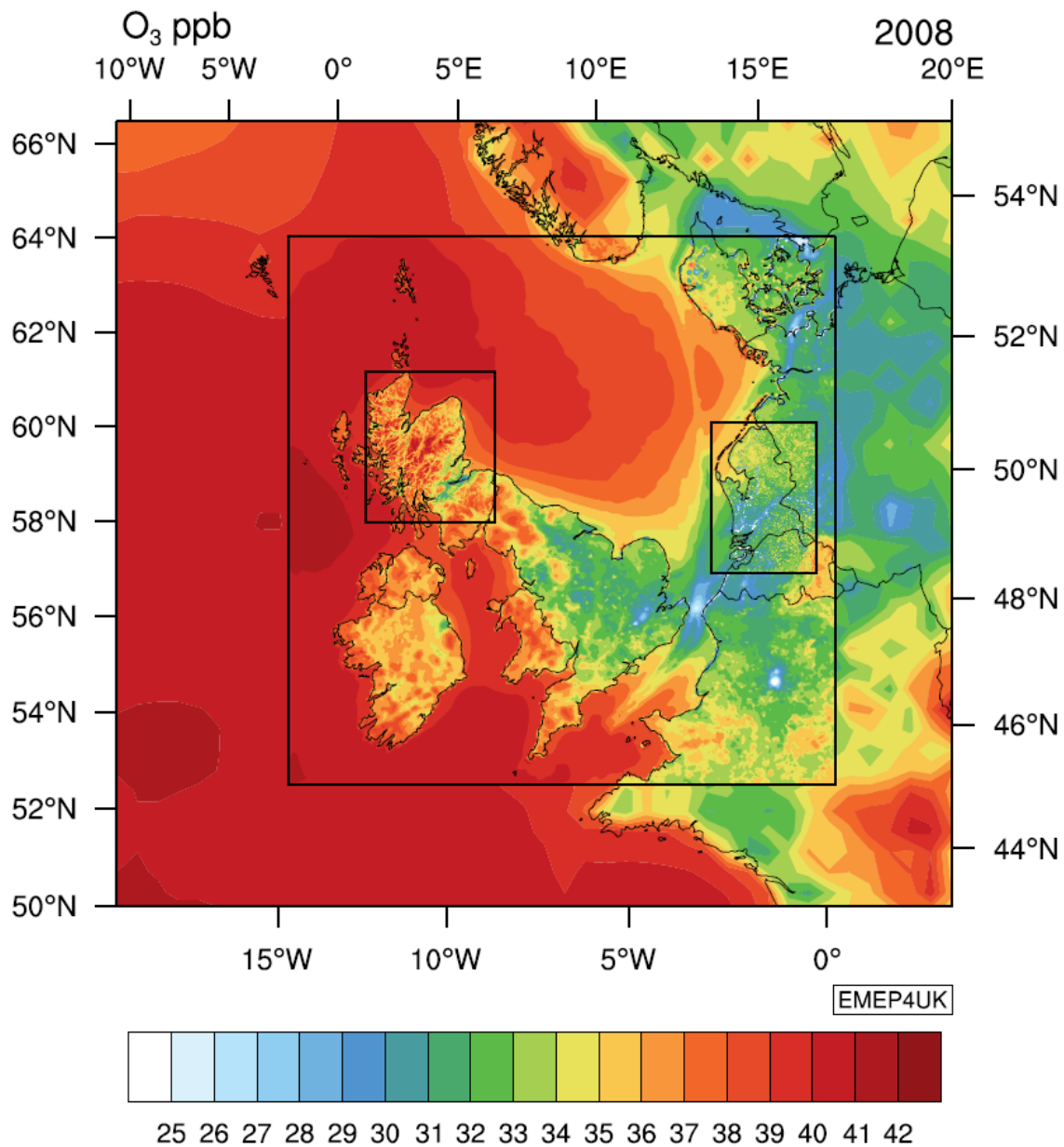


Further Development of the UK Application of the EMEP model (EMEP4UK) 2011-2014 – AQ0727



Massimo Vieno, Stefan Reis, Tony Dore, Riinu Ots, Rachel Beck, and Mark Sutton

NERC, Centre for Ecology and Hydrology (Edinburgh Research Station)

Bush Estate, Penicuik, Midlothian, EH26 0QB, UK.

Client: DEFRA	
Client Project number: AQ0727	CEH Project number: NEC04822
Project Title: Further Development of the UK Application of the EMEP model (EMEP4UK) 2011-2014	
Start date: 01/12/2011	Completion date: 30/11/2014
Client Project Officers: Daniel Waterman	
CEH Project Officer: Massimo Vieno	
Main authors: Massimo Vieno, Stefan Reis, Tony Dore, Riinu Ots, Rachel Beck, and Mark Sutton	
Reporting period: Final project report	
Report date: 01/04/2015	
Report Number: 03	

This report is a confidential document prepared under contract between Defra and the Natural Environment Research Council (NERC). It should not be distributed or quoted without the permission of the Centre for Ecology & Hydrology (CEH).

1. Introduction

The Defra and CEH partnership-funded project EMEP4UK (Defra AQ0702 – 2007-2011) developed and delivered an off-line atmospheric-chemistry transport model (ACTM) based on the EMEP MSC-W model, using the Weather Research Forecast (WRF) model as the main meteorological driver. The model, termed EMEP4UK, is able to represent UK atmospheric composition in greater detail than European-scale models, with a capability to simulate hourly air pollution interactions over decadal time scales using a 5 km grid or finer.

The current version of the EMEP4UK is rv4.4 (Vieno et al., 2010; Vieno et al., 2014) and it is based on the EMEP MSC-W rv4.4 model, for which the model code is open source and available for download from the EMEP web site¹ (Simpson et al., 2012). The EMEP MSC-W model is currently used by the UNECE Convention on Long-range Transboundary Air Pollution (CRLTAP) and the European Commission to support European policy development, e.g. for modelling surface concentrations of atmospheric pollutants such as oxidised nitrogen, sulphur dioxide, particulate matter and ozone. Dry and wet deposition of sulphates, oxidised and reduced nitrogen is routinely calculated by the EMEP4UK model. The EMEP4UK is an ideal tool to analyse the impact of policies on the UK with the benefit of higher resolution which is, for example, critical to account for the spatial allocation of wet deposition (Fournier et al., 2004).

This document covers the results and deliverables of the EMEP4UK project. Key results and policy applications are presented here.

¹ <http://www.emep.int>

2. EMPE4UK model description

2.1. EMEP4UK atmospheric chemistry transport model description.

The EMEP4UK model framework consists of an atmospheric chemistry transport model (ACTM) which simulates hourly to annual average atmospheric composition and deposition of various pollutants; including PM₁₀, PM_{2.5}, secondary organic aerosols (SOA), elemental carbon (EC), secondary inorganic aerosols (SIA), SO₂, NH₃, NO_x, and O₃, and the weather research and forecast model (WRF) which is used to calculate the required meteorological input data for the ACTM (Vieno et al., 2009; Vieno et al., 2010; Vieno et al., 2014). Dry and wet depositions of pollutants are also calculated by the model.

EMEP4UK operates at horizontal resolutions ranging from 50 to 1 km x 1 km covering the UK and Europe in a one-way nesting approach. The default vertical domain ranges from ~90 m (thickness of the first layer at the surface) up to ~16 km (at the top of the vertical domain), however since version rv4.4 of the EMEP4UK model, setting flexible vertical resolution is possible. A typical model domain setup is shown in **Figure 1**; the European domain is modelled at a horizontal resolution of 50 km x 50 km (133 x 171 grid cells) and the British Isles domain at a horizontal resolution of 5 km x 5 km (220 x 270 grid cells), however, the model setup is very flexible and can be changed for the need of scientific and/or policy applications. As an example the domain setup used in the EU FP7 project ECLAIRE² is shown in **Figure 1**, with 1 km x 1 km resolution applied over central Scotland and The Netherlands to investigate detailed spatial aspects of air pollution concentrations and deposition in relation to ecosystem effects.

All the results and analyses included in this report are based on EMEP4UK rv4.4, however, the model has since been further updated to a more recent rv4.6 version, which has been released after the project funding period was completed and also includes a thinnest lower surface layer of 50 m instead of the previous default of 90 m.

The default chemical scheme is the *EmChem09* (Simpson et al., 2012) as it has been extensively validated at the European scale (<http://www.emep.int>). It has 72 species and 137 reactions. Full details of the chemical scheme are given by Simpson et al. (2012). Other chemical schemes are available for the EMEP4UK model and more details can be found in Simpson et al. (2012). The *Common Representative Intermediates* (CRI) (Watson et al., 2008) chemical mechanism has been successfully applied in the EMEP4UK model and the

² <http://www.eclair-fp7.eu>

results published in (von Schneidemesser et al., 2014). Moreover, an ad-hoc EMEP4UK-CRI model run has also been submitted to the Defra Model Inter-comparison Exercise (MIE).

Anthropogenic emissions of NO_x , NH_3 , SO_2 , primary $\text{PM}_{2.5}$, primary $\text{PM}_{\text{coarse}}$ (the difference between PM_{10} and $\text{PM}_{2.5}$), CO , and non-methane volatile organic compounds (NMVOC) are included from different sources at respective spatial resolutions. For the UK, emissions values are taken from the National Atmospheric Emission Inventory (NAEI³) at 1 km x 1 km resolution and aggregated to 5 km x 5 km resolution. For the rest of the European domain, the model uses the EMEP 50 km x 50 km resolution emission estimates provided by the EMEP Centre for Emission Inventories and Projections (CEIP⁴). Emissions estimates for international shipping (ENTEC, 2010) are aggregated to 5 km x 5 km for those emissions within the British Isles domain. Forest fires are also included in the model, as well as biogenic emissions of isoprene and (when required) monoterpenes, which are calculated in the model for every grid-cell, and at every model time-step, using near-surface air temperature.

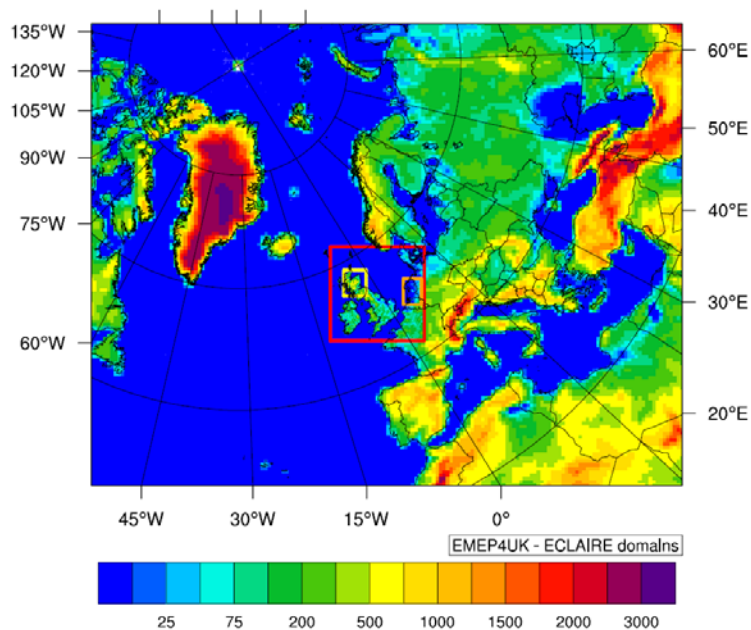


Figure 1. EMEP4UK model domain as used for the ECLAIRE project. The largest domain has a horizontal resolution of 50 km x 50 km, a nested 5 km x 5 km domain covering the British Isles, part of France, The Netherlands, Belgium (Red box), and two nested domains at 1km² horizontal resolution covering most of Scotland (yellow box) and the Netherlands (orange box) for detailed analyses.

³ <http://naei.defra.gov.uk>

⁴ <http://www.ceip.at/>

2.2. Weather and Research Forecast model (WRF) as meteorological driver.

The WRF model version 3 is the default meteorological driver for the EMEP4UK model; however, other meteorology may be used, for example European Centre for Medium-Range Weather Forecast (ECMWF⁵) meteorology is compatible with the EMEP4UK model. The WRF model output consists of a 3-hourly (resolution only limited by available disk space) instantaneous value. The WRF domain is matched to the EMEP4UK model in both horizontal and vertical dimensions. The WRF model version 3.6.1 is used as the default meteorological driver for EMEP4UK model versions rv4.4 or newer.

The WRF model setup is described in detail here: www.wrf-model.org. A summary of the main physical schemes used specifically for the EMEP4UK is show below.

- Microphysics: Lin Purdue
- Cumulus Parameterization: Grell-3
- Radiation Physics: Goddard Chou and Suarez
- PBL Physics: YSU
- Nudging: Re-analysis nudged for wind, temperature (and specific humidity 2001-2012)

The nudging of the WRF model with reanalysis data has been currently reviewed for the EMEP4UK model. The WRF model runs done for the year 2013 (and newer) do not include the specific humidity nudging. This was the result of the collaboration between CEH and the University of Edinburgh under the NERC AWSOME project (Heal et al., 2013). It has been found that nudging the specific humidity led to a substantial underestimation of surface rainfall. The surface rainfall for the year 2001-2012, where the specific humidity was nudged, has been normalised to match the UK annual average for each year to avoid bias in rainfall affecting wet deposition results.

The WRF meteorological and EMEP4UK atmospheric chemistry data are available on request; however, each calculated year requires ~500 GB of disk space. Currently no automatic download facility via web access is provided due to the large data volume, however the data can be easily shared using physical means such as portable hard disks. CEH is currently exploring ways to make all large EMEP4UK and WRF generated datasets web-

⁵ <http://www.ecmwf.int/>

accessible, utilising THREDDS server technology and the CEH Environmental Informatics programme capabilities to enable users to visualise, explore and download.

3. EMEP4UK A0727 project deliverables

This document covers the deliverables of the AQ0727 “Further Development of the UK Application of the EMEP model (EMEP4UK) 2011-2014”. Originally, only four simple emission scenario runs had been planned for policy assessment (basically assessing the effect of SNAP sector-level emissions perturbations). However, during the runtime of the project, a series of additional and more complex scenarios runs were required by Defra and conducted in close communication with policy officers (Dr Daniel Waterman and Dr Peter Coleman) and in support of the Defra Air Quality Expert Group (AQEG) PM_{2.5} report:

- Support for an AQEG review of policy options to reduce PM_{2.5} concentrations over the UK (7 runs).
- Analysis of future shipping emissions scenarios for the year 2020 (4 runs).
- Extension of the critical load analysis for the Defra MIE (3 runs).
- Analysis of episode with elevated PM₁₀ during the spring of 2014 (2 runs).
- Analysis of emission scenarios for 2030 NECD and CLE2030 (3 runs).

For all these additional model assessment runs, which required input data preparation, setting up and conducting the model runs and post-processing/data analysis, the required time has been re-allocated from other tasks in agreement with Defra (this specifically affected Tasks 6 and 10).

3.1. Evaluation of how UK atmospheric composition is affected by shipping (Task 1).

Shipping emissions contribute significantly to some of the pollutants which degrade UK air quality, particularly SO₂. Shipping emission effects occur around the whole of the coastline of the UK, but are a dominant source in the south and east of England. The MARPOL Annex VI regulation on marine pollution came in to force in 2005, designating the North Sea and English Channel as a Sulphur Emissions Control Area (SECA) within which the sulphur content of bunker fuel used for international shipping should not exceed 1.5% by mass. Shipping activities in these areas were required to use low sulphur fuel from November 2007 onwards. In 2008 an amendment to MARPOL Annex VI was adopted by the International Marine Organisation (IMO) requiring further reductions in sulphur fuel content for shipping to 1% by 2010 and 0.1% by 2020 in the same SECAs.

This study used the EMEP4UK model to address the contribution of shipping emissions to the UK SO₂ and NO₂ surface concentrations. As discussed with Defra the focus of this work was to identify suitable areas where the shipping contribution to surface concentrations of SO₂ and NO_x is sufficiently significant that a decrease in shipping emissions would be observable in measurements, allowing observation and attribution of any future changes.

An EMEP4UK model experiment has been performed to simulate the consequences of a 30% reduction in SO₂ and NO_x emissions from shipping relative to other land-based sources of these pollutants. The model was then used to show the effect on UK air concentrations of SO₂ and NO₂, relative to the reference situation in 2008. The results were assessed in the context of the validity of the model, where the modelled air concentrations of SO₂ are compared with measurements at two southern English sites, which are strongly influenced by shipping emissions.

A reduction of 30% of shipping emissions had the effect of reducing surface SO₂ concentrations at all coastal areas around the UK, on average by approximately 10%, with up to 18% in some areas of the south and east of England. Overall the reduction in shipping emissions caused small decreases across most of the UK (**Figure 2** – right panel). The reductions in shipping emissions also led to reductions in NO₂ concentrations, with values of up to 12% reduction for NO₂ (not shown here) again in south and east England. The effect across the whole of the UK landmass for NO₂ is smaller, due to the relatively larger contribution of road transport sources to ground level NO₂ concentrations compared to those from marine sources.

The purple column in **Figure 3** shows the percentage change of SO₂ surface concentrations at the Automatic Urban Rural Network AURN (rural and sub-urban) and Acid Gas and Aerosol Network (AGANet Conolly et al. (2011)) monitoring sites estimated by the EMEP4UK model when shipping emissions are reduced by 30%, respectively with the sites being ranked for latitude. **Figure 3** shows an increase of shipping influence to SO₂ surface concentration as a function of latitude as sites closest to the English Channel are subject to a larger influence from shipping.

A short report “Long term modelling of shipping emissions in the UK as a basis to select a possible location for monitoring sites” with all details of this analysis was produced for Defra.

3.2. Participation in Defra's modelling inter-comparison exercise (Task 1).

A set of 4 perturbation runs have been carried out to provide the EMEP4UK data for phase 2 of the Defra model inter comparison.

- Reduce total anthropogenic NO_x and VOC by 30% across the UK + Europe
- Reduce total anthropogenic NO_x and VOC by 30% across the UK only
- Reduce anthropogenic NO_x by 30% across UK + Europe
- Reduce anthropogenic VOC by 30% across UK + Europe

The full set of emissions reduction scenario simulations have been delivered to David Carslaw to be analysed.

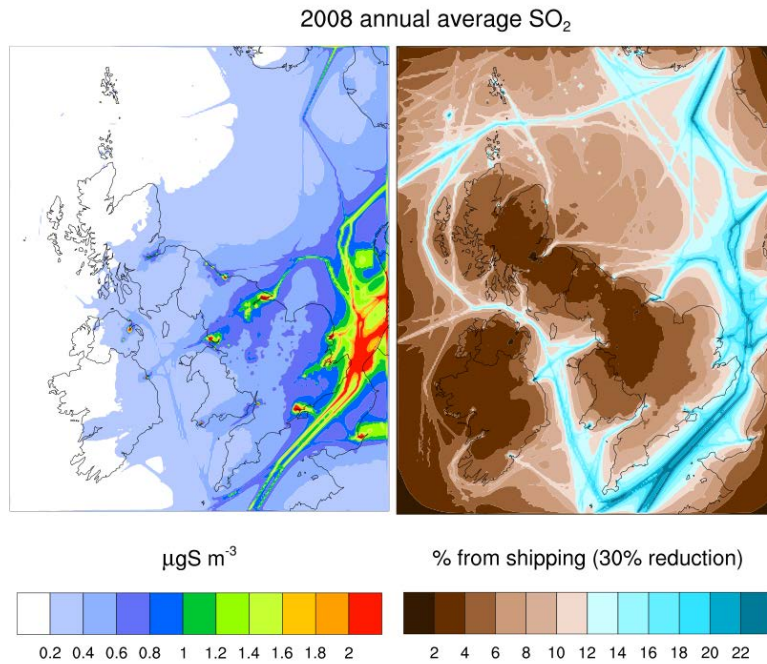


Figure 2. EMEP4UK surface concentration of SO₂-S for the year 2008 (left panel) and percentage of reduction (right panel) of surface SO₂ concentration due to a 30% emissions reduction in shipping-emitted species.

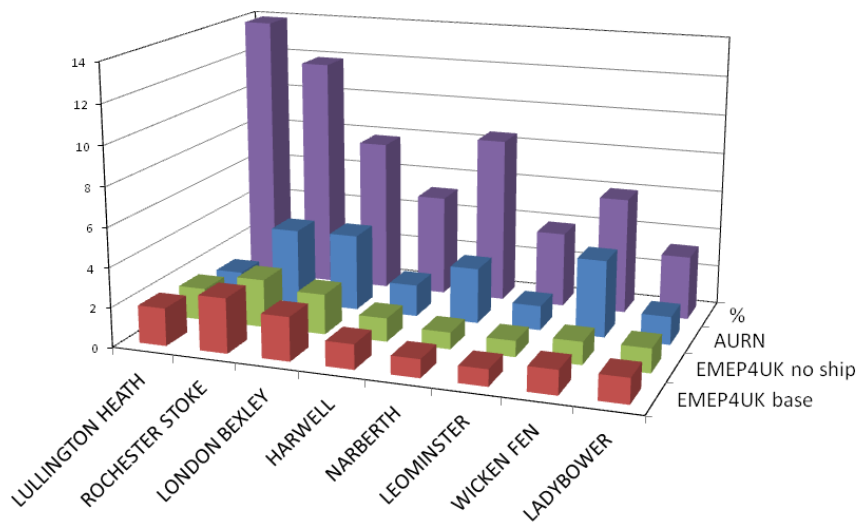


Figure 3. Annual average surface SO₂ concentrations in 2008 for all available rural and sub-urban AURN sites; EMEP4UK base run in RED, EMEP4UK with a 30% shipping emissions reduction in GREEN and AURN observation in BLUE: units are in µg m⁻³. The purple column shows the percentage change in SO₂ surface concentration at each site estimated by the EMEP4UK model when shipping emissions are reduced by 30%. The sites are ranked for latitude where *Lullington Heath* is the lowest latitude and *Ladybower* the highest latitude of the available AURN sites for SO₂.

3.3. EMEP4UK 12 years of model output (Task 2)

The EMEP4UK default gridded output includes yearly, monthly, daily and hourly values of more than 300 parameters. The routine output variables of the EMEP4UK mode rv4.4 are shown in Table 1. Exemplary annual average output for the year 2001 to 2012 is shown in:

- **Figure 4 to Figure 9:** annual average surface concentration of O₃, SO₂, NO₂, NH₃, SO₄²⁻ and NO₃⁻, respectively
- **Figure 10 to Figure 12:** annual dry deposition of SO_x, NO_y and NH_x, respectively
- **Figure 13 to Figure 15:** annual wet deposition of SO_x, NO_y and NH_x, respectively

Figure 16 shows an example of PM_{2.5} components included in the EMEP4UK model rv4.4. For the year 2008 the annual average surface concentrations of PM_{2.5} and all the PM_{2.5} components included in the EMEP4UK model rv.4.4 are shown in **Figure 16**. All PM_{2.5} and PM₁₀ components included in the EMEP4UK rv4.4 model are available for the years 2001-2012 but not displayed here.

Table 1. Selected species included in EMEP4UK. For hourly data only a selection of pollutants is included in the routine output to minimise the usage of disk space (~1 GB yr⁻¹ of disk space is currently required for each hourly output species). Any variables labelled as 'on request' does imply the need to re-run the model.

1.	EMEP4UK output	averaging time				
		yearly	monthly	daily	hourly	
	Wet deposition SO _x	mgS m ⁻²	yes	yes	yes	on request
	Wet deposition OXN	mgN m ⁻²	yes	yes	yes	on request
	Wet deposition RDN	mgN m ⁻²	yes	yes	yes	on request
	Dry deposition SO _x	mgS m ⁻²	yes	yes	yes	on request
	Dry deposition OXN	mgS m ⁻²	yes	yes	yes	on request
	Dry deposition RDN	mgN m ⁻²	yes	yes	yes	on request
	SOMO35	ppb.day	yes	yes	yes	n/a
	POD1_DF	mmole m ⁻²	yes	yes	n/a	n/a
	POD1_CF	mmole m ⁻²	yes	yes	n/a	n/a
	POD3_TC	mmole m ⁻²	yes	yes	n/a	n/a
	EUAOT40_Crops	ppb.h	yes	yes	n/a	n/a
	EUAOT40_Forests	ppb.h	yes	yes	n/a	n/a
	SO ₂	µg m ⁻³	yes	yes	yes	yes
	NH ₃	µg m ⁻³	yes	yes	yes	yes
	HNO ₃	µg m ⁻³	yes	yes	yes	yes
	NO ₂	µg m ⁻³	yes	yes	yes	yes
	NO	µg m ⁻³	yes	yes	yes	yes
	SO ₄ ²⁻ _F	µg m ⁻³	yes	yes	yes	yes
	NO ₃ ⁻ _F	µg m ⁻³	yes	yes	yes	yes
	NO ₃ ⁻ _C	µg m ⁻³	yes	yes	yes	yes
	NH ₄ ⁺ _F	µg m ⁻³	yes	yes	yes	yes
	SEASALT_F	µg m ⁻³	yes	yes	yes	on request
	SEASALT_C	µg m ⁻³	yes	yes	yes	on request
	DUST_ROAD_F	µg m ⁻³	yes	yes	yes	on request
	DUST_ROAD_C	µg m ⁻³	yes	yes	yes	on request
	DUST_WB_F	µg m ⁻³	yes	yes	yes	on request
	DUST_WB_C	µg m ⁻³	yes	yes	yes	on request
	DUST_SAH_F	µg m ⁻³	yes	yes	yes	on request
	DUST_SAH_C	µg m ⁻³	yes	yes	yes	on request
	EC_F	µg m ⁻³	yes	yes	yes	on request
	EC_C	µg m ⁻³	yes	yes	yes	on request
	ASOA_OM	µg m ⁻³	yes	yes	yes	on request
	BSOA_OM	µg m ⁻³	yes	yes	yes	on request
	FFIRE_BC	µg m ⁻³	yes	yes	yes	on request
	FFIRE_OM	µg m ⁻³	yes	yes	yes	on request
	PM _{2.5}	µg m ⁻³	yes	yes	yes	yes
	PM ₁₀	µg m ⁻³	yes	yes	yes	yes

Note:

SOMO35 - The Sum of Ozone Means Over 35 ppb is the indicator for health impact assessment

POD_Y - Phyto-toxic ozone dose, is the accumulated stomatal ozone flux over a threshold Y

AOT40 - is the accumulated amount of ozone over the threshold value of 40 ppb

F – fine modelled fraction for size < PM_{2.5}

C – coarse modelled fraction for PM_{2.5} < size < PM₁₀

OM – organic matter

EC – elemental carbon

BC – black carbon

FIRE – forest fire

ASOA – anthropogenic secondary organic aerosols

BSOA – biogenic secondary organic aerosols

SHA – Sahara dust

WB – windblown dust

ROAD – road dust

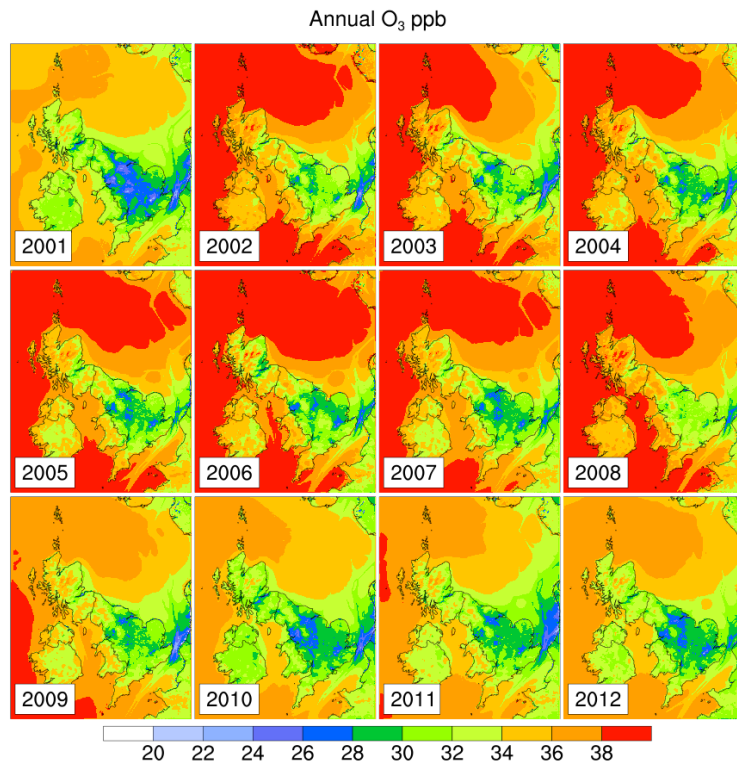


Figure 4. EMEP4UK rv4.4 annual average surface concentrations of O₃ for the years 2001 to 2012, illustrating inter-annual variability of ozone due to meteorological conditions, as well as longer term trends in background ozone in contrast to emission reductions.

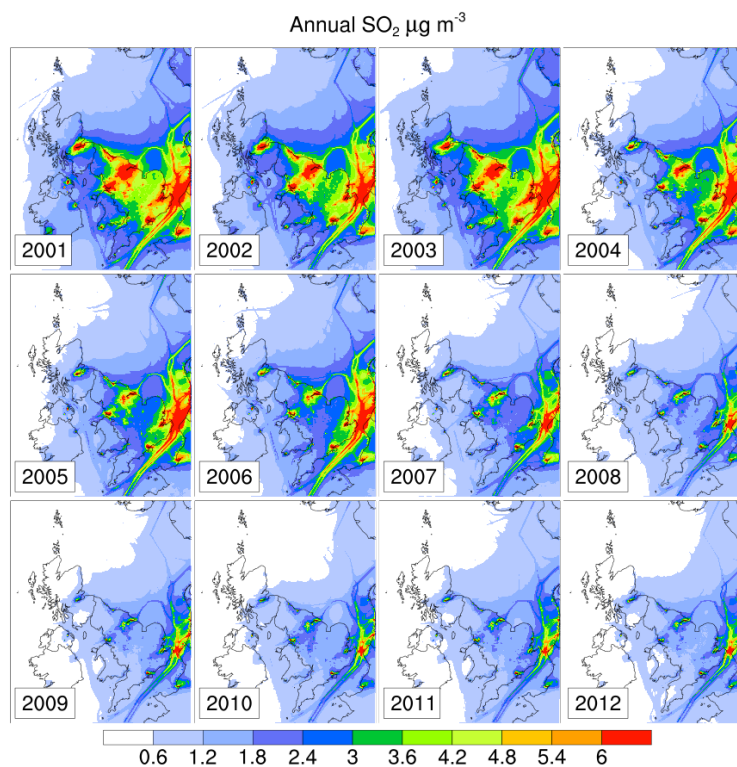


Figure 5. EMEP4UK rv4.4 annual average surface concentrations of SO₂ for the years 2001 to 2012, highlighting the effect of continuous reductions of emissions from UK land-based sources, as well as the effect of the MARPOL agreement establishing a SECA.

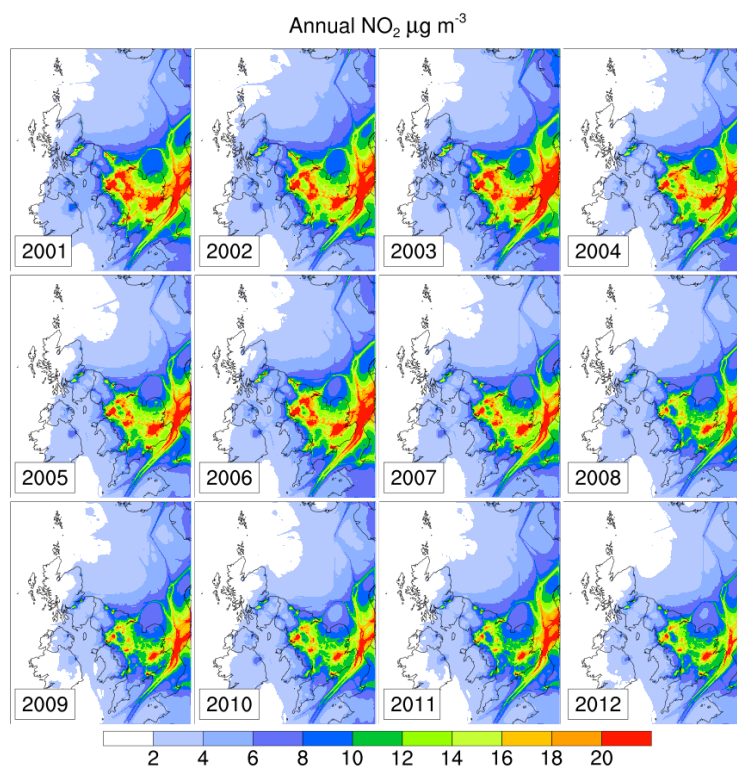


Figure 6. EMEP4UK rv4.4 annual average surface concentrations of NO₂ for the years 2001 to 2012, showing the decadal trend of reductions in land-based emissions from large point sources and road transport.

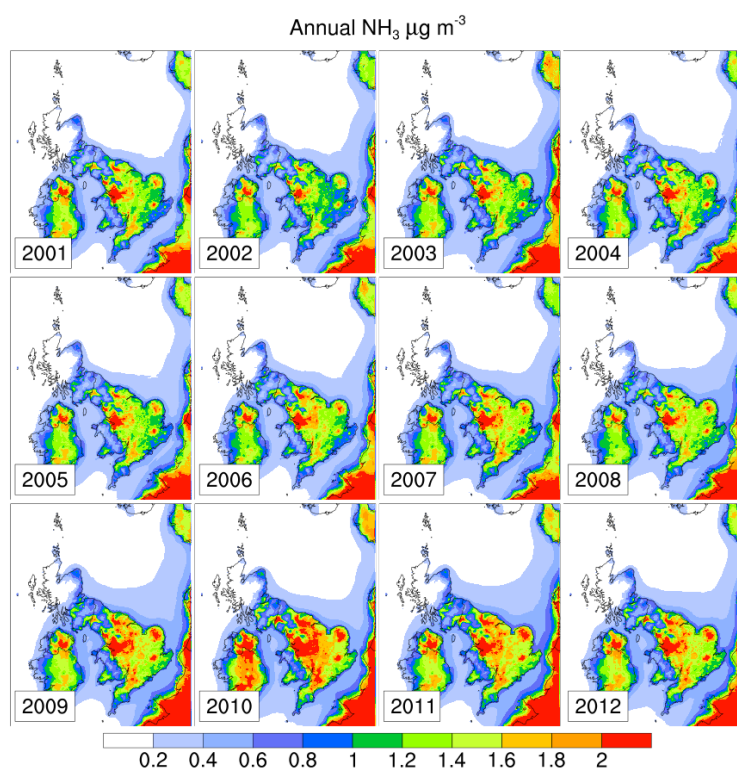


Figure 7. EMEP4UK rv4.4 annual average surface concentrations of NH₃ for the years 2001 to 2012, illustrating inter-annual variability due to meteorological/climatological drivers of ammonia volatilisation from fertiliser and manure applications. At the same time, this figure highlights that no substantive reduction in agricultural emissions have occurred since 2001.

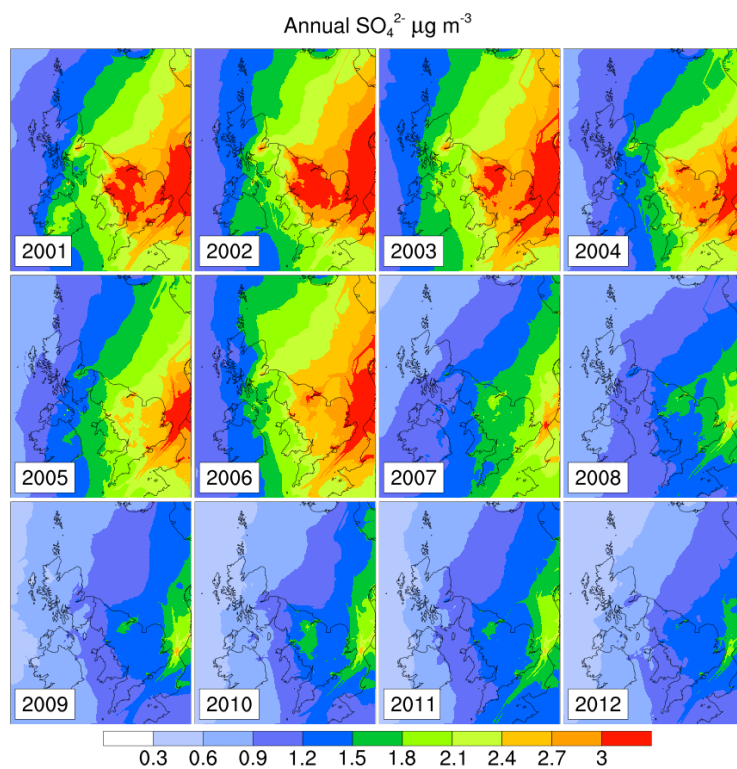


Figure 8. EMEP4UK rv4.4 annual average surface concentrations of SO_4 for the years 2001 to 2012, showing the direct result of reductions of SO_2 emissions achieved from UK land based sources and the effect of the implementation of the SECA.

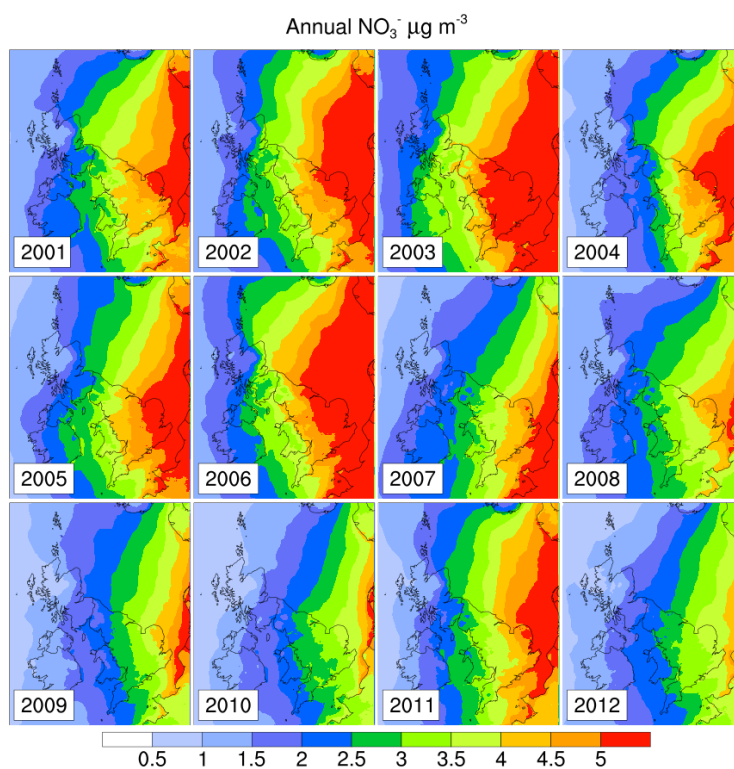


Figure 9. EMEP4UK rv4.4 annual average surface concentrations of NO_3 for the years 2001 to 2012.

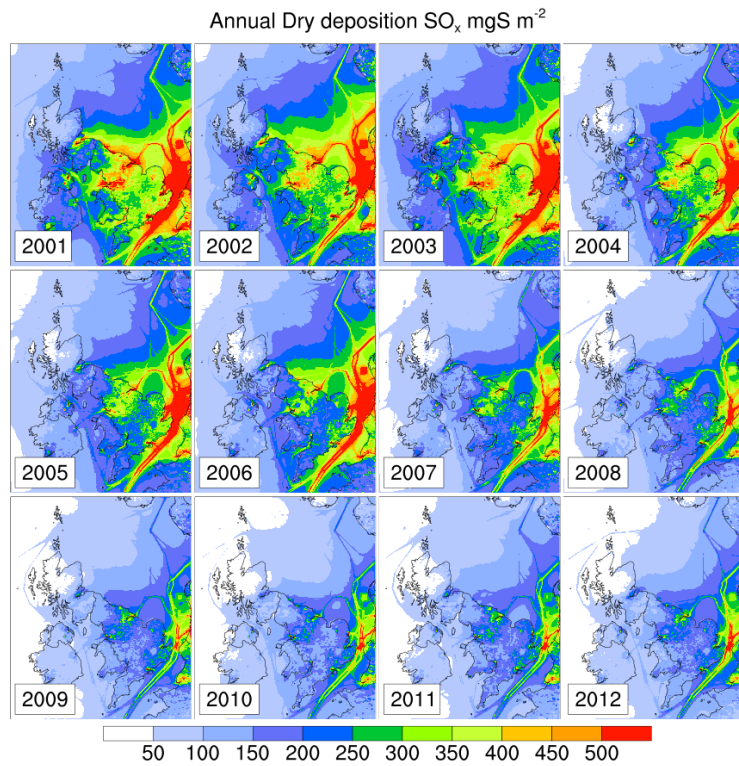


Figure 10. EMEP4UK rv4.4 annual dry deposition of SO_x for the years 2001 to 2012.

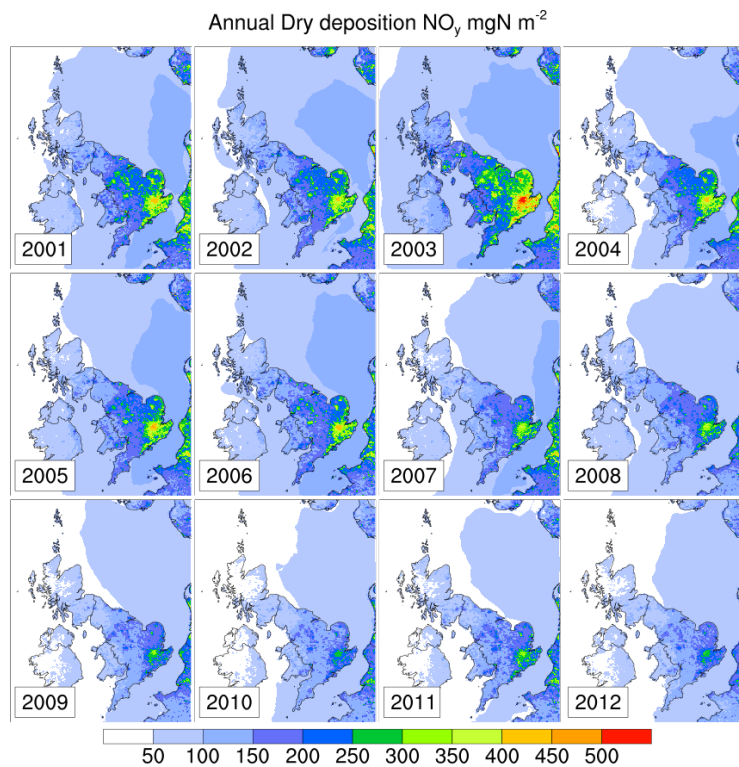


Figure 11. EMEP4UK rv4.4 annual dry deposition of NO_y for the years 2001 to 2012

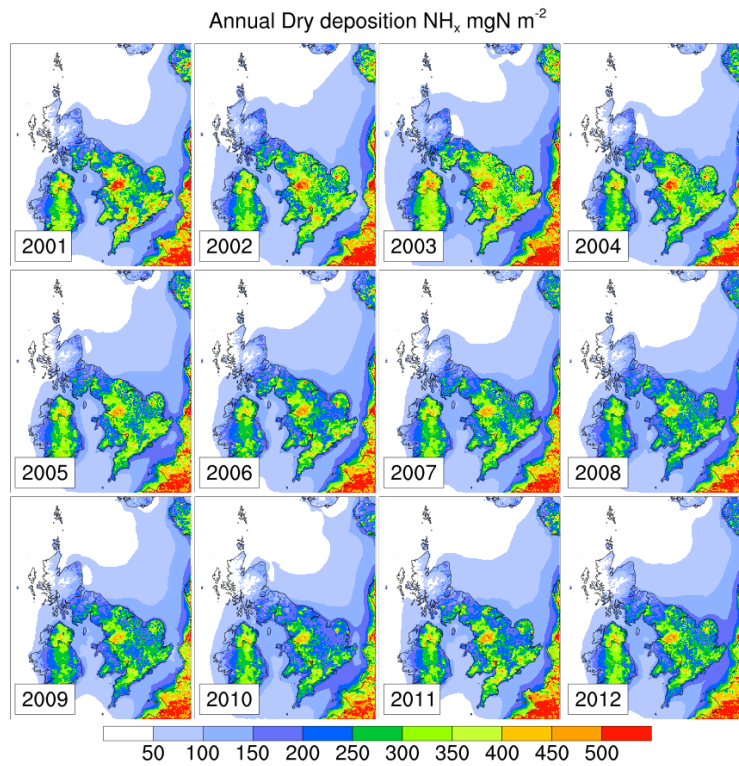


Figure 12. EMEP4UK rv4.4 annual dry deposition of NH_x for the years 2001 to 2012

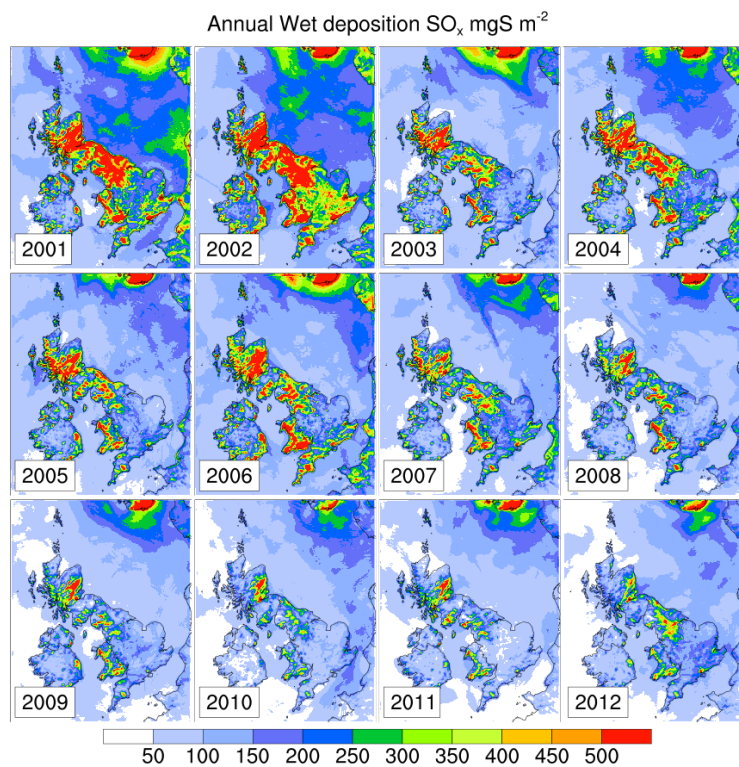


Figure 13. EMEP4UK rv4.4 annual wet deposition of SO_x for the years 2001 to 2012

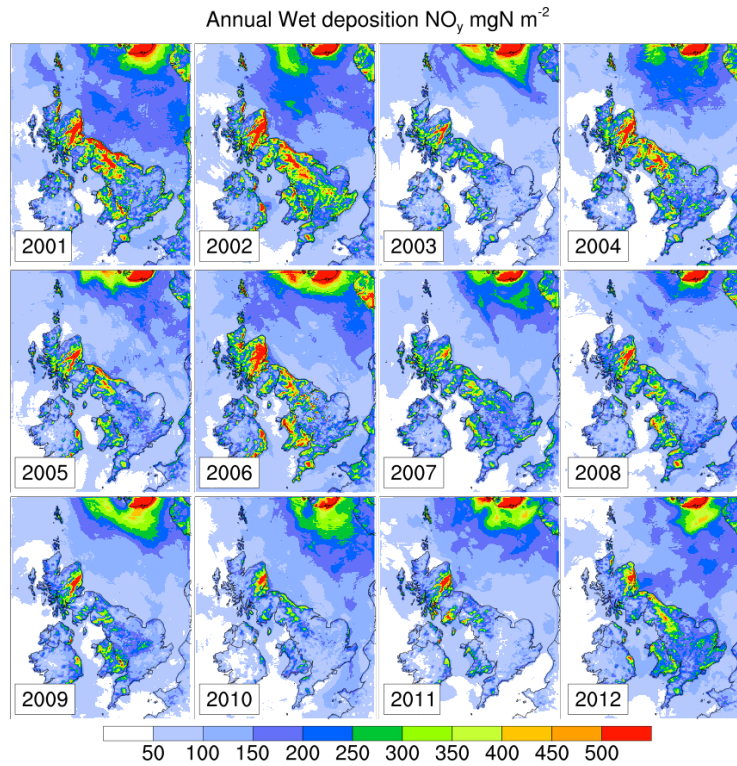


Figure 14. EMEP4UK rv4.4 annual wet deposition of NO_y for the years 2001 to 2012

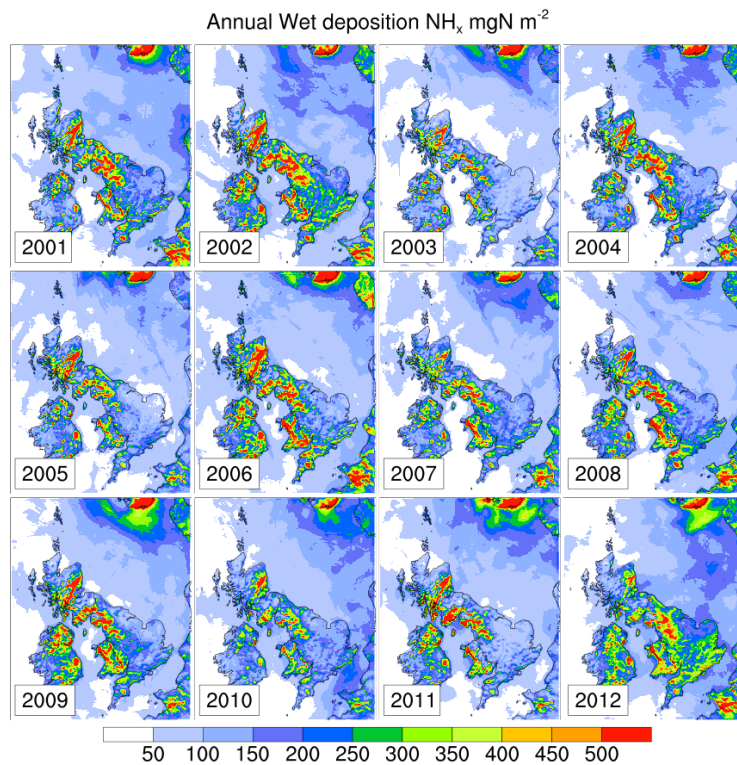


Figure 15. EMEP4UK rv4.4 annual wet deposition of NH_x for the years 2001 to 2012

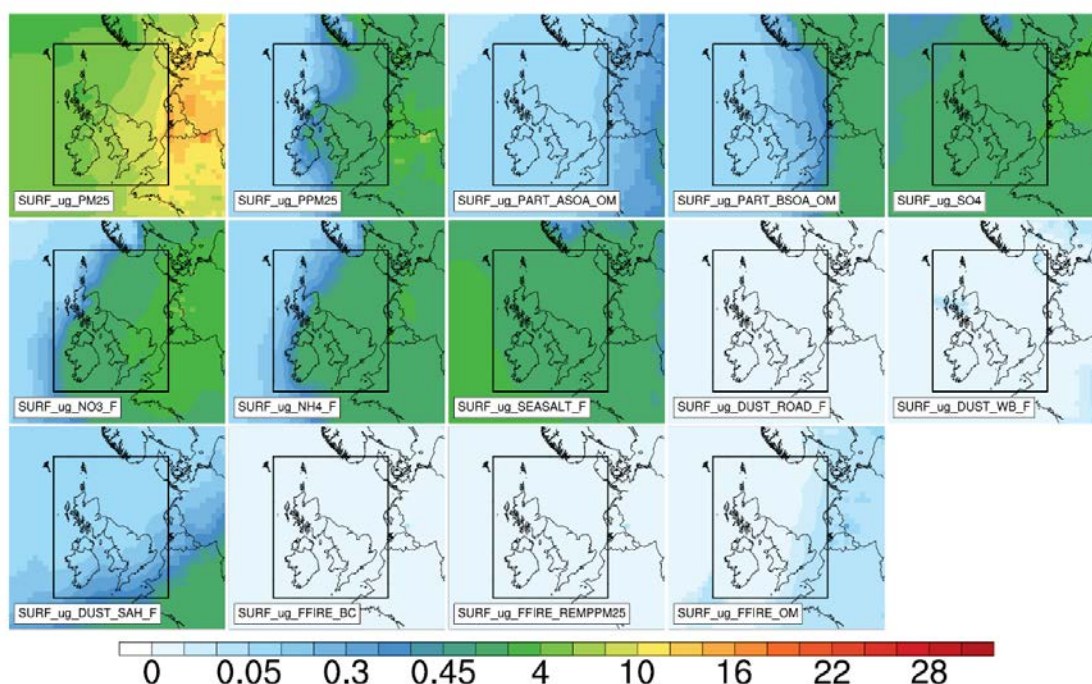


Figure 16. 2008 surface concentrations of $PM_{2.5}$ components calculated with the EMEP4UK model for a horizontal resolution of 50 km x 50 km outside the black box, and 5 km x 5 km within the black box. The panels are (from top left to bottom right): total $PM_{2.5}$, primary $PM_{2.5}$, anthropogenic secondary organic aerosols, biogenic secondary organic aerosols, sulphate, fine nitrate, coarse nitrate, ammonium, fine sea salt, fine road dust, fine windblown dust, fine Sahara dust, forest fire black carbon, forest fire primary $PM_{2.5}$, and forest fire organic matter. This figure highlights that UK $PM_{2.5}$ concentrations are primarily influenced by UK and European emissions of primary $PM_{2.5}$, as well as secondary inorganic aerosol (SIA) components.

3.4. EMEP4UK model verification (Task 3)

EMEP4UK is routinely validated against observations. A set of routines have been developed to automatically compare the EMEP4UK model with observation data from the AURN network. These scripts are written in NCL⁶ and R⁷ languages and use some of the functions from the Openair (Carslaw and Ropkins, 2012) R package to calculate the fraction of predictions within a factor 2 (FAC2), mean bias (MB), mean gross error (MGE), normalised mean bias (NMB), root mean square error (RMSE) and the correlation coefficient (r). A summary of the analysis for hourly modelled vs. observed surface concentration of O_3 , NO_2 and $PM_{2.5}$ for all AURN rural sites for the year 2010 is shown in **Table 2**, **Table 3**, and **Table 4**, respectively. The analysis is available for any of the available meteorological years modelled in the EMEP4UK project (2001-2012). Moreover, the EMEP4UK model analysis

⁶ The NCAR Command Language (Version 6.2.1) [Software]. (2014). Boulder, Colorado: UCAR/NCAR/CISL/VETS. <http://dx.doi.org/10.5065/D6WD3XH5>

⁷ <http://www.r-project.org/>

is available for all AURN sites including road side, urban and suburban sites and can be provided on request.

For each site of the AURN a summary analysis is also provided. In **Figure 17**, **Figure 18**, and **Figure 19** the 2010 surface concentrations are compared with the EMEP4UK model at the Harwell AURN site for NO₂, O₃ and PM_{2.5}, respectively. The analysis is available on request for all AURN sites and meteorological years included in this contract.

The EMEP4UK model monthly output is also compared with the observed data from the AGANeT and National Ammonia Monitoring network (NAMN) for NH₃, HNO₃, SO₂, NO₃, NH₄, and SO₄ (Vieno et al., 2014).

The EMEP4UK model results have been also extensively evaluated in the Defra's modelling inter-comparison exercise (MIE⁸), demonstrating how well the model performs for the species and removal processes included in this exercise (i.e. ozone, nitrate, and sulphate) as shown in Carslaw (2011a) and Carslaw (2011c).

The EMEP4UK model was compared with measurements of sulphur and nitrogen concentration in air (SO₂, NO₂, NH₃, HNO₃, SO₄²⁻, NO₃⁻, and NH₄⁺) and precipitation (SO₄²⁻, NO₃⁻, and NH₄⁺) as part of the Defra deposition model inter-comparison. EMEP4UK gave good agreement with measurements and performed well when compared with other models of variable complexity, generally obtaining lower values for normalised mean bias and root mean square error and higher values for the 'factor of two' and correlation coefficient than the average for the group of models. This demonstrates that the EMEP4UK model is fit for purpose to be used as a tool to support policy on nitrogen and acid deposition. The correlation statistics are illustrated in **Table 5**. The EMEP4UK is based on the EMEP MSC-w model which is annually reviewed by the Convention on Long Range Transport of Air Pollution (CLRTAP) and has been compared with leading ACTMs in several studies such as Simpson et al. (2014). As the EMEP4UK is developed and applied in close collaboration with the EMEP MSC-w team at met.no and reported to UNECE e.g. at TFMM and TFHTAP meetings, these studies are directly relevant to the EMEP4UK model further validation. More publications and verification of the EMEP MSC-w model can be found here: <http://www.emep.int>.

⁸ <http://uk-air.defra.gov.uk/research/air-quality-modelling?view=intercomparison>

Table 2. Summary comparison of the EMEP4UK calculated surface O₃ and observations from the AURN network for the year 2010.

Rural AURN site	lat	lon	alt	FAC2	MB	MGE	NMB	NMGE	RMSE	r
ASTON HILL	52.5	-3.0	370	0.9	2.7	13.6	0.0	0.2	17.4	0.6
AUCHENCORTH MOSS	55.8	-3.2	260	1.0	5.4	11.3	0.1	0.2	14.6	0.7
BUSH ESTATE	55.9	-3.2	180	0.9	3.5	11.6	0.1	0.2	15.6	0.7
CHARLTON MACKRELL	51.1	-2.7	54	0.9	3.2	14.0	0.1	0.3	17.9	0.7
ESKDALEMUIR	55.3	-3.2	269	0.9	8.2	13.6	0.1	0.2	17.6	0.6
FORT WILLIAM	56.8	-5.1	5	0.8	13.4	17.8	0.3	0.4	23.6	0.6
GREAT DUN FELL	54.7	-2.5	847	0.9	4.5	12.9	0.1	0.2	17.1	0.5
HARWELL	51.6	-1.3	137	0.9	4.6	14.2	0.1	0.3	18.6	0.7
HIGH MUFFLES	54.3	-0.8	267	0.9	3.3	12.0	0.1	0.2	15.5	0.7
LADYBOWER	53.4	-1.8	420	0.9	2.2	11.8	0.0	0.2	15.6	0.7
LERWICK	60.1	-1.2	85	0.9	2.0	12.1	0.0	0.2	16.8	0.5
LOUGH NAVAR	54.4	-7.9	130	0.8	14.9	17.4	0.3	0.4	22.5	0.7
LULLINGTON HEATH	50.8	0.2	125	0.7	15.2	18.2	0.4	0.5	22.4	0.6
MARKET HARBOROUGH	52.6	-0.8	145	0.8	11.4	17.2	0.2	0.4	21.2	0.7
NARBERTH	51.8	-4.7	169	0.9	2.4	13.2	0.0	0.2	17.1	0.6
ROCHESTER STOKE	51.5	0.6	14	0.7	-3.5	19.1	-0.1	0.5	24.5	0.2
SIBTON	52.3	1.5	46	0.9	6.9	14.2	0.1	0.3	18.1	0.7
ST OSYTH	51.8	1.0	8	0.9	10.5	15.7	0.2	0.3	20.3	0.7
STRATH VAICH	57.7	-4.8	266	0.9	2.2	13.3	0.0	0.2	17.0	0.6
WEYBOURNE	53.0	1.1	16	1.0	-1.8	13.6	0.0	0.2	18.4	0.6
WICKEN FEN	52.3	0.3	5	0.8	13.7	17.5	0.3	0.4	21.8	0.8
YARNER WOOD	50.6	-3.7	119	0.9	2.9	12.8	0.0	0.2	17.0	0.7

Table 3. Summary comparison of the EMEP4UK calculated surface NO₂ and observations from the AURN network for the year 2010.

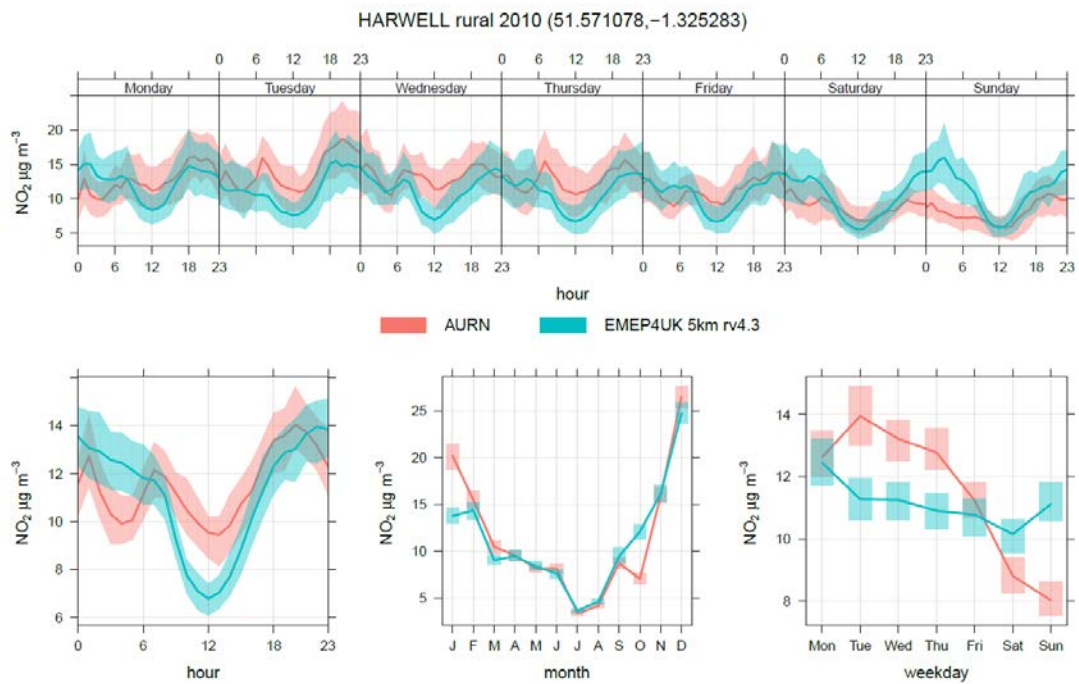
Rural AURN site	lat	lon	alt	FAC2	MB	MGE	NMB	NMGE	RMSE	r
ASTON HILL	52.5	-3.0	370	0.5	0.5	3.9	0.1	0.7	6.8	0.7
BUSH ESTATE	55.9	-3.2	180	0.5	-2.1	5.1	-0.3	0.6	8.6	0.6
CHARLTON MACKRELL	51.1	-2.7	54	0.7	-1.8	5.5	-0.2	0.5	8.3	0.7
ESKDALEMUIR	55.3	-3.2	269	0.4	0.9	2.3	0.3	0.9	4.1	0.6
FORT WILLIAM	56.8	-5.1	5	0.1	-11.5	11.6	-0.9	0.9	16.2	0.3
HARWELL	51.6	-1.3	137	0.6	-0.4	6.0	0.0	0.5	9.6	0.7
HIGH MUFFLES	54.3	-0.8	267	0.5	1.2	4.4	0.2	0.8	7.8	0.6
LADYBOWER	53.4	-1.8	420	0.6	0.5	5.8	0.1	0.6	9.3	0.5
LULLINGTON HEATH	50.8	0.2	125	0.6	0.3	5.6	0.0	0.6	9.1	0.5
MARKET HARBOROUGH	52.6	-0.8	145	0.7	-0.9	5.5	-0.1	0.5	8.6	0.7
NARBERTH	51.8	-4.7	169	0.5	-0.4	2.9	-0.1	0.7	5.6	0.5
ROCHESTER STOKE	51.5	0.6	14	0.7	-5.0	10.5	-0.2	0.4	14.5	0.6
ST OSYTH	51.8	1.0	8	0.6	-2.8	6.8	-0.2	0.5	11.0	0.6
WICKEN FEN	52.3	0.3	5	0.6	-0.6	5.1	-0.1	0.5	7.6	0.7
YARNER WOOD	50.6	-3.7	119	0.5	0.3	3.1	0.1	0.7	5.8	0.5

Table 4. Summary comparison of the EMEP4UK calculated surface PM_{2.5} and observations from the AURN network for the year 2010.

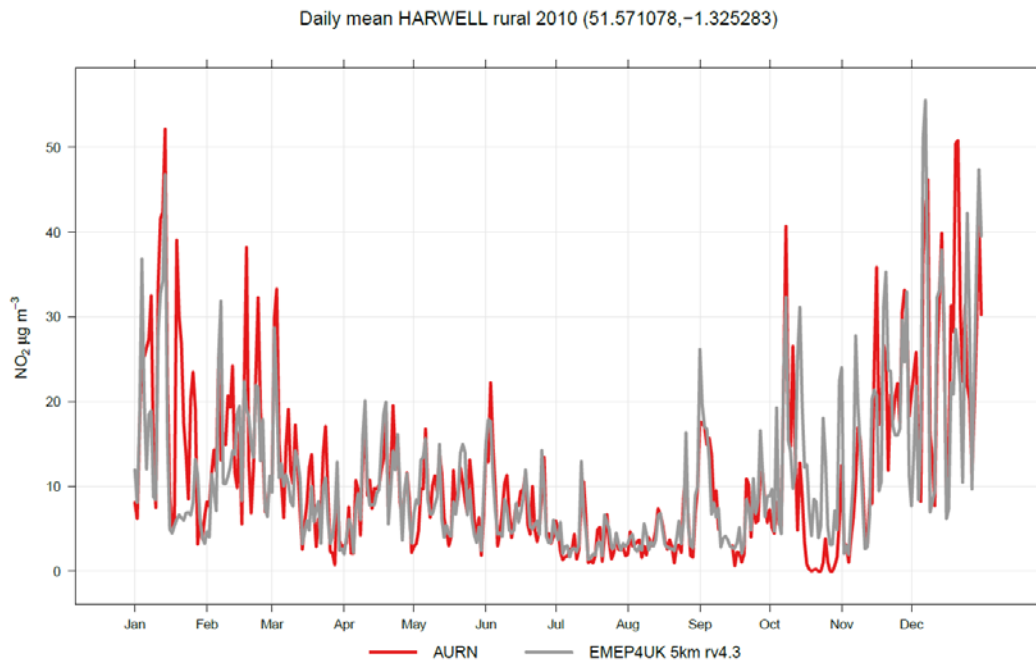
Rural AURN site	lat	lon	alt	FAC2	MB	MGE	NMB	NMGE	RMSE	r
AUCHENCORTH MOSS	55.8	-3.2	260	0.4	2.9	4.6	0.7	1.1	6.6	0.5
HARWELL	51.6	-1.3	137	0.8	-0.3	4.6	0.0	0.5	7.1	0.6
ROCHESTER STOKE	51.5	0.6	14	0.7	1.4	5.0	0.1	0.5	8.2	0.6

Table 5. Model performance statistics for EMEP4UK for the year 2003, for comparison with measurements: FAC2: fraction of points greater than 0.5x and less than 2x the measured value; NMB: normalised mean bias; NMGE: normalised mean gross error; RMSE: root mean square error; r: Pearson correlation coefficient (units for NMGE and RMSE: $\mu\text{g m}^{-3}$ – gas and aerosol; $\mu\text{.equiv L}^{-1}$ – aqueous)

Group	Phase	FAC2	NMB	NMGE	RMSE	r
SO ₂	Gas	0.86	0.22	0.39	1.00	0.66
NO ₂	Gas	1.00	-0.18	0.24	2.52	0.94
NH ₃	Gas	0.75	-0.24	0.41	1.25	0.66
SO ₄ ²⁻	Aerosol	1.00	0.00	0.09	0.18	0.97
NO ₃ ⁻	Aerosol	0.92	0.15	0.19	0.66	0.95
NH ₄ ⁺	Aerosol	1.00	-0.17	0.26	0.47	0.93
SO ₄ ²⁻	Aqueous	1.00	-0.12	0.22	7.13	0.83
NO ₃ ⁻	Aqueous	0.73	-0.42	0.43	11.67	0.79
NH ₄ ⁺	aqueous	0.84	-0.23	0.35	10.62	0.70



a)



b)

Figure 17. EMEP4UK 2010 surface concentrations of NO_2 : a) temporal variation of modelled and observed hourly concentrations at Harwell and b) daily average of observation and modelled. The temporal variability and the resulting agreement with observations heavily depends on the temporal emission profiles applied in the model, which are based on generic sectoral profiles developed for the EMEP model. UK-specific temporal profiles currently being developed by the NAEI team for Defra will likely improve the temporal match (in

particular for diurnal and weekly time scales) with observations and will be tested in EMEP4UK as soon as they become available.

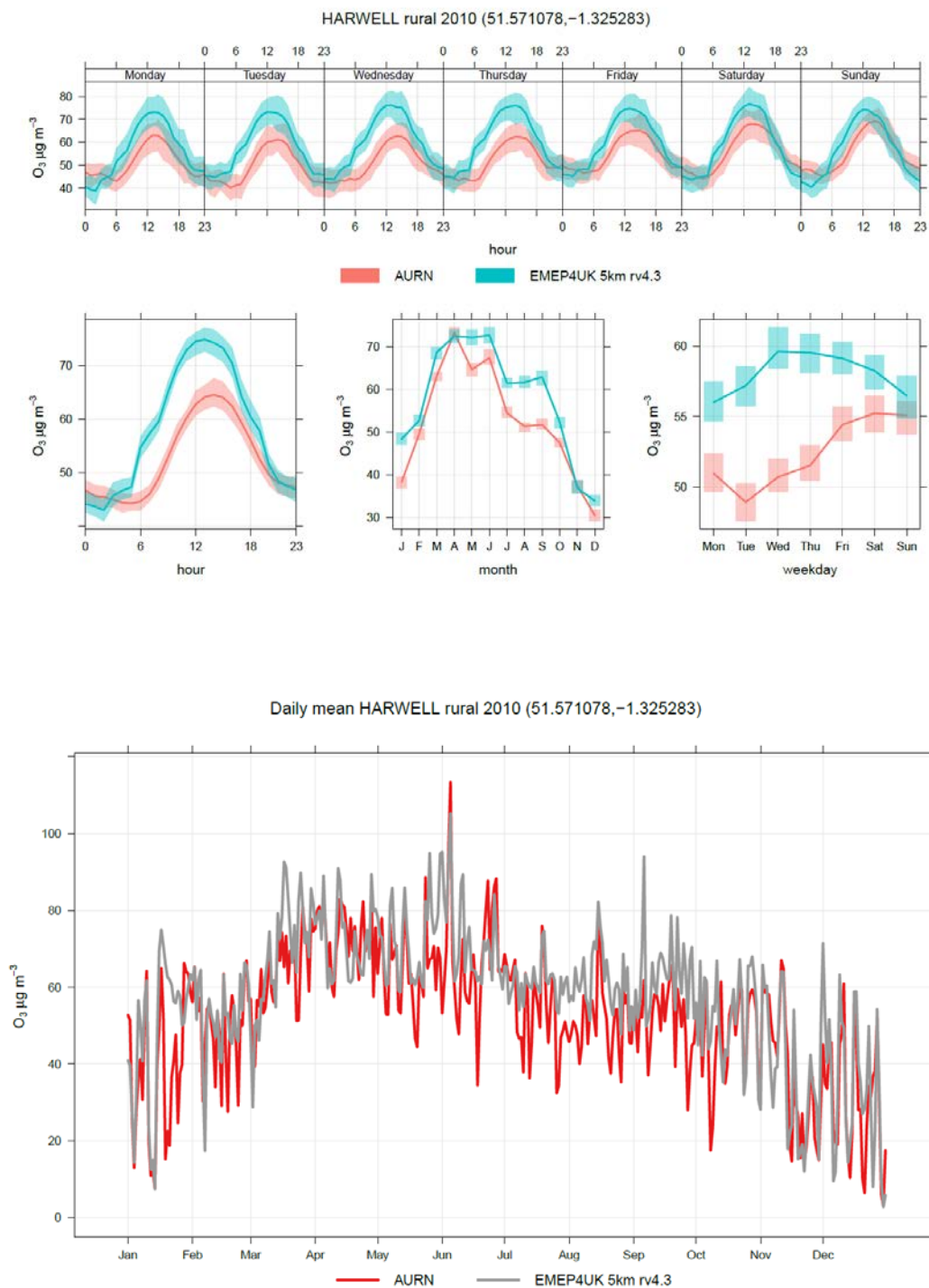
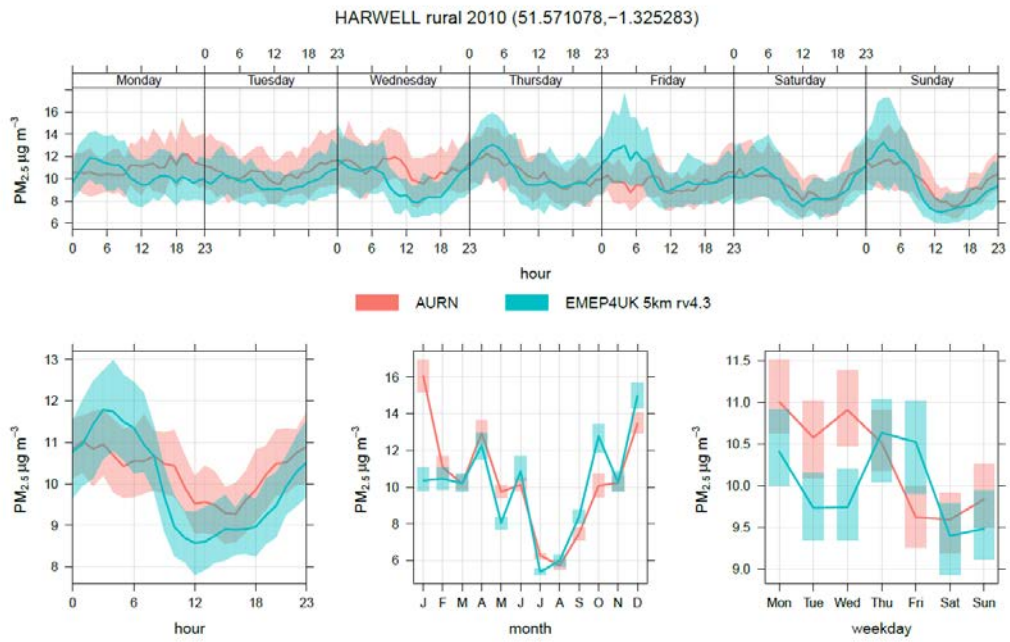


Figure 18. EMEP4UK 2010 surface concentration of O_3 : a) temporal variation of modelled and observed hourly concentrations at Harwell and b) daily average of observation and modelled.



Daily mean HARWELL rural 2010 (51.571078,-1.325283)

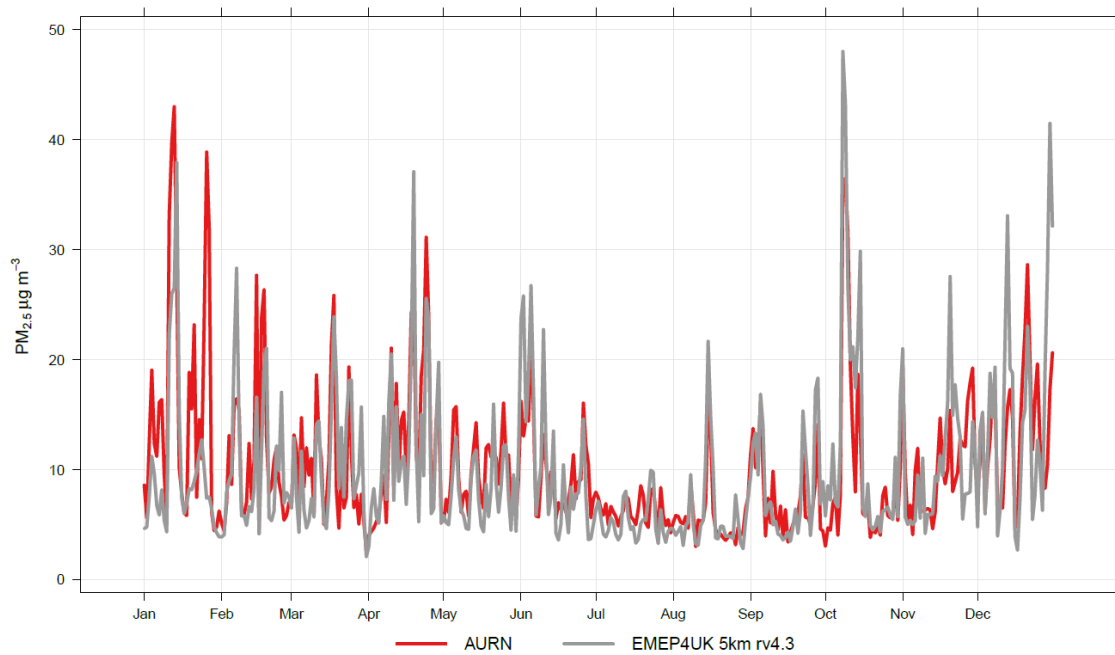


Figure 19. EMEP4UK 2010 surface concentration of PM_{2.5}: a) temporal variation of modelled and observed hourly concentrations at Harwell and b) daily average of observation and modelled.

3.5. Ad-hoc analysis (Task 4)

3.6. Two emissions reduction scenarios for the year 2030

Two emission scenarios for the year 2030 and a baseline emission for the year 2005 have been used to evaluate the effect of projected European and UK emissions reductions by 2030 over the UK. The meteorological year chosen to run all three simulations was 2005.

3.6.1. 2030 emissions scenarios used

The EMEP 2005 emissions are used as baseline to create the two 2030 emission estimate scenario used here called NECD2030 and CLE2030+UEP45.

The CLE2030 scenario is derived from the 2012 IIASA TSAP report #1 “Future emissions of air pollutants in Europe – Current legislation baseline and the scope for further reductions” (IIASA, 2012), however, for the United Kingdom the UEP45 central case emission estimate was applied as requested by Defra. Emission reduction for both scenarios (NECD2030 and CLE2030) of NO_x and PM_{2.5} are used as a proxy for CO and PM_{co} emissions reduction (coarse fraction of PM), respectively. The NECD2030 is based on the reductions resulting from the revised UNECE CLRTAP Gothenburg Protocol.

The EU28 total annual national emission figures for 2005, NECD2030 and CLE2030+UEP45UK are summarised in **Figure 20**, **Figure 21**, **Figure 22**, **Figure 23**, **Figure 24**, **Figure 25** for SO_x, NO_x, CO, NMVOCs, NH₃ and primary PM_{2.5}, respectively.

The gridded emissions were calculated using the EMEP 2005 baseline applying the appropriate scaling factor to match the respective 2030 emissions scenario. This approach retains the spatial distribution unchanged from the 2005 baseline. Moreover, no change in emissions from domestic and international shipping were assumed. The gridded emissions used in this work are shown in **Figure 26**, **Figure 27**, **Figure 28**, **Figure 29**, **Figure 30**, and **Figure 31** for SO_x, NO_x, CO, NMVOCs, NH₃ and primary PM_{2.5}, respectively.

3.6.2. 2030 scenario results

Results from modelling the 2030 scenarios for the EMEP4UK rv4.3 UK domain at 5 km x 5 km horizontal resolution and a portion of the EU domain at 50 km x 50 km horizontal resolution, for the base 2005, NECD2030 and CLE2030 are shown in panel a) of **Figure 32**, **Figure 33**, **Figure 34**, **Figure 35**, **Figure 36** and **Figure 37**, for the surface concentrations of SO₂, NO₂, NH₃, SO₄, NO₃, and PM₁₀, respectively.

Figure 32 b) and c) shows the changes in surface concentration of SO₂ between the year 2005 base case emission, the CLE2030 and NECD emissions reduction. Both emission scenarios included here shows a similar UK reduction of up to 5 µg m⁻³ in surface concentration.

Figure 33 b) and c) shows the changes in surface concentration of NO₂ between the year 2005 base case emission, the CLE2030 and NECD emissions reduction. The NECD emissions scenario shows a higher reduction in NO₂ surface concentrations of up to 20 µg m⁻³ and when compared with CLE2030, the NECD shows a higher reduction by up to ~4 µg m⁻³.

Figure 34 b) and c) shows the changes in surface concentrations between the year 2005 base case emission, the CLE2030 and NECD emissions reduction. The NECD emissions scenario shows no change in NH₃ surface concentrations (no emissions changes between 2005 and NECD) whereas for CLE2030 a small reduction of up to 0.5 µg m⁻³ is shown.

Figure 35 b) and c) shows the changes in surface concentration of SO₄ between the year 2005 base case emission, the CLE2030 and NECD emissions reduction. The NECD emissions scenario and CLE2030 shows similar changes in surface concentration up to 1.2 µg m⁻³ however, the NECD has a higher reduction up to 0.4 µg m⁻³.

Figure 36 b) and c) shows the changes in surface concentration of NO₃ between the year 2005 base case emission, the CLE2030 and NECD emissions reduction. The NECD emissions scenario shows changes in surface concentration up to 3 µg m⁻³ and the CLE2030 scenario shows changes of up to 2.1 µg m⁻³.

Figure 37 b) and c) shows the changes in surface concentration of PM₁₀ between the year 2005 base case emission, the CLE2030 and NECD emissions reduction. The NECD emissions scenario shows changes in surface concentration up to 8 µg m⁻³ and the CLE2030 scenario shows changes of up to 6 µg m⁻³.

3.7. Impact of 3 future shipping emissions scenarios

EMEP4UK was applied to simulate the future concentrations of PM₁₀ and PM_{2.5} for the UK for three different scenarios of emissions of SO₂ and particulate matter from shipping.

The study considered the costs and benefits associated with three policy scenarios in 2020:

- Policy Scenario 1: no change in policy with 2020 activity. In this scenario, sulphur in fuel limits of 1% are taken to apply in SECAs (as per MARPOL requirements since

2010). The fuel used outside SECAs is assumed to be residual oil (RO) with a sulphur content of 2.7%

- Policy Scenario 2: partial implementation of the Directive’s requirements, limited to the introduction of 0.1% sulphur limits in SECAs from 2015;
- Policy Scenario 3: full implementation of the Directive’s requirements, consisting of the introduction of 0.1% sulphur limits in SECAs from 2015, followed by 0.5% sulphur limits outside SECAs from 2020.

Further details are included in (Brutus, 2014). The impact on human health of implementing emissions reductions was assessed by calculation of the Population Weighted Mean Concentrations (PWMC) of PM₁₀ and PM_{2.5}. As well as emissions reductions in primary particulate matter, emissions reduction of SO₂ can also lead to a lowering in particulate concentrations due to a lower rate of formation of ammonium sulphate aerosol in the atmosphere. Emissions scaling factors for predicted future emissions provided by Ricardo-AEA for SO₂, NO_x, VOC, NH₃, CO, PM₁₀ and PM_{2.5} were used to define UK emissions for the year 2020.

The PWMC were calculated for the UK using spatially disaggregated population data for the UK supplied by Ricardo-AEA at a 1 km resolution. The results for the three scenarios are illustrated in **Table 6**. The PM concentrations calculated include all chemical components included in the EMEP4UK model. In the case of PM₁₀ there is a significant contribution from sea salt. As the sea salt component is the same for all three scenarios, the effect of abatement of PM and SO₂ emissions from shipping is shown to result in a smaller % decrease in PM₁₀ concentrations than that for PM_{2.5}. For scenario 3, a 1.4% reduction in PWMC for PM_{2.5} was calculated relative to scenario 1.

Table 6. Population-weighted mean concentrations for the UK for PM₁₀ and PM_{2.5} for scenarios 1-3. Percentage reduction in population-weighted mean concentrations for scenarios 2 and 3 relative to scenario 1.

	Scenario 1	Scenario 2	Scenario 3
PM ₁₀	15.23	15.15	15.10
PM _{2.5}	8.40	8.33	8.28
PM ₁₀ % reduction	-	0.48	0.83
PM _{2.5} % reduction	-	0.79	1.41

3.8. Deposition data for calculation of the exceedance of critical loads 2010-2012.

The deposition data submitted to the model inter-comparison was used to calculate the exceedance of critical loads for both nitrogen and acid deposition and used to compare the results obtained using the different ACTMs with those from the CBED data set. As the simulation year (2003) for the deposition model inter-comparison was somewhat anomalous (due to low precipitation and a high incidence of flow of polluted continental air from the south-east), it was decided to update the data used in the model inter-comparison. Deposition data calculated with the EMEP4UK for the three year period 2010-2012 has been supplied to Jane Hall, CEH Bangor and this will be used to calculate the exceedance of critical loads and make a comparison with the results obtained with the CBED and FRAME data for annual deposition averaged over the same three year period.

3.9. April/March 2014 UK PM episode

The EMEP4UK version rv4.4, driven by the Weather and Research Forecast model (WRF) version 3.6.1 was used to simulate the March-April 2014 PM pollution event over Europe and for the UK. EMEP4UK model simulates the concentration of PM with size less than 10 μm (PM_{10}) and with size less than 2.5 μm ($\text{PM}_{2.5}$). In the model version used here $\text{PM}_{2.5}$ is the sum of the fine fraction of: particulate ammonium (NH_4^+), particulate sulphate (SO_4^{2-}), particulate nitrate (NO_3^-), elemental carbon (EC), organic matter (OM), sea salt (SS), and mineral dust. PM_{10} is the sum of $\text{PM}_{2.5}$ plus the coarse fractions of EC, OM, NO_3^- , SS, and dust. The EMEP4UK model includes a suite of anthropogenic emissions in addition to dust emissions from Sahara and from other dust sources, such as roads.

Figure 38 shows the EMEP4UK modelled daily surface concentration of PM_{10} components and the daily average AURN observations (solid line) for two AURN sites. All AURN sites are included in the analysis and although not shown here are available on request. Secondary inorganic aerosols (SIA) are the cause of the elevated UK PM_{10} for most of the episode. Saharan dust contributed to the elevated UK PM_{10} only towards the end of the episode (2nd-4th of April).

The imported SIA are clearly visible in **Figure 39**. The SIA and their precursors originating from Germany/Denmark are advected to the UK for the whole duration of the episode. These events occur regularly, with notable effects in 2003, 2014 and again in 2015, depending on the timing of the spring manure and fertiliser application in Europe and the UK and meteorological conditions during that period. Due to its profound impacts on ambient

concentrations on $PM_{2.5}$ over the UK and its transboundary nature, the model results illustrating the source regions and the contributing emissions are of high policy relevance, in particular in the context of the CLRTAP.

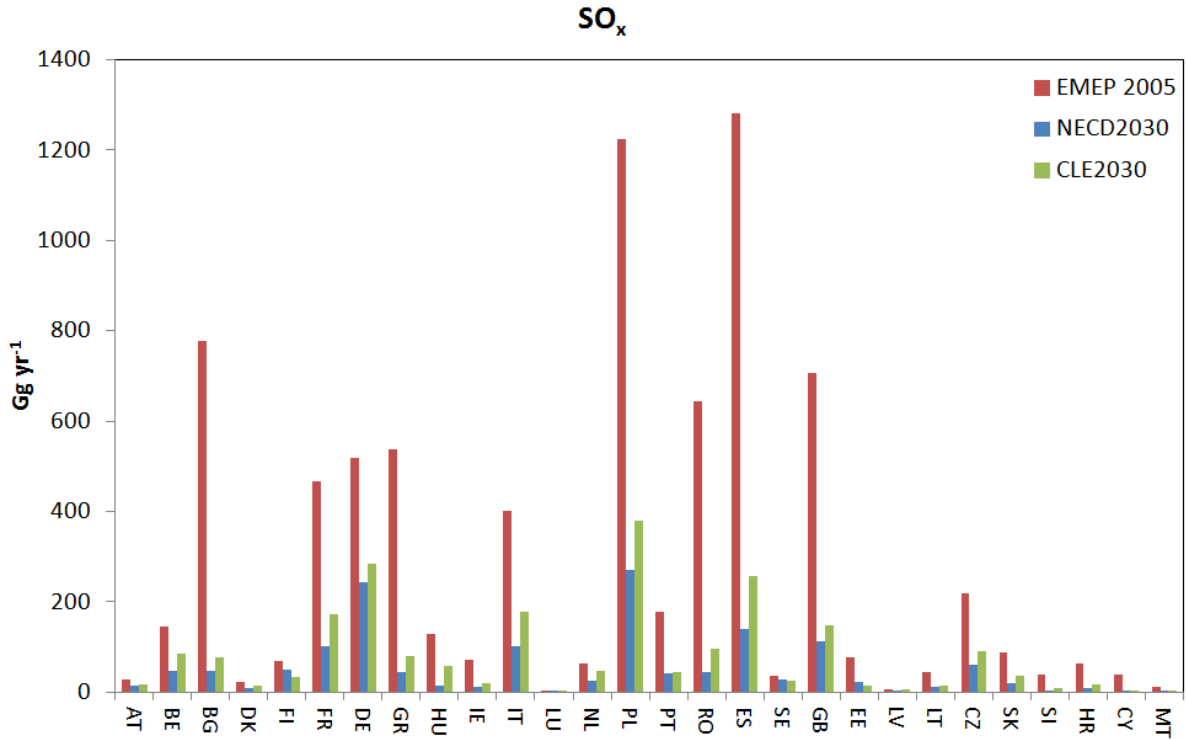


Figure 20. SO_x annual emissions estimate for each country included in the EU28: red EMEP baseline 2005, blue NECD2030 and green CLE2030/UEP45 (UK only).

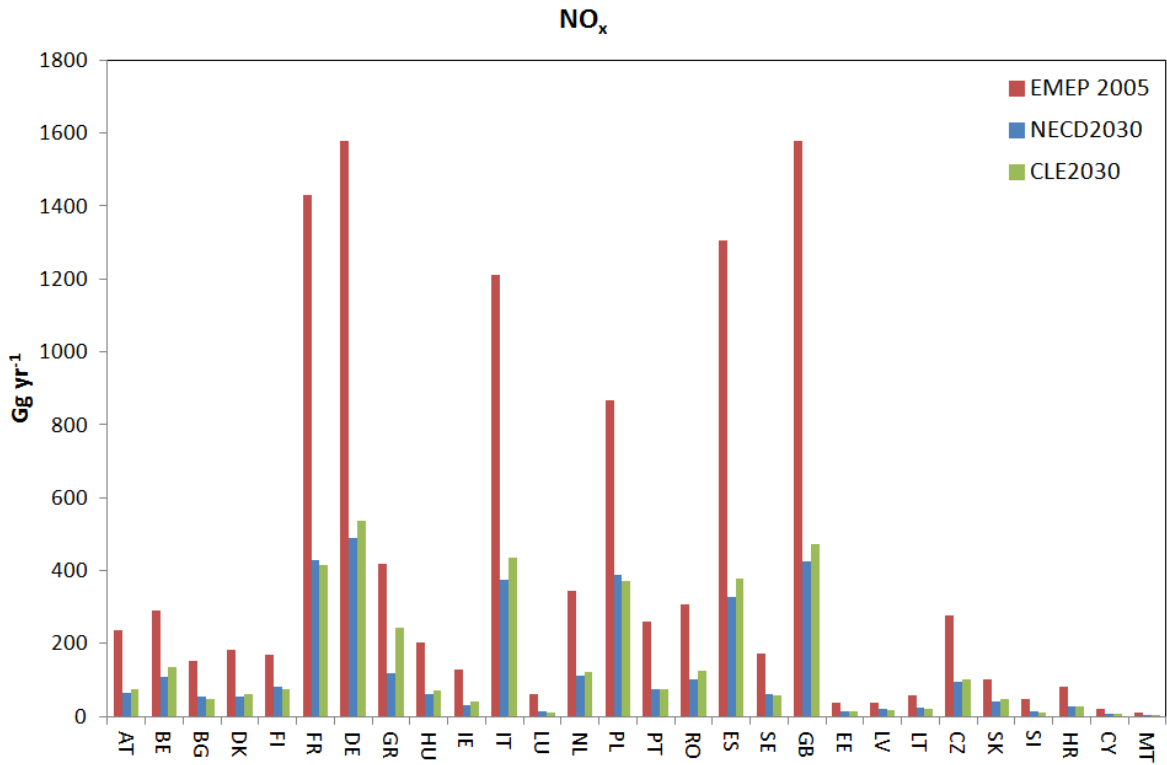


Figure 21. NO_x annual emissions estimate for each country included in the EU28: red EMEP baseline 2005, blue NECD2030 and green CLE2030/UEP45 (UK only).

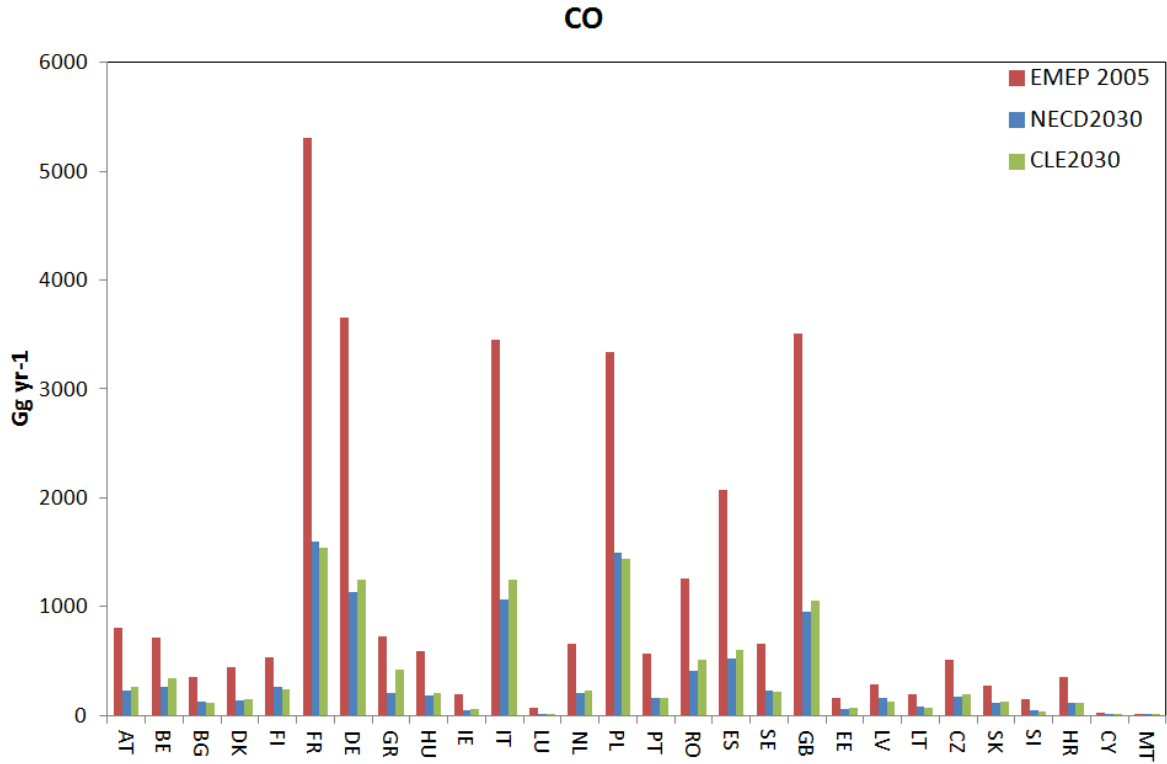


Figure 22. CO annual emissions estimate for each country included in the EU28: red EMEP baseline 2005, blue NECD2030 and green CLE2030/UEP45 (UK only).

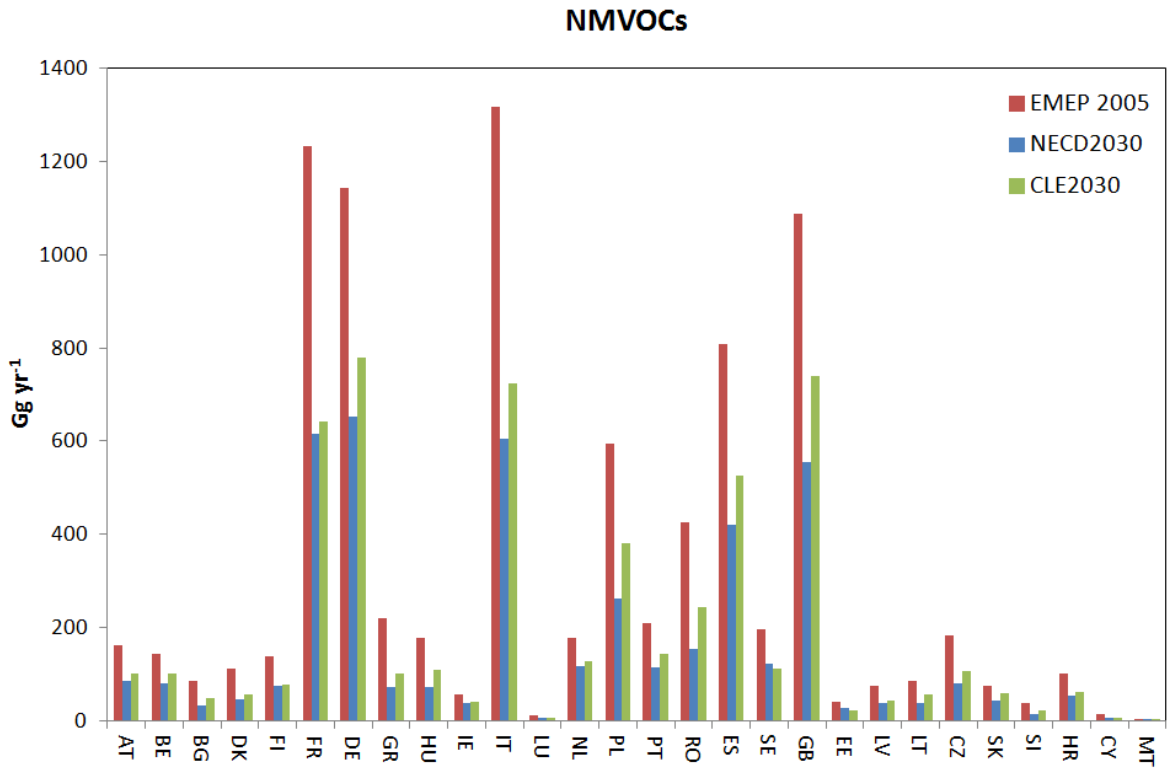


Figure 23. NMVOCs annual emissions estimate for each country included in the EU28: red EMEP baseline 2005, blue NECD2030 and green CLE2030/UEP45 (UK only).

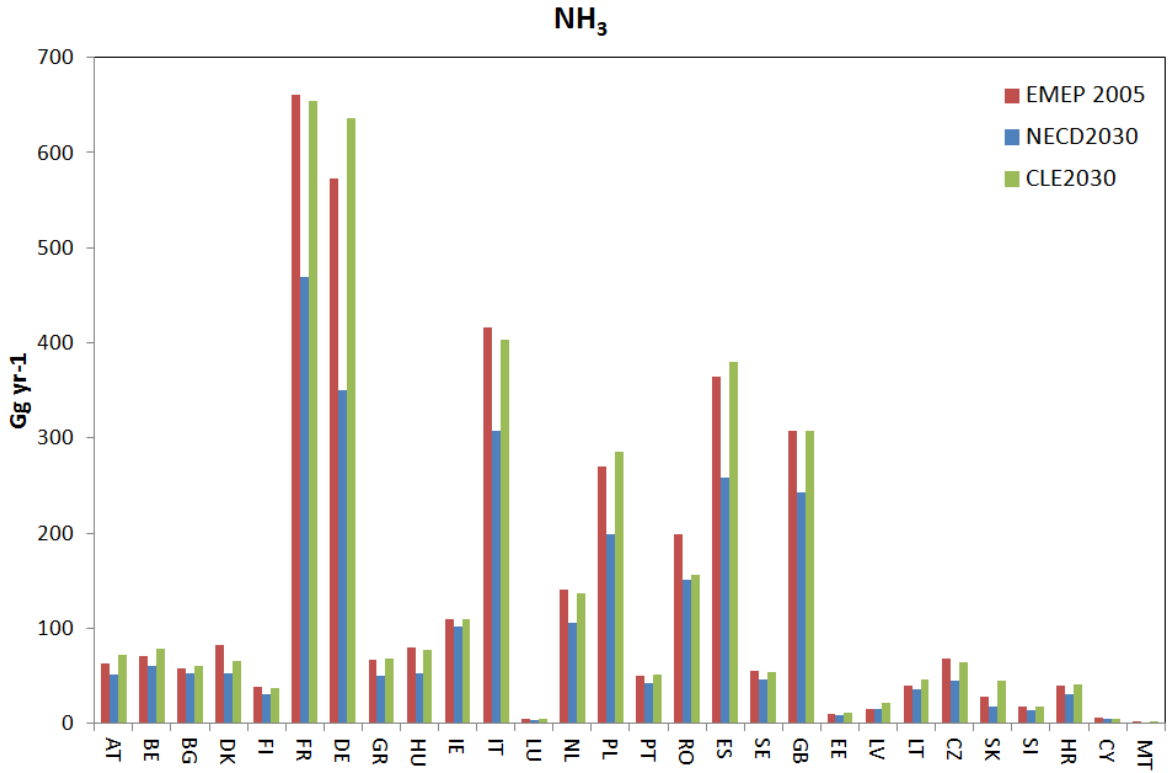


Figure 24. NH₃ annual emission estimates for each country included in the EU28: red EMEP baseline 2005, blue NECD2030 and green CLE2030/UEP45 (UK only).

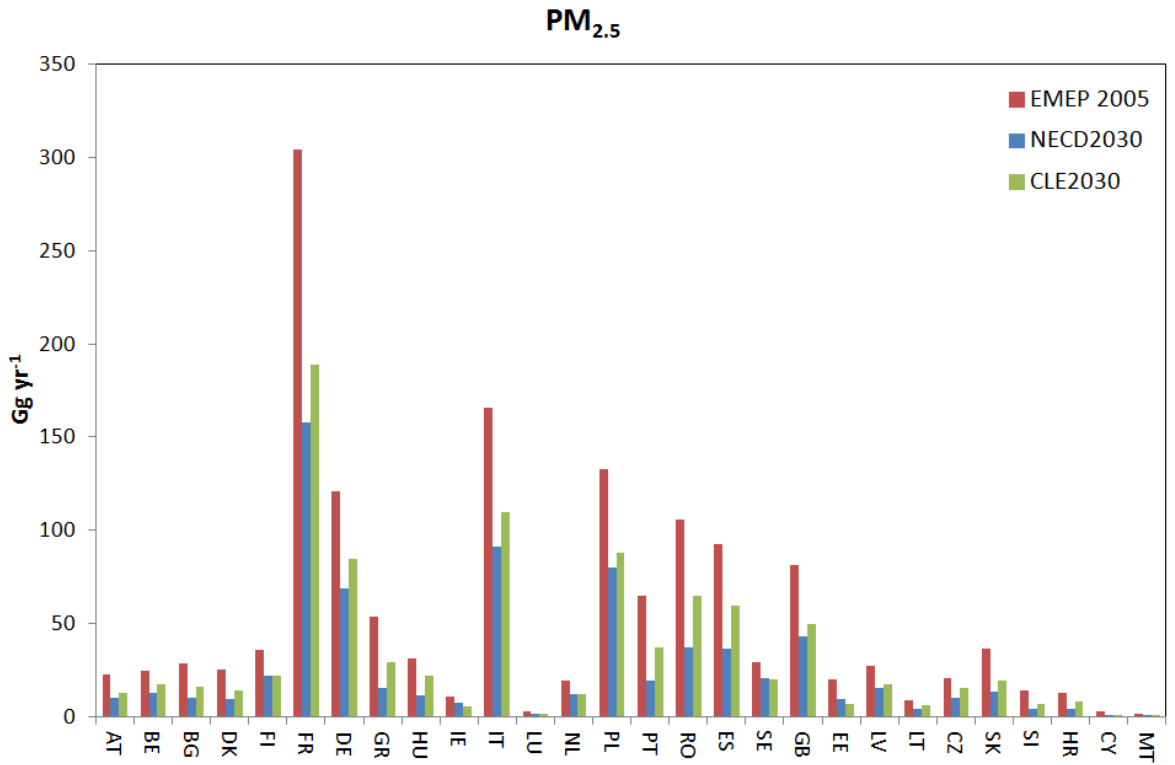


Figure 25. Primary PM_{2.5} annual emissions estimate for each country included in the EU28: red EMEP baseline 2005, blue NECD2030 and green CLE2030/UEP45 (UK only).

EMEP4UK 2005

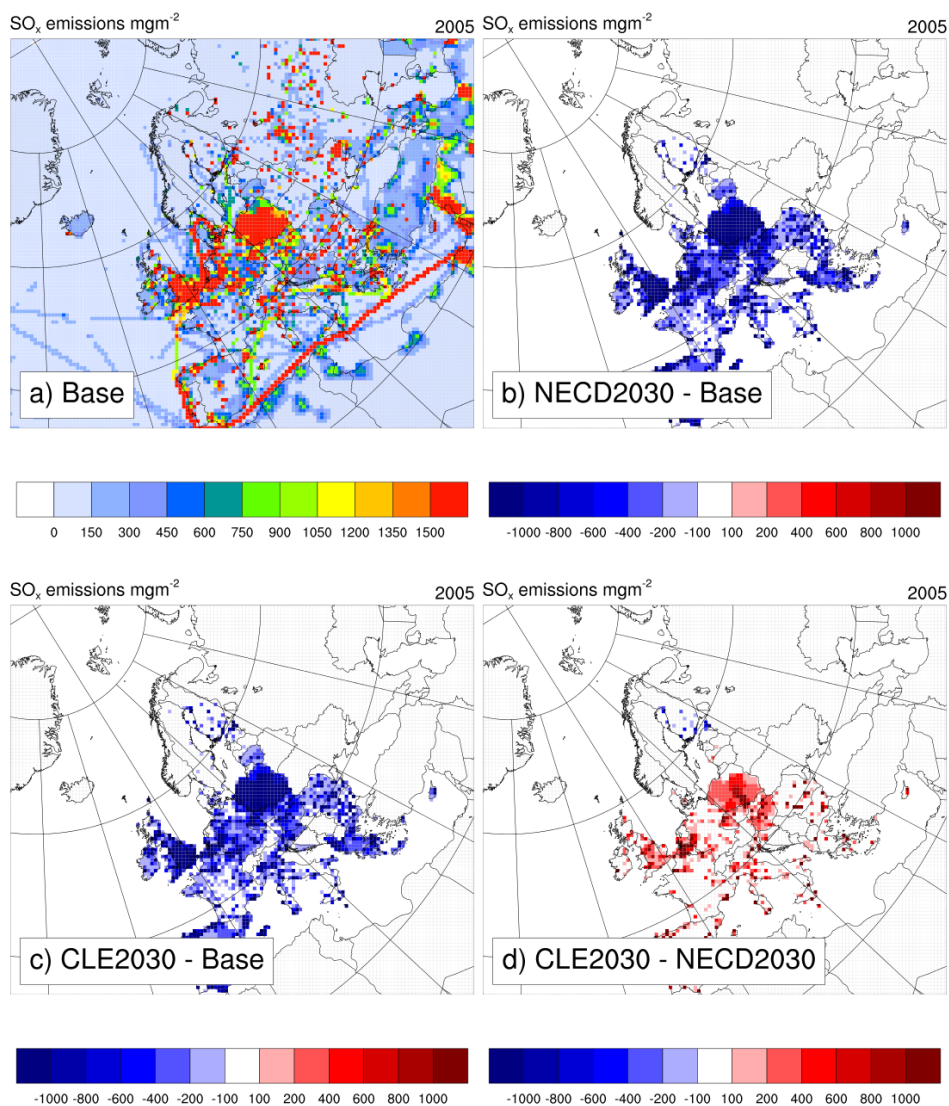


Figure 26. SO_x gridded emissions used in the EMEP4UK model: a) total annual emissions, b) difference between the NECD2030 and baseline 2005, c) difference between CLE2030/UEP45 (UK only) and baseline 2005, and d) difference between CLE2030 and NECD2030.

EMEP4UK 2005

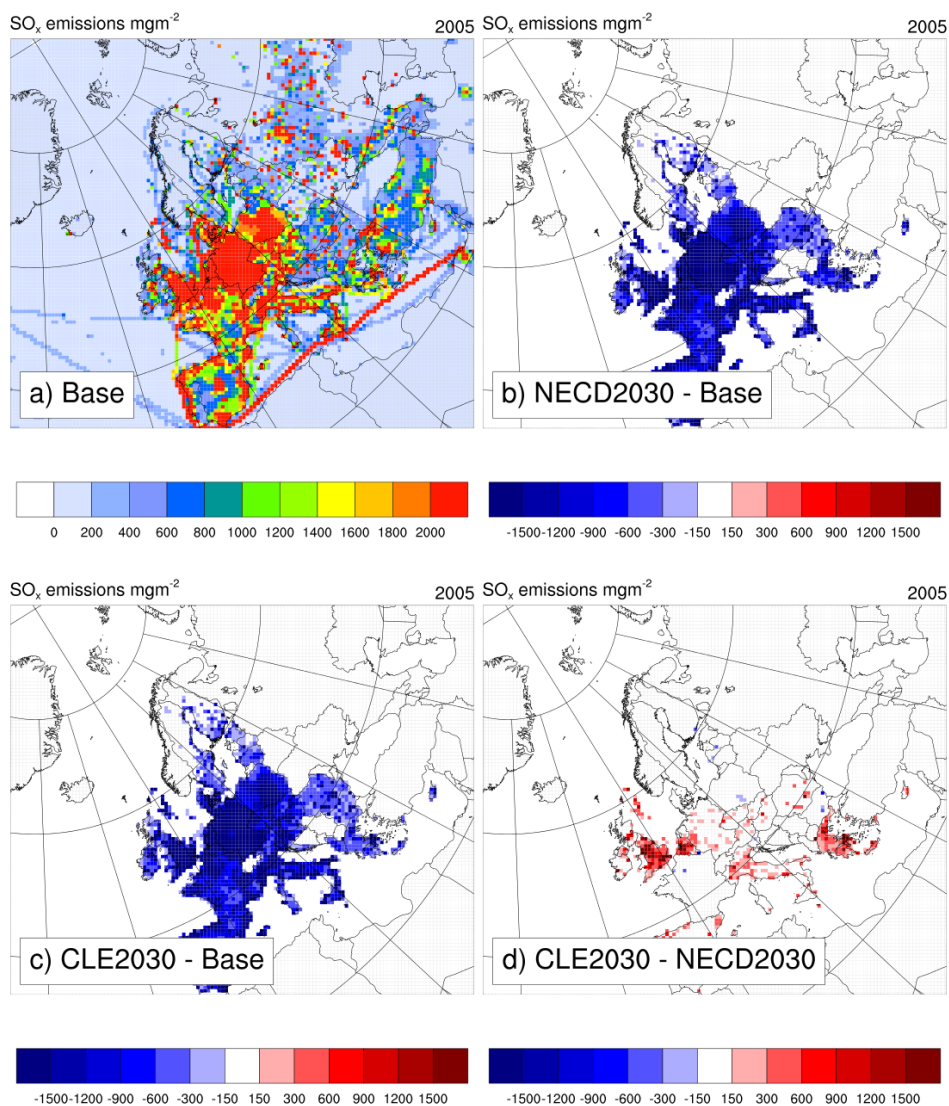


Figure 27. NO_x gridded emissions used in the EMEP4UK model: a) total annual emissions, b) difference between the NECD2030 and baseline 2005, c) difference between CLE2030/UEP45 (UK only) and baseline 2005, and d) difference between CLE2030 and NECD2030.

EMEP4UK 2005

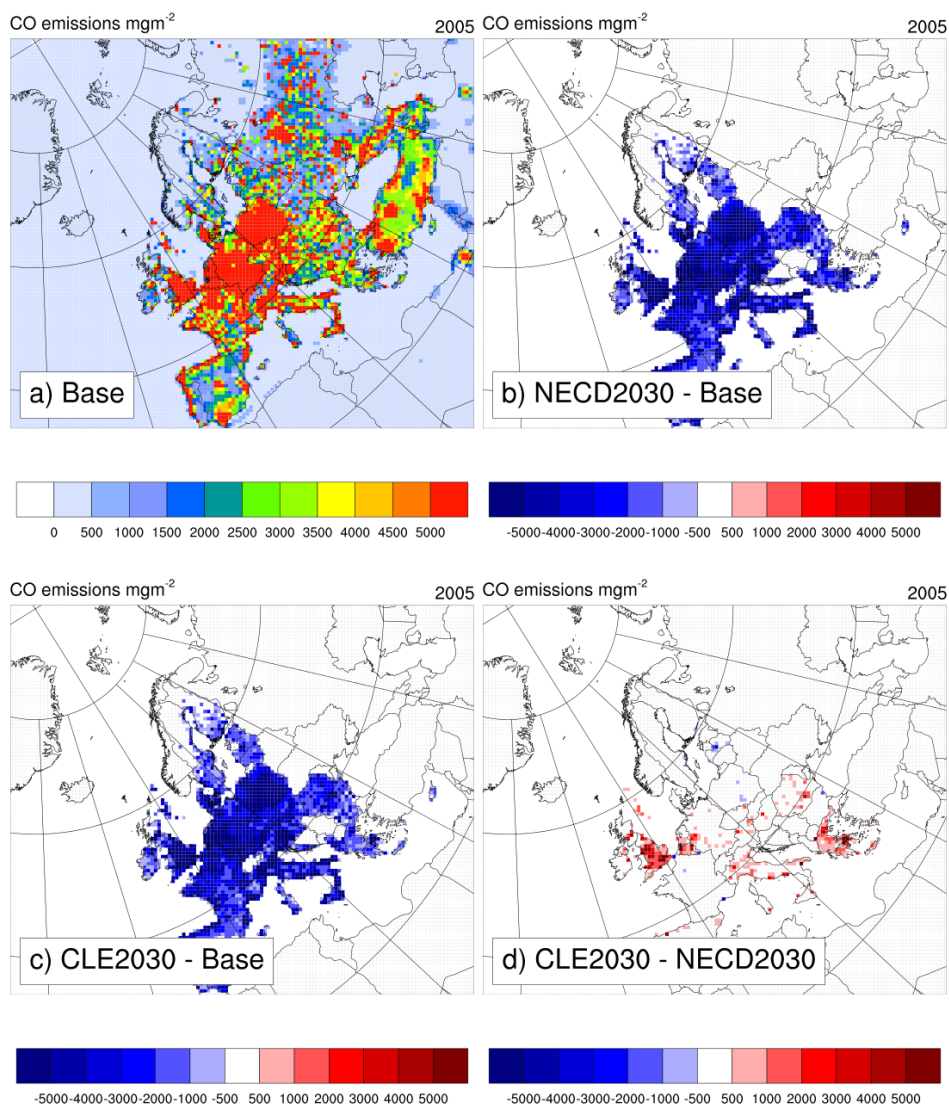


Figure 28. CO gridded emissions used in the EMEP4UK model: a) total annual emissions, b) difference between the NECD2030 and baseline 2005, c) difference between CLE2030/UEP45 (UK only) and baseline 2005, and d) difference between CLE2030 and NECD2030.

EMEP4UK 2005

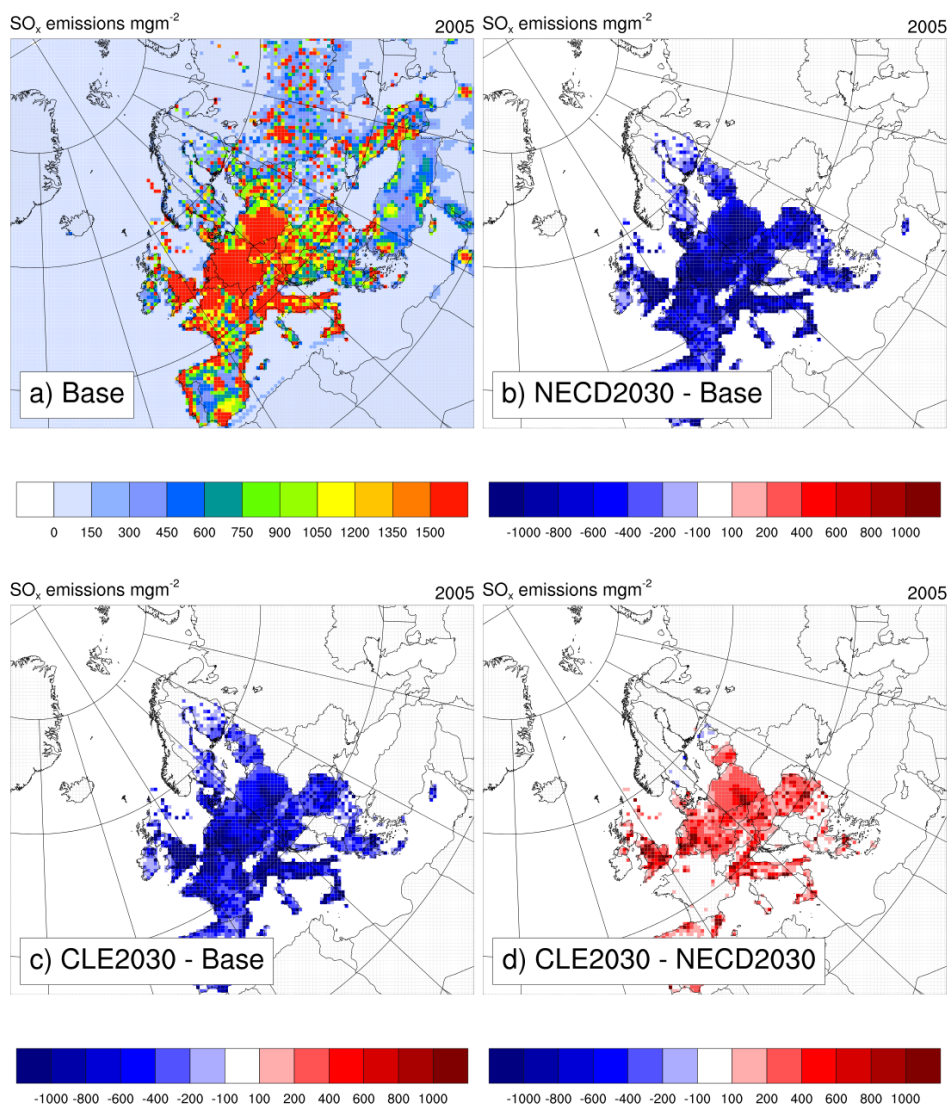


Figure 29. NMVOCs gridded emissions used in the EMEP4UK model: a) total annual emissions, b) difference between the NECD2030 and baseline 2005, c) difference between CLE2030/UEP45 (UK only) and baseline 2005, and d) difference between CLE2030 and NECD2030.

EMEP4UK 2005

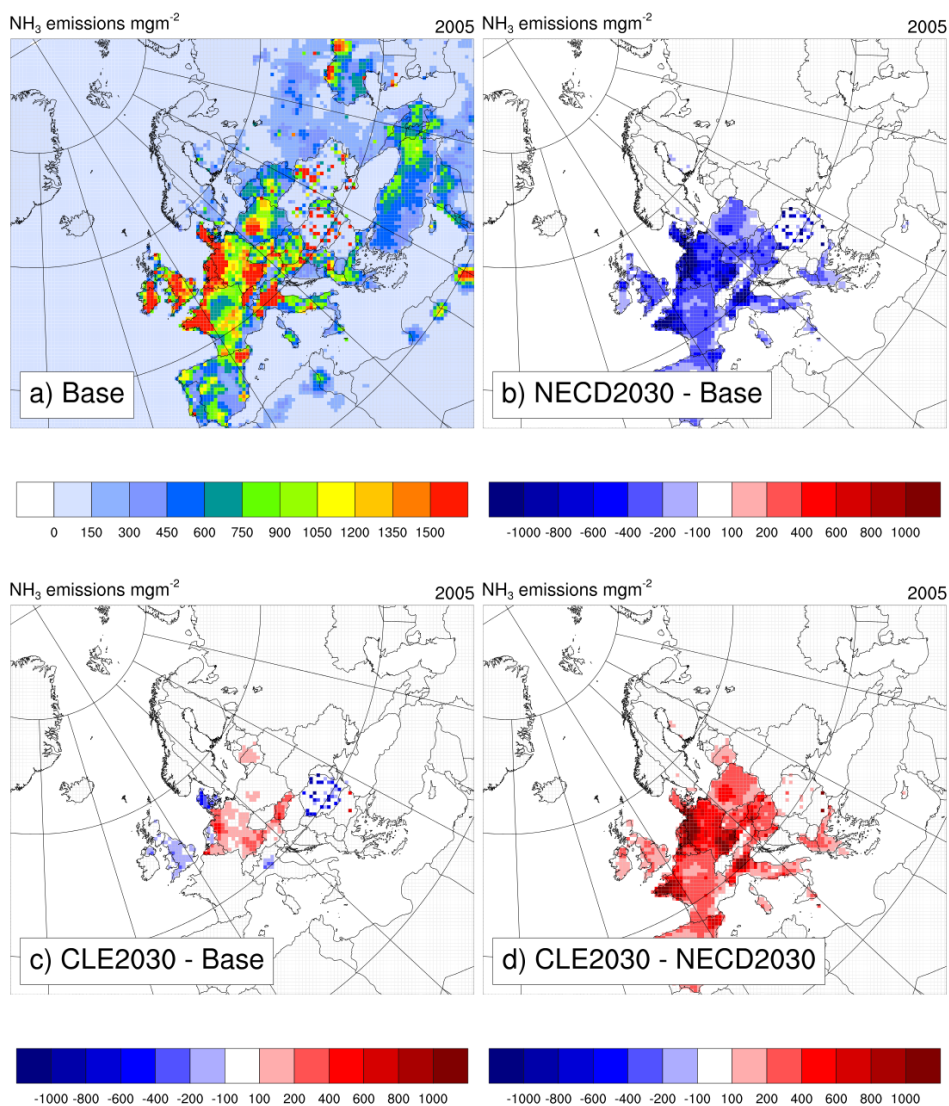


Figure 30. NH₃ gridded emissions used in the EMEP4UK model: a) total annual emissions, b) difference between the NECD2030 and baseline 2005, c) difference between CLE2030/UEP45 (UK only) and baseline 2005, and d) difference between CLE2030 and NECD2030.

EMEP4UK 2005

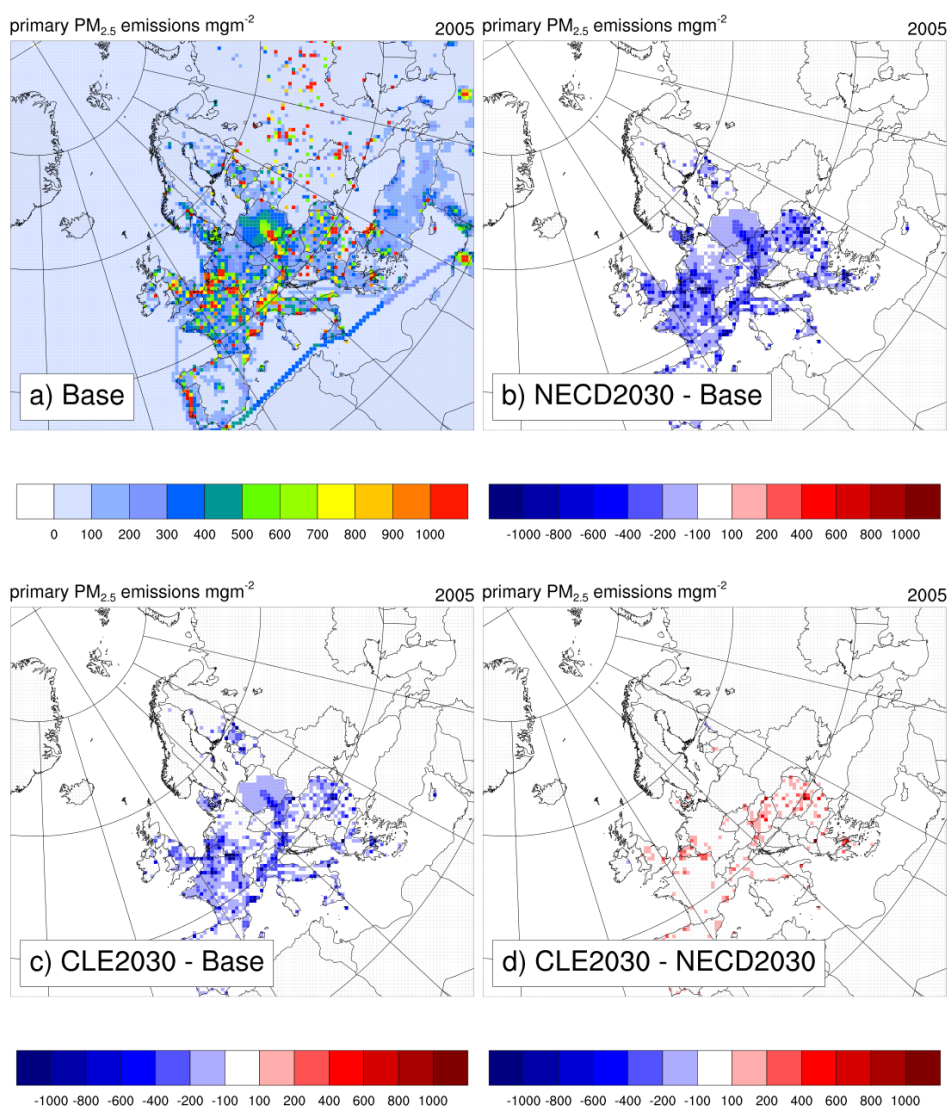


Figure 31. Primary PM_{2.5} gridded emissions used in the EMEP4UK model: a) total annual emissions, b) difference between the NECD2003 and baseline 2005, c) difference between CLE2030/UEP45 (UK only) and baseline 2005, and d) difference between CLE2030 and NECD2030.

EMEP4UK 2005

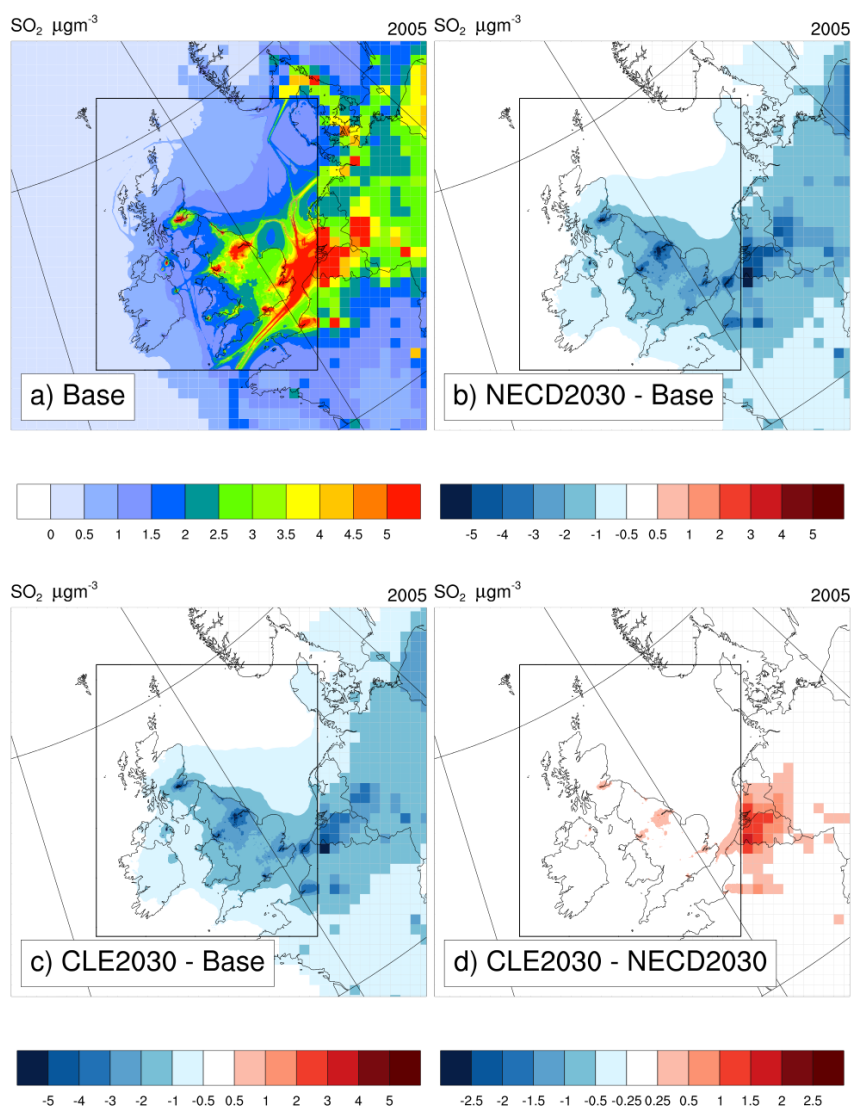


Figure 32. EMEP4UK annual average surface concentration of SO₂ for the UK 5 km x 5 km domain (inside black box) and the European 50 km x 50 km domain (part of the domain shown) used as boundary and initial condition for the UK domain for: a) 2005, b) NECD2030 minus 2005, c) CLE2030 minus 2005, and d) CLE2030 minus NECD2030.

EMEP4UK 2005

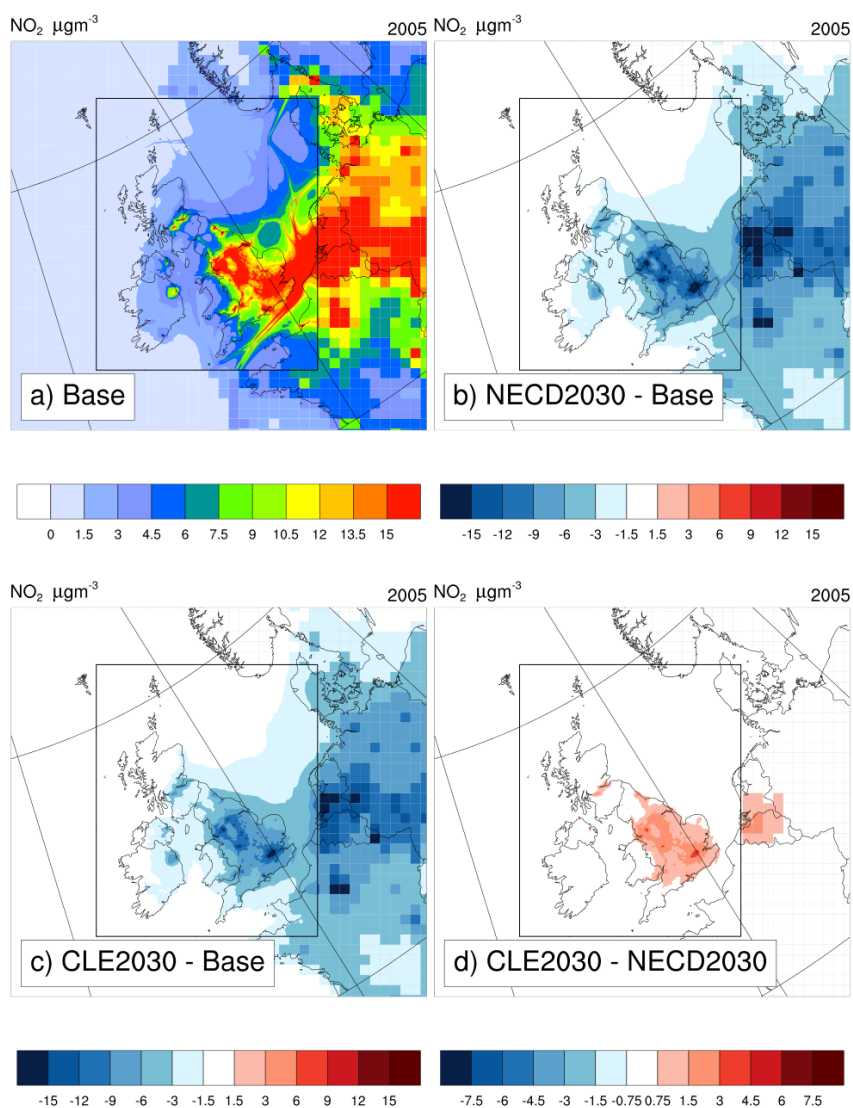


Figure 33. EMEP4UK annual average surface concentration of NO₂ for the UK 5 km x 5 km domain (inside black box) and the European 50 km x 50 km domain (part of the domain shown) used as boundary and initial condition for the UK domain for: a) 2005, b) NECD2030 minus 2005, c) CLE2030 minus 2005, and d) CLE2030 minus NECD2030. All simulations are using a common 2005 meteorology.

EMEP4UK 2005

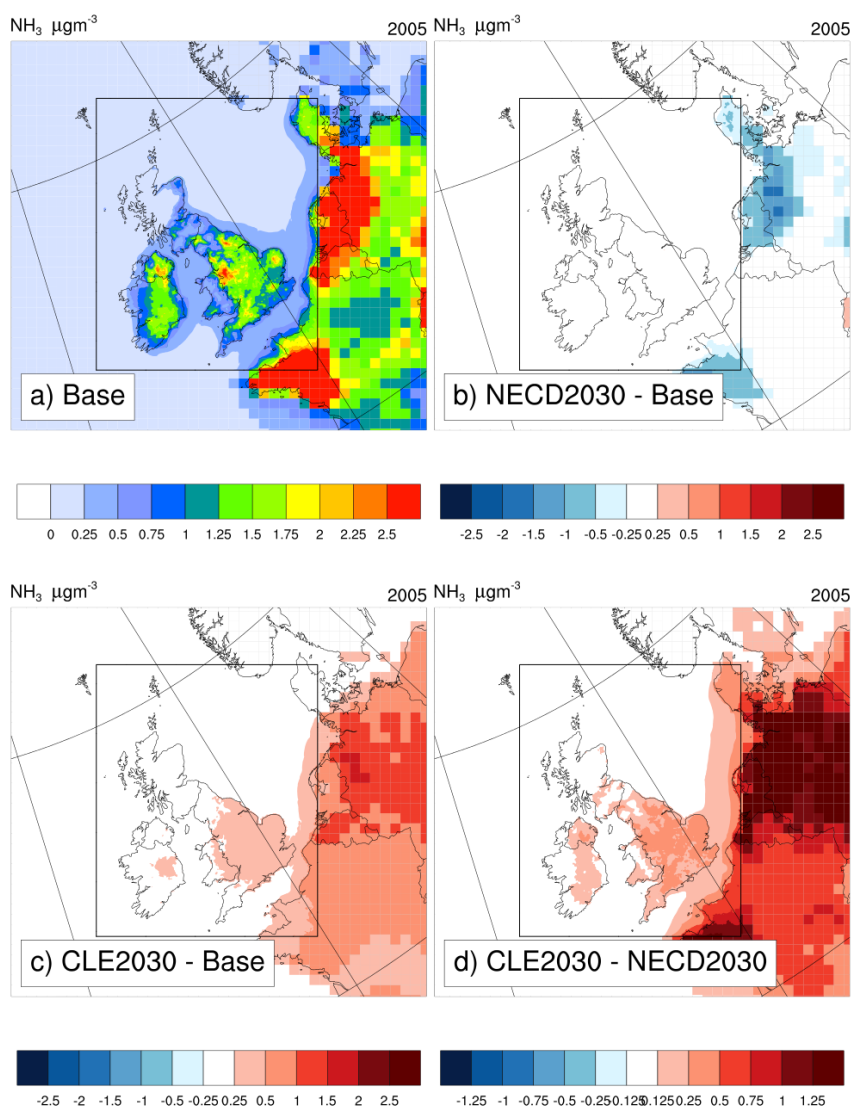


Figure 34. EMEP4UK annual average surface concentration of NH_3 for the UK 5 km x 5 km domain (inside black box) and the European 50 km x 50 km domain (part of the domain shown) used as boundary and initial condition for the UK domain for: a) 2005, b) NECD2030 minus 2005, c) CLE2030 minus 2005, and d) CLE2030 minus NECD2030. All simulations are using a common 2005 meteorology.

EMEP4UK 2005

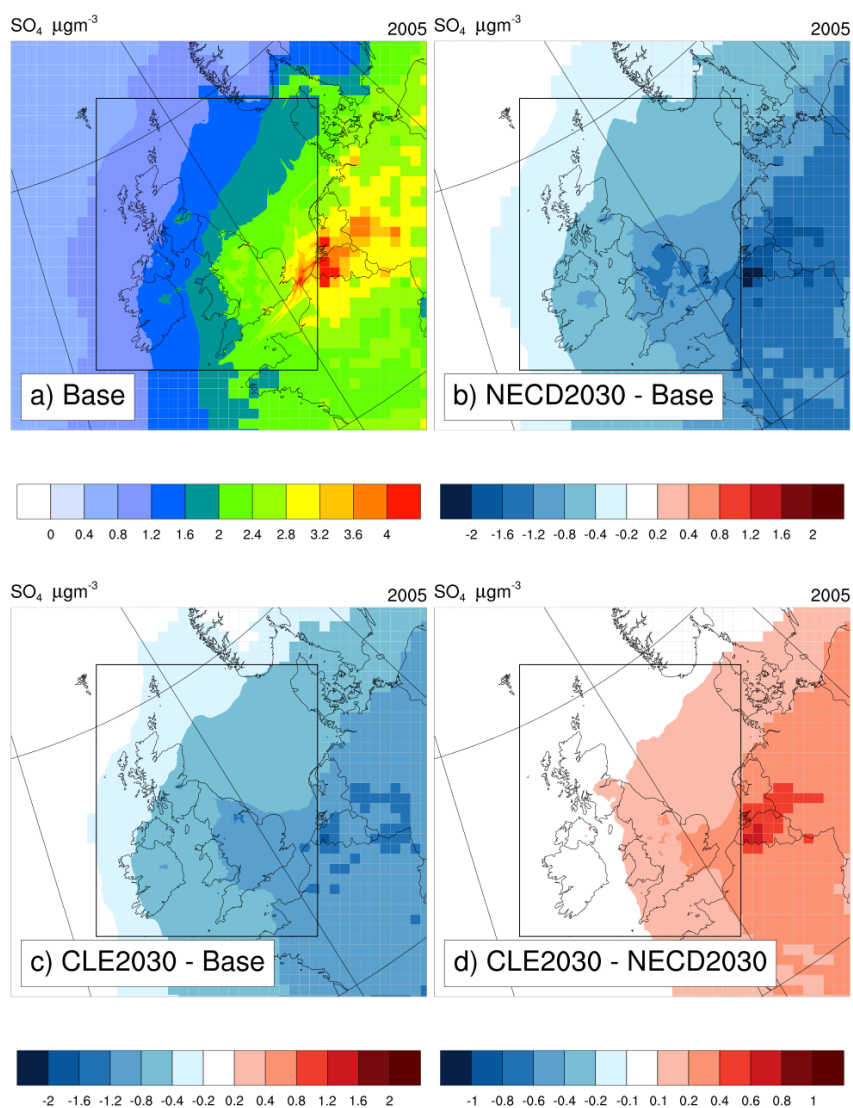


Figure 35. EMEP4UK annual average surface concentration of SO_4 for the UK 5 km x 5 km domain (inside black box) and the European 50 km x 50 km domain (part of the domain shown) used as boundary and initial condition for the UK domain for: a) 2005, b) NECD2030 minus 2005, c) CLE2030 minus 2005, and d) CLE2030 minus NECD2030. All simulations are using a common 2005 meteorology.

EMEP4UK 2005

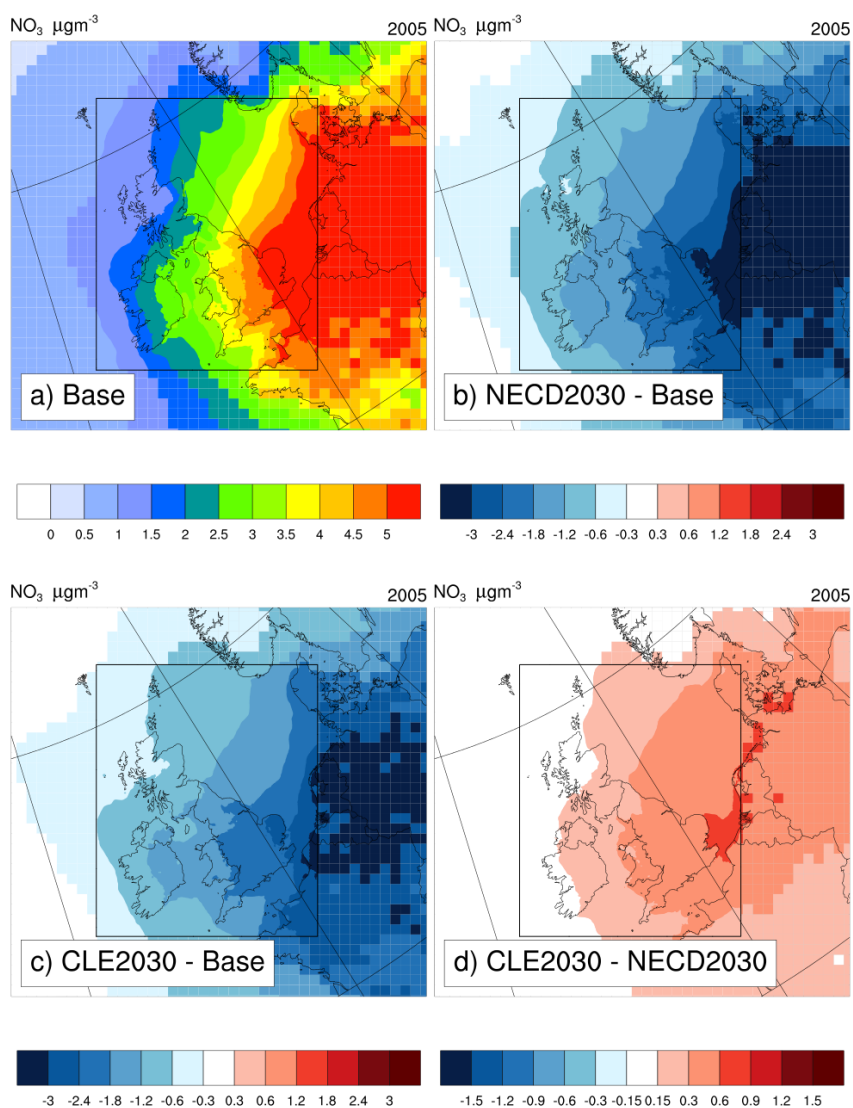


Figure 36. EMEP4UK annual average surface concentration of NO₃ for the UK 5 km x 5 km domain (inside black box) and the European 50 km x 50 km domain (part of the domain shown) used as boundary and initial condition for the UK domain for: a) 2005, b) NECD2030 minus 2005, c) CLE2030 minus 2005, and d) CLE2030 minus NECD2030. All simulations are using a common 2005 meteorology.

EMEP4UK 2005

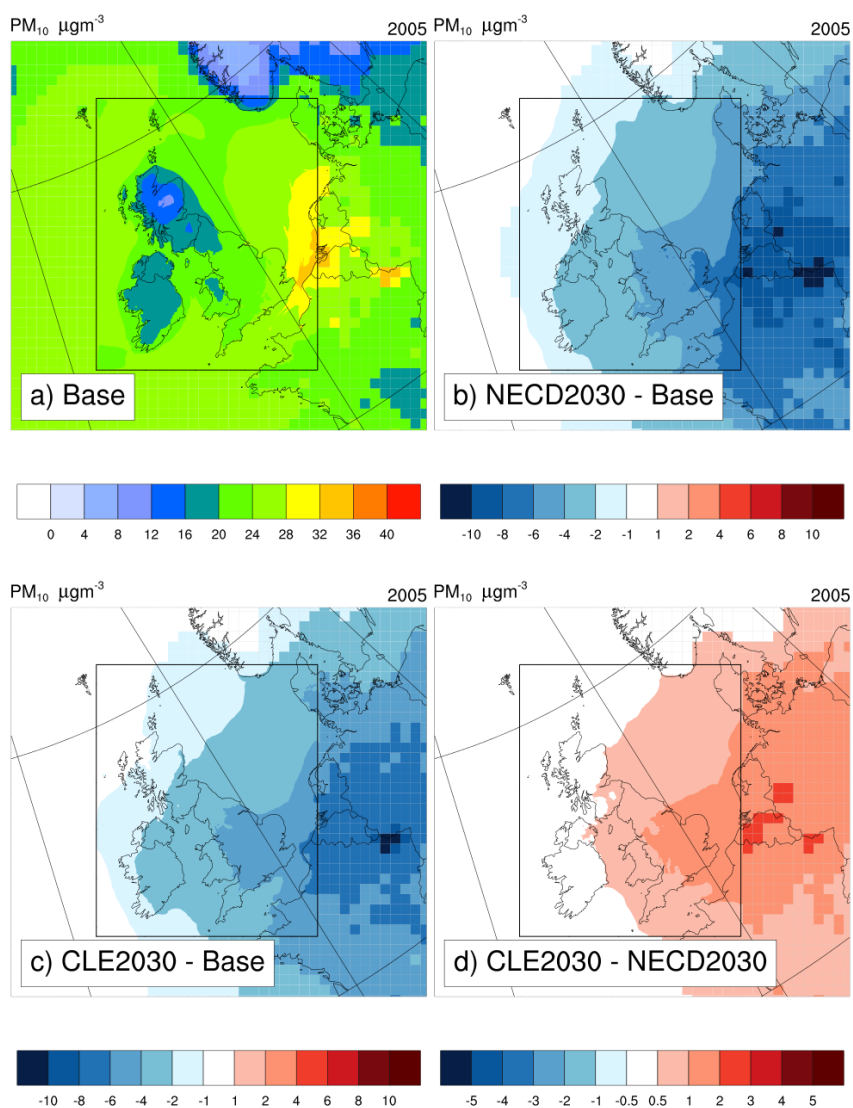
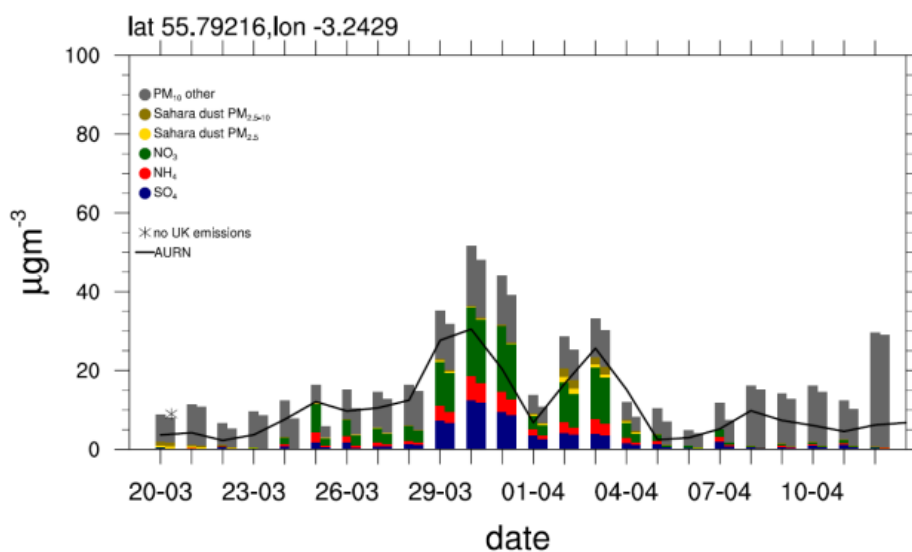


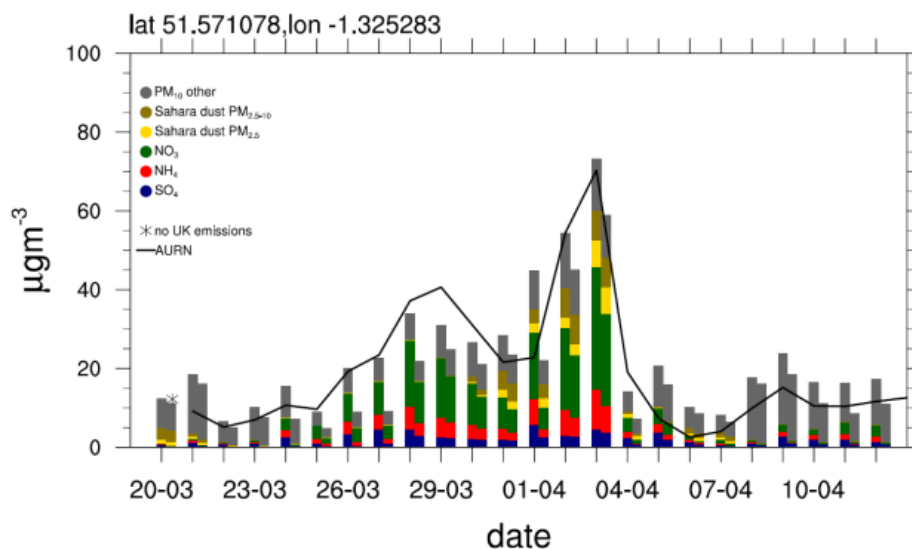
Figure 37. EMEP4UK annual average surface concentration of PM₁₀ for the UK 5 km x 5 km domain (inside black box) and the European 50 km x 50 km domain (part of the domain shown) used as boundary and initial condition for the UK domain for: a) 2005, b) NECD2030 minus 2005, c) CLE2030 minus 2005, and d) CLE2030 minus NECD2030. All simulations are using a common 2005 meteorology.

PM₁₀ Auchencorth Moss



a)

PM₁₀ Harwell



b)

Figure 38. Modelled components of PM₁₀ for two selected AURN sites (all sites available but not shown here): a) Auchencorth Moss (Scotland) and b) Harwell (England). The solid black line shows the observed AURN daily PM₁₀ concentration, whereas the bars show the EMEP4UK daily PM₁₀ components: dark blue SO₄, red NH₄, green NO₃, yellow fine Sahara dust, dark yellow coarse Sahara dust and grey the remaining PM₁₀ components. The period covered is from the 20-Mar-14 until 12-Aug-2014

March - April high PM UK episode daily PM_{2.5} μgm⁻³

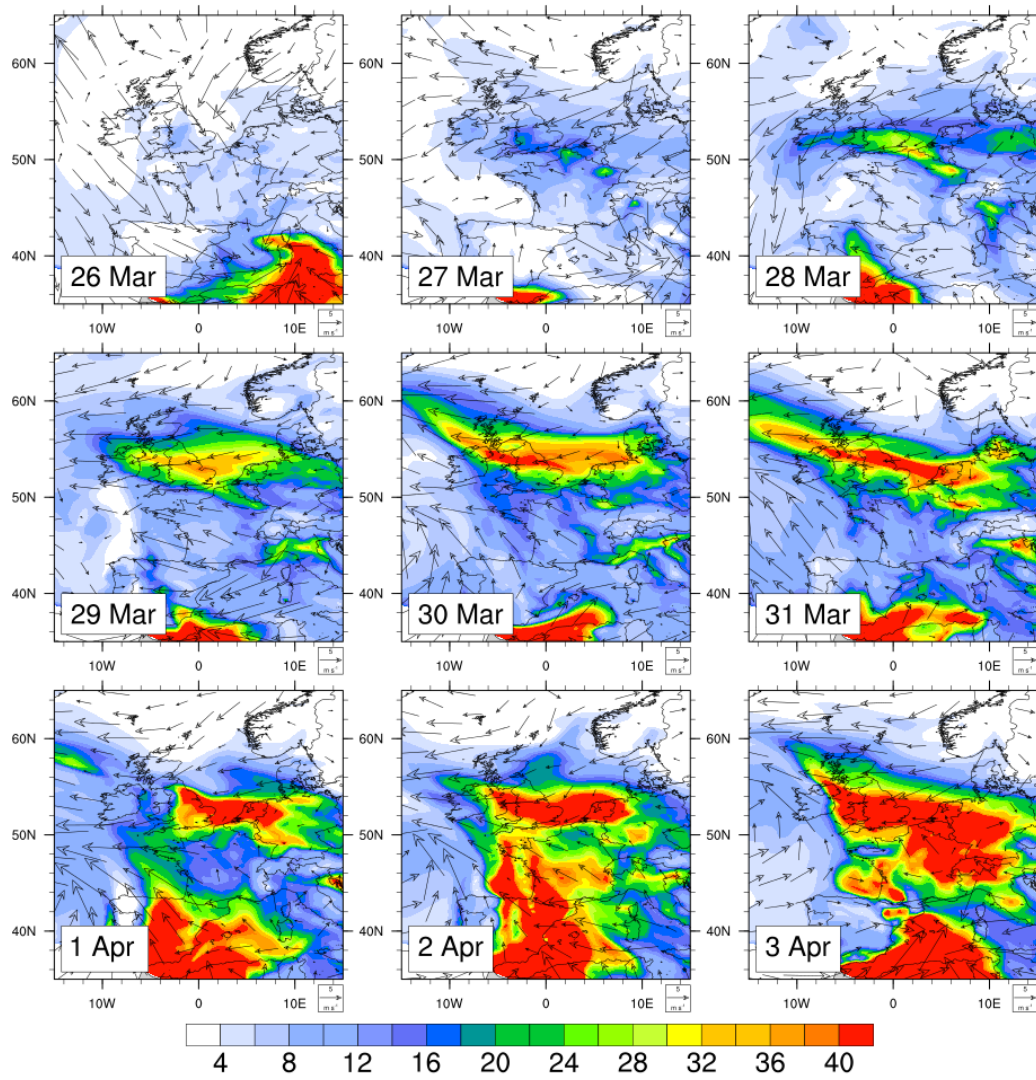


Figure 39. Spring 2014 daily surface concentrations of PM_{2.5} as calculated by the EMEP4UK model rv4.4 50km². The wind speed and direction at 12:00 is shown for each day of the episode. The transboundary contribution and its origins are clearly visible, as well as the varying contributions during the formation of the episode.

3.10. 2011 and 2012 WRF meteorological data (Task 5 and Task 7)

The meteorological data for the years 2011 and 2012 have been calculated with the WRF model version 3.1.1. Any years from 2001-2012 can currently be simulated by the EMEP4UK model, as for all these years WRF meteorological simulations have been run, evaluated and are stored locally, allowing a detailed analysis of changes of atmospheric composition and deposition under several different meteorological conditions as shown in Vieno et al. (2014) and to conduct scenario analyses for policy applications at short notice.

3.11. Uncertainty/Sensitivity analysis of the EMEP4UK model framework (Task 6)

The run time of the most recent production version (rv4.4) of the EMEP4UK model (using 126 processors on the CEH NEMESIS High Performance Computing cluster) is currently ~12 hours for a full simulated year and for the UK domain at a horizontal resolution of 5 km x 5 km. This is very fast in comparison to other Eulerian ACTMs with matching complexity and allows for fast response times and simulation of multiple years, which is essential for the assessment of interannual variability for instance. It does however restrict the ability to perform uncertainty analysis using standard techniques such as Monte Carlo simulations, requiring 100s to 1000s of model simulations to yield robust results. However, the ability of the model to run at different resolutions does allow investigation of how the model calculated surface concentrations and deposition of various pollutants are affected by the change of the model resolution which itself is a key source of uncertainty.

One important aspect of uncertainty in modelling is the choice of grid resolution. To estimate this uncertainty in the EMEP4UK model two approaches are used; perturbation experiments and investigating the changes of model results solely determined by resolution changes. We have explored this and focused on using different horizontal resolutions, as described in the following section.

With funding support both by Defra and the EU FP7 ECLAIRE project, we extended the EMEP4UK model domain to calculate surface concentrations and/or deposition at horizontal scales ranging from 50, 5 to 1km² for the UK, as well as other regions in Europe, e.g. The Netherlands and the Po Valley in Italy. Model resolution has a large influence on pollutants such as ozone where the non-linearity of the NO_x and VOC chemistry may lead to different results when the model is applied at different resolution. In a similar way, the spatio-temporal heterogeneity of NH₃ emission patterns and the high rate of near-source deposition results in a strong influence of grid resolution on effects, e.g. the deposition of S and N on SSSIs or

SACs. **Figure 40** shows the NO_x emissions for Central Scotland to highlight the difference in horizontal distribution of emission patterns when the model is applied at these different scales.

The implication of this is that even before any chemical conversion takes place in the model, large parts of the model domain overestimate (in rural areas, predominantly) or, in the case of urban areas, underestimate the NO_x emissions which affects not only the surface concentrations of NO_x, but also the generation or titration of surface ozone. In **Figure 41** EMEP4UK calculated annual average surface ozone concentrations are shown for each model resolution. The Scottish central belt shows a change in annual average ozone surface concentrations from ~70 µg m⁻³ (at 50 km x 50 km) to ~ 60 µg m⁻³ (at 1 km x 1 km). This difference of up to 10 µg m⁻³ is a direct result of a change in model resolution and may have clear implications for the model-based assessment of attainment of limit values, or human exposure and health effects.

Moreover, the higher resolution better represented the mountain terrain in the Scottish highlands. The 1km² Scottish domain better resolved the higher ozone concentrations over elevated terrain with a difference of up to 20 µg m⁻³.

Figure 42 shows the annual average surface concentration of ozone calculated at the 3 horizontal resolutions (50, 5 and 1km²). The EMEP4UK model and WRF model version are kept unchanged for each model resolution. All the interpolation and extrapolation needed for the model input file are calculated using either online conversion in the EMEP4UK model or the ad-hoc pre-processors developed for the EMEP4UK model.

The major driver of differences in the model results when applied at different resolutions are: the changes of chemical regime (ozone non linearity vs. NO_x), emissions spatial distribution, and meteorological parameters such as rainfall.

The changes in surface concentrations can help to quantify the uncertainties of the operational EMEP4UK model when run at 5 km x 5 km horizontal resolution. **Figure 43** shows two important issues when a model, such as the EMEP4UK or any other gridded model, is used to extract the surface concentration from a specific location. Firstly the site location may be in the corner of a grid making the site location not necessarily the most representative for the grid average, and secondly the resolution affects the model calculated surface ozone as demonstrated above.

Figure 43 shows a zoomed in section for **Figure 42** for the 2008 annual average surface concentration of ozone covering an area of $\sim 20 \text{ km}^2$ near Aberdeen, UK. In this specific example the surface concentration changes from about 34 to 31 and 32 $\mu\text{g m}^{-3}$, for the 50, 5 and 1 km x 1 km model resolution, respectively.

The removal process such as dry deposition, for example for NH_x , is fairly similar at the different scales (in the same zoomed area) as shown **Figure 44**. However, wet deposition can change drastically between resolutions as the high rainfall is as expected to be co-located with the mountain terrain which is better resolved as the model resolution increases (**Figure 45**).

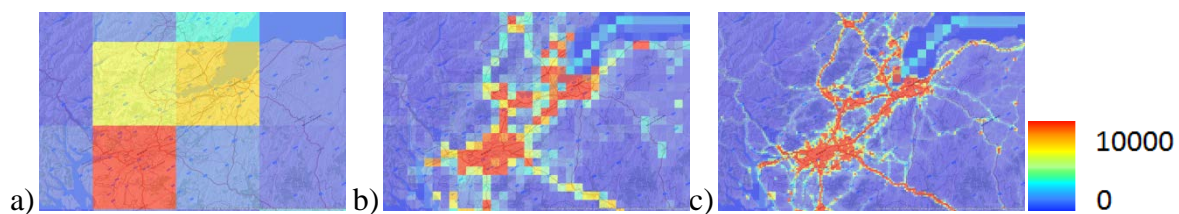


Figure 40. 2008 NO_x emission estimates for the Scottish central belt (Glasgow–Edinburgh) aggregated to different model resolutions, resp. at original NAEI resolution: a) 50 km x 50 km, b) 5 km x 5 km and c) 1 km x 1 km. Units are mgN m^{-2} .

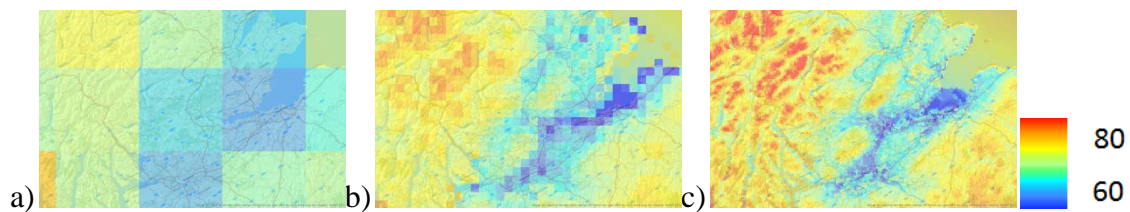


Figure 41. Surface ozone calculated by the EMEP4UK model at different resolutions: a) 50 km x 50 km, b) 5 km x 5 km and c) 1 km x 1 km. Units are ppb.

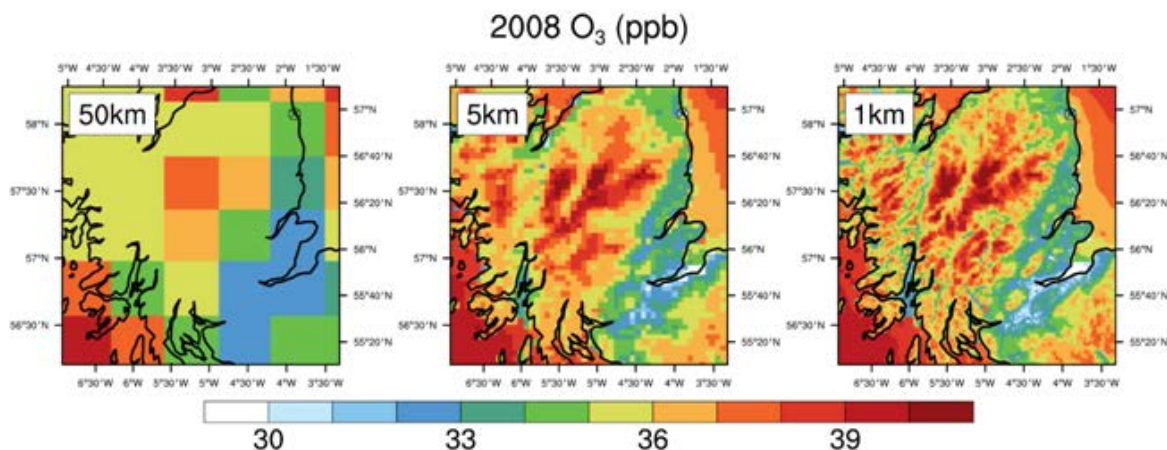


Figure 42. 2008 annual average surface concentration of ozone for an area of the EMEP4UK model for three horizontal resolutions: 50, 5 and 1 km x 1 km.

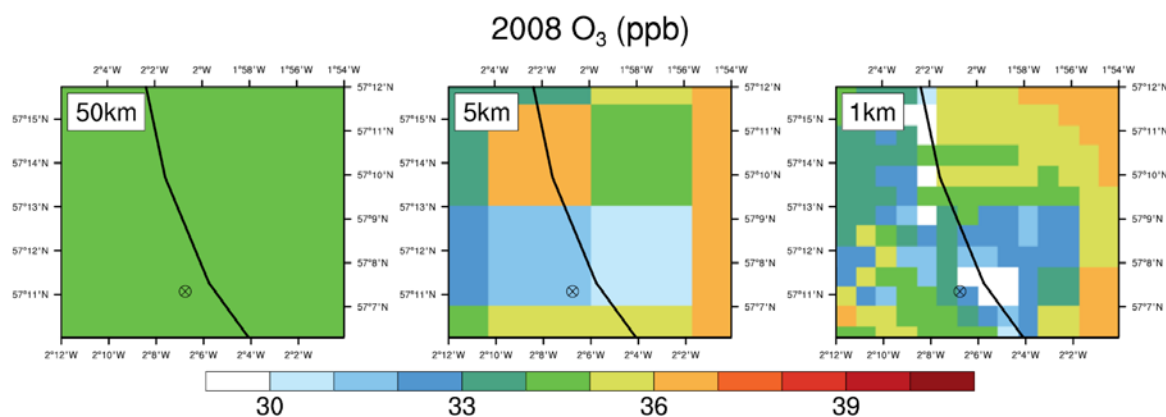


Figure 43. 2008 annual average surface concentration of ozone for an area of the EMEP4UK model covering 20 km² near Aberdeen UK (black crossed circle) for three horizontal resolutions: 50, 5 and 1 km x 1 km. The black line crossing the figure is the coast line (sea on the right in each panel).

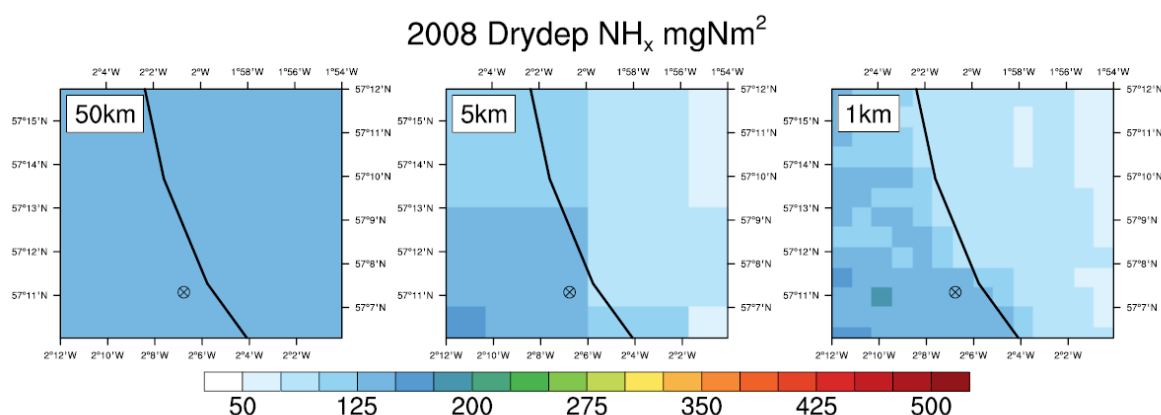


Figure 44. 2008 annual total dry deposition for reduced nitrogen for an area of the EMEP4UK model covering 20 km² near Aberdeen UK (black crossed circle) for three horizontal resolutions: 50, 5 and 1 km x 1 km

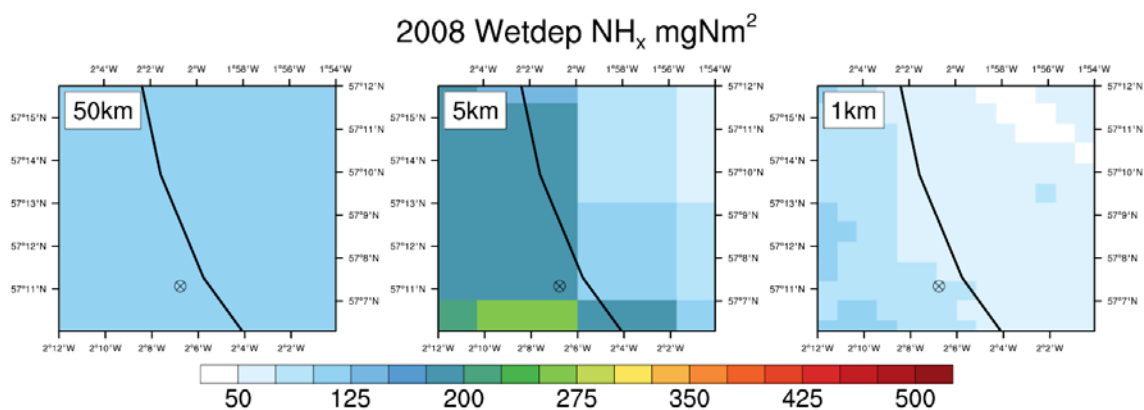


Figure 45. 2008 annual total wet deposition for reduced nitrogen for an area of the EMEP4UK model covering 20 km² near Aberdeen UK (black crossed circle) for three horizontal resolutions: 50, 5 and 1 km x 1 km

3.12. Extending the EMEP4UK model to secondary organic aerosols and exploring other chemical schemes (i.e. CRI). This will include inter-comparison and knowledge exchange with other UK and international research groups which use different model such as WRF-CHEM, CMAQ and UKCA (Task 8 and Task 9)

3.12.1. The EMEP4UK model to secondary organic aerosols scheme

The update of the EMEP4UK model to version rv4.4 includes the secondary organic aerosol (SOA) scheme. In this version (rv4.4) the model is now capable to explicitly simulate all major components of PM_{2.5} (primary PM_{2.5}, secondary inorganic aerosols, secondary organic aerosols, Sahara dust, road dust, and PM_{2.5} from forest fires). Emissions from forest fires have also been included in the EMEP4UK model in order to better represent the atmospheric chemistry in Europe and specifically in the UK. Daily emissions from forest and vegetation are taken from the Fire INventory from NCAR version 1.0 (FINN). The forest fire emissions are available for the years 2002-2013.

The primary PM emissions are speciated into elemental carbon (EC), primary organic aerosols (POA) from fossil fuel combustion, POA from domestic combustion, and the remaining primary PM are assigned as primary PM by emission source sectors as detailed by Kuenen et al. (2014). Biogenic emissions of VOCs such as isoprene and monoterpenes are calculated online by the model for every grid cell and time-step (Simpson, 1995).

The SOA formation in the EMEP4UK model is computed using the *volatility basis set* (VBS) approach (Donahue et al., 2006), which was first implemented into the EMEP MSC-W model by Bergström et al. (2012). The default EMEP MSC-W model only includes non-

volatile POA emissions and the SOA production is only from anthropogenic and biogenic VOCs using the 5 VBS for the semi-volatility VOCs (SVOC) as shown in **Figure 46**.

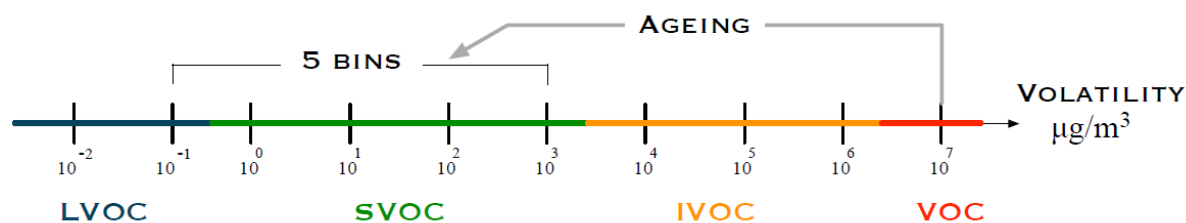


Figure 46. Volatility basis set used in the EMEP4UK model.

The EMEP4UK model has since been further extended to use 9 VBS (Bergström et al., 2012) with saturation concentrations ranging from $10^{-2} \mu\text{g m}^{-3}$ for low volatility VOCs (LVOC) to $10^6 \mu\text{g m}^{-3}$ for the intermediate volatility VOCs (IVOC) $\mu\text{g m}^{-3}$ as shown in **Figure 46**. In the 9 VBS the POA are emitted into the LVOC, SVOC and IVOC as discussed in Shrivastava et al. (2008).

The EMEP4UK model SOA default 5 VBS schemes called “Base” and an experimental application which uses the extended 9 VBS SOA scheme, plus an emission scenario where the NAEI emissions from solid fuel combustion are redistributed into urban areas to simulate urban solid fuel emissions. This approach has been taken since the official NAEI data for solid fuel emissions show low emissions in urban areas reflecting compliance with the enforced smoke free areas in most city centres. The experiment has been labelled as “SF_SV-POA” (Solid Fuel/Semi-volatile POA). The model has been used to calculate the annual average surface concentration of organic aerosols (OA) at the ClearFlo London site (Young et al., 2015; Bohnenstengel et al., 2014); which is deemed representative of an atmospheric chemical composition at an urban background site.

In **Figure 47** the measurement data from the aerosol mass spectrometer (AMS) deployed in ClearFlo are compared with EMEP4UK model annual average results for 2012 for a) the Base scenario and b) the SF_SV-POA scenario.

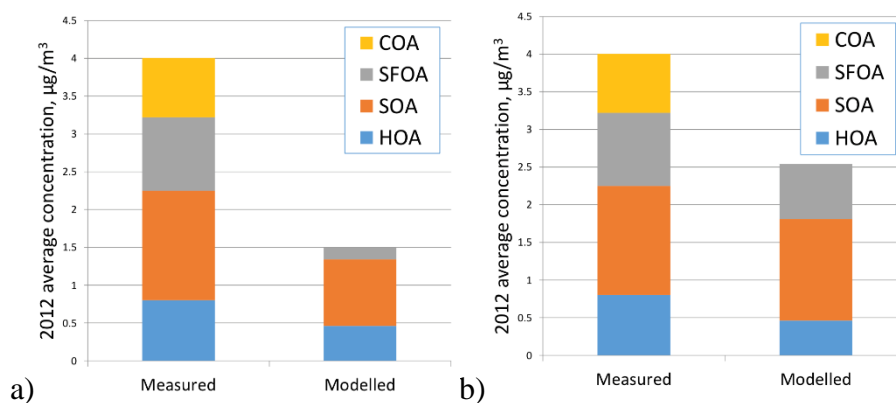


Figure 47. Annual average concentrations of measured and modelled OA components at an urban background site in London. COA - cooking organic aerosol, SFOA - solid fuel organic aerosol, SOA - secondary organic aerosol and HOA - hydrocarbon-like organic aerosol (corresponding to POA from fossil fuel combustion).

The preliminary results of the SF_SV-POA experimental model run shows a better agreement with the observations, however, the OA created by emissions from cooking operations (COA) (Figure 47) is to date not included in the official NEAI emissions inventory and therefore not represented by the model.

3.12.2. The EMEP4UK CRI scheme implementation

The Common Representative Intermediates (CRI) mechanism (Jenkin et al., 2008) version 2 R5 (CRIV2 R5) (Watson et al., 2008) has been implemented into the EMEP4UK model version rv4.4. The EMEP4UK with the CRIV2R5 chemical mechanism has been used to analyse changes in surface ozone between the year 1998 and 2008 (von Schneidmesser et al., 2014).

The EMEP4UK results calculated using the CRIV2 R5 chemical scheme have been submitted to the Defra MIE as an additional EMEP4UK model run. In the MIE the EMEP4UK base run and the CRI dataset have been compared with the WRF-Chem, CMAQ, NAME and AQUM. The results of the inter-comparison have been published in the Defra MIE⁹ reports (Carslaw, 2011a).

A set of model tests have been set up to verify differences in the predicted ozone surface concentrations between the standard EMEP chemistry and when using the enhanced CRIV2 R5. The run time of the EMEP4UK-CRI is in the order of ~2-3 times slower, compared with the standard EMEP4UK setup, which is still very fast for a regional ACTM (for a one year simulation, the EMEP4UK-CRI the run time is ~18 hours compared to 7 hours for a standard EMEP4UK run, using 128 processors).

⁹ <http://uk-air.defra.gov.uk/research/air-quality-modelling?view=intercomparison>

As an example, **Figure 48**, **Figure 49**, and **Figure 50** show the results of the model run using the CRIrv2 chemical scheme compared with the default chemical scheme used in the EMEP MSC-W and EMEP4UK model. From this first analysis the enhanced CRI scheme seems to show higher reactivity compared with the default scheme and is better at representing lower ozone concentrations.

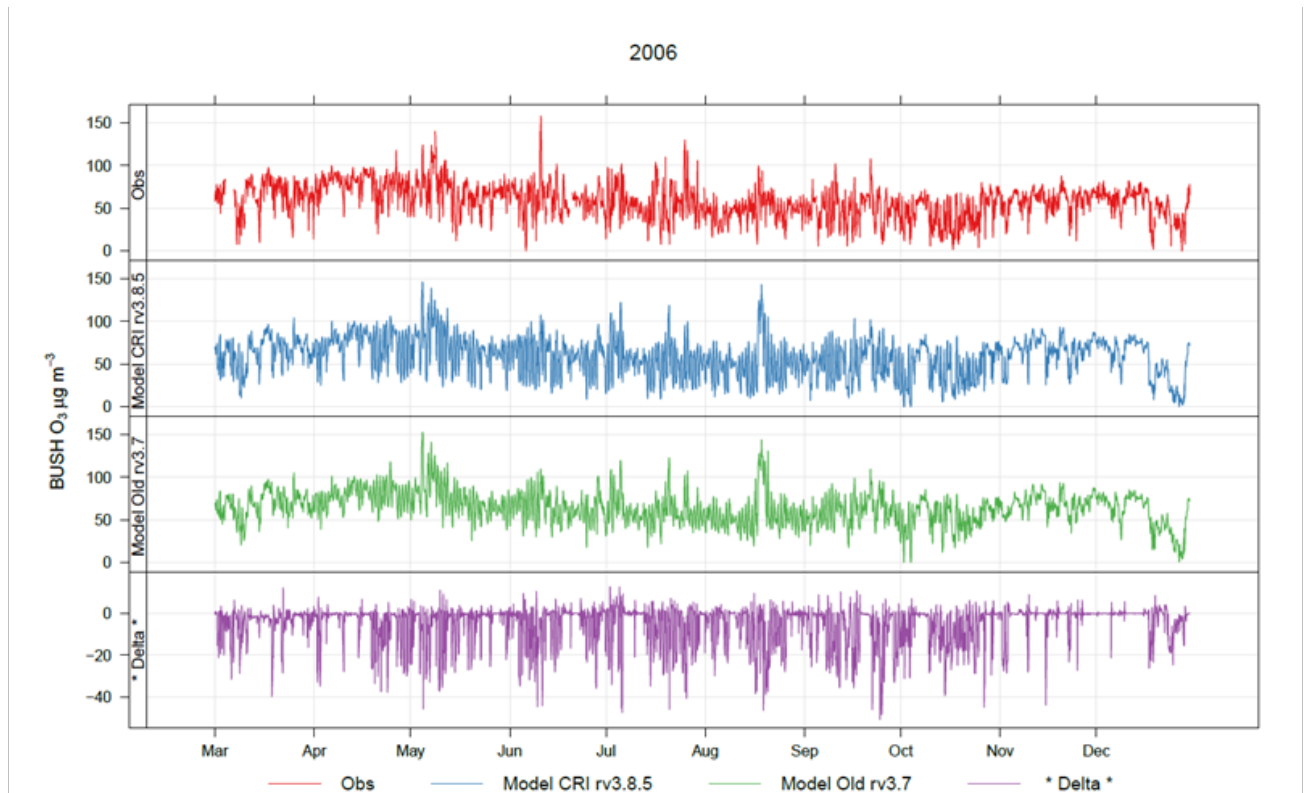


Figure 48. EMEPUK calculated ozone surface concentration for the year 2006 (Mar-Dec) and the AURN observations for the *Bush Estate* site. Where Model CRI rv3.8.5 is the new updated EMEP4UK model with the CRIrv2 chemical scheme and model “Old rv3.7” is the default EMEP4UK model

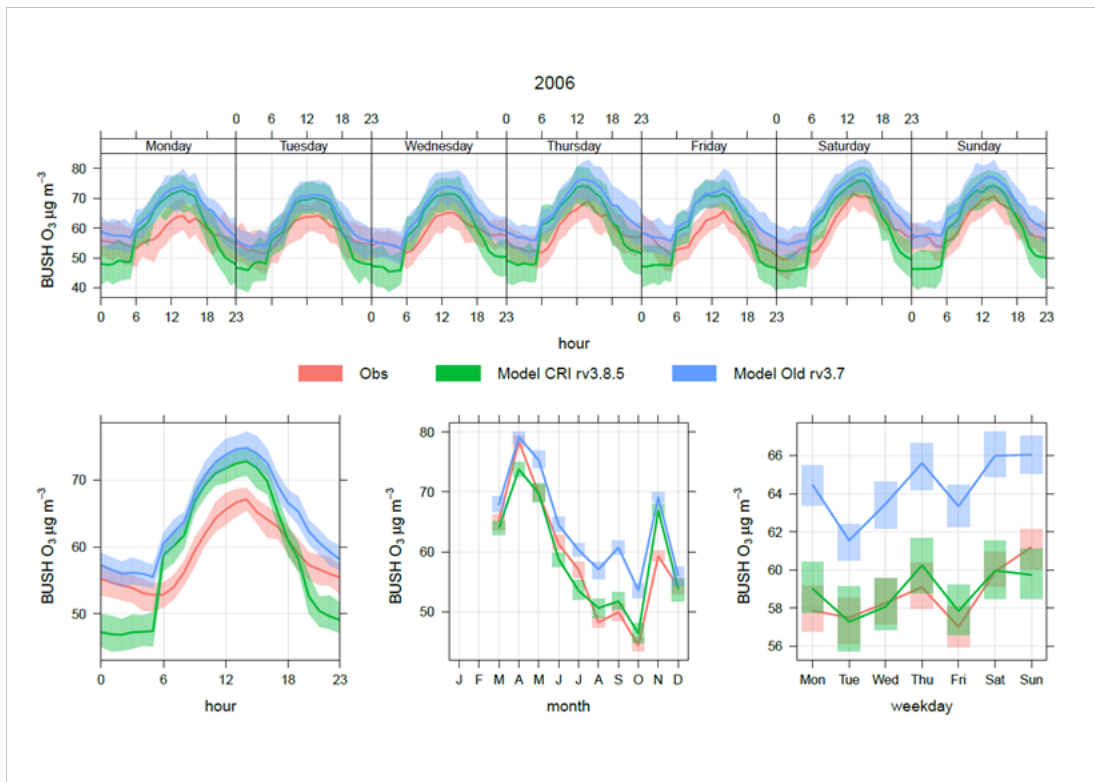


Figure 49. EMEPUK (CRI and standard model) calculated ozone surface concentration for the year 2006 (Mar-Dec) and the AURN observations for the *Bush Estate* site. Daily, monthly and weekly analysis.

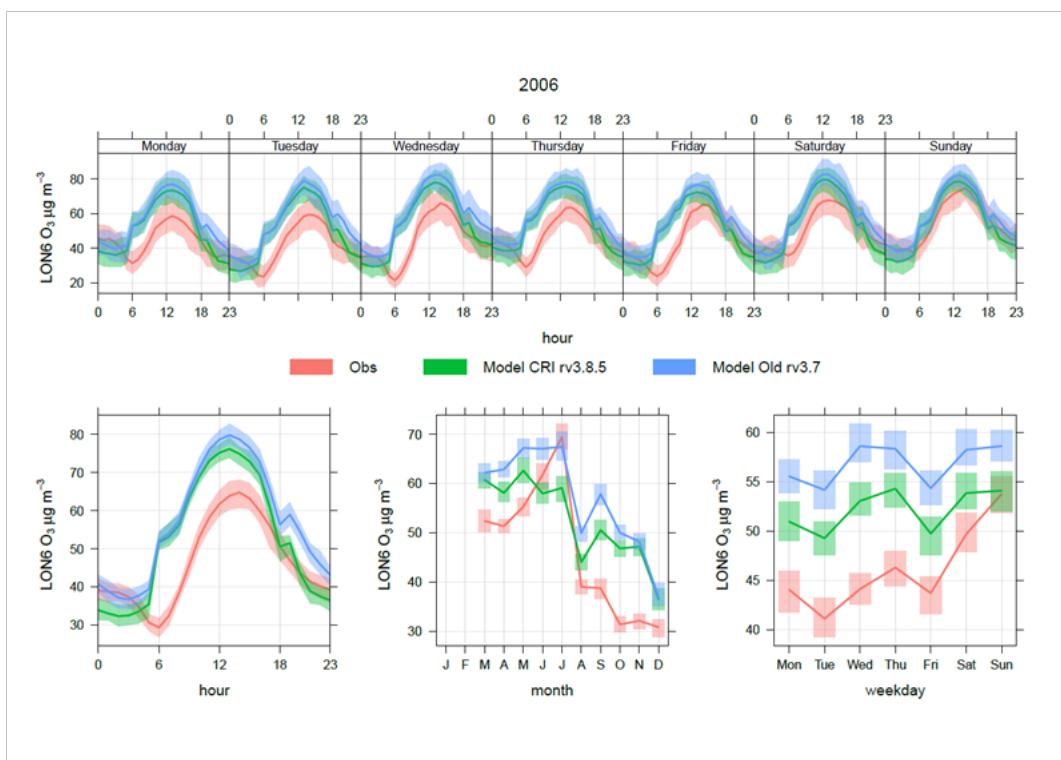


Figure 50. EMEPUK (CRI and base standard model) calculated ozone surface concentration for the year 2006 (Mar-Dec) and the AURN observations for the *London Eltham* site. Daily, monthly and weekly analysis.

3.13. UK air quality Sensitivity to long-range transport of secondary pollutant including secondary inorganic aerosols and secondary organic aerosols (Task 9)

Over the UK, the modelled contribution of the secondary organic aerosol component to total PM_{2.5} concentrations is found to be small in comparison to the contribution from secondary inorganic aerosols. Evidence from observations, however, suggest missing emissions from solid fuel and cooking oils (see section 3.10). The focus here is on the contribution from secondary inorganic aerosols.

The EMEP4UK surface concentrations of a number of pollutants, including NO₂, SO₂ and NH₃, and particle NO₃⁻, SO₄²⁻ and NH₄⁺, were simulated for the whole decade 2001-2010. To quantify the influence of long-range (i.e. non-UK, or ‘transboundary’) and short-range (UK, ‘domestic’) emissions on the UK surface concentrations of these components, a perturbation experiment was carried out by setting UK land emissions to zero for the respective year.

The observations from the UK Acid Gases and Aerosols Network (AGANet) are used to validate the model for the pollutants listed above. The size cut-off of the DELTA sampler has been estimated to be ~4.5 µm (Tang et al., 2009), therefore the measured concentrations are between the PM_{2.5} and PM₁₀ size fractions. The EMEP4UK model assigns all SO₄²⁻ and NH₄⁺ components to PM_{2.5}. Modelled NO₃⁻ is assigned to both PM_{2.5} and PM_{coarse} which leads to potential negative bias in modelled versus measured concentrations for NO₃⁻.

Surface concentrations of secondary inorganic particle components over the UK have been analysed for 2001-2010 using the EMEP4UK regional atmospheric chemistry transport model and evaluated against measurements. The model simulations were able to accurately represent both the long-term decadal surface concentration trends of particle sulphate and nitrate and an episode in early 2003 of substantially elevated nitrate concentrations measured across the UK by the AGANet network.

The 2003 episode was identified as consisting of three separate phases, each of less than 1 month duration, in February, March and April. The primary cause of the elevated nitrate levels across the UK was meteorological: a persistent high pressure system with varying location impacted on the relative importance of transboundary versus domestic emissions. Whilst long-range transport dominated the elevated nitrate levels in February, domestic emissions mainly contributed to the March phase, while for the April phase, both domestic emissions and long-range transport contributed. A prolonged episode such as the one in early

2003 can have substantial impact on annual average concentrations and the exceedance of daily limit values set by the European Commission. The episode led to annual concentration differences at the regional scale of similar magnitude to those driven by long-term changes in precursor emissions over the full decade investigated here. This work has been published in Vieno et al. (2014) and *Figure 51* shows an extract this publication where the AGANet monthly observations are compared with the EMEP4UK monthly average surface concentrations of NO_3^- for 2001-2010 at four sites of the AGANet network: *Strathvaich Dam* (north-west Scotland), *Bush* (central Scotland), *Rothamsted* (south-east England), and *Yarner Wood* (south-west England). All available AGANET site are compared with the EMEP4UK model, but not shown here and are available on request.

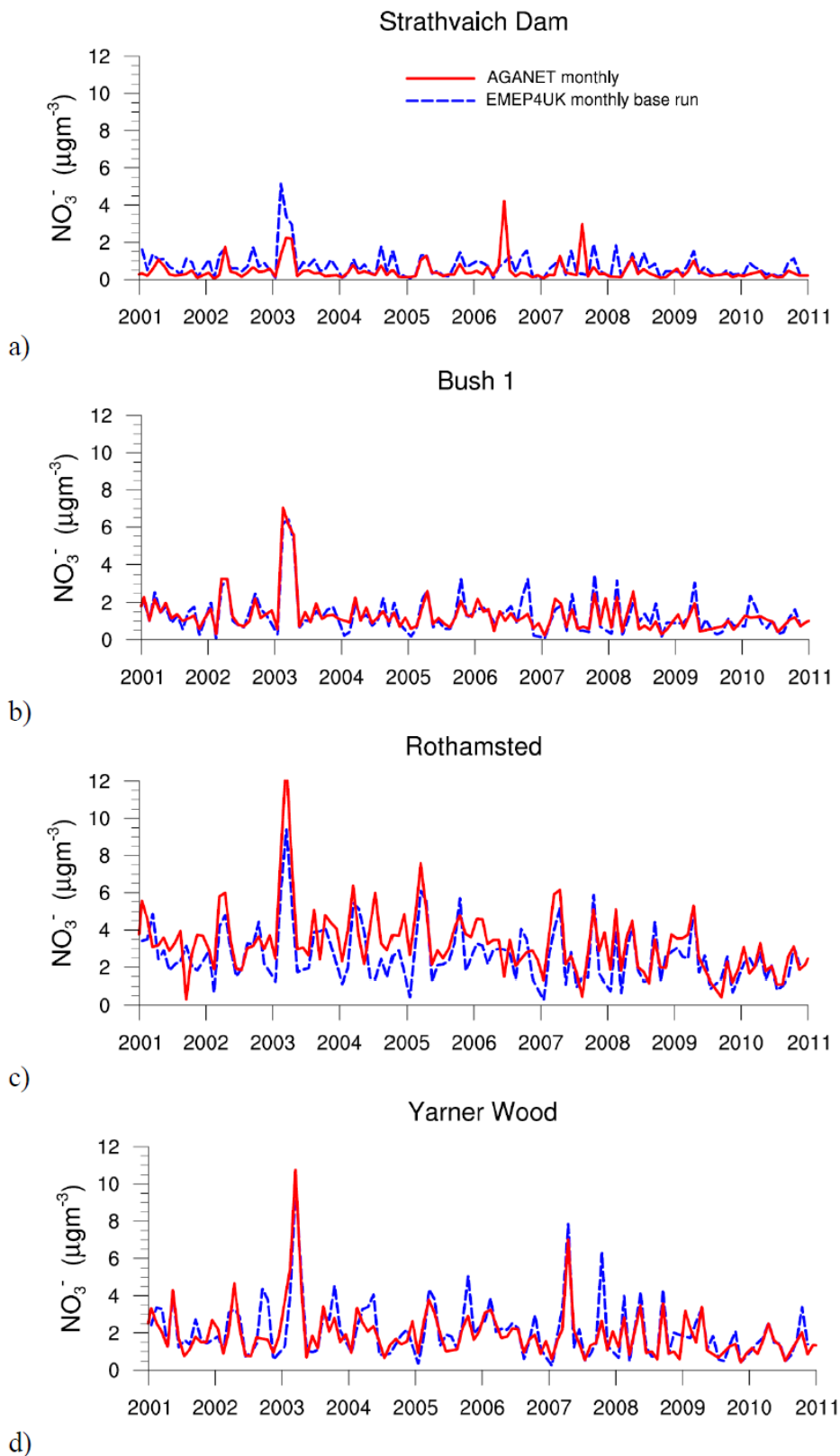


Figure 51. Monthly average surface concentration of particulate matter nitrate, observed (RED) and modelled (BLUE), for 2001-2010 at four sites of the AGANet network: Strathvaich Dam (north-west Scotland), Bush (central Scotland), Rothamsted (south-east England), and Yarner Wood (south-west England). For full information, see Vieno et al. (2014)

3.14. Examination of multi-pollutant and co-benefit approach to investigate emissions reduction scenarios, changes in agricultural practice and human behaviour, which together may give a larger improvement in air quality and ecosystem recovery than each implementation alone (Task10)

Task 10 was focused on evaluating the effectiveness of policy measures controlling primary PM_{2.5} versus components of SIA, including NH₃, emissions in support of the Air Quality Expert group analysis. The results of this will be published in the 2015 PM_{2.5} AQEG report.

The EMEP4UK model has been used to calculate hourly surface concentrations of PM_{2.5} for the year 2010 to investigate the contribution of changes in different PM_{2.5} components to UK concentrations and population exposure. The emissions used for this analysis were derived from the EMEP inventory (for the European domain), the NAEI inventory (for the UK) and the ENTEC inventory for shipping.

A base run and a set of 5 variation experiments were carried out. The experiments applied 30% reductions to UK emissions for each of the following pollutants contributing to particulate matter formation individually:

1. NH₃
2. NO_x
3. SO_x
4. Anthropogenic NMVOC
5. Primary PM_{2.5}

This 30% perturbation was applied to land-based emissions only; shipping emissions (both domestic and international) have remained constant.

The 2010 annual average surface concentrations for PM_{2.5} (in $\mu\text{g m}^{-3}$) are shown at the 50 km x 50 km resolution of the European domain in **Figure 52a** (only a portion of the whole domain is shown) and at the 5 km x 5 km resolution of the domain covering the British Isles in **Figure 52b**.

The results of the emissions reduction experiments 1 to 5 are shown in **Figure 53** to **Figure 55**. The units are % reduction in total PM_{2.5} concentration (compared with the baseline, i.e. the model run based on unperturbed 2010 emissions) for each grid square resulting from the

emissions reduction in the specified pollutant derived using the following equation (ij are the 2-D grid square indexes $i=1\dots220$, $j=1\dots270$):

$$(Experiment_{ij} - Base_{ij}) \div Base_{ij} \times 100$$

For example **Figure 53a** shows the % reduction in surface $PM_{2.5}$ concentrations compared with the base case for a 30% reduction in UK NH_3 emissions and **Figure 55** shows the % reduction in surface $PM_{2.5}$ compared with the base case for a 30% reduction in UK primary $PM_{2.5}$ emissions.

The annual average surface concentrations of $PM_{2.5}$ in 2010 over the UK are generally lower compared with neighbouring continental countries such as France, the Netherlands and Germany. Overall, $PM_{2.5}$ concentrations in the UK are relatively insensitive to UK-only reductions in emissions of individual components or precursors. The maximum reduction in UK $PM_{2.5}$ concentrations is ~6% for a 30% change in UK emissions and in most locations the reductions are considerably lower. This implies that $PM_{2.5}$ in the UK (as far as annual average concentrations are concerned) is substantially influenced by import of primary $PM_{2.5}$, and/or by $PM_{2.5}$ formed from emissions of precursors emitted outside of the UK (and, in the case of SIA, by non-linearities in the SIA chemistry). The strong influence of continental Europe is revealed by the consistently decreasing $PM_{2.5}$ concentrations away from the continent in **Figure 52**.

On average across the UK, the effectiveness of reducing UK emissions by 30% on UK $PM_{2.5}$ concentrations declines in the order NH_3 , primary $PM_{2.5}$, SO_x , NO_x and VOC, but see below key points about geographical differences in the $PM_{2.5}$ reductions:

- NO_x : The 30% reductions in NO_x yield a maximum of 3% reduction in $PM_{2.5}$ over some rural areas, and generally a maximum of 1.5% reductions in $PM_{2.5}$ over other rural areas (**Figure 53b**). A key observation is that reductions in $PM_{2.5}$ concentrations over urban centres are smaller than in rural areas for these NO_x reductions due to the larger relative local contribution of primary $PM_{2.5}$ emissions from urban sources.
- SO_x : The 30% reductions in SO_x yield up to 5% reductions in $PM_{2.5}$ in the Trent valley and around 3% reductions in $PM_{2.5}$ over quite wide areas of central and northern England and central Scotland (**Figure 54a**). Again, the $PM_{2.5}$ benefit is not, in general, associated with the major urban areas except where these also have major SO_x sources in the vicinity.

- VOC: The 30% reductions in VOC yield a maximum of 1.5% reductions in $PM_{2.5}$ in central & northern England and central Scotland (**Figure 54b**).
- NH_3 and $PM_{2.5}$: The 30% reductions in NH_3 and primary $PM_{2.5}$ yield the greatest percentage reductions in $PM_{2.5}$ concentrations (up to ~6%, see **Figure 53a** and **Figure 55**) but the key observation is the inverse relationship in the geographic patterns of $PM_{2.5}$ sensitivity to these two components. The reductions in NH_3 emissions result in the largest $PM_{2.5}$ concentration decreases in rural areas, whereas the reductions in primary $PM_{2.5}$ give the largest decrease in areas of high population density. This reflects the geographical pattern of the sources and that, through the short atmospheric lifetime of NH_3 , UK emissions of NH_3 generally have short range impact.

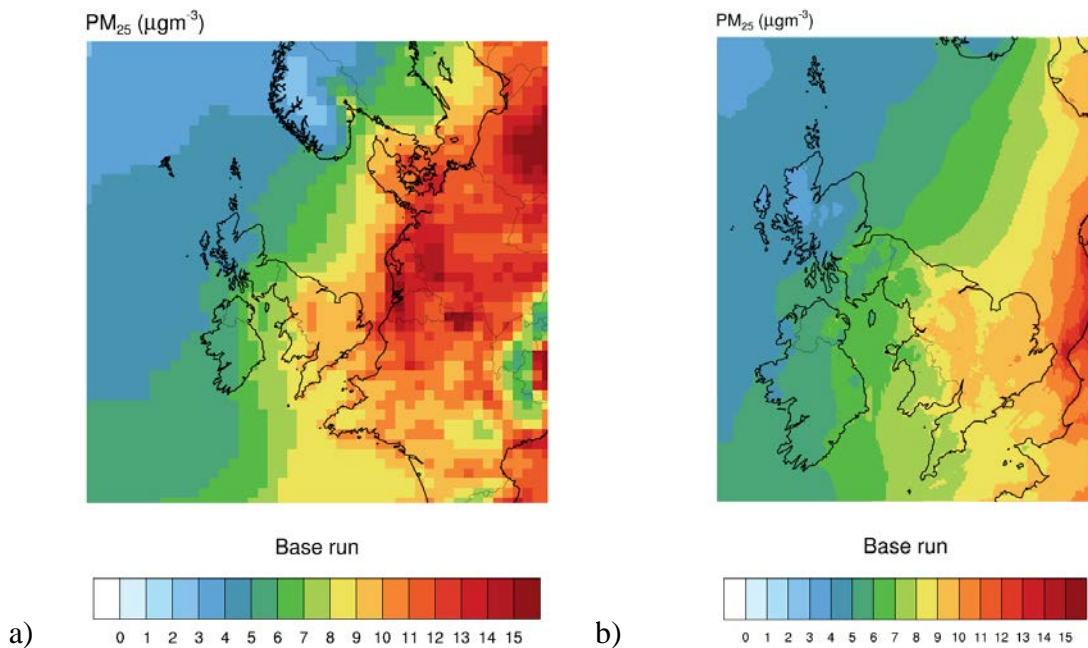
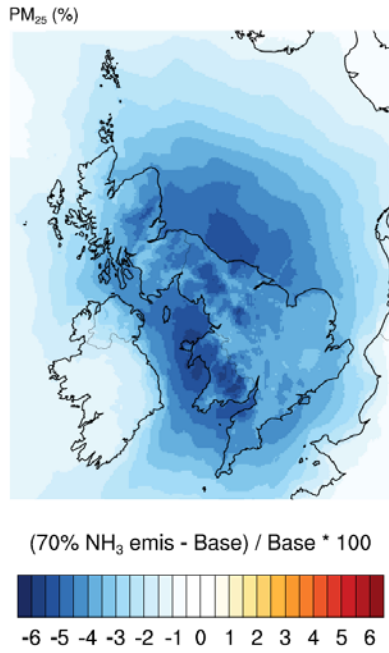
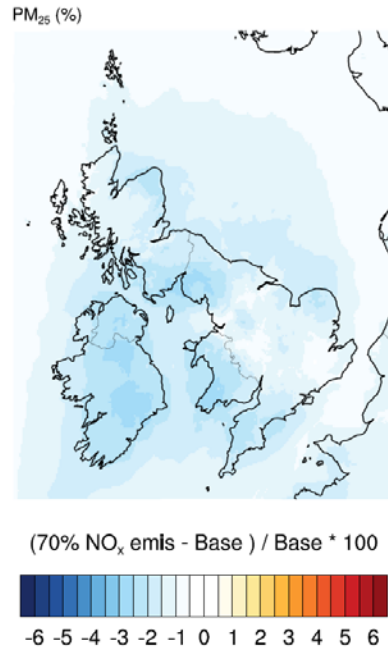


Figure 52. 2010 annual average EMEP-WRF surface concentrations of $PM_{2.5}$ at horizontal resolution of: a) 50 km x 50 km (a portion of the European model domain is shown) and b) nested 5 km x 5 km for the British Isles.

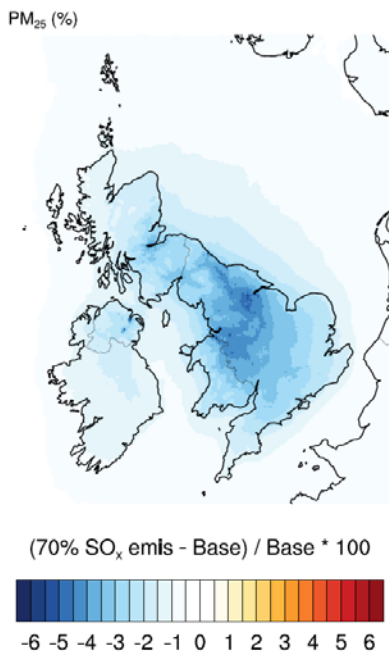


a)

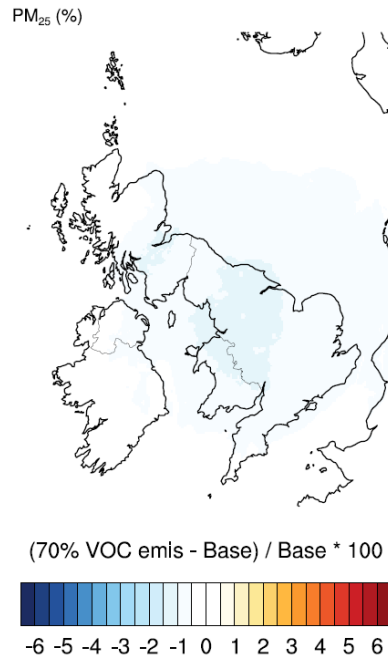


b)

Figure 53. Percentage change in PM_{2.5} simulated by the EMEP4UK model for: a) 30% emissions reduction of UK NH₃ emissions and b) 30% emissions reduction of UK NO_x emissions.



a)



b)

Figure 54. Percentage change in PM_{2.5} simulated by the EMEP4UK model for: a) 30% emissions reduction of UK SO_x emissions and b) 30% emissions reduction of UK anthropogenic VOC emissions.

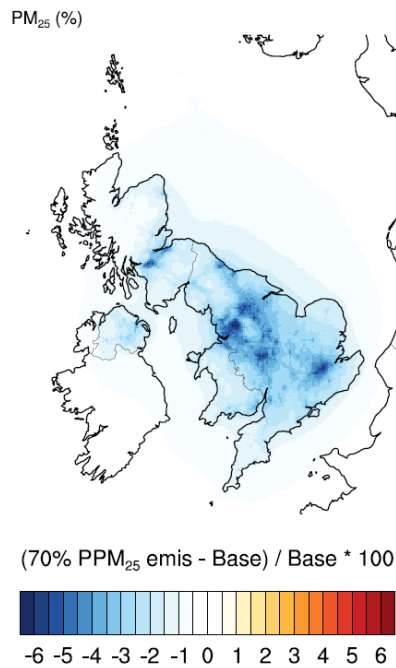


Figure 55. Percentage change in PM_{2.5} simulated by the EMEP4UK model for 30% emissions reduction primary PM_{2.5} emissions.

4. Summary

The EMEP4UK model has been demonstrated to be fully operational and applied as a tool to perform both basic scientific research and policy applications. The EMEP4UK model default meteorological driver is the WRF model. Both model source codes are open source and can be freely downloaded from their respective web sites (www.emep.int and www.wrf-model.org). The additional pre-processors for the meteorology and other input parameters developed within this contract are available on request. The EMEP4UK developments carried out in previous and this current Defra and CEH partnership-funded project have been reported back to the EMEP MSC-w model development team and included in the official EMEP MSC-W model (Simpson et al., 2012;Schulz et al., 2013). This input constitutes national support and in-kind contributions to EMEP and the value of this collaboration has been widely acknowledged by the EMEP modelling team and CLRTAP task forces.

The EMEP4UK model version rv4.4 has been extensively validated both in the UK (Vieno et al., 2014) and in Europe performing in a similar or better way than other air quality models (Simpson et al., 2014) in terms of representing atmospheric conditions and air pollution levels, while having demonstrated a high degree of computational efficiency resulting in runtimes which are several times faster than comparable models.

The EMEP4UK model combines an appropriate level of complexity to deliver up-to-date scientific analysis capability of atmospheric composition with the speed required to perform hundreds of simulations, for instance for the generation of source-receptor relationships for integrated assessment modelling applications.

Additionally to the work carried out with Defra and CEH partnership funding and described in this report, the model has been further updated and expanded, e.g. in the context of NERC and EU funding to allow, for instance, the use of a flexible vertical domain. Preliminary tests have been carried out with an additional layer near the surface, with the new surface layer now at ~50 m.

Further explorative developments have been conducted to run the EMEP4UK model in forecast mode. The tests carried out shows that the runtime for a 3 day forecast using 64 processors for both the meteorology and the chemistry is well within 2 hours. This provides a good basis for the development of high resolution air quality information systems.

References

Bergström, R., van der Gon, H. A. C. D., Prevot, A. S. H., Yttri, K. E., and Simpson, D.: Modelling of organic aerosols over Europe (2002-2007) using a volatility basis set (VBS) framework: application of different assumptions regarding the formation of secondary organic aerosol, *Atmospheric Chemistry and Physics*, 12, 8499-8527, DOI 10.5194/acp-12-8499-2012, 2012.

Bohnenstengel, S. I., Hamilton, I., Davies, M., and Belcher, S. E.: Impact of anthropogenic heat emissions on London's temperatures, *Quarterly Journal of the Royal Meteorological Society*, 140, 687-698, Doi 10.1002/Qj.2144, 2014.

Brutus, J., Grebot, B., Tsagatakis, I., Murrells, T., Dore, A.J., Vieno, M., Hall, J., Kryza, M., Carnell, E., Dragosits, U.: Update to Existing Impact Assessment for the Revised Annex VI of MARPOL to Support Assessment of Impacts of Revised EU Directive on Sulphur Content of Marine Fuels, Report to the Department for Transport, AMEC Environment & Infrastructure UK Limited 2014.

Carslaw, D. C.: Report: Defra regional and transboundary model evaluation analysis - Phase 1 http://uk-air.defra.gov.uk/library/reports?report_id=653, 2011a.

Carslaw, D. C.: Report: Defra deposition model evaluation analysis - Phase 1, http://uk-air.defra.gov.uk/library/reports?report_id=652, 2011c.

Carslaw, D. C., and Ropkins, K.: openair - An R package for air quality data analysis, *Environ Modell Softw*, 27-28, 52-61, DOI 10.1016/j.envsoft.2011.09.008, 2012.

Conolly, C., Lawrence, H., Vincent, K., Donovan, B., Davies, M., Colbeck, C., Cape, J. N., Tang, Y. S., Bealey, W. J., Leaver, D., Poskitt, J., Beith, S., Thacker, S., Hockenhull, K., Woods, C., Simmons, I., Braban, C. F., van Dyke, N., Rowland, P., Fowler, D., and Sutton, M. A.: UK Eutrophying and Acidifying Atmospheric Pollutants (UKEAP) Annual Report 2010, 2011.

Donahue, N. M., Robinson, A. L., Stanier, C. O., and Pandis, S. N.: Coupled partitioning, dilution, and chemical aging of semivolatile organics, *Environ Sci Technol*, 40, 2635-2643, Doi 10.1021/Es052297c, 2006.

ENTEC: Defra, UK ship emissions inventory, final report, http://uk-air.defra.gov.uk/reports/cat15/1012131459_21897_Final_Report_291110.pdf, Crown copyright, 2010.

Fournier, N., Dore, A. J., Vieno, M., Weston, K. J., Dragosits, U., and Sutton, M. A.: Modelling the deposition of atmospheric oxidised nitrogen and sulphur to the United Kingdom using a multi-layer long-range transport model, *Atmospheric Environment*, 38, 683-694, 10.1016/j.atmosenv.2003.10.028, 2004.

Heal, M. R., Heaviside, C., Doherty, R. M., Vieno, M., Stevenson, D. S., and Vardoulakis, S.: Health burdens of surface ozone in the UK for a range of future scenarios, *Environ Int*, 61, 36-44, <http://dx.doi.org/10.1016/j.envint.2013.09.010>, 2013.

IIASA: Future emissions of air pollutants in Europe – Current legislation baseline and the scope for further reductions, [http://ec.europa.eu/environment/air/pdf/TSAP-BASELINE-20120613\[1\].pdf](http://ec.europa.eu/environment/air/pdf/TSAP-BASELINE-20120613[1].pdf), access 01/12/2014, 2012.

Jenkin, M. E., Watson, L. A., Utembe, S. R., and Shallcross, D. E.: A Common Representative Intermediates (CRI) mechanism for VOC degradation. Part 1: Gas phase mechanism development, *Atmospheric Environment*, 42, 7185-7195, DOI 10.1016/j.atmosenv.2008.07.028, 2008.

Kuenen, J. J. P., Visschedijk, A. J. H., Jozwicka, M., and van der Gon, H. A. C. D.: TNO-MACC_II emission inventory; a multi-year (2003-2009) consistent high-resolution European emission inventory for air quality modelling, *Atmospheric Chemistry and Physics*, 14, 10963-10976, DOI 10.5194/acp-14-10963-2014, 2014.

Schulz, M., Gauss, M., Anna Benedictow, Jan Eiof Jonson, Svetlana Tsyro, Agnes Nyiri, David Simpson, Birthe Marie Steensen, Heiko Klein, ´A lvaro Valdebenito, Peter Wind, Alf Kirkevaag, Jan Griesfeller, Jerzy Bartnicki, Dirk Olivie, Alf Grini, Trond Iversen, Øyvind Seland, Valiyaveetil S. Semeena, Hilde Fagerli, Wenche Aas, Anne-Gunn Hjellbrekke, Katarina Mareckova, Robert Wankmuller, Philipp Schneider, Sverre Solberg, Tove Svendby, Li Liu, Maximilian Posch, Massimo Vieno, Stefan Reis, Maciej Kryza, Malgorzata Werner, and Walaszek, K.: Transboundary Acidification, Eutrophication and Ground Level Ozone in Europe in 2011, 2013.

Shrivastava, M. K., Lane, T. E., Donahue, N. M., Pandis, S. N., and Robinson, A. L.: Effects of gas particle partitioning and aging of primary emissions on urban and regional organic aerosol concentrations, *J Geophys Res-Atmos*, 113, Doi 10.1029/2007jd009735, 2008.

Simpson, D.: Biogenic Emissions in Europe .2. Implications for Ozone Control Strategies, *J Geophys Res-Atmos*, 100, 22891-22906, 1995.

Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L. D., Fagerli, H., Hayman, G. D., Gauss, M., Jonson, J. E., Jenkin, M. E., Nyíri, A., Richter, C., Semeena, V. S., Tsyro, S., Tuovinen, J. P., Valdebenito, Á., and Wind, P.: The EMEP MSC-W chemical transport model – Part 1: Model description, *Atmos. Chem. Phys. Discuss.*, 12, 3781-3874, 10.5194/acpd-12-3781-2012, 2012.

Simpson, D., Andersson, C., Christensen, J. H., Engardt, M., Geels, C., Nyiri, A., Posch, M., Soares, J., Sofiev, M., Wind, P., and Langner, J.: Impacts of climate and emission changes on nitrogen deposition in Europe: a multi-model study, *Atmos. Chem. Phys. Discuss.*, 14, 6663-6720, 10.5194/acpd-14-6663-2014, 2014.

Vieno, M., Dore, A. J., Wind, P., Di Marco, C., Nemitz, E., Phillips, G., Tarrason, L., and Sutton, M. A.: Application of the EMEP Unified Model to the UK with a Horizontal Resolution of 5 x 5 km(2), *Atmospheric Ammonia*, 367-372, 2009.

Vieno, M., Dore, A. J., Stevenson, D. S., Doherty, R., Heal, M. R., Reis, S., Hallsworth, S., Tarrason, L., Wind, P., Fowler, D., Simpson, D., and Sutton, M. A.: Modelling surface ozone during the 2003 heat-wave in the UK, *Atmospheric Chemistry and Physics*, 10, 7963-7978, DOI 10.5194/acp-10-7963-2010, 2010.

Vieno, M., Heal, M. R., Hallsworth, S., Famulari, D., Doherty, R. M., Dore, A. J., Tang, Y. S., Braban, C. F., Leaver, D., Sutton, M. A., and Reis, S.: The role of long-range transport and domestic emissions in determining atmospheric secondary inorganic particle concentrations across the UK, *Atmos. Chem. Phys.*, 14, 8435-8447, 10.5194/acp-14-8435-2014, 2014.

von Schneidmesser, E., Vieno, M., and Monks, P. S.: The changing oxidizing environment in London – trends in ozone precursors and their contribution to ozone production, *Atmos. Chem. Phys. Discuss.*, 14, 1287-1316, 10.5194/acpd-14-1287-2014, 2014.

Watson, L. A., Shallcross, D. E., Utembe, S. R., and Jenkin, M. E.: A Common Representative Intermediates (CRI) mechanism for VOC degradation. Part 2: Gas phase mechanism reduction, *Atmospheric Environment*, 42, 7196-7204, DOI 10.1016/j.atmosenv.2008.07.034, 2008.

Young, D. E., Allan, J. D., Williams, P. I., Green, D. C., Harrison, R. M., Yin, J., Flynn, M. J., Gallagher, M. W., and Coe, H.: Investigating a two-component model of solid fuel organic aerosol in London: processes, PM1 contributions, and seasonality, *Atmos. Chem. Phys.*, 15, 2429-2443, 10.5194/acp-15-2429-2015, 2015.