenic emissions	NAEI	No change
Natural emissions	MEGAN/Biomass burning	No change

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[38] J. Bieser, A. Aulinger, V. Matthias, M. Quante, H.A.C. Denier van der Gon, Vertical emission profiles for Europe based on plume rise calculations , Environmental Pollution 159 (2011)

The new WRF Phase 2 configuration was tested using sensitivity analysis for January and July 2006. Subsequent use of the new configuration, alongside emissions and boundary conditions in 2006 (Jan and July) has shown that NO_x concentrations have reduced by approximately 30-40%. The meteorological sensitivity analysis in Phase 2 has also shown that the new WRF configuration improves the model's ability to predict the observed diurnal profile of surface meteorological parameters such as wind speed, temperature and relative humidity, with a resulting increase in the mixing of pollutants in the afternoon – evening period.

These changes have improved the magnitude of NO_x concentrations in the afternoon and evening period, especially in summer, leading the improvement of diurnal profiles of NO_x, NO_2 and O_3 concentrations (see Figure 24), with further details available in the supplement report entitled "*CMAQ Development for UK National Modelling: WRF Optimisation*" (version 07 July 2013).

Figure 24: Temporal profiles of observed, Phase 1 modelled (mod-p1), and Phase 2 modelled (mod-p2) NO_x and O_3 concentrations in January and July 2006
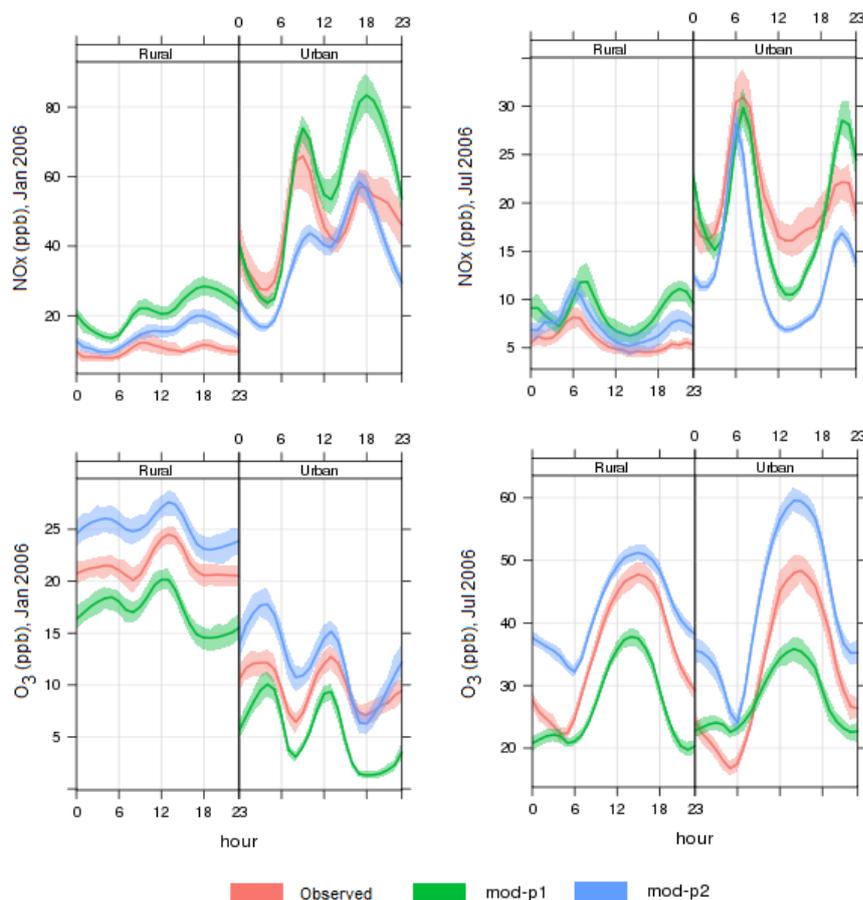
Use of the Phase 2 configuration has resulted in a reduction in the magnitude of NO_x and O_3 bias, with NO_x concentrations overestimated at rural sites and underestimated at urban locations in both January and July 2006 (Figure 24). One possible reason for the bias in NO_x predictions is an error in the spatial distribution of NO_x emissions.

O_3 concentrations are overestimated at both rural and urban sites in January and July with this behaviour only partially explained by the predictions of NO_x. Specifically, the over prediction of both O_3 and NO_x at the rural sites suggests a combination of mechanisms influencing O_3 predictions, with the analysis of surface O_3 concentrations at Mace Head indicating that boundary conditions may be partly responsible for the positive bias in O_3 at rural locations.

Also during Phase 2, CMAQ version 5 was released and so a comparison was made with predictions using v4.7.1 of the model in 2009. A statistical analysis using FAC2, MGE, NMGE, RMSE, r and COE, indicates that the v4 results of NO_2, NO_x, O_3, PM_{10} and $PM_{2.5}$ are in better agreement with the measurements than v5 (see dashboards). However, the differences for O_3, PM_{10} and $PM_{2.5}$ are small, for example, the differences between RMSE values of v4 and v5 is less than 1 µg m^{-3} for all three species.

The temporal profiles of modelled results and the measurements in Figure 25 and Figure 26 suggest a small reduction of O_3 due to the increase of NO_x using v5. The CMAQ O_3 predictions for 2009, 2010 and 2011 at Mace Head suggests that CMAQ may have inherited the bias from MACC boundary conditions, and that changes in v5 of the model have very small effects on regional pollutants like O_3 at remote locations where local influences are negligible.

Figure 25 and Figure 26 also show that the differences between PM_{10} and $PM_{2.5}$ from v4 and v5 are very small and that overall both models have a tendency to under predict both PM_{10} and $PM_{2.5}$ concentrations. The mean bias in $PM_{2.5}$ predictions tends to be smaller than for PM_{10} suggesting that improving coarse mode PM emissions may improve the model's performance.

Figure 25: Monthly profiles of 2009 concentrations of NO₂ and NOₓ derived from CMAQ v4 (top) and v5 (bottom).
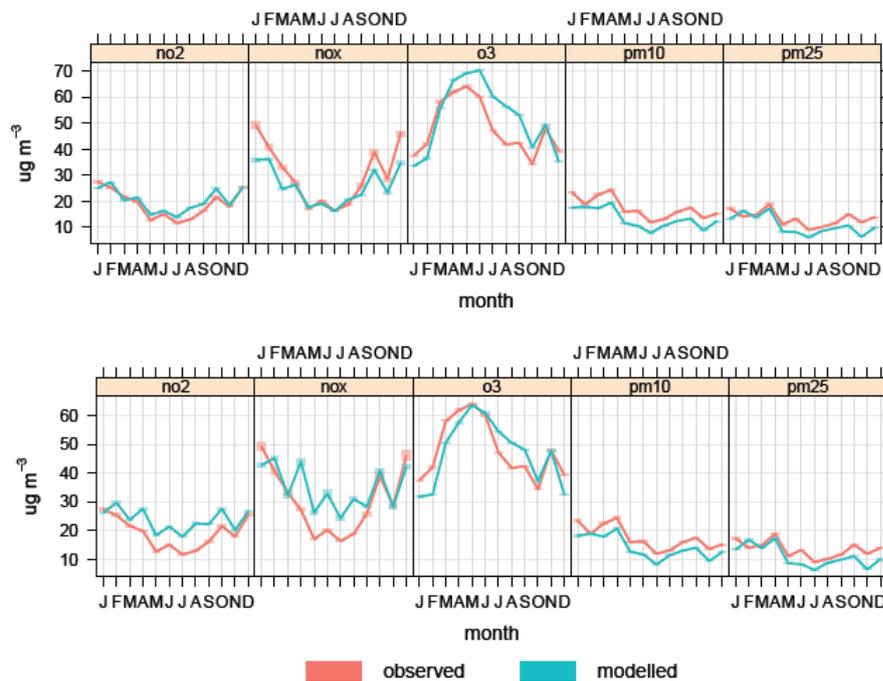
Figure 26: Diurnal profiles of 2009 concentrations of NO₂ and NOₓ derived from CMAQ v4 (top) and v5 (bottom).
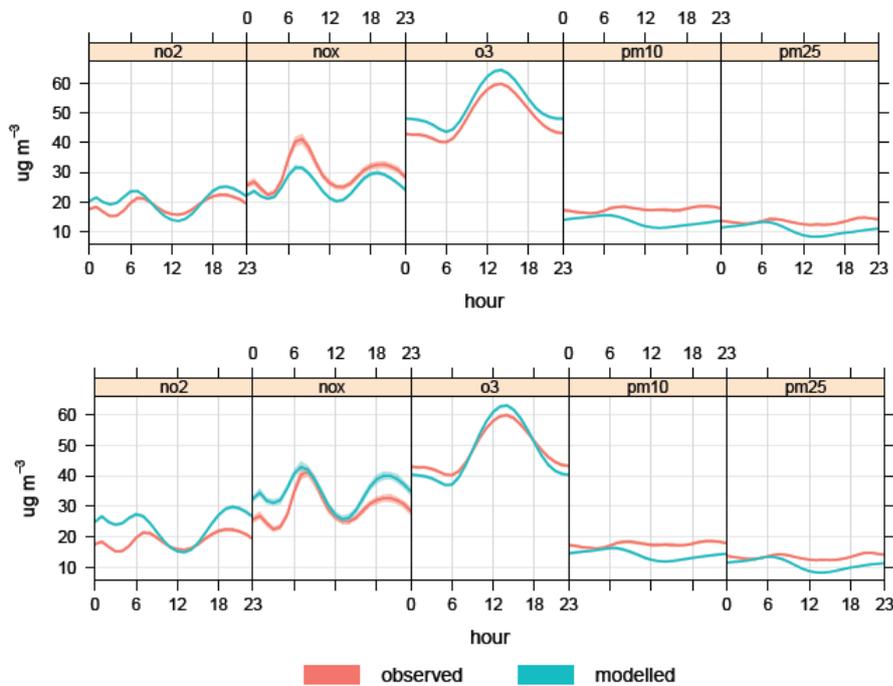
The differences between RMSE values of v4 and v5 are larger for NOₓ predictions and can be as high as 14 µg m⁻³. The v5 NOₓ predictions are biased between April-August months whereas NO₂ is biased between February-November (Figure 25). The diurnal profiles show that the differences between v4 and v5 predictions are greatest overnight and early morning (Figure 26). The positive bias of primary pollutants such as NOₓ is believed to be strongly influenced by the modification of Kzmin parameterisation in v5, which yields weaker mixing conditions particularly at rural sites.

B. CMAQ-UK model performance 2009-2011

CMAQ-UK model

Gas and particles model performance

pollutant	n	obs	mod	FAC2	MB	MGE	NMB	NMGE	RMSE	r	COE
no2	433877	19	22	0.57	3.23	11.7	0.169	0.61	19	0.61	0.27
nox	433877	30	31	0.54	0.78	22.3	0.025	0.73	51	0.42	0.24
o3	569535	48	48	0.76	-0.62	15.2	-0.013	0.31	20	0.71	0.25
pm10	117751	18	14	0.69	-4.27	7.8	-0.238	0.43	11	0.59	0.11
pm25	169696	14	11	0.63	-3.59	6.7	-0.250	0.47	10	0.59	0.13

Model: CMAQ v5.0.1   ICBCs: MACC
Grid: UK 10km   Chem: cb05clae5aq
Met: WRF(w02)   Observation: DEFRA MIE
Emissions: NAEI,TNO,ENTEC   Date: 2009001-2011365
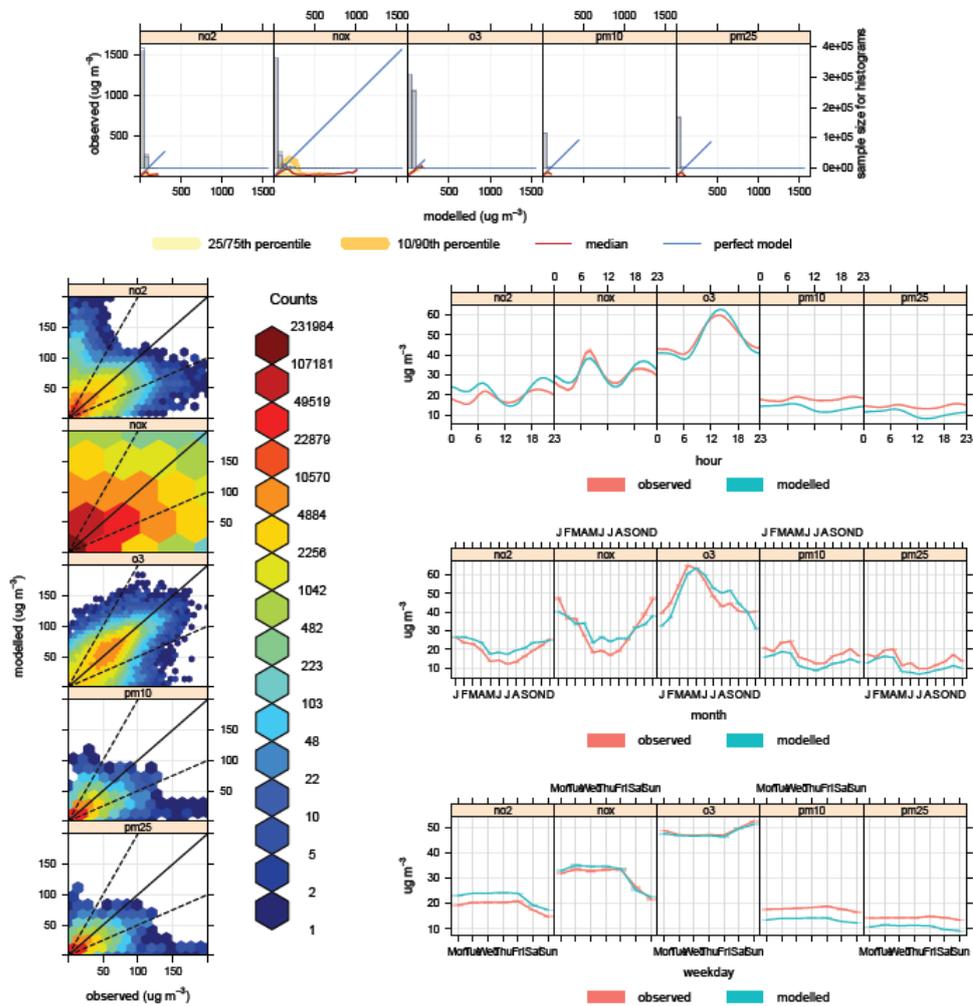
C. Sample of the model performance dashboards of NO_2, O_3, PM_{10} and $PM_{2.5}$ and PM components in 2011

CMAQ–UK O3 model

v5.0.1 O3 model performance

Annual O$_3$ Concentration

ug m^{-3} 25 35 45 55
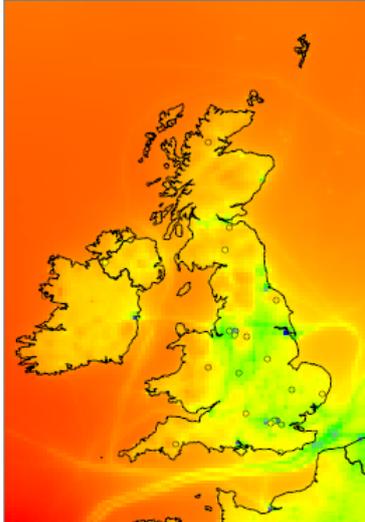
Statistic	type	n	obs	mod	FAC2	MB	MGE	NMB	NMGE	RMSE	r	COE
8h	RU	133526	54.8	53.7	0.88	−1.11	13.23	−0.0202	0.24	17.5	0.69	0.195
8h	SU	17282	43.2	44.1	0.82	0.93	12.27	0.0215	0.28	15.6	0.77	0.303
8h	UB	42323	37.2	40.9	0.75	3.69	13.77	0.0993	0.37	17.6	0.73	0.272
8hDx	SU	730	59.5	62.7	0.95	3.20	12.36	0.0538	0.21	15.4	0.79	0.199
8hDx	UB	1796	53.4	60.1	0.89	6.63	14.78	0.1240	0.28	18.7	0.76	0.225
8hDx	RU	5650	67.6	67.9	0.96	0.38	11.66	0.0056	0.17	15.7	0.68	0.182
8hDx100	UB	5	13.2	18.2	0.60	5.00	9.40	0.3788	0.71	13.1	0.29	0.045
8hDx100	SU	2	13.5	20.5	0.50	7.00	10.00	0.5185	0.74	12.2	1.00	−3.000
8hDx100	RU	16	17.5	12.9	0.62	−4.62	8.25	−0.2643	0.47	9.7	0.55	−0.784
AOT40	UB	5	3.2	4.6	0.60	1.41	2.35	0.4406	0.74	2.9	0.41	0.023
AOT40	RU	16	3.7	3.9	0.94	0.17	0.96	0.0460	0.26	1.2	0.64	−0.034
AOT40	SU	2	2.8	4.8	0.50	2.07	2.13	0.7464	0.77	3.0	−1.00	−45.808

UB=Urban background,SU=Suburban,RU=Rural
8h=8h mean,8hDx=8h Daily max, 8hDx100=8hDx>100ugm3

Odds ratio skill score (Moderate or above: 100ug m^{-3})

% ■ correct rejection ■ false alarm ■ hit ■ miss

8hr daily maximum

● RU ● SU ● UB

● RU ● SU ● UB
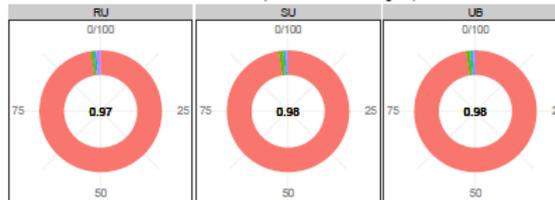
KING'S College LONDON   **RICARDO-AEA**   University of Hertfordshire UH   **rdScientific**   Department for Environment Food & Rural Affairs
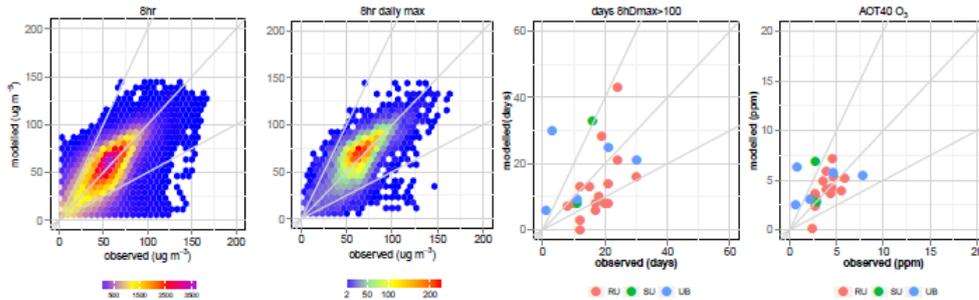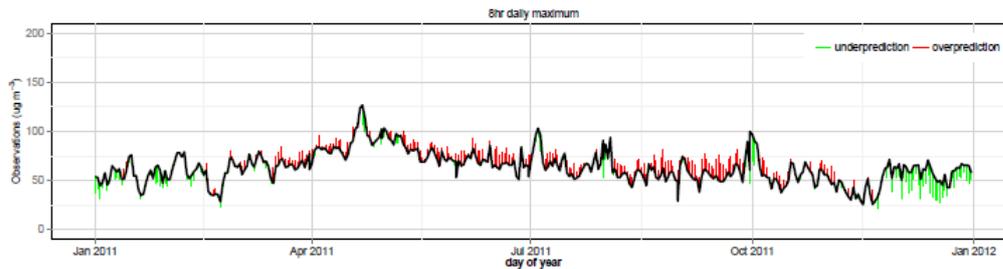
CMAQ–UK PM10 model

v5.0.1 PM10 model performance

Annual PM₁₀ Concentration
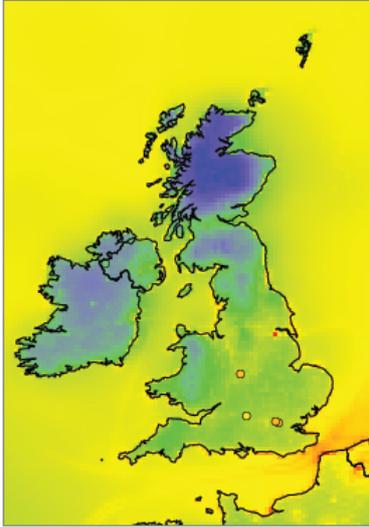
Statistic	type	n	obs	mod	FAC2	MB	MGE	NMB	NMGE	RMSE	r	COE
Daily PM10	RU	791	14	12.7	0.84	−1.8	5.0	−0.12	0.35	7.1	0.74	0.287
Daily PM10	UB	963	23	15.4	0.78	−7.7	8.5	−0.33	0.37	11.3	0.78	0.105
Hourly PM10	RU	18532	14	12.7	0.69	−1.8	6.7	−0.12	0.46	10.0	0.61	0.157
Hourly PM10	UB	22563	23	15.4	0.66	−7.7	9.9	−0.33	0.43	14.4	0.63	0.069
Monthly PM10	RU	28	15	13.0	1.00	−1.7	3.1	−0.12	0.21	3.8	0.85	0.399
Monthly PM10	UB	34	23	15.2	1.00	−7.7	7.7	−0.33	0.33	8.3	0.93	−0.235
PM10 DPM10>50	RU	3	5	1.3	0.50	−3.7	3.7	−0.73	0.73	5.3	0.33	−0.100
PM10 DPM10>50	UB	3	18	2.3	0.00	−16.0	16.0	−0.87	0.87	16.0	0.94	−35.000

UB=Urban background, SU=Suburban, RU=Rural
DPM10=Daily mean, DPM10>50=Number of Days DPM10>50

Odds ratio skill score (Moderate or above: 50ug m⁻³)
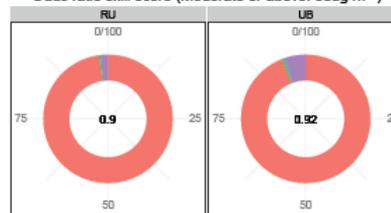
% ■correct rejection ■false alarm ■hit ■miss

ug m⁻³ ○ 10 ○ 20 ○ 30

Daily mean PM₁₀ concentration

KING'S College LONDON   **RICARDO-AEA**   University of Hertfordshire UH   **rdScientific**   Department for Environment Food & Rural Affairs
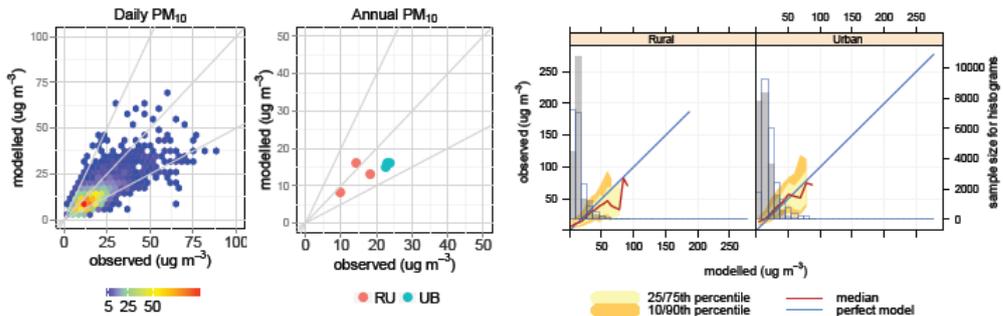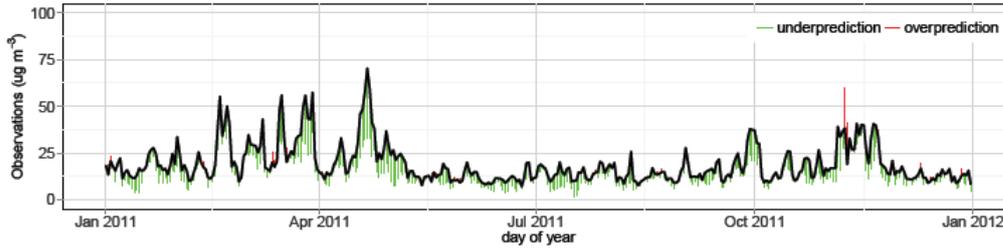
CMAQ-UK PM2.5 model

v5.0.1 PM2.5 model performance

Annual PM$_{2.5}$ Concentration
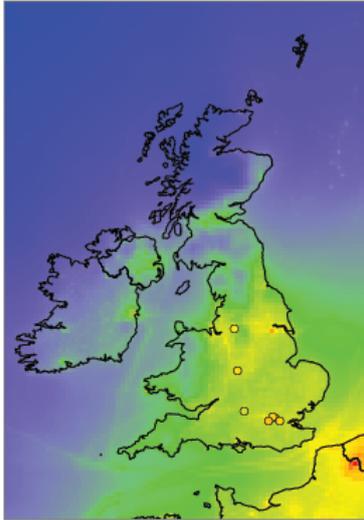
Statistic	type	n	obs	mod	FAC2	MB	MGE	NMB	NMGE	RMSE	r	COE
Daily PM2.5	RU	658	13	10	0.79	−2.6	4.3	−0.20	0.33	6.2	0.83	0.37
Daily PM2.5	UB	1630	16	12	0.78	−4.5	5.8	−0.28	0.36	9.1	0.75	0.30
Daily PM2.5	SU	305	16	11	0.74	−5.5	6.2	−0.34	0.39	8.9	0.82	0.23
Hourly PM2.5	RU	15592	13	10	0.64	−2.6	5.8	−0.20	0.45	8.6	0.70	0.22
Hourly PM2.5	SU	6924	16	11	0.61	−5.5	7.4	−0.35	0.46	10.9	0.70	0.14
Hourly PM2.5	UB	38230	16	12	0.63	−4.5	7.3	−0.28	0.45	12.4	0.62	0.21
Monthly PM2.5	RU	23	13	11	1.00	−2.7	2.9	−0.20	0.22	3.5	0.94	0.40
Monthly PM2.5	UB	57	16	12	1.00	−4.5	4.6	−0.28	0.29	5.3	0.92	0.16
Monthly PM2.5	SU	12	17	11	1.00	−6.3	6.3	−0.36	0.36	7.7	0.92	0.12

UB=Urban background, SU=Suburban, RU=Rural

Odds ratio skill score (Moderate or above: 36ug m^{-3})
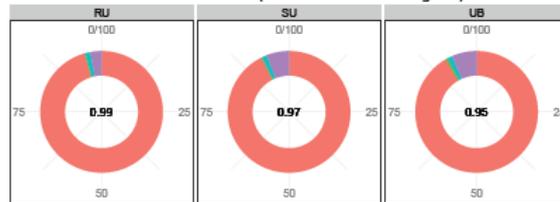
% ■correct rejection ■false alarm ■hit ■miss

ug m^{-3} ○ 10 ○ 20

Daily mean PM$_{2.5}$ concentration

Daily mean PM2.5

AnnualPM2.5

● RU ● SU ● UB

Rural   Suburban   Urban Background

25/75th percentile
10/90th percentile

median
perfect model
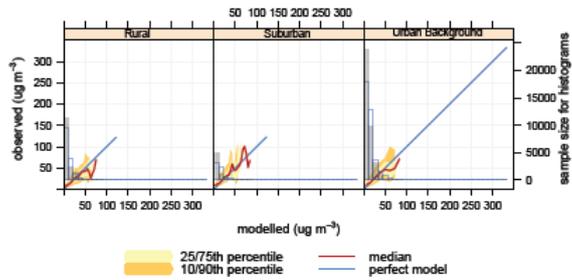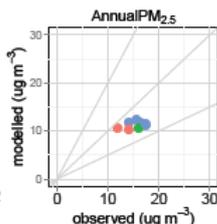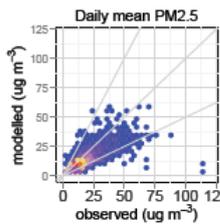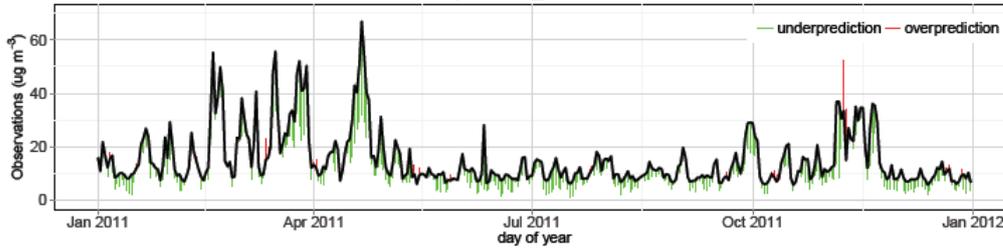
CMAQ-UK model

v5.0.1 fine particles model performance

				pollutant	n	obs	mod	FAC2	MB	MGE	NMB	NMGE	RMSE	r	COE
Model:	CMAQ v5.0.1	ICBCs:	MACC	pm25_nh4	1044	0.74	0.71	0.47	−0.028	0.46	−0.038	0.62	0.76	0.70	0.369
Grid:	UK 10km	Chem:	cb05clae5aq	pm25_no3	1044	1.05	0.71	0.29	−0.341	0.78	−0.324	0.74	1.47	0.68	0.317
Met:	WRF(w02)	Observation:	DEFRA MIE	pm25others	1044	2.73	1.79	0.45	−0.933	1.83	−0.342	0.67	3.21	0.24	0.068
Emissions:	NAEI,TNO,ENTEC	Date:	2011001−2011365	pm25_so4	1044	1.22	1.57	0.54	0.350	0.92	0.287	0.75	1.43	0.47	0.008
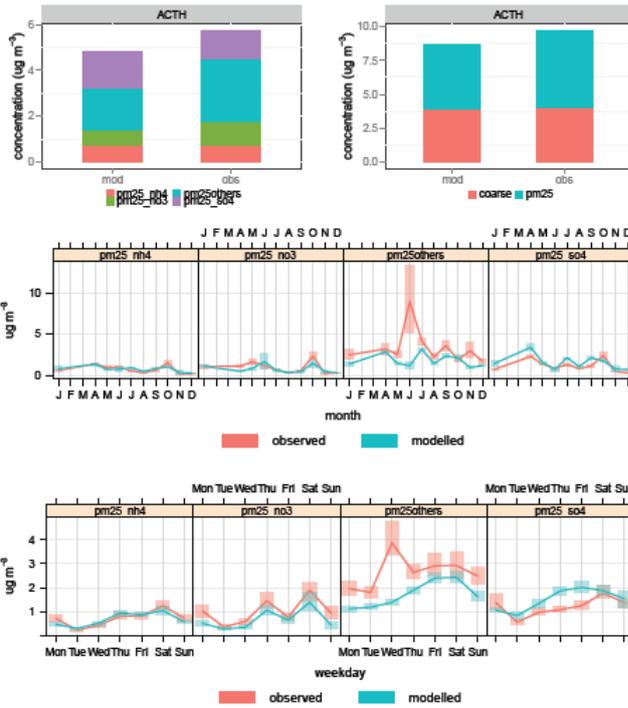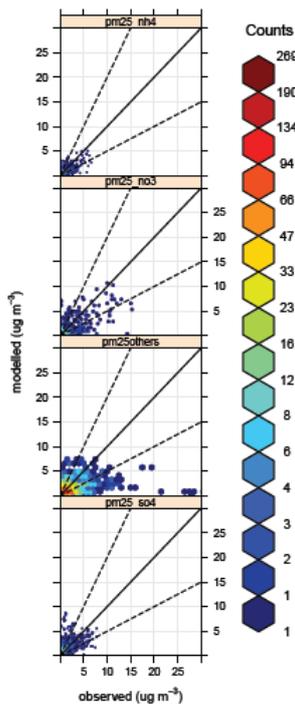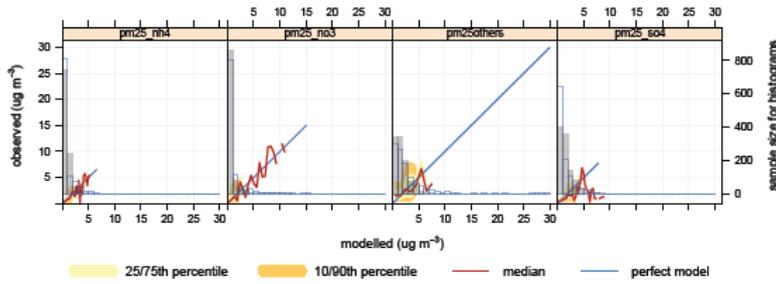
D. WRF3.4.1 model performance dashboard (2009-2011)

E. European species specific emission scaling factors from 2009 to future years

(Selected countries)

Country	CO 2019	CO 2020	CO 2021	NH3 2019	NH3 2020	NH3 2021	NOX 2019	NOX 2020	NOX 2021	PM10 2019	PM10 2020	PM10 2021	PM2.5 2019	PM2.5 2020	PM2.5 2021	SO2 2019	SO2 2020	SO2 2021	VOC 2019	VOC 2020	VOC 2021
Albania	0.86	0.86	0.85	1.16	1.17	1.16	1.11	1.12	1.13	1.00	1.00	0.99	0.96	0.96	0.95	0.80	0.82	0.85	0.86	0.85	0.84
Austria	0.78	0.76	0.75	1.07	1.07	1.08	0.58	0.53	0.51	0.90	0.90	0.89	0.82	0.81	0.80	0.84	0.82	0.81	0.84	0.83	0.81
Belarus	0.92	0.92	0.91	1.07	1.08	1.08	0.98	0.99	0.99	1.02	1.02	1.02	1.01	1.02	1.02	1.13	1.13	1.12	0.87	0.86	0.84
Belgium	1.37	1.43	1.35	0.91	0.91	0.93	0.74	0.70	0.68	0.88	0.87	0.87	0.78	0.77	0.77	0.77	0.77	0.77	0.84	0.84	0.82
Denmark	0.48	0.44	0.43	0.83	0.82	0.81	0.65	0.61	0.59	0.73	0.70	0.69	0.64	0.60	0.59	0.69	0.68	0.67	0.69	0.66	0.65
Estonia	0.76	0.73	0.73	0.89	0.90	0.91	0.77	0.75	0.73	0.68	0.64	0.63	0.74	0.70	0.69	0.42	0.40	0.39	0.90	0.90	0.89
Finland	0.74	0.71	0.69	0.94	0.94	0.94	0.67	0.64	0.63	0.84	0.82	0.82	0.79	0.77	0.76	0.76	0.76	0.76	0.74	0.72	0.70
France	0.76	0.75	0.74	0.99	0.99	0.99	0.59	0.55	0.53	0.90	0.89	0.89	0.80	0.78	0.77	0.50	0.47	0.47	0.76	0.74	0.73
Germany	0.82	0.80	0.80	0.98	0.99	0.99	0.64	0.60	0.58	0.86	0.85	0.85	0.79	0.77	0.77	0.76	0.74	0.73	0.84	0.83	0.82
Greece	0.82	0.80	0.79	0.90	0.89	0.90	0.68	0.65	0.61	0.71	0.69	0.69	0.72	0.70	0.70	0.35	0.35	0.32	0.67	0.65	0.62
Iceland	1.52	1.52	1.52	1.05	1.05	1.05	0.83	0.83	0.84	1.37	1.37	1.37	1.35	1.35	1.35	1.11	1.14	1.17	0.71	0.70	0.70
Ireland	1.12	1.12	1.13	0.97	0.97	0.97	0.84	0.82	0.78	1.00	1.00	1.00	0.95	0.95	0.94	0.76	0.75	0.71	0.91	0.90	0.88
Italy	0.69	0.67	0.67	1.01	1.01	1.01	0.64	0.60	0.58	0.83	0.82	0.81	0.79	0.77	0.76	0.66	0.65	0.63	0.70	0.69	0.68
Luxemburg	0.48	0.46	0.46	0.95	0.95	0.94	0.48	0.45	0.42	0.74	0.74	0.74	0.64	0.64	0.63	0.71	0.72	0.71	0.68	0.67	0.66
Netherlands	0.78	0.76	0.75	0.88	0.87	0.87	0.66	0.62	0.60	0.89	0.89	0.89	0.79	0.78	0.77	0.89	0.89	0.88	0.85	0.85	0.84
Norway	1.10	1.12	1.13	1.03	1.04	1.05	0.85	0.83	0.81	0.96	0.95	0.96	0.93	0.92	0.93	0.98	0.98	0.98	0.67	0.65	0.65
Spain	0.92	0.92	0.92	1.05	1.05	1.05	0.66	0.61	0.59	0.96	0.96	0.96	0.93	0.92	0.92	0.48	0.44	0.44	0.82	0.81	0.80
Sweden	0.71	0.69	0.68	0.93	0.92	0.93	0.60	0.56	0.54	0.93	0.93	0.94	0.87	0.87	0.87	0.89	0.87	0.87	0.81	0.80	0.79
Switzerland	0.66	0.63	0.62	1.00	1.00	1.00	0.68	0.63	0.60	0.86	0.84	0.83	0.77	0.74	0.73	0.78	0.76	0.75	0.82	0.81	0.81

F. Three year mean PM$_{2.5}$ concentrations

Table 16 Three year mean PM$_{2.5}$ concentrations at 41 urban background sites

Site ID	Site name	Measured (µgm^{-3})	Modelled (µgm^{-3})	
		2009-2011	2009-2011	2019-2021
ABD	Aberdeen	7.5	6.1	3.2
BEL2	Belfast Centre	13.3	8.4	4.6
BIR1	Birmingham Tyburn	16.8	11.0	5.8
BLC2	Blackpool Marton	10.6	8.6	4.8
BRS8	Bristol St Paul's	14.1	11.0	6.5
CARD	Cardiff Centre	13.2	9.0	5.4
CHS6	Chesterfield	13.6	11.0	6.1
CLL2	London Bloomsbury	16.7	11.1	5.9
COV3	Coventry Memorial Park	11.8	10.8	5.7
DERY	Derry	13.9	5.9	3.7
EB	Eastbourne	13.3	9.6	5.1
ED3	Edinburgh St Leonards	9.9	6.4	3.2
GLA3	Glasgow Centre	11.4	6.7	3.7
HR3	London Harrow Stanmore	14.1	10.5	5.6
HUL2	Hull Freetown	12.1	11.8	6.1
KC1	London N. Kensington	14.7	10.9	5.9
LEAM	Leamington Spa	14.9	9.8	5.1
LEED	Leeds Centre	14.9	10.9	5.8
LEIC	Leicester Centre	15.0	10.1	5.1
LVP	Liverpool Speke	11.8	10.7	6.7
MAN3	Manchester Piccadilly	14.4	11.1	6.1
MID	Middlesbrough	9.9	10.0	5.0
NEWC	Newcastle Centre	11.1	7.9	4.2
NO12	Norwich Lakenfields	12.8	10.3	5.5
NOTT	Nottingham Centre	14.4	11.0	5.6
NPT3	Newport	14.5	9.4	5.4
OX8	Oxford St Ebbes	12.5	10.3	5.8
PLYM	Plymouth Centre	10.7	6.9	3.9
PMTH	Portsmouth	13.1	11.8	6.4
PRES	Preston	11.6	9.7	5.7
REA1	Reading New Town	13.2	10.6	6.0
SEND	Southend-on-Sea	13.3	12.1	6.4
SHE2	Sheffield Centre	15.7	10.7	5.6
SOUT	Southampton Centre	14.2	11.8	7.0
STOK	Stoke-on-Trent Centre	15.9	10.8	5.5
SUN2	Sunderland Silksworth	11.9	7.4	3.8
TED	London Teddington	15.0	10.7	5.9
TRAN	Wirral Tranmere	9.2	10.3	6.1
WAR	Warrington	12.7	11.5	7.3
WIG5	Wigan Centre	17.1	10.6	6.2
YK10	York Bootham	14.2	10.6	5.4
Average	**All sites AEI**	**13.2**	**9.9**	**5.4**

G. Supplementary maps and tables of comparative statistics between CMAQ, PCM and OSRM

Table 17: Maps of annual mean concentration of the sulphate component of PM10, CMAQ-UK and PCM compared. Note the difference in scale: the scale extends to 5.0 μg m-3 for the CMAQ-UK plots, but 2.0 μg m-3 for the PCM plots. The black numbers in the PCM maps indicate the location of measurement sites and the measured concentrations at the sites.
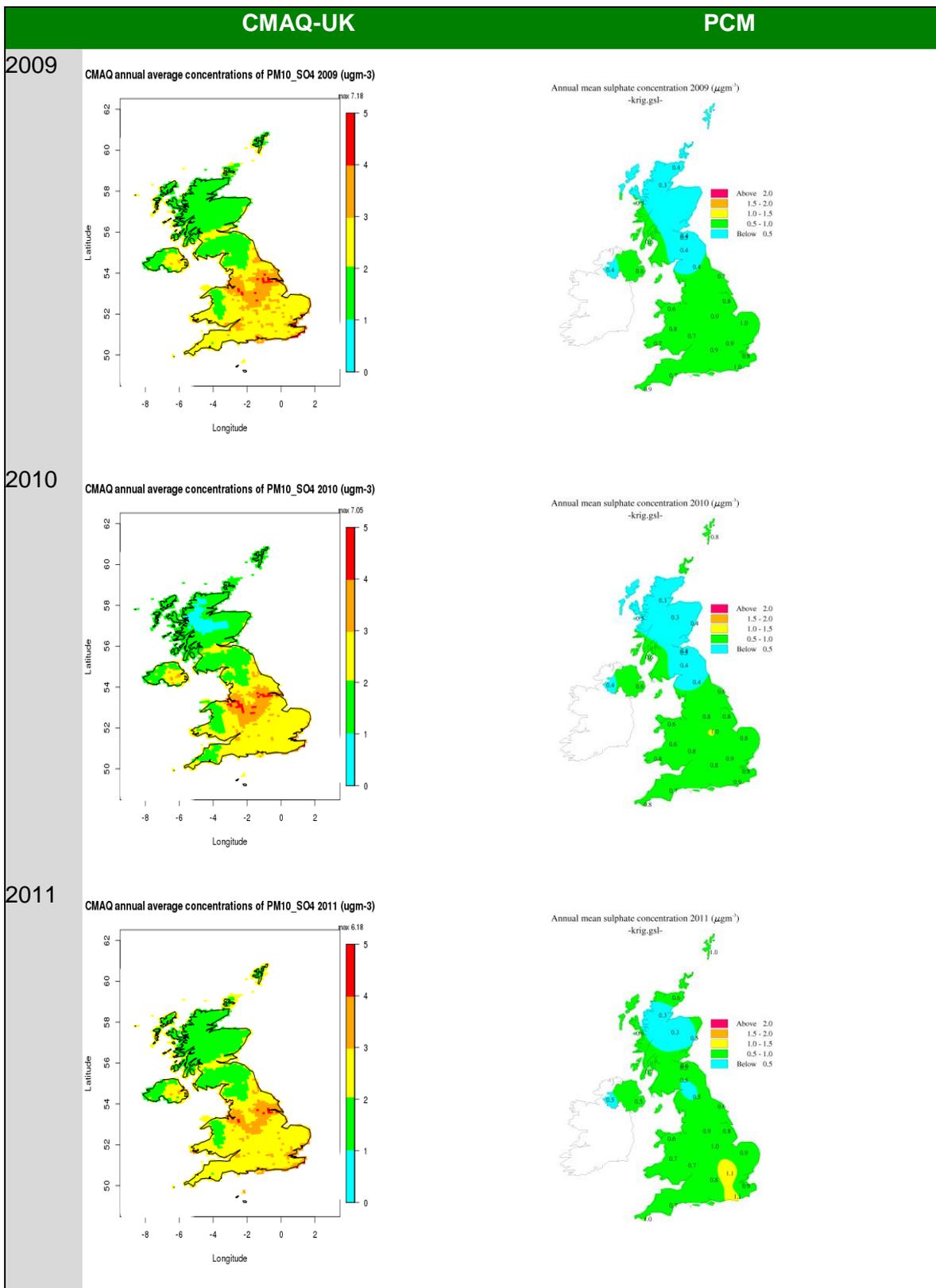
Table 18: Maps of annual mean concentration of the ammonium component of PM10, CMAQ-UK and PCM compared
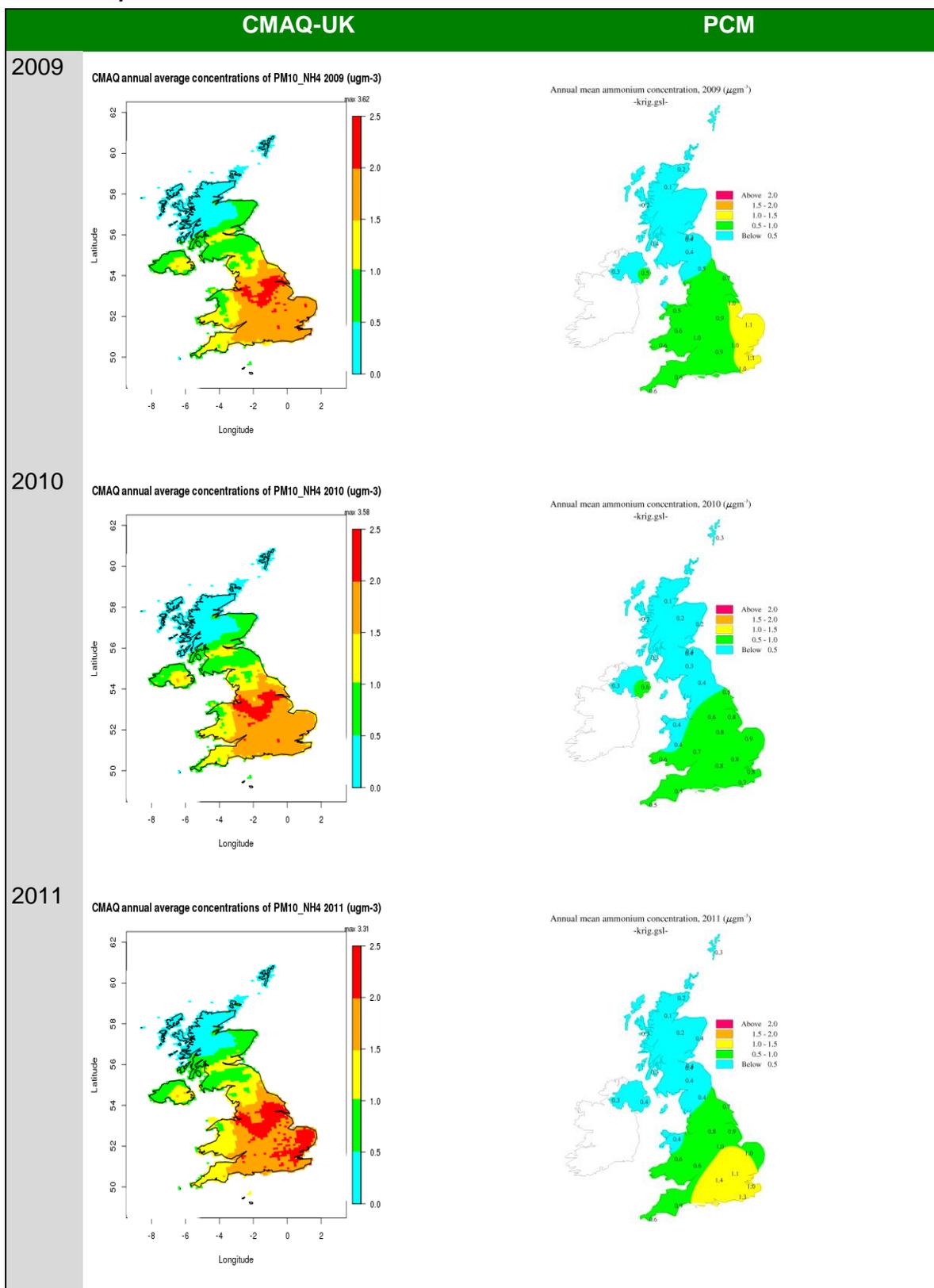
Table 19: Maps of annual mean concentration of the nitrate component of PM10, CMAQ-UK and PCM compared.
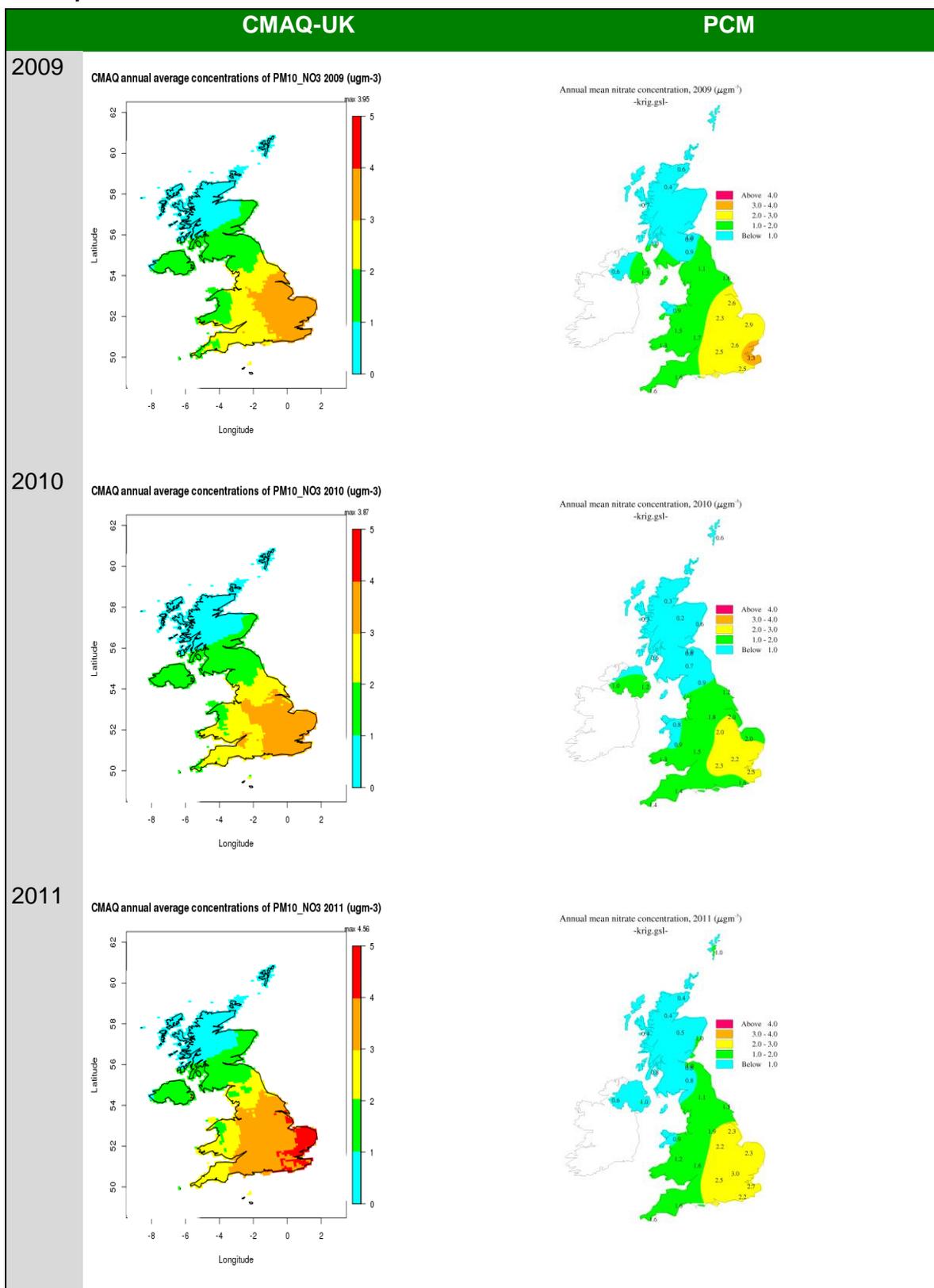
Table 20: Maps of annual mean concentration of the chloride component of PM10, CMAQ-UK and PCM compared.
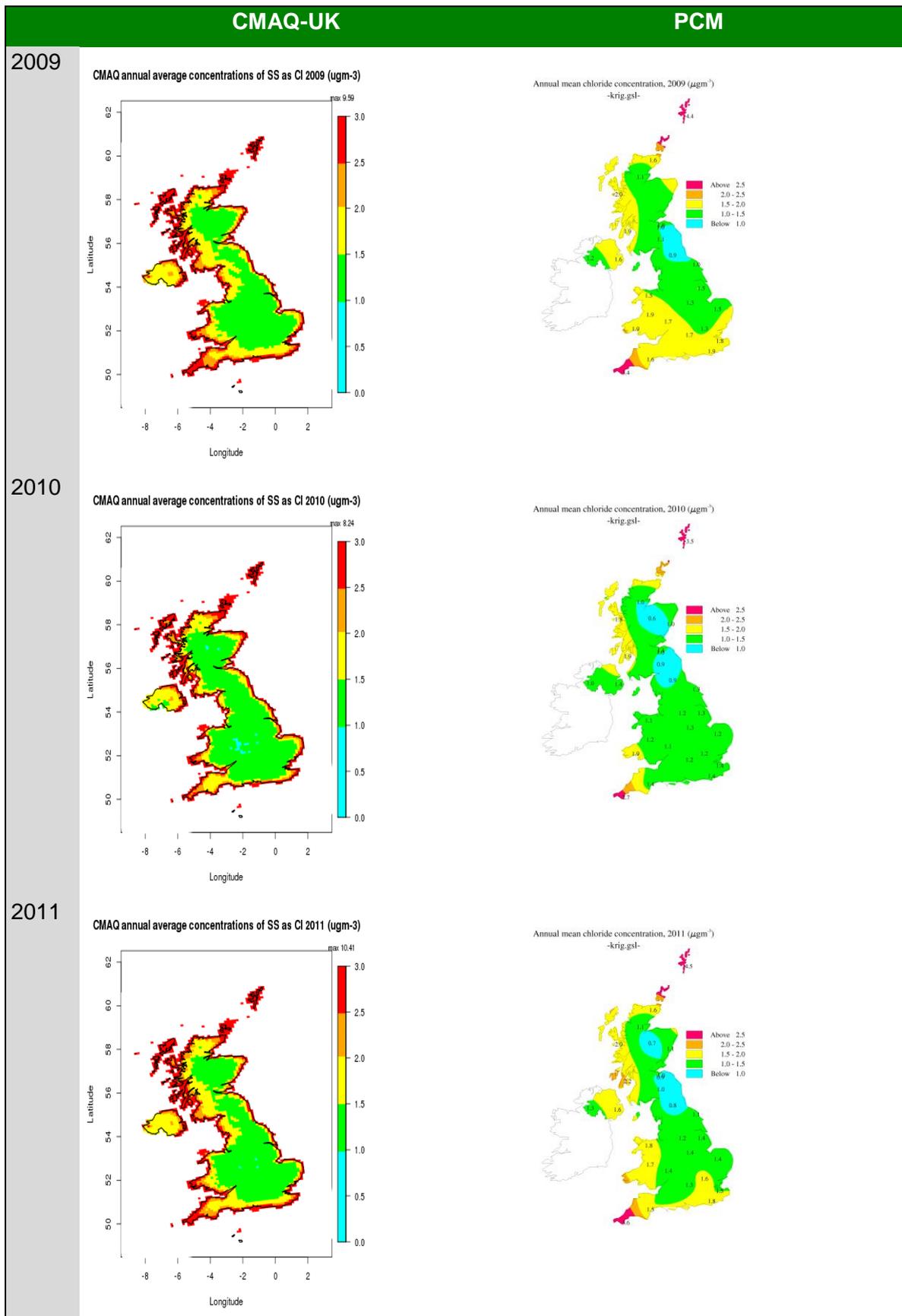
Table 21: Maps of SO2 annual mean concentrations from CMAQ-UK and PCM. In the PCM maps an ecosystems and vegetation mask has been applied (see text for details.
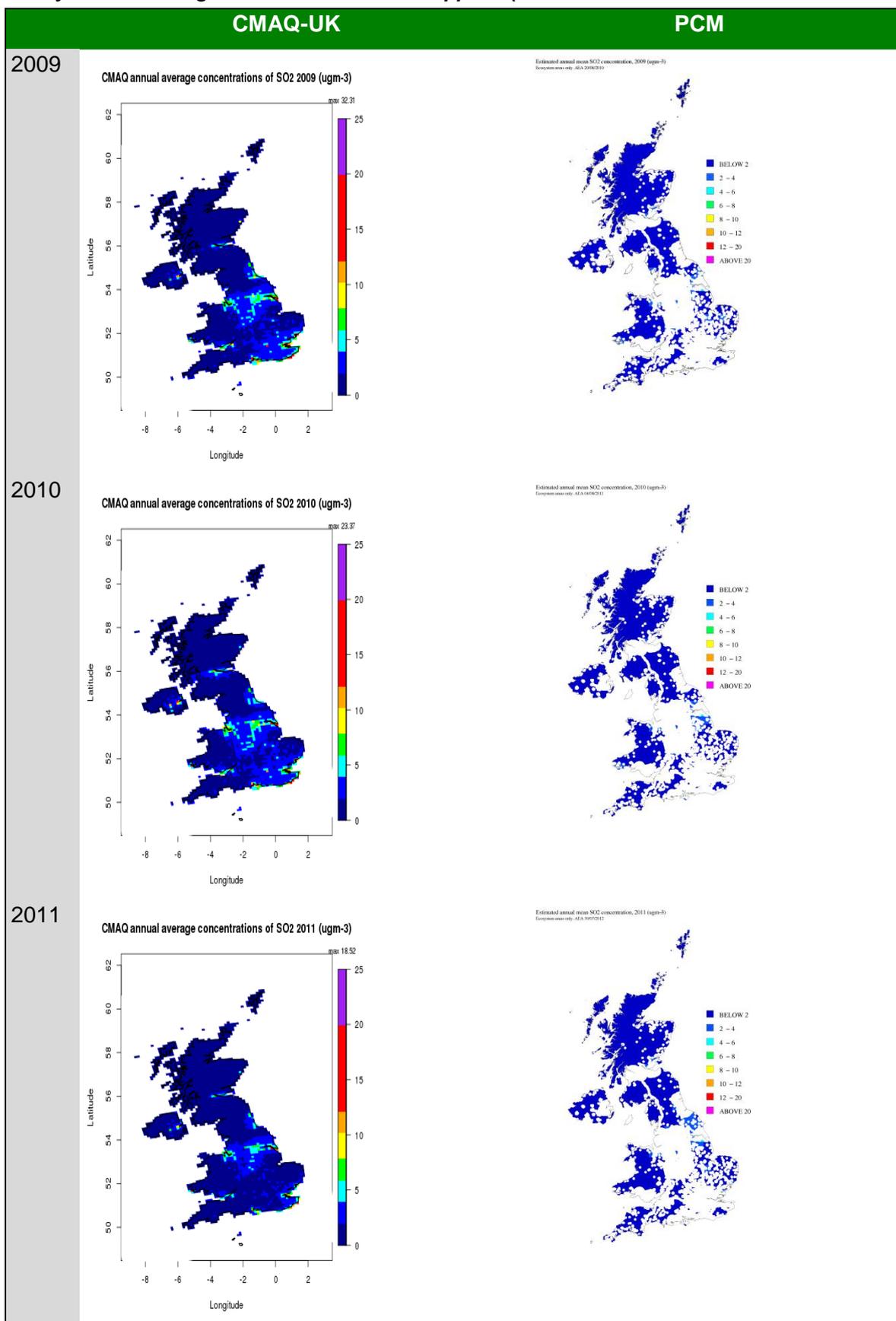
Table 22: Maps of 99.18 percentile of 24-hour mean SO2 concentrations from CMAQ-UK and PCM.

CMAQ-UK	PCM
2009	

| **2010** | |

| **2011** | |
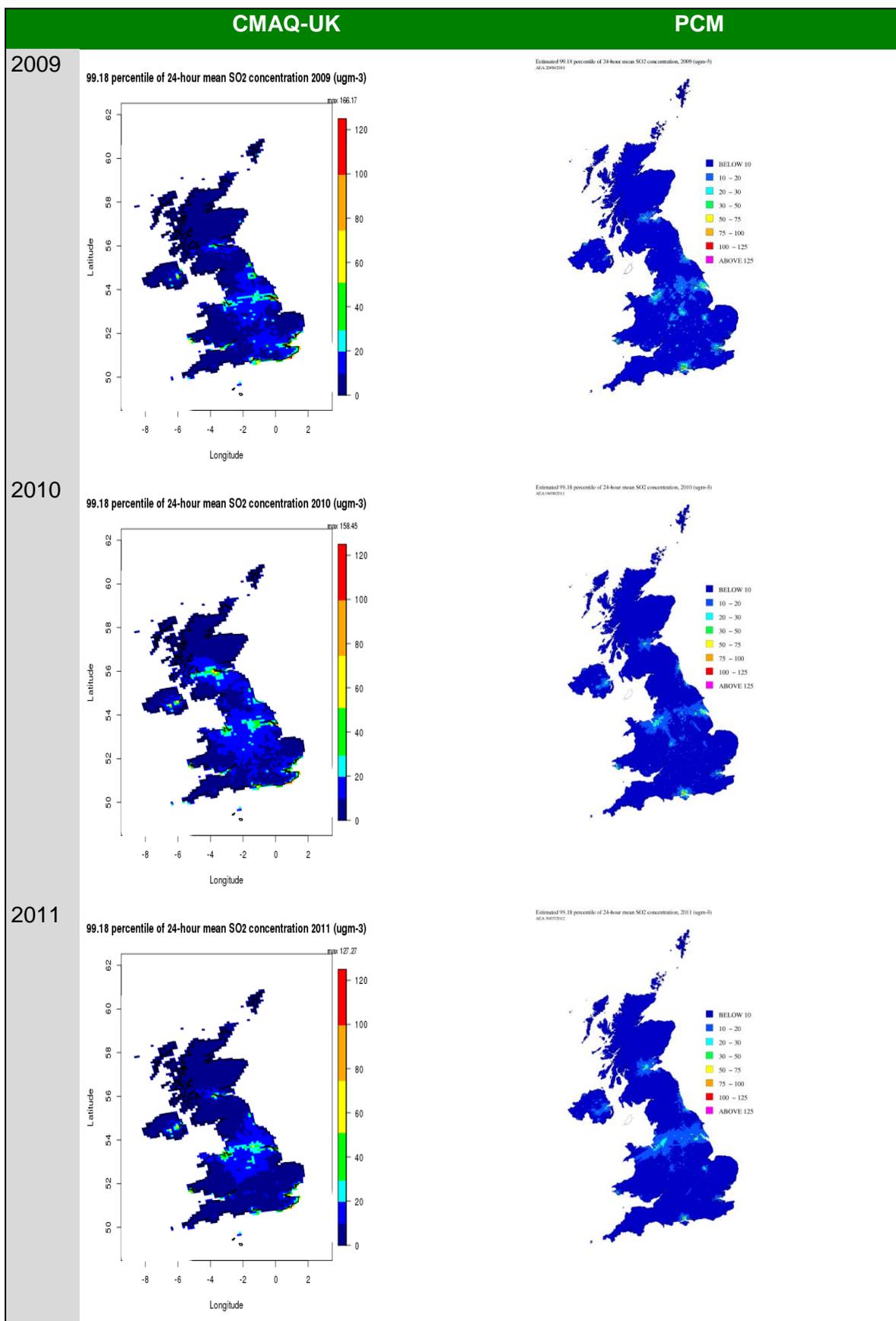
H. Comparative statistics between CMAQ, PCM and OSRM

Ozone metrics

Table 23: OSRM verification summary – AOT40 metrics

OSRM AOT 40 metric	Year	Mean of measurements (ugm^{-3}.hours)	Mean of model estimates (µgm^{-3}.hours)	R^2	% outside DQO	No. sites used in assessment
National network	2009	3182	4274	0.12	36	67
Verification sites	2009	2738	3818	0.03	70	20
National network	2010	2244	4404	0.11	61	57
Verification sites	2010	2518	4150	0.17	50	12
National network	2011	2333	2171	0.20	42	72
Verification sites	2011	2627	2158	0.21	23	13

Table 24: PCM empirical model verification summary – AOT40 metric

PCM AOT 40 metric	Year	Mean of measurements (ugm^{-3}.hours)	Mean of model estimates (µgm^{-3}.hours)	R^2	% outside DQO	No. sites used in assessment
National network	2009	3182	3262	0.60	24	67
Verification sites	2009	2738	3463	0.21	45	20
National network	2010	2244	2620	0.41	25	57
Verification sites	2010	2518	2619	0.02	50	12
National network	2011	2333	2294	0.35	32	72
Verification sites	2011	2627	2339	0.03	54	13

Table 25: CMAQ-UK model verification summary – AOT40 metric

CMAQ-UK AOT 40 metric	Year	Mean of measurements (ugm^{-3}.hours)	Mean of model estimates (µgm^{-3}.hours)	R^2	% outside DQO	No. sites used in assessment
National network	2009	3182	4554	0.09	55	67
Verification sites	2009	2738	5358	0.01	65	20
National network	2010	2244	3383	0.22	49	57
Verification sites	2010	2518	3552	0.04	50	12
National network	2011	2333	3782	0.20	60	72
Verification sites	2011	2627	5154	0.04	54	13

Table 26: OSRM verification summary – number of days exceeding 120 µgm-3.

OSRM DGT120 metric	Year	Mean of measurements (days)	Mean of model estimates (days)	R^2	% outside DQO	No. sites used in assessment
National network	2009	1.3	4.8	0.18	71	72
Verification sites	2009	1.1	4.5	0.00	91	11
National network	2010	0.9	3.7	0.05	74	77
Verification sites	2010	1.5	5.1	0.09	92	13
National network	2011	2.6	2.4	0.09	62	73
Verification sites	2011	3.2	1.8	0.30	54	13

Table 27: PCM verification summary – number of days exceeding 120 µgm-3.

PCM DGT120 metric	Year	Mean of measurements (days)	Mean of model estimates (days)	R^2	% outside DQO	No. sites used in assessment
National network	2009	1.3	1.5	0.53	33	72
Verification sites	2009	1.1	1.7	0.14	91	11
National network	2010	0.9	1.1	0.47	34	77
Verification sites	2010	1.5	0.8	0.01	57	14
National network	2011	2.6	2.6	0.24	42	73
Verification sites	2011	3.2	2.6	0.04	77	13

Table 28: CMAQ-UK verification summary – number of days exceeding 120 µgm-3.

CMAQ-UK DGT120 metric	Year	Mean of measurements (days)	Mean of model estimates (days)	R^2	% outside DQO	No. sites used in assessment
National network	2009	1.3	1.2	0.002	54	72
Verification sites	2009	1.1	1.1	0.01	73	11
National network	2010	0.9	0.8	0.01	47	77
Verification sites	2010	1.5	0.5	0.01	57	14
National network	2011	2.6	1.1	0.06	60	73
Verification sites	2011	3.2	1.2	0.09	77	13

PM$_{10}$ Metrics

Table 29: PCM verification summary – average annual mean PM10.

PCM Mean PM10	Year	Mean of measurements (µg.m^{-3})	Mean of model estimates (µg.m^{-3})	R^2	% outside DQO	No. sites used in assessment
National network	2009	18.4	18.1	0.13	0	21
Verification sites	2009	19.5	17.2	0.04	0	8
National network	2010	18.0	17.7	0.21	0	22
Verification sites	2010	19.1	16.5	0.29	0	10
National network	2011	19.3	18.3	0.30	0	27
Verification sites	2011	19.4	18.0	0.45	0	13

Table 30: CMAQ-UK verification summary – average annual mean PM10.

CMAQ-UK Mean PM10	Year	Mean of measurements (µg.m^{-3})	Mean of model estimates (µg.m^{-3})	R^2	% outside DQO	No. sites used in assessment
National network	2009	18.4	14.5	0.03	0	21
Verification sites	2009	15.4	9.4	0.27	20	5
National network	2010	18.0	13.3	0.11	0	22
Verification sites	2010	19.1	13.0	0.04	0	10
National network	2011	19.3	13.9	0.38	0	27
Verification sites	2011	19.4	13.9	0.26	0	13

PM_{2.5} Metrics

Table 31: PCM verification summary – average annual mean PM2.5.

PCM Mean PM2.5	Year	Mean of measurements ($\mu g.m^{-3}$)	Mean of model estimates ($\mu g.m^{-3}$)	R^2	% outside DQO	No. sites used in assessment
National network	2009	12.9	12.6	0.23	0	21
Verification sites	2009	13.6	12.4	0.02	0	5
National network	2010	13.0	12.7	0.27	5	22
Verification sites	2010	13.6	12.4	0.02	0	5
National network	2011	13.0	12.7	0.55	4	27
Verification sites	2011	14.1	13.1	0.66	0	5

Table 32: CMAQ-UK verification summary – average annual mean PM2.5.

CMAQ-UK Mean PM2.5	Year	Mean of measurements ($\mu g.m^{-3}$)	Mean of model estimates ($\mu g.m^{-3}$)	R^2	% outside DQO	No. sites used in assessment
National network	2009	12.9	10.4	0.18	0	21
Verification sites	2009	13.6	11.8	0.67	0	5
National network	2010	13.0	9.6	0.43	0	22
Verification sites	2010	13.6	11.8	0.67	0	5
National network	2011	13.0	10.0	0.60	0	27
Verification sites	2011	14.1	10.3	0.48	0	5

(FDMS sites included in the national network sites only).

SO₂ Metrics

Table 33: CMAQ-UK verification summary – average annual mean SO2.

CMAQ-UK Mean SO2	Year	Mean of measurements ($\mu g.m^{-3}$)	Mean of model estimates ($\mu g.m^{-3}$)	R^2	% outside DQO	No. sites used in assessment
National network	2009	2.0	3.3	0.26	58	65
Verification sites	2009	4.1	6.2	0.38	56	39
National network	2010	2.4	3.1	0.48	59	73
Verification sites	2010	3.8	5.2	0.15	56	41
National network	2011	1.9	2.4	0.19	54	72
Verification sites	2011	3.5	3.2	0.00	62	37

Table 34: PCM verification summary – average annual mean SO2.

PCM Mean SO2	Year	Mean of measurements ($\mu g.m^{-3}$)	Mean of model estimates ($\mu g.m^{-3}$)	R^2	% outside DQO	No. sites used in assessment
National network	2009	1.0	1.3	0.28	64	67
Verification sites	2009	3.5	4.1	0.03	70	40
National network	2010	2.4	2.4	0.28	68	73
Verification sites	2010	3.9	3.0	0.03	71	41
National network	2011	1.9	2.2	0.22	69	72
Verification sites	2011	3.5	2.4	0.00	73	37

Table 35: CMAQ-UK verification summary – 99.18 percentile of 24-hr mean SO2.

CMAQ-UK 99.73 percentile of 1-hr mean	Year	Mean of measurements (μg.m⁻³)	Mean of model estimates (μg.m⁻³)	R^2	% outside DQO	No. sites used in assessment
National network	2009	19.5	20.9	0.13	32	37
Verification sites	2009	21.6	29.9	0.28	49	43
National network	2010	14.8	15.4	0.37	40	43
Verification sites	2010	23.5	28.8	0.43	40	41
National network	2011	10.7	15.8	0.17	57	44
Verification sites	2011	17.4	17.1	0.08	58	31

Table 36: PCM verification summary – 99.18 percentile of 24-hr mean SO2.

PCM 99.73 percentile of 1-hr mean	Year	Mean of measurements (μg.m⁻³)	Mean of model estimates (μg.m⁻³)	R^2	% outside DQO	No. sites used in assessment
National network	2009	12.5	10.3	0.19	33	39
Verification sites	2009	21.1	17.8	0.12	57	44
National network	2010	14.8	13.6	0.21	40	43
Verification sites	2010	23.5	16.3	0.07	41	41
National network	2011	10.7	11.6	0.19	36	44
Verification sites	2011	17.4	13.4	0.19	26	31

Table 37: CMAQ-UK verification summary – 99.73 percentile of 1-hr mean SO2.

CMAQ-UK 99.73 percentile of 1-hr mean	Year	Mean of measurements (μg.m⁻³)	Mean of model estimates (μg.m⁻³)	R^2	% outside DQO	No. sites used in assessment
National network	2009	29.2	45.2	0.29	57	37
Verification sites	2009	46.7	62.4	0.35	60	43
National network	2010	33.1	40.4	0.73	53	43
Verification sites	2010	45.5	62.8	0.61	49	41
National network	2011	16.6	31.9	0.40	77	44
Verification sites	2011	39.5	34.9	0.31	39	31

Table 38: PCM verification summary – 99.73 percentile of 1-hr meanSO2.

PCM 99.73 percentile of 1-hr mean	Year	Mean of measurements (μg.m⁻³)	Mean of model estimates (μg.m⁻³)	R^2	% outside DQO	No. sites used in assessment
National network	2009	18.4	26.8	0.10	44	39
Verification sites	2009	39.4	44.0	0.23	52	44
National network	2010	33.1	34.4	0.20	44	43
Verification sites	2010	45.5	44.9	0.14	46	41
National network	2011	16.6	29.3	0.30	68	44
Verification sites	2011	39.5	37.1	0.43	32	31

NO/NOx Metrics

Table 39: PCM verification summary – average annual mean NOx.

PCM annual mean NOX	Year	Mean of measurements (µg.m⁻³)	Mean of model estimates (µg.m⁻³)	R²	% outside DQO	No. sites used in assessment
National network	2009	36.9	35.8	0.77	16.9	65
Verification sites	2009	44.6	41.7	0.79	29.3	92
National network	2010	42.8	40.7	0.73	22.4	67
Verification sites	2010	50.8	50.0	0.61	39.2	74
National network	2011	32.9	32.0	0.79	25.0	71
Verification sites	2011	46.8	41.8	0.48	38.8	67

Table 40: CMAQ-UK verification summary – average annual mean NOx.

CMAQ-UK annual mean NOX	Year	Mean of measurements (µg.m⁻³)	Mean of model estimates (µg.m⁻³)	R²	% outside DQO	No. sites used in assessment
National network	2009	36.9	34.2	0.32	26.2	65
Verification sites	2009	44.6	45.3	0.25	27.5	80
National network	2010	42.8	33.0	0.30	29.9	67
Verification sites	2010	50.8	43.4	0.17	25.7	74
National network	2011	32.9	28.8	0.32	19.7	71
Verification sites	2011	46.8	37.2	0.24	26.9	67

Table 41: PCM verification summary – average annual mean NO2.

PCM annual mean NOX	Year	Mean of measurements (µg.m⁻³)	Mean of model estimates (µg.m⁻³)	R²	% outside DQO	No. sites used in assessment
National network	2009	22.6	22.1	0.82	12.5	64
Verification sites	2009	28.6	25.3	0.67	20.7	92
National network	2010	25.1	24.2	0.80	13.4	67
Verification sites	2010	29.2	28.4	0.75	18.9	74
National network	2011	21.4	21.1	0.82	11.3	71
Verification sites	2011	27.8	25.9	0.61	20.9	67

Table 42: CMAQ-UK verification summary – average annual mean NO2.

CMAQ-UK annual mean NOX	Year	Mean of measurements (µg.m⁻³)	Mean of model estimates (µg.m⁻³)	R²	% outside DQO	No. sites used in assessment
National network	2009	22.6	23.2	0.50	14.1	64
Verification sites	2009	28.6	28.9	0.42	10.9	92
National network	2010	25.1	23.6	0.51	11.9	67
Verification sites	2010	29.2	28.7	0.41	10.8	74
National network	2011	21.4	21.6	0.51	15.5	71
Verification sites	2011	27.8	26.3	0.40	13.4	67

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