Improving the temporal profiles of emission input data for high resolution atmospheric transport modelling – a case study for the UK

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ABSTRACT

Atmospheric transport models (ATMs) are widely applied, ranging from basic research into processes of atmospheric transport, chemical transformation and deposition to ex-ante and ex-post evaluations of air quality related policy decisions. In a policy context, the drive for an accurate quantification of the uncertainties of model output is particularly strong.

ATMs are typically evaluated by model intercomparisons, both assessing the relative performance in reproducing selected atmospheric parameters and comparing model results to atmospheric measurements conducted on the ground, airborne or by remote sensing. And to a certain extent, sensitivity analyses have been conducted to identify the key drivers of the models' output, for instance looking into the quality of meteorological input data such as wind speed and direction, ambient temperature, spatial and temporal distribution of rainfall and suchlike.

While emission inventories have been subject to significant improvements over the last decades and efforts to both quantify and reduce the uncertainties of inventory datasets, both the sensitivity to and the processing of emission input data in currently applied atmospheric transport models has not been investigated in sufficient depth. This is in particular the case for the temporal resolution of emissions, which may have a significant impact on the match between model results and measurement data in particular when applying ATMs with exceptionally high spatial resolution on a national or below scale (5×5 or 1×1 km, in the case of the UK).

This paper aims to conduct first analyses of the sensitivity of atmospheric transport models based on data for the United Kingdom to make a case for the improvement of temporal resolutions of emission input data into state-of-the-art models. Further to that, the authors would like to derive recommendations and priorities for the improvement of emission processing routines in the view of limited resources.

1 Introduction

Emissions of substances to the atmosphere both from anthropogenic and natural and biogenic sources are the main drivers of atmospheric transport models (ATMs) with regard to their resulting ambient concentrations of primary and secondary pollutants. While meteorological input data have been subject to continuous improvement, using sophisticated models such as MM5, ECWMF or WRF, and other model parameters of ATMs have been improved over time (temporal and spatial resolution of the output, substance splits, aerosol formation, chemistry etc.), anthropogenic emission data input in particular is often handled at a comparatively coarse level.

This paper aims at giving a brief overview of how different ATMs currently applied in Europe and the US process emission datasets as input, with a focus on anthropogenic emissions, with specific regard to spatial, temporal and sectoral/chemical resolution. In a second step, it discusses some of the key problems and the development of solutions with regard to the spatial and temporal resolution of emission data input. Finally, the paper will discuss initial results of running the model with improved temporally resolved emission data and derive conclusions for further research needs.

1.1 Overview on emission handling in atmospheric transport models

The models discussed in Section 2 in some detail are:

- EMEP Unified Model (EMEP, MSC-West)
- EMEP MSC-East HM Model (EMEP, MSC-East)
- CHIMERE Model (IPSL, France)
- FRAME (Centre for Ecology & Hydrology, UK)
- Models-3, resp. CMAQ/SMOKE (US EPA, USA)
- EMEP4UK (Centre for Ecology & Hydrology , application of the EMEP-UNI to the UK)

This review cannot be and does not aim to be comprehensive. Its objective is to identify gaps and shortcomings of current approaches to process emission datasets as input to ATMs and to derive, where possible, priorities for improvement. Furthermore, the review takes into account current state-of-the art emission inventories, identifying, where possible, shortcomings in their structure and ways to overcome these. The following inventories are taken into account:

- EMEP (WEBDAB)
- The UK National Emissions Inventory (UK NAEI)

2 Review of the current state of emission processing

2.1 EMEP MSC-W EMEPUNI

The emission routines of the EMEP Unified model (EMEP-UNI) are described in detail in the EMEP Report 1/2003 (August 2003), which is available online[1]. The emission datasets originate from officially reported datasets submitted to EMEP by parties to the CLRTAP. These can be accessed via the EMEP Centre on Emission Inventories and Projections (CEIP) system[2].

2.1.1 Temporal and spatial distribution

Anthropogenic emissions are spatially distributed using a static table by SNAP (Selected Nomenclature for Air Pollutants) category only, which to some extent reflects average stack heights and includes calculations on plume-rise, conducted for different stability conditions. There is no information given if and how Large Point Sources (LPS) are included on an individual, exact coordinate base, including stack heights or other parameters that may be available by type of source. In any case, the height profiles of different source categories below SNAP-sector level in particular for *Combustion in Energy and Transformation Industries* (SNAP 1) and *Combustion in Manufacturing Industry* (SNAP 3) cannot be properly reflected by the current coarse structure.

The temporal distribution of emissions is conducted based on time factors provided by IER, University of Stuttgart (http://www.ier.uni-stuttgart.de), which have been calculated for a specific year and are different for individual pollutants, sources and countries. The document mentions, that simple day and night factors are applied as well, but does not give any details for which purpose, sector or pollutant this might be the case.

2.1.2 VOC speciation

EMEP-UNI uses a VOC speciation based on work conducted in the UK and documented in the report PORG, 1993. For each of the 10 SNAP sectors, a different split is applied; however, SNAP sectors 1, 2, 3 and 10 seem to use the same split factors. The speciation distinguishes 11 model species.

2.2 EMEP MSC-East HM

The HM Model of the EMEP MSC-East uses a combination of officially reported datasets and expert estimates for gap filling purposes. The procedures are described in detail here: http://www.msceast.org/hms/emission.html Gap-filling is even more important than in the case of "classic" air pollutants, as completeness and level of detail of officially reported data less for HMs.

2.2.1 Temporal and spatial distribution

The spatial distribution of emissions to the EMEP 50×50 km grid is conducted on the basis of nationally reported distributions (18 countries have reported at least once a spatial pattern since 1990), for the remaining countries, the distribution of national totals was based on expert estimates by Berdowski et al. (1997).

A coarse approach to distribute emissions to different model layers across a vertical profile, based on sectoral information provided by countries, but not taking into account yet plume rise. Emissions are allocated to the three lowest model layers, i.e. 0-70, 70-150 and 150-300.

[1] http://www.emep.int/UniDoc/index.html
[2] http://www.ceip.at/

A seasonal variation based on assumptions for the main emitting sectors has been derived by Ryaboshapko et al. (1999), and takes into account individual countries and their specific cycles, but does not take into account factors such as temperature, or differences below the sector level (e.g. base load and peak load power plants).

2.2.2 HM Speciation

While Lead, Cd and other metals have a very low volatility, mercury is regarded both in gaseous and in particulated forms, with the gaseous species containing elemental and oxidised forms. Emission inventories currently do not report information on speciation, thus MSC-East applies a mercury split based on Axendfeld et al. (1991) and Pacyna and Muench (1991).

2.3 CHIMERE

The CHIMERE model developed by IPSL is taking an approach to anthropogenic emission input that is less closely connected with the emission inventories compared to the EMEP models. The model requires input data for 14 substances:

- NO, NO_2
- SO_2
- CO
- CH_4 *(currently assumed zero)*
- Ethan, n-Butane, Propene, Isoprene, x-Xylene, Formaldehyde, Acetaldehyde, Methyl ethyl Ketone
- Ethanol, Methanol *(not used yet, but in a new model version will be)*

Emission datasets are directly taken from the EMEP CEIP datasets.

2.3.1 Temporal and spatial distribution

EMEP emissions are taken on a 50×50 km EMEP grid and are interpolated onto the CHIMERE grid. For traffic NMVOC emissions, a spatial aspect is implicitly taken into account by applying different splits between gasoline and diesel consumption for different countries.

Monthly, daily and hourly variations are taken into account based on *Society 1994* data.

2.3.2 VOC Speciation

In order to distribute total NMVOC emissions taken from EMEP, CHIMERE does not use the sectoral information available on CEIP, but distributes emissions to main sectors according to an approach prepared by IER, University of Stuttgart (GENEMIS, 1994). For each sector, NMVOC emissions are then split into 32 classes based on Middleton et al. (1990) and standard VOC profiles again by IER (Society, 1994). These are then aggregated into the 9 CHIMERE classes, weighted by mass and reactivity again according to Middleton et al. (1990). The VOC profiles are assumed to be the same for the all countries, with the exception of road transport (see above).

2.4 FRAME

The FRAME model is taken into account as well, as even though it is not operating on the same regional scale or substance scope as for instance the EMEP models, it has already some procedures for the handling of emission data input, which could be further developed and implemented in other models. The emission input for FRAME is based on the UK inventory (NAEI) and Fournier (2002), in particular for LPS emissions.

FRAME uses two types of emission datasets, gridded emissions for NH_3, NO_x and SO_2 at 5×5 km resolution, and individual Large Point Source (LPS) emissions (NO_x and SO_2). NH_3 gridded emissions have a very simplified daily profile, but a detailed vertical distribution which reflects the emission sector (mainly agriculture, distinguishing between pig, poultry, cattle etc.). For SO_2 and NO_x, gridded data does not have a daily profile, but similar to NH_3 is distributed vertically according to the SNAP code. (e.g. road transport, commercial & residential combustion, ...).

2.4.1 Spatial distribution

In the case of LPS data, SO_2 and NO_x are emitted at the effective stack height since FRAME version 4.7 (including real stack height and plume rise, see *Vieno, 2005*). Point sources (with stack height, stack diameter, exit velocity and exit temperature for the 20 most contributing LPS) are reported in the NAEI as an individual source group, along with the following sectors:

- Combustion in energy production and transfer
- Combustion in commercial, institutional, residential and agricultural boilers
- Combustion in industry
- Production processes
- Extraction and distribution of fossil fuel
- Road transport
- Other transport and machinery
- Waste transport and disposal

These sectors match the SNAP structure of the EMEP/CORINAIR inventory. For the year 1999, a 1×1 km^2 gridded version of the NAEI has been compiled, which is used as the basis for the spatial distribution of NO_x and SO_2 emissions in FRAME to 5×5 km^2. Point source data for 1999 was made available by AEA Technology on a 100×100 m^2 grid. Plume rise is calculated based on parameters derived from Seinfeld and Pandis (1997), see as well Vieno (2005).

Starting with version 5.3 of FRAME, NH_3 emission handling, which previously had been all at ground level, as well as low level SO_2 and NO_x emissions have been improved.

NH_3 emissions are split into 6 individual categories:

- Cattle
- Sheep
- Pigs
- Poultry
- Fertiliser
- Non-agricultural

For these categories, the spatial distribution is derived from geospatial datasets allowing for detailed distribution of source contributions to each 5 km grid cell, e.g. assuming specific times for cattle being on the field or in house, allocating grazing emissions on surface level, while housing emissions are allocated to an emission height of 10 m and so on. The improvement of using this more sophisticated

method of emission height attribution for low sources is measurable, e.g. increasing the correlation between measured and modelled NH_3 concentrations for the year 1999 from R^2=0.50 to R^2=0.55.

Further investigations by Vieno (2005) led to the conclusion, that for other sectors as well the correct emission height attribution and thus injection into the proper model layer led to improved correlation with measurements and thus inventories should take emission heights for each relevant source group into account.

2.4.2 Temporal distribution

FRAME, as a statistical model, operates on an annual time scale and provides annual statistics on concentrations and depositions. At this stage, FRAME does not require emission input data with detailed temporal patterns.

2.5 CMAQ/SMOKE

SMOKE is the emission processor developed by the US EPA to process emission inventory data in varying formats to generate input for CMAQ. It supports a variety of inventory formats for criteria, particulate, toxics, and activity data inventories. SMOKE does not provide conversion utilities to process files, but rather expects text files to be provided in the correct format to be processed. SMOKE is flexible, enabling users to upload inventory pollutants in a wide range of formats, distinguishing the following main categories:

- Non-point/stationary area sources
- Non-road mobile sources
- On-road mobile sources
- Point sources (SMOKE has formats for annual or average-day inventories, for day-specific inventories, and for hour-specific inventories

The structures and nomenclatures used are all tailored to the US EPA reporting and inventory procedures and are not directly applicable to European inventories, e.g. EMEP. In this context, SMOKE is unlikely to be useful as an emission processor to drive European ATMs, as the core step is to determine the correct temporal and spatial distribution of national sectoral annual totals. This step needs to take into account the specific situation of European sources and with the effort of adapting the inventory structure to EMEP or other reporting nomenclatures, SMOKE is inherently not able to be superior to any pre-processing method develop specifically for the current European inventory structure.

SMOKE and other emission models (e.g. EMS-95[3], MEPPS[4], EPS2.0[5]) are applied in the US to:

- spatially allocate aggregated emissions to a pre-defined model grid,
- temporally allocate annual emissions to hour-by-hour emissions based on activity data; and
- disaggregate lumped pollutant species (e.g., VOCs) into the individual chemical species or groups of species needed to model chemical processes in the atmosphere.

There appears to be a suite of models available and under further development to pre-process anthropogenic emissions in particular as input to CMAQ, in contrast to the current situation in Europe.

[3] Emissions Modeling System-95, http://64.27.125.175/tech/emis/ems95_guide/ems95.html
[4] Models-3 Emission Processing and Projection System
[5] Emissions Preprocessing System, http://www.imaqs.uh.edu/epsmain.htm

2.6 EMEP4UK

The EMEP4UK model is directly derived from the EMEP unified model described in Section 2.1. Hence, all aspects described above are valid for EMEP4UK as well. The main difference is the use of UK NAEI emission inventory datasets on a 1x1 km spatial resolution for the UK modelling domain, producing output on a 5x5 km grid. For a full description of the EMEP4UK model, see Vieno et al., 2009.

3 Comparative analysis

3.1 Spatial resolution

On a regional scale, the EMEP models are supplied with a coarse sectoral split (10 SNAP sectors) on a 50×50 km^2 grid resolution. This implicitly applies as well to CHIMERE, where EMEP datasets are used, only transformed to fit the different CHIMERE grid. Large Point Sources are currently not treated explicitly, based on exact coordinates and including relevant information such as stack height, exhaust gas temperature, stack diameter or measurement data from the plants. The reason for this can be mainly seen in the lack of comprehensive datasets, as the last collection of data on LPS through CORINAIR/EMEP was conducted in 1990/94 and has not been updated. Other datasets, such as compiled by the European Pollutant Emission Register[6] (EPER) could deliver vital information on the location and emissions of individual facilities, but face different problems. On the one hand, data relevant for modelling (stack heights etc.) do not belong to reporting obligations, on the other hand comparisons of emissions reported to EPER and sectoral emissions for the same country reported to EMEP resulted in significant discrepancies. In how far the future European Pollutant Release and Transfer Register[7] (EPRTR) will be able to harmonise reporting between sectors and facilities, and more important, provide information relevant to modelling, is yet unclear. First steps to integrate international shipping routes as line sources, respectively allocating them to 50×50 km^2 grid cells, have been taken for the EMEP Unified Model, but motorways and major roads are not taken into account separately. At a 50×50 km^2 scale, the effect of major line sources such as motorways could be negligible, however, similar to some LPS, their contribution to local to sub-regional ambient air pollution could be significant. In case the model resolutions are refined in the future, this would need to be carefully assessed, e.g. in the case of EMEP4UK.

Where methods for the spatial allocation of sectoral emissions to grid cells are concerned, input datasets such as land use data, agricultural statistics and suchlike have been improved over time and most of these datasets would allow for a much finer resolution of emissions than the 50 km scale without changes in methodology. Further to that, SNAP sectors are comparatively coarse and often include sources grouped together that are in fact inhomogeneous and marked by different spatial patterns. A careful assessment of the sectoral structure underlying the spatial distribution methods is paramount.

Finally, where countries are providing gridded datasets directly, methodologies to generate these are likely to differ, as the EMEP/CORINAIR Emission Inventory Guidebook offers only coarse guidelines for the spatial distribution of emissions. Different assumptions and the use of other surrogate datasets, for instance different or outdated land use data or activity data, could lead to a spatial distortion of emissions within countries. The magnitude of such effects could be significant, in particular for pollutants such as ground level ozone, where the location of NO_x and NMVOC limited regions for ozone formation may be prone to such distortions.

3.2 Temporal resolution

Both EMEP models and CHIMERE use methodology and time factors derived from datasets generated in the course of the EUROTRAC subproject GENEMIS[8] and provided by IER, University of Stuttgart in the frame of different levels of collaboration. While these time factors are of high quality and offer hourly fractions of emissions for each pollutant individually, it needs to be stated that they have been calculated for a specific year (1994) and thus temporal patterns (e.g. energy use, traffic counts etc.) are,

[6] http://www.eper.cec.eu.int/eper/default.asp
[7] see http://europa.eu/scadplus/leg/en/lvb/l28149.htm
[8] http://genemis.ier.uni-stuttgart.de

again, specific for this very year. Their application to split annual total emissions down to seasonal, or even hourly values does present an improvement to applying less sophisticated datasets. However, they are outdated and more advanced methods have been or are currently being developed, incorporating for instance temperatures and other meteorological data in high temporal resolution to reflect the influence of these parameters on emissions e.g. from power and heat generation, road transport (evaporation, re-suspension of road dust) and agriculture. In the case of agriculture, the need for a more process-oriented approach to allocate emissions in space and time has already been acknowledged and currently groups at the Norwegian Meteorological Office, CEH and IIASA are working on methodologies to incorporate these findings into the processing of emission data. In parallel, the EU funded research project NATAIR[9] aims at the development of improved methodologies and parameters to calculate natural and biogenic emissions for ATMs.

3.3 Substance resolution/chemical speciation

The VOC speciation applied in the EMEP Unified Model dates back to a 1993 report and allocates VOCs to 11 substance groups, based a detailed UK inventory described in PORG (1993) as well as in Andersson-Skoeld and Simpson (1997). The allocation provides a percentage share for each of the 11 substance classes for each SNAP sector. One major issue could be the use of the same Split for each country, in particular with regard to applying the same split to SNAPs 1, 2 and 3, thus assuming the same speciation for emissions from large power plants and for small residential and commercial boilers. This does not properly reflect the different fuel types and burning conditions in these quite different appliances, e.g. the use of fuel wood and coal/gas etc. Further to that, shares of different fuels have changed, some significantly so, over time, for instance the increase of natural gas in power generation vs. the use of coal, or the relative shares of petrol and diesel engines in road transport. CHIMERE uses 9 reactivity classes and allocates VOCs based on Middleton et al. 1990, using a similar approach to EMEP-UNI. In both cases, the VOC speciation is based on splits that have been generated in the early 1990s and many of the major source categories have undergone profound changes in the 13-16 years since they ware published. The comprehensive introduction of three-way catalysts in vehicles for instance has had a massive impact on the VOC split emitted from this source group. A new paper on VOC speciation has been published recently by Theloke et al. (2007) and should be reviewed with the aim to potentially update the splits in both models.

The speciation of mercury in the MSC-East HM model is based on publications from 1991. Here it would be worthwhile to investigate the impact of more advanced control technologies to reduce emissions of particulate matter as well as the comprehensive fitting of $DeNO_x$/$DeSO_x$-technologies to power plants on the emission of particulated forms as well as the oxidised and elemental Hg emissions both on the amount emitted and on the shares of each species. Recent findings, e.g. by Kakareka et al. (2000) could provide better insight.

[9] http://natair.ier.uni-stuttgart.de

4 Emission inventory datasets

4.1 Overview

The main central data sources for emission datasets on a European or global scale are CORINAIR/EMEP and the UK National Atmospheric Emissions Inventory. Both offer access to their datasets via web portals, which can be found at

- http://www.ceip.at/ (Centre on Emission Inventories and Projections, Austria) and
- http://www.naei.org.uk/ (UK NAEI)

The EMEP dataset covers air pollutants, referring to UNFCCC for greenhouse gases (GHGs), while UK NAEI covers all main gaseous substances, including GHGs, as well as particulate matter (PM), heavy metals (HM) and persistent organic pollutants (POPs).

Both inventory datasets will be briefly discussed with regard to their usability and fitness for purpose to provide input data to ATMs.

4.2 CORINAIR/EMEP

Countries signing and ratifying the different protocols to the Convention on Long Range Transboundary Air Pollution (CLRTAP) are subject to mandatory emission reporting according to the reporting guidelines provided by EMEP. Their individual reporting requirements depend on the protocols signed and thus the number of reporting countries varies between pollutants. In addition to that, completeness and correctness of submissions is subject to substantial variations. Efforts are taken to conduct comprehensive reviews and assessments to improve the quality and completeness of reporting, but at this stage a fully complete dataset based on official emissions should not expected to be available in the near future.

Substances covered have evolved over time, starting with the key acidifying pollutants SO_2 and NO_x, then adding CO, NMVOCs and NH_3, different species of Particulate Matter, and finally extending towards POPs and Heavy Metals.

4.2.1 Sectoral structure: SNAP, NFR and CRF

The sectoral structure for reporting has been changed in recent years to harmonise reporting requirements with those according to IPCC, which has led to a transition from SNAP[10] to NFR (Nomenclature for Reporting, currently NFR02)[11], which is closely linked to IPCC CRF[12]. While CRF was mainly developed for reporting greenhouse gas emissions in the context of the UNFCCC, the SNAP structure had been better suited to account for emissions in particular from non-energy related sources. The perceived loss of detail in reporting when moving from SNAP to NFR, where the most detailed level 3 of SNAP had a far better resolved sectoral structure, than level 2 of NFR, was to some extent academic, as few countries actually reported on this detailed level. However, the discussion between modellers and inventory compilers is still ongoing, if it is possible to conduct meaningful modelling, for instance of emission control options, on a sectoral level as aggregated as NFR level 2. The emerging need to accommodate “new” sources, e.g. in the case of tyre and break wear, re-suspension of road dust,

[10] Selected Nomenclature for Air Pollutants
[11] Nomenclature for Reporting
[12] Common Reporting Format

has made revisions of the NFR structure necessary and further elaboration in the area of heavy metals and POPs might create a need for a further breakdown of the sector structure. A discussion on the issue of the most meaningful level of detail is ongoing and some experts believe that in order to compile the inventory with sufficient quality, it is necessary anyways to calculate emissions on a highly detailed level and to then aggregate according to the reporting format level. For both atmospheric transport modelling and Integrated Assessment Modelling, this highly detailed level holds vital information, which is lost in the aggregation process and needs to be either collected again, or replaced by second-best surrogate datasets. A positive side effect of harmonising reporting formats between NFR and CRF has been that it reduces the effort to merge emissions of air pollutants and greenhouse gases (GHGs) into a common framework, which is of particular importance for Integrated Assessment Modelling to account for synergies and trade-offs between air pollution control strategies and reducing emissions of greenhouse gases.

4.2.2 Expert estimates

Dealing with incomplete submissions and countries not reporting (in time), gap filling is a vital task to compile a comprehensive dataset for the modelling obligations that both EMEP centres have to fulfil. For all pollutants, so-called "expert estimates" are used to complement available official submissions, respectively fill obvious gaps in incomplete datasets. These are most often generated in the frame of research projects or specific studies of sectors and substances and are, in contrast to official submissions, typically based on centralised approaches using activities and emission factors to derive emission figures.

In the view of the political relevance of issues such as compliance with emission targets and air quality limit values, the question of which datasets to use for modelling is far from easy to address. From a modellers point of view, the best available datasets should of course be used as these are the main input factors determining the quality of modelling results, e.g. to assess compliance with limit values for atmospheric concentrations, deposition fields and such like. On the other hand, officially reported emission data cannot be disregarded without a sound and thorough review process, which would need to be empowered to clearly identify inconsistencies, spot obvious errors and review activity rates and emission factors used to derive emission figures. This issue could be resolved in the future by current plans to extend the scope of the CORINAIR/EMEP Emission Inventory Guidebook to be linked to an Emission Factor database and – potentially – encouraging parties to report activity rates on a more regular basis. A comprehensive review of heavy metal emission inventories in the EMEP report 1/2006 has thoroughly discussed the issue of expert estimates and their value for the review of official submissions.

4.2.3 Spatial and temporal distribution and gridding

The requirements for providing gridded datasets are outlined in the document ECE/EB.AIR/80[13]. Basically, countries are obliged to report gridded national totals as well as sectoral data according to NFR (01, 02a, 02b, 03, 04, 05, 06, 07, 08, 09 or Natural) every 5 years from 1990. Starting from 2000, LPS data should be reported including lat/long coordinates according to a format given by ECE/EB.AIR/80, too. The guidance on how to distribute emissions spatially for area, point and line sources given in the CORINAIR/EMEP Emission Inventory Guidebook is allocated to numbers 12 (spatial disaggregation) and 13 (temporal disaggregation) of each sectoral chapter. The individual chapters provide mostly general methodological suggestions on how to distribute emissions spatially and which datasets to use as surrogates to derive geographical patterns.

The same generally applies to the temporal resolution.

[13] http://www.unece.org/env/eb/Air_Pollutionwithcover_15_ENG.pdf

4.3 UK National Atmospheric Emissions Inventory (UK NAEI)

The UK NAEI (http://www.naei.org.uk/) provides emission data for the United Kingdom (comprising the devolved administrations England, Scotland, Northern Ireland and Wales) mainly fulfilling the UKs reporting obligations under the UNECE CLRTAP and the UNFCCC.

The datasets are online available in 1×1 km^2 spatial resolution as annual total emissions, distributed in high sectoral detail for all relevant air pollutants (acidifying substances, ozone precursors, particulate matter, heavy metals, persistent organic pollutants and greenhouse gases).

At this stage, there are no temporal profiles available, however, the NAEI data warehouse already provides some information on activity data, which may be a first response to the growing needs for supporting data for modelling purposes.

NAEI emission datasets are further described in the subsequent sections, as they are the key emission dataset used for the application of the EMEP4UK model.

5 The application of high resolution datasets and modelling

5.1 Approach

For the UK, the development of the EMEP4UK model (Vieno et al., 2009) has introduced the unique opportunity to use emission datasets with a high spatial resolution (1 × 1 km) from the UK National Atmospheric Emission Inventory (NAEI) and high-resolution meteorological data (calculated using the WRF model) to model ambient concentrations of air pollutants on a fine scale for the whole of the UK.

In order to deliver these high resolution model results, the availability of input datasets in sufficient resolution is essential. In the following sections, the focus will be on the emission datasets used for modelling and the evaluation, in how far these steps have led to improvements in the way model results can be validated against observations. One of the drivers for the development of EMEP4UK has been the concentration and deposition patterns for instance of acidifying substances modelled on a 50 x 50 km scale with the European EMEP Unified model, which were viewed by UK experts as not suitable to reflect the real situation in the UK. And as the deposition patterns are crucial to determine for instance the exceedance of critical levels and loads, one of the core research tasks for the application of EMEP4UK was the integration of local and regional aspects, such as orographic rainfall over highlands in the UK and better land cover information.

Clearly, the 50 km resolution of the EMEP Unified model is not suited to reproduce specific aspects on the local or regional scale, not only because of the model's spatial resolution, but as well because of the coarse 50 km resolution of the emission data it receives.

5.2 Spatial resolution

Where the spatial resolution of emission data is concerned, the UK NAEI provides high resolution emissions on 1x1 km for the whole of the UK and a wide range of substances. Figs. 1 to 4 display a comparison for the year 2005 of the key gases taken into account in this paper between UK NAEI and EMEP emission maps generated using the EMEP WEBDAB[14] application. As official submissions of gridded data for the year 2005 are not available at this stage, expert emissions were selected to produce the EMEP maps, which are based on 2005 country sectoral totals submitted and gridded in the same spatial pattern as the 2000 maps submitted by national experts.

[14] http://www.emep-emissions.at/emission-data-webdab/

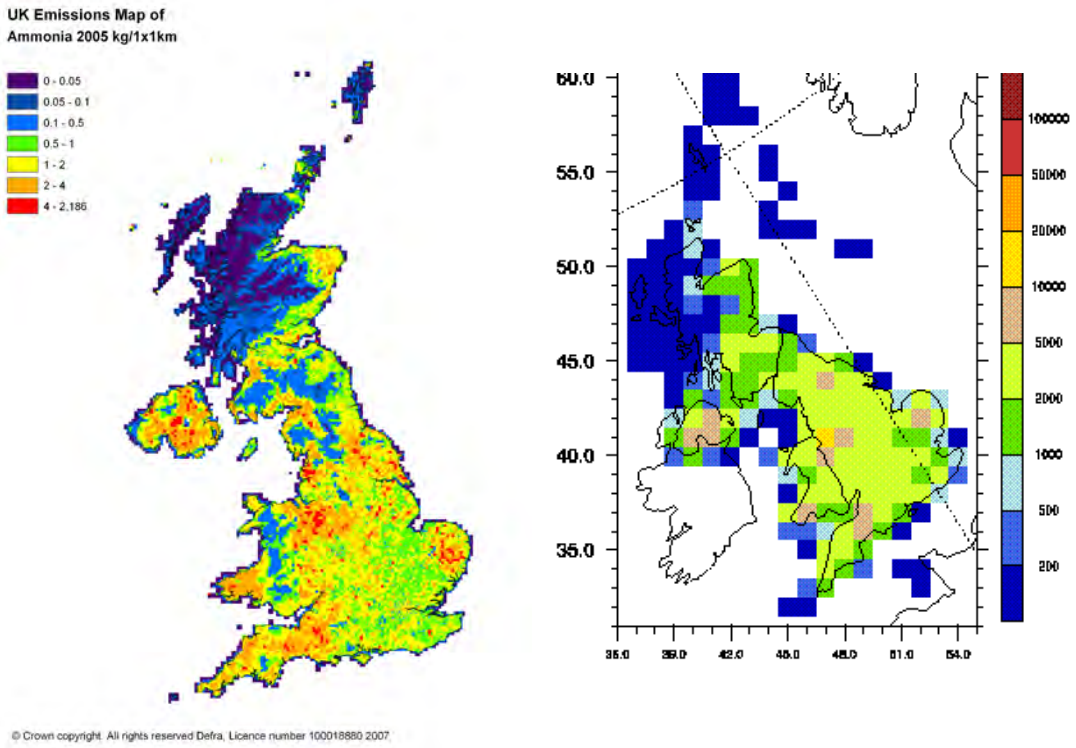

Fig. 1. Comparison of UK NAEI (left) and EMEP (right) emission maps for NH_3 for the year 2005

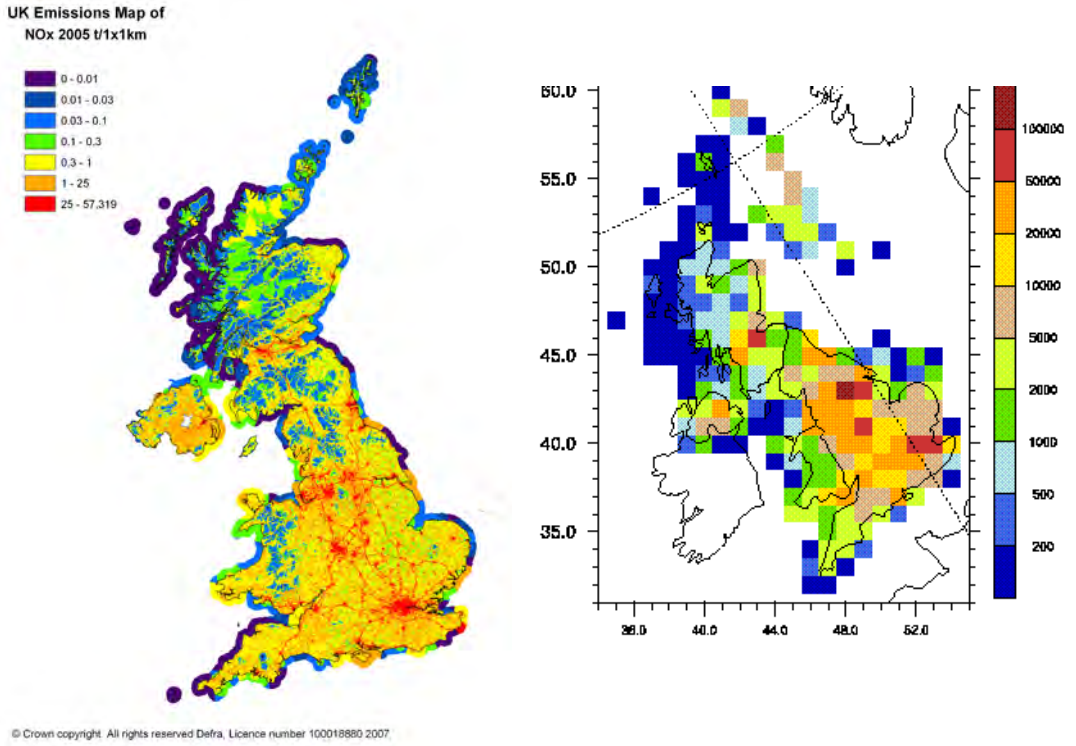

Fig. 2. Comparison of UK NAEI (left) and EMEP (right) emission maps for NO_x for the year 2005

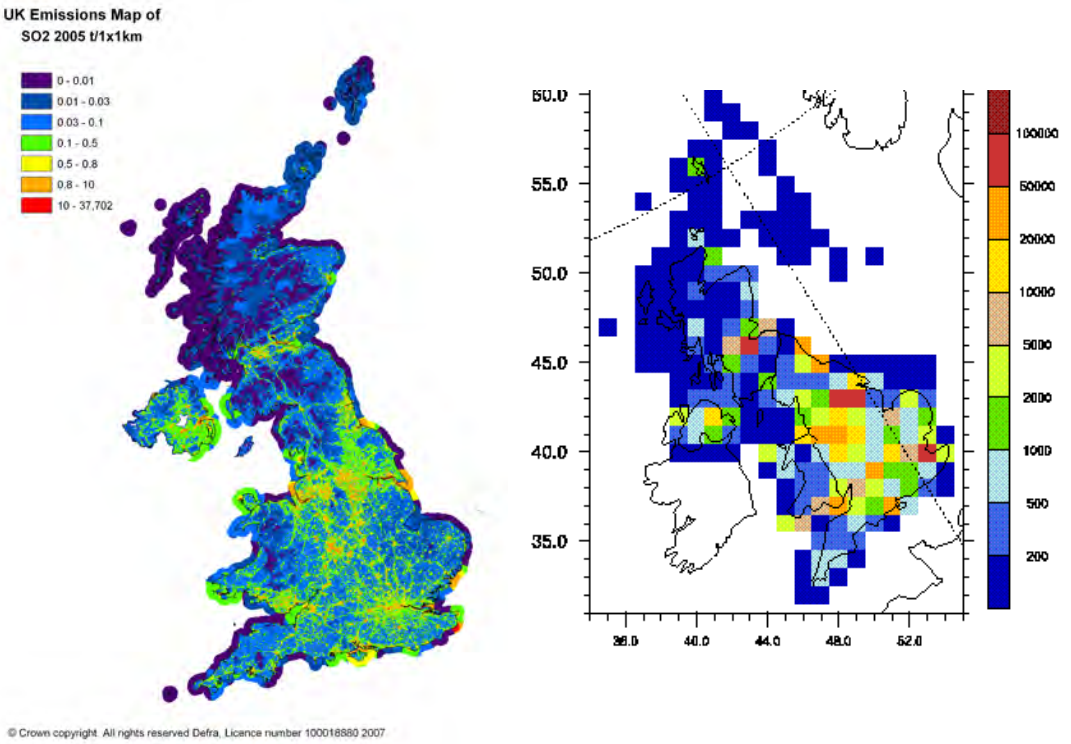

Fig. 3. Comparison of UK NAEI (left) and EMEP (right) emission maps for SO_2 for the year 2005

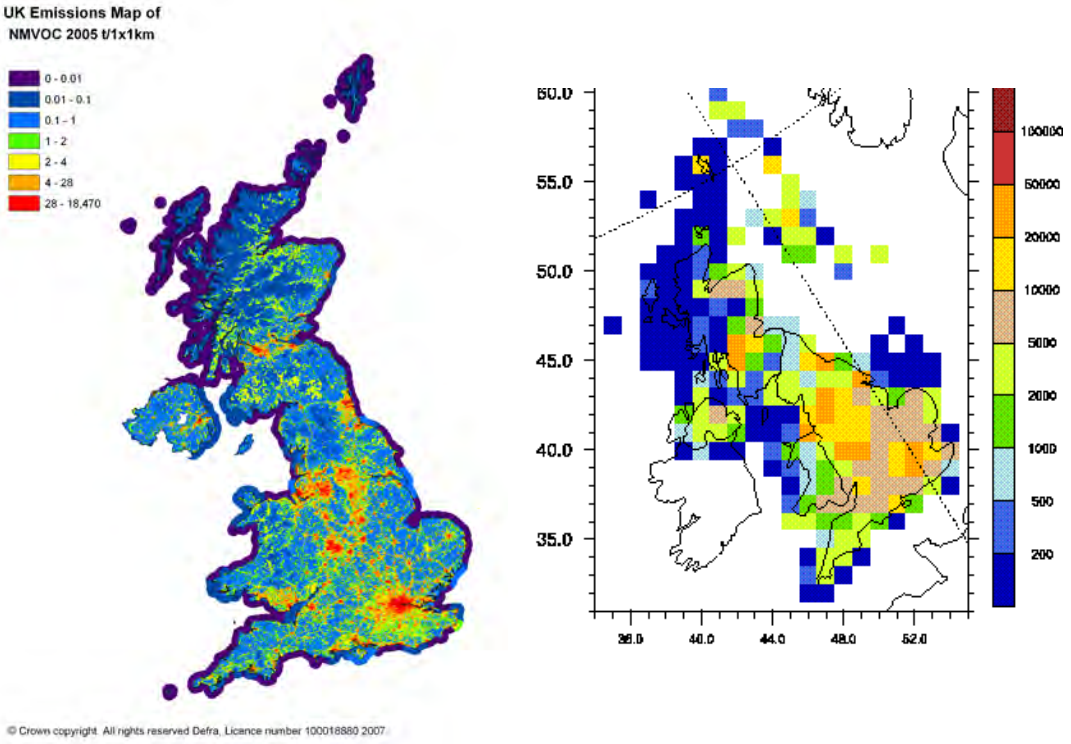

Fig. 4. Comparison of UK NAEI (left) and EMEP (right) emission maps for NMVOC for the year 2005

The maps shown in Fig. 1 for ammonia highlight a problem in the distribution and level of emissions for instance in Northern Ireland and parts of Eastern England and Wales. This is of particular importance, since ammonia is often deposited close to source areas and the different distribution patterns will be reflected in quite different deposition fields for acid deposition. NO_x emission maps in Fig. 2 are not massively different in the way the densely populated areas and major transport networks are

mapped. EMEP WEBDAB includes off-short sources, which are not displayed in the UK NAEI map. For SO_2, the influence of coastal shipping and ports can be seen nicely in the NAEI map in Fig. 3, and for this pollutant, the importance of large point sources is very high, which is obviously difficult to map accurately in the 50 km grid resolution, which often combines several individual sources and becomes a dominant grid cell with very high emissions in this process. NMVOC emissions again do not seem to differ substantially in both maps in Fig. 4.

A word of caution has to be added, however, with regard to the very high resolution presented in the UK NAEI maps: the maps look very plausible, reflecting patterns of high population density, agricultural lands, road networks etc., but their quality heavily depends on the spatial data used to generate them and the methodology applied. Unless inverse modelling methods or remote sensing can be applied on a large scale, these high resolution maps are very difficult to verify, and may add a certain level of uncertainty to the modelling, that is likely to be small, but nevertheless needs to be taken into account when comparing to observations.

6 Temporal resolution

Accounting for the temporal distribution of emissions on a national scale requires a vast amount of data. Thus, a first step is to identify the main drivers of the temporal patterns of the relevant emissions and strike a balance between a perfect, real-time temporal allocation and a feasible, cost-effective approach which will deliver a substantial improvement over the current situation (i.e. no distinct temporal distribution) while being manageable on a regular basis.

The following Table 1 lists the main influencing factors for the temporal distribution of emissions for selected source sectors and pollutants, being by no means comprehensive and complete, but showing that a few factors can actually inform about the distribution of different pollutants across sectors. For this study, meteorological parameters such as temperature, rainfall and suchlike have been excluded, keeping the focus of the sensitivity runs on the influence of purely anthropogenic activities on the emission distribution.

Table 1. Overview of main influencing factors for the temporal distribution of emissions for selected source sectors and pollutants

Source sector	**NH_3**	**NO_x**	**SO_2**	**NMVOC**
Large Combustion Plants	-	power/heat demand, *temperature*	power/heat demand, *temperature*	-
Residential and Commercial Combustion	-	power/heat demand, *temperature*	power/heat demand, *temperature*	-
Industrial Production Processes	-	shift routines, seasonality	shift routines, seasonality	shift routines, seasonality
Solvent Use	-	-	-	paint/solvent application routines, *temperature*
Road Transport	-	daily/weekly/seasonal transport demand patterns	-	daily/weekly/seasonal transport demand patterns
Other Mobile Sources	-	daily/weekly/seasonal transport demand patterns		daily/weekly/seasonal transport demand patterns
Agriculture	fertiliser & manure application, *temperature*	fertiliser & manure application, *temperature*	-	-
Natural and Biogenic Sources	*temperature*	*temperature*	-	-

Text in *italics*: parameter not taken into account in this study

Two main sectors contributing to the bulk of NO_x, SO_2 and NMVOC emissions have been identified to serve as a test case, *Large Combustion Plants* and *Road Transport*. To complement these, the sector Residential and Commercial Combustion is briefly discussed afterwards.

6.1 Road transport

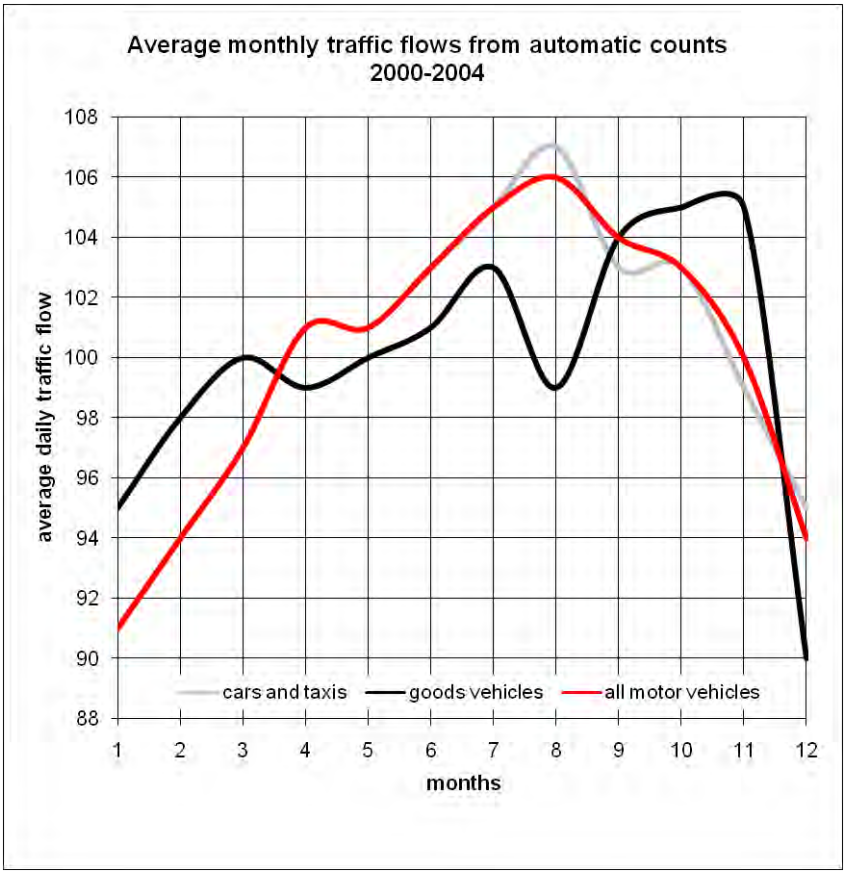

Fig. 5. Average traffic distribution over the year split into passenger and goods transport for the UK based on statistical data for the period 2000-2004

Fig. 5. displays the hourly distribution of transport activities, distinguishing between cars and taxis (i.e. passenger transport) and goods vehicles (freight), as the temporal patterns are quite different. This differentiation is important, as heavy goods vehicles in particular emit a large share of NO_x from transport sources, while passenger cars and light duty vehicles typically emit NO_x and NMVOCs at the same time, which is of relevance for groundlevel ozone formation. This graph identifies a clear diurnal pattern throughout the week, with lower traffic volumes on the weekends, and – for passenger transport – a morning and afternoon rush-hour peak, while freight transport is marked by strong activities during mid day.

Emissions, resulting from traffic, depend on several facts, like the composition of the fleet and the temporal and seasonal influence of the road traffic leading to different driving patterns on weekends, working days, school holidays or bank holidays.

For this study data are derived from manual and automatic traffic counts conducted at a number of fixed sites on major and minor road. The advantage of manual counts is the complete coverage of major road sites and the fairly good coverage of minor roads, however typically no information about traffic at night, during weekends, or holiday periods, is provided due to the fact that traffic is counted for only 12 hours on each count on so-called “neutral-weeks”, weeks without any distinct seasonal influence. These counting weeks of the yearly *Road Traffic Statistics* of the *Department for Transport* take place in March, April, May, June, September and October and are normally at the same time of the year to be directly comparable between years.

Automatic counts fill the gaps left by manual counters using automatic sensors for a continuous monitoring of traffic, and are even capable of distinguishing different vehicle types. On the down side,

these automatic counts have some problems for example to identify special types of good vehicles, coaches and buses, because of having similar axle spacing and chassis height.

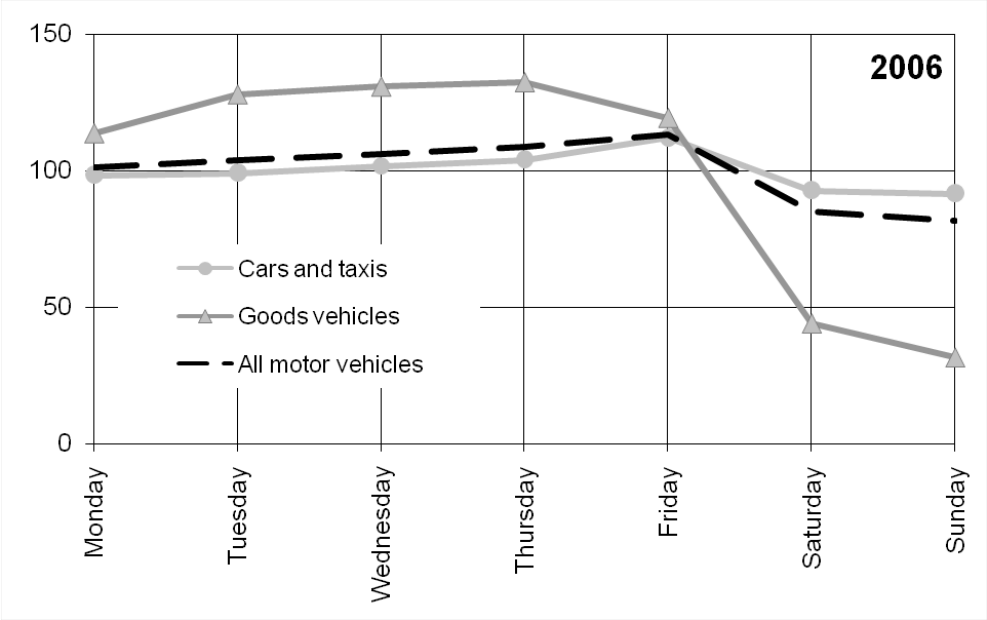

Fig. 6. Average traffic distribution over a week split into passenger and goods transport for the UK based on statistical data in the year 2006 (not including any bank holidays)

For the *National Road Traffic Surveys* by the *Department for Transport* data of both manual and automatic counts are used. For the annual average daily flow, data from automatic counts on similar roads are combined with manually counted data in neutral months. The raw counted data have to be multiplied by factors derived from automatic traffic counts of the same year, which do not highly vary from year to year, expect when bad weather condition restrict traffic. For cars this factor is usually between 1.00 and 1.25 while for goods vehicles, due to less traffic at weekends, the factor is lower, approximately between 0.75 and 1.25.

In 2004 UK road traffic amounted to 498.6 billion vehicle kilometres. This is an increase of 1.67 % compared to 2003. The traffic for 2005 has increased by 0.16% relating to 2004. The used data of traffic distribution refer to all roads, accordingly major and minor roads are aggregated. The data are split up into three different traffic users: cars and taxis, goods vehicles and, the aggregate, all motor vehicles. All motor vehicles include both cars and taxis, goods vehicles and the other road users like motorcycles, buses and coaches and light vans. The determination of the classification can be found in the glossary of *Road Traffic Statistics (2004).*

As Fig. 6. indicates, the main difference between goods and passenger transport occurs on the weekend. Heavy goods vehicles and light duty vehicles mainly operate during working ours and during weekdays, showing a steep decline on Friday and significantly lower activity levels on Saturday/Sunday. For cars and taxis, a peak on Friday is followed by reduced levels on the weekend, but the difference smaller.

The index used in Table 2 is always referring to an average day with the figure 100. All 52 weeks of the year are represented by one average week whereas days of the week contain working days, as well as bank holidays. For the purpose of this study, bank holiday days are replaced by a Saturday profile.

Table 2. Daily traffic flow in the UK for a general week in 2004 and 2006 on all roads for cars and taxis, goods vehicles and all motor vehicles. 100 is the index of an average day.

	2004 All roads			2006 All roads		
Day of the week	Cars and taxis	Goods vehicles	All motor vehicles	Cars and taxis	Goods vehicles	All motor vehicles
Monday	99	116	102	99	114	101
Tuesday	99	128	103	99	128	104
Wednesday	100	130	105	102	131	106
Thursday	104	132	109	104	132	109
Friday	113	120	115	112	119	113
Saturday	92	43	84	93	44	85
Sunday	91	30	82	92	32	82

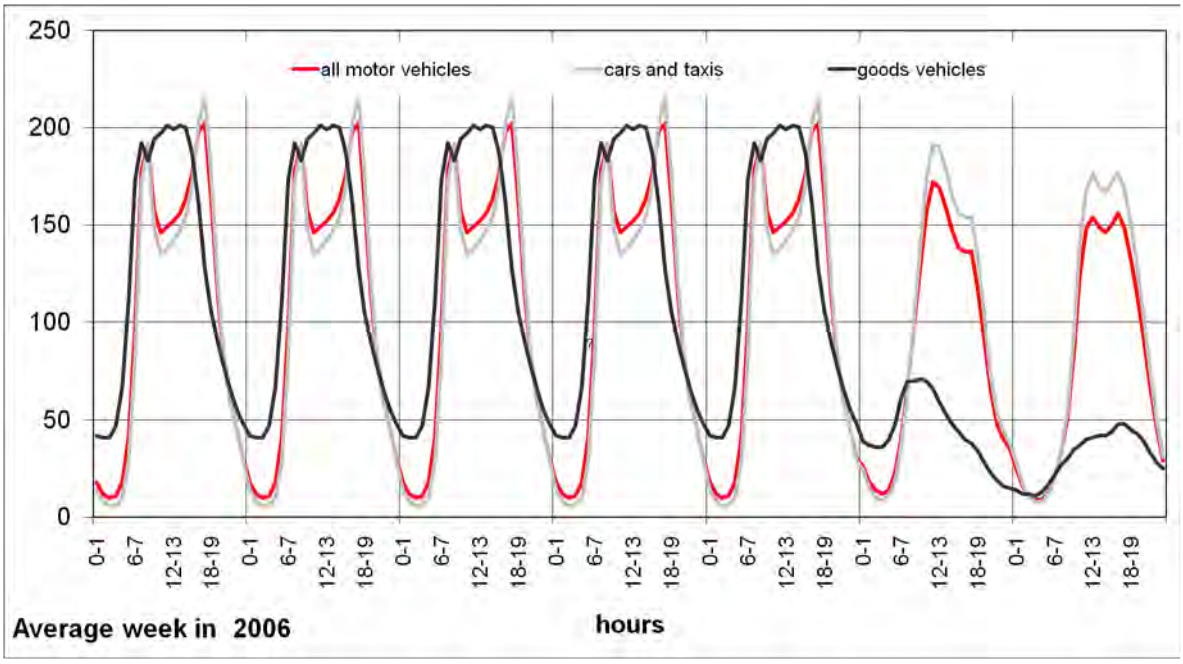

Fig. 7. Average hourly traffic distribution over a week split into passenger and goods transport for the UK based on statistical data in the year 2006 (not including any bank holidays)

Fig. 7. finally displays an average hourly resolution of road traffic emissions distinguishing between cars and taxis (passenger transport) and goods vehicles (freight transport). While passenger transport is marked by two explicit peaks (morning and afternoon rush hour due to commuting), freight transport matches working hours quite well. The curve presented for all motor vehicles demonstrates that the overall traffic volume is by far dominated by passenger transport. However, this cannot be directly applied to temporal emission patterns, as emission factors for heavy goods vehicles are significantly different from those of passenger cars.

Using these figures, hourly profiles for the whole year of 2006 have been generated and, for the purpose of this study, aggregated to reflect the temporal distribution of the annual national total emissions from road transport on the 1 km resolution provided by the UK NAEI.

6.2 Power generation in Large Combustion Plants

The figures of the UK Department of Business, Enterprise and Regulatory Reform (BEER) provide information on energy supply. The data are available as well in unadjusted as seasonally adjusted and

temperature corrected data. Within adjusted data coal, petroleum and natural gas consumption are temperature corrected. Seasonal and temperature adjustment factors were reassessed. For details of temperature correction see BERR energy statistics website[15].

For this study, the focus is on gas and coal fired power plants, with the former reflecting the temporal patterns for peak-load plants, and the latter for base-load generation units. Oil fired power plants are not commonly used for electricity generation in the UK. The relevance of distinguishing peak-load and base-load plants is the different temporal behaviour and the way they are dependent on external factors, such as for instance temperature. Fig. 8. displays the monthly energy consumption, split into gas and coal for the years 2003 and 2006. Electricity generation from coal is significantly lower in the summer, while the operation of gas fired power plants shows an indifferent variation throughout the year.

Fig. 8. nicely illustrates as well, how the influence of inter-annual variation of ambient temperatures can affect energy consumption patterns. In addition to that, the temporal pattern for gas fired power plants in 2003 shows a lot of variation, whereas in 2006, it is only marked by a slight steady increase towards the end of the year. As a general indicator, the temporal profile for total conventional power generation has been derived as well, which is mainly driven by the pattern for generation based on coal fired plants.

In general, a clear statistical dependency of monthly energy consumption with temperature can be identified (see Fig. 9.), but the correlations for coal ($R^2 = 0.79$) and gas ($R^2 = 0.19$) further show, that this is only the case for base load operated power plants (i.e. mainly coal fired units). Gas fired power plans often provide energy for peak load situations, which are typically not directly temperature dependant. The correlation for the sum of conventional thermal power generation is close to that of coal ($R^2=0.79$) due to the fact that - even though the amount of electricity produced in the by coal or gas fired plans is not significantly different (40.7 % compared to 36.5 %) – the level produced by gas does not vary significantly throughout the year, but is comparatively “flat”. Hence, the distinct temporal profile of coal based power generation drives the shape of the temporal profile for total conventional power generation.

[15] www.berr.gov.uk/files/file19317.pdf

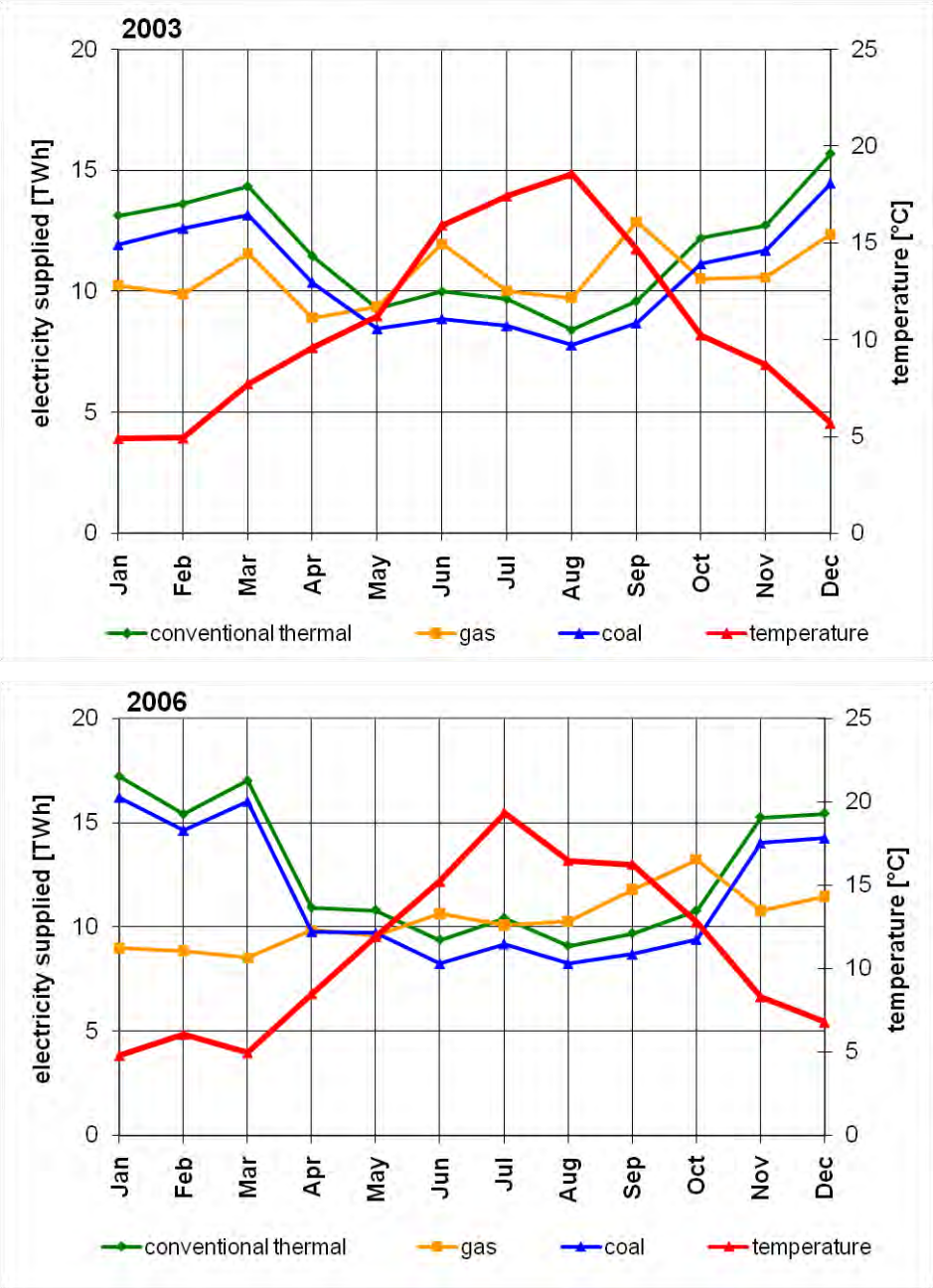

Fig. 8. Total monthly average energy consumption in the UK over the years 2001 – 2008 and split into gas and coal consumption for a year, not adjusted for temperature

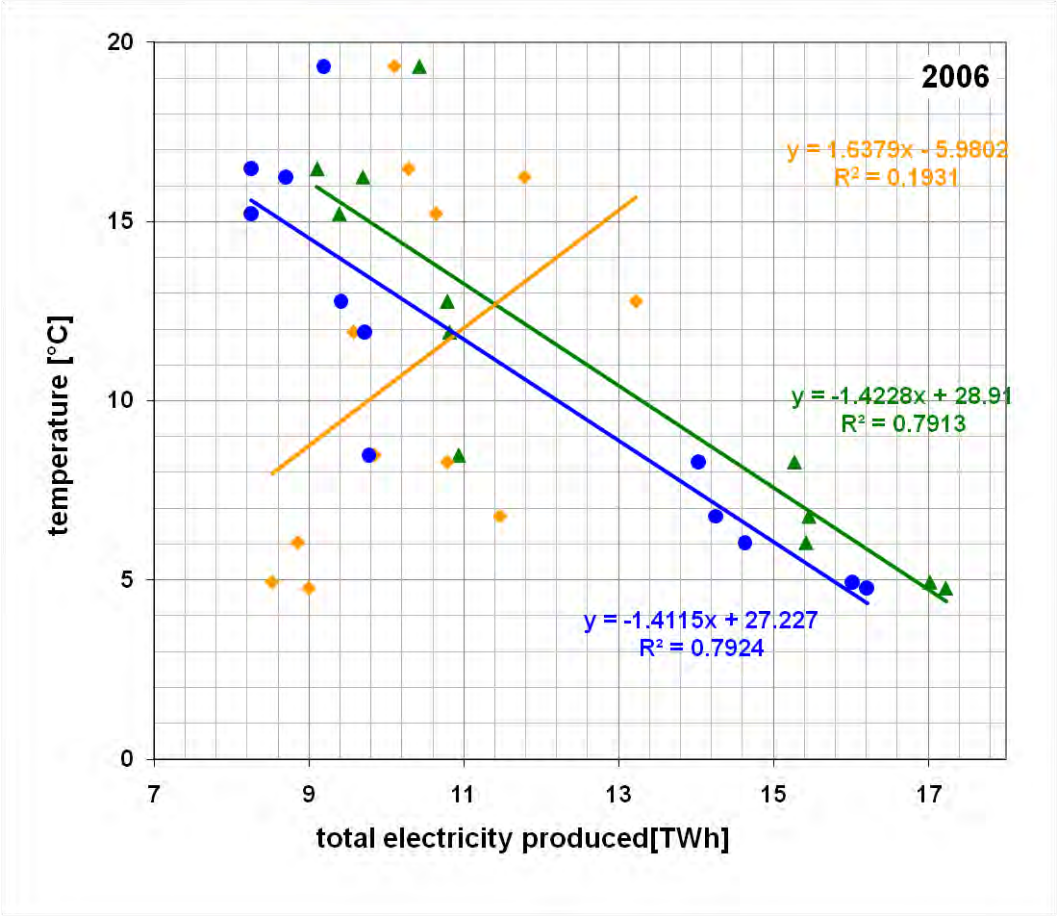

Fig. 9. Analysing the correlation between average monthly temperature and electricity production in public power plants split by fuel type (coal [blue circles], natural gas [orange diamonds] and conventional thermal power generation [green triangles]) for the UK in 2006

Different to road transport, a daily or even hourly resolution of emissions from power generation is not the objective. This is one the one hand not as important, due to the fact that emissions from these stationary sources are less variable in these shorter time spans, but the main reason is the general lack of data. Since the deregulation of European energy markets, the operation of power plants has become more dependant on the current market price developments, and operating hours, capacities etc. have become critical business information, that is not readily disclosed for the purpose of statistical analysis as in our case. We thus use monthly values available from official statistics and distribute NAEI emissions from large combustion plants according to the monthly shares in annual total emissions, and – where information is available – applied to individual point sources.

6.3 Residential and Commercial Combustion

The sources summarised under the label *residential and commercial combustion* comprise household heating and gas-cooking, heating and process energy for small businesses or public buildings and similar applications. The contribution of this sector to total emissions of the main pollutants is comparatively small, however, the spatial dispersion of individual sources and the distinct temporal profiles – again driven by a clear temperature dependence (see Fig. 10) – make this sector interesting to look into. In addition to that, individual sources such as wood combustion could present a significant source in rural areas, where the typically more important sources such as power plants and road transport are less abundant.

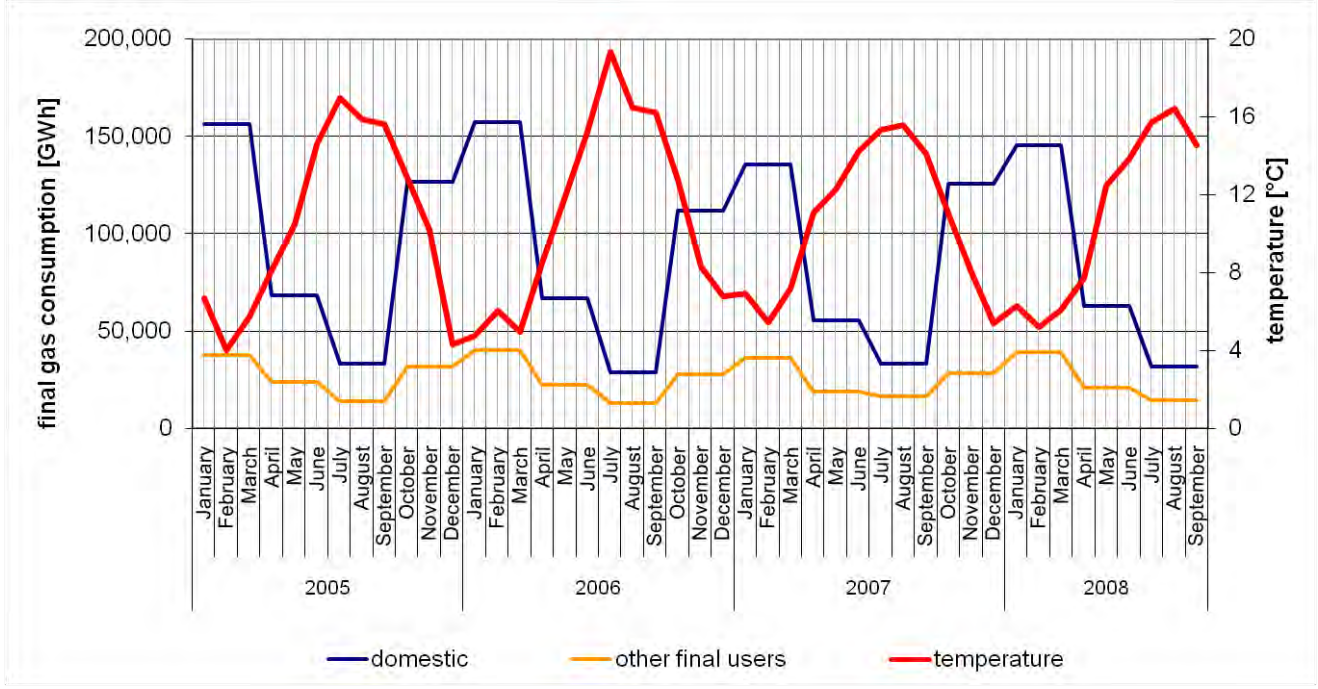

Fig. 10. Overview on final gas consumption (quarterly values) of households, other final energy users in relation to monthly mean temperature in the UK for the years 2004 and 2008

Fig. 10. displays annual mean temperatures over domestic and other final end user consumption of natural gas. Domestic gas consumption shows a strong (negative) correlation (R^2=-0.877) with temperature throughout the 4 year period. The same applies to other final users (-0.881), albeit the variation is much less pronounced. For the purpose of this study, domestic gas consumption figures have been used to create temporal profiles for this sector for the UK in 2006.

7 Model results and comparison with observations

7.1 Comparison with measurements

As a first indicator, the model performance both with the outdated, existing temporal profiles ("BASE") and the new temporal resolution are compared with measurement stations. Figs. 11, 12 and 13 show a preliminary analysis, comparing modelled data with an urban measurement station in the city of London (Eltham). Further analyses to be conducted will investigate the influence on how well modelled results match different types of stations (e.g. urban, rural and background stations) and conduct a comparative assessment of how different temporal patterns are for individual pollutants and sectors.

In Fig. 11., modelled and measured concentrations of NO_2 are compared for the year of 2006. In general, the model is capable of producing measured concentrations well, with a correlation of R^2=0.5854 with the old temporal resolution. This correlation is only slightly improved (R^2=0.5866) by 0.0012 when applying the new temporal resolution to the emission input data.

The overall correlation for sulphur dioxide (Fig. 12) between measured and modelled concentrations is comparatively poor (R^2=0.2874), which hints at one of the problems when modelling concentrations with a very high spatial and temporal resolution. SO_2 emissions mainly originate from large point sources and diesel operated heavy goods vehicles. At the same time, the formation of ammonium sulphates with ammonia significantly affects the presence of ambient SO_2 concentrations, especially in the view of the fact, that SO2 emissions have been dramatically reduced in recent times and the chemical mechanisms within the EMEP4UK model (as well as the EMEP unified model) regarding the formation of secondary aerosols (sulfates and nitrates) will need to be critically reviewed in this context.

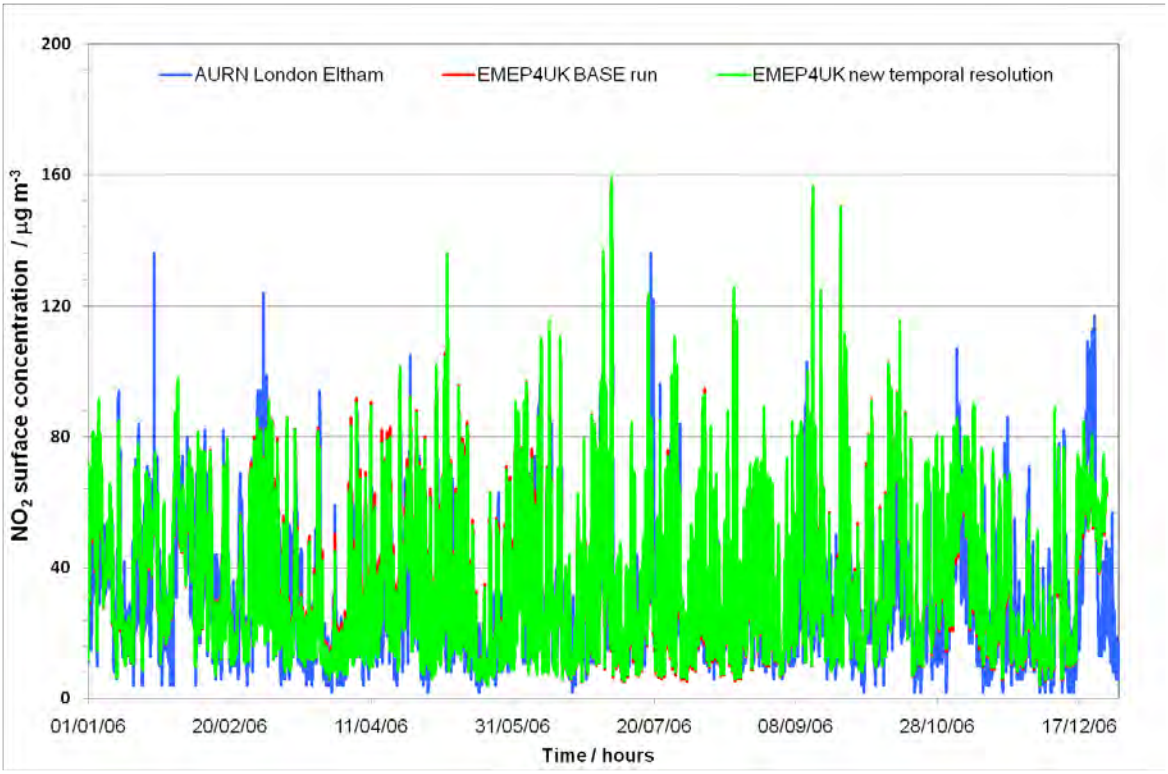

Fig. 11. Comparison of EMEP4UK model runs (base run with old temporal resolution and run with new temporal resolution) with a measurement station of the UK automatic measurement network (based in London Eltham) for NO_2 surface concentrations

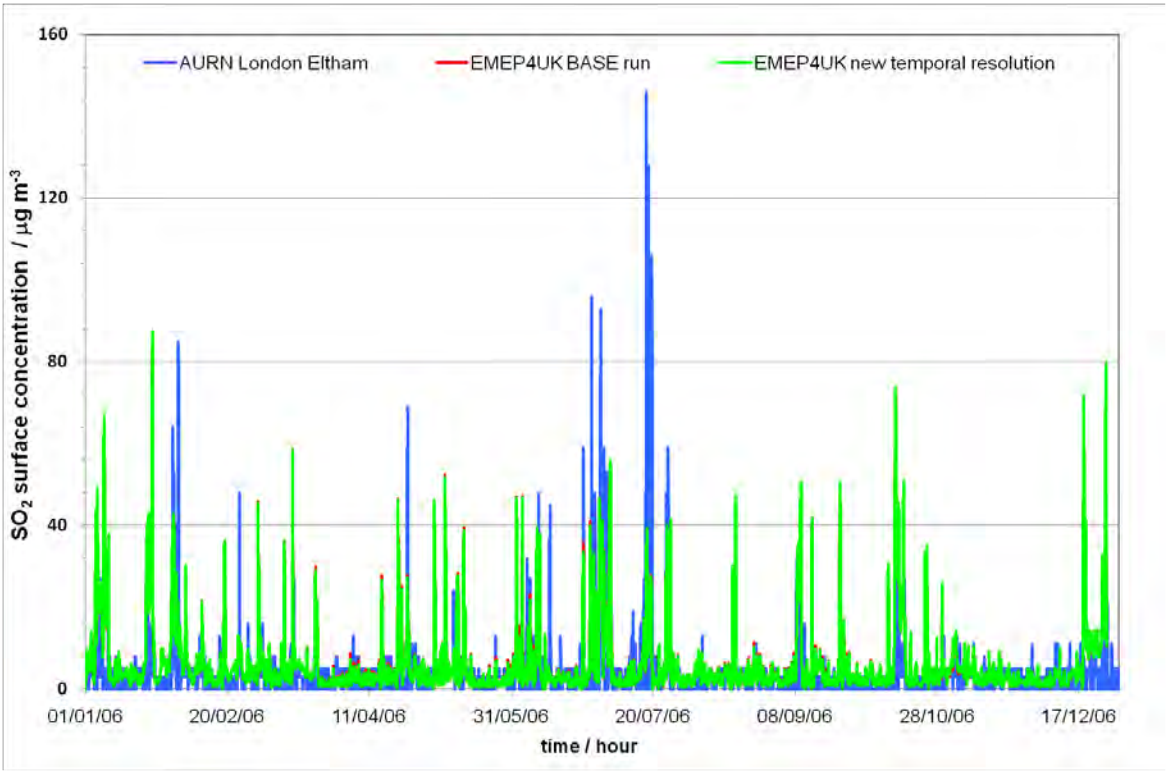

Fig. 12. Comparison of EMEP4UK model runs (base run with old temporal resolution and run with new temporal resolution) with a measurement station of the UK automatic measurement network (based in London Eltham) for SO_2 surface concentrations

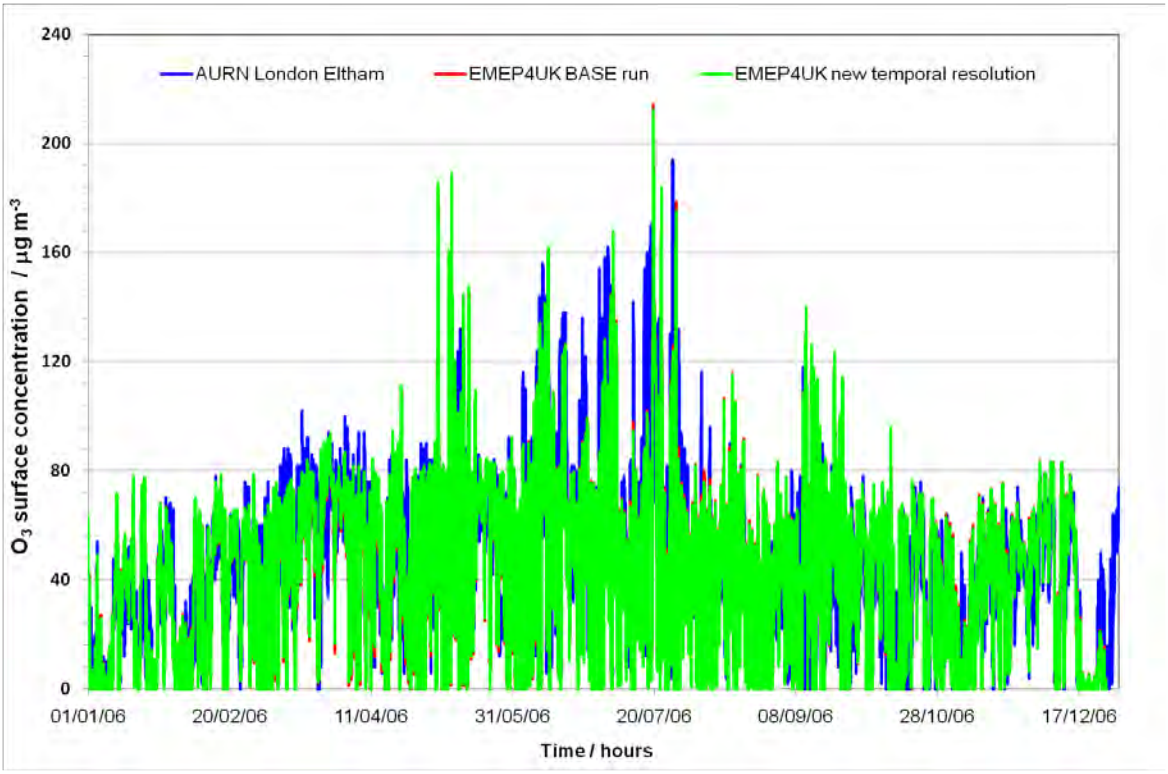

Fig. 13. Comparison of EMEP4UK model runs (base run with old temporal resolution and run with new temporal resolution) with a measurement station of the UK automatic measurement network (based in London Eltham) for O_3 surface concentrations

Fig. 13. Finally shows how well the EMEP4UK model is able to reproduce ambient concentrations of tropospheric ozone levels. With an R^2 of 0.7668, modelled concentrations manage to follow the trends and show peaks with satisfactory accuracy. However, the use of the new temporal profiles does not have a noticeable effect on the correlation. The most likely explanation for this is that for this study, only weekly and monthly resolutions have been applied, while ozone formation and in particular peak concentrations of ozone are marked by episodes which build within hours to a few days. A further improvement of especially emissions from road transport is probably going to further improve the fit between measured and modelled concentrations, e.g. for measurement stations that are strongly influenced by road transport emissions.

7.2 Direct comparison of model results - old vs. new temporal resolution

The comparison in Section 7.1 indicated only small signals regarding the fit of modelled and measured concentrations for selected substances. A different way of assessing the effect of the new temporal profiles applied is to directly compare the modelled concentrations on an hourly basis for the same substances. The results of this comparison are depicted in Fig. 15. for NO_2, SO_2 and O_3.

The graphs for NO_2 and SO_2 show a similar effect, with changes of ± 5 $\mu g/m^3$ overall, and a few peaks of up to ± 5 $\mu g/m^3$. What is remarkable is the difference in effects on both pollutants on a seasonal scale. The new temporally resolved emissions result in lower concentrations of NO_2 in the spring and slightly higher concentrations in summer and winter. In July, a few substantial peaks can be observed, where the concentrations calculated with the new temporal profile are up to 10 $\mu g/m^3$ higher than before. The situation for SO_2 is different and a mirror-image in July/August, albeit with a different orientation of the peaks. A comprehensive explanation of this will require further analysis of e.g. aerosol formation and dry/wet deposition of sulfates and nitrates during that period.

For tropospheric ozone, the changes look quite different (Fig. 15. bottom). Concentrations in spring, autumn and winter are on average higher, in the summer, however, somewhat lower. The overall signal is small compared to SO_2 or NO_2 discussed above, owing to the fact that ozone formation is strongly affected by meteorological factors as well as the spatial and temporal resolution of NMVOCs, the second relevant compound for the formation of ozone formation, which have not been addressed in this study yet.

In general, the sometimes modest reactions of modelled concentrations to changes in temporal profiles give an indication that it is important to focus on those sectors with both a high contribution to overall emissions and a substantial temporal variation.

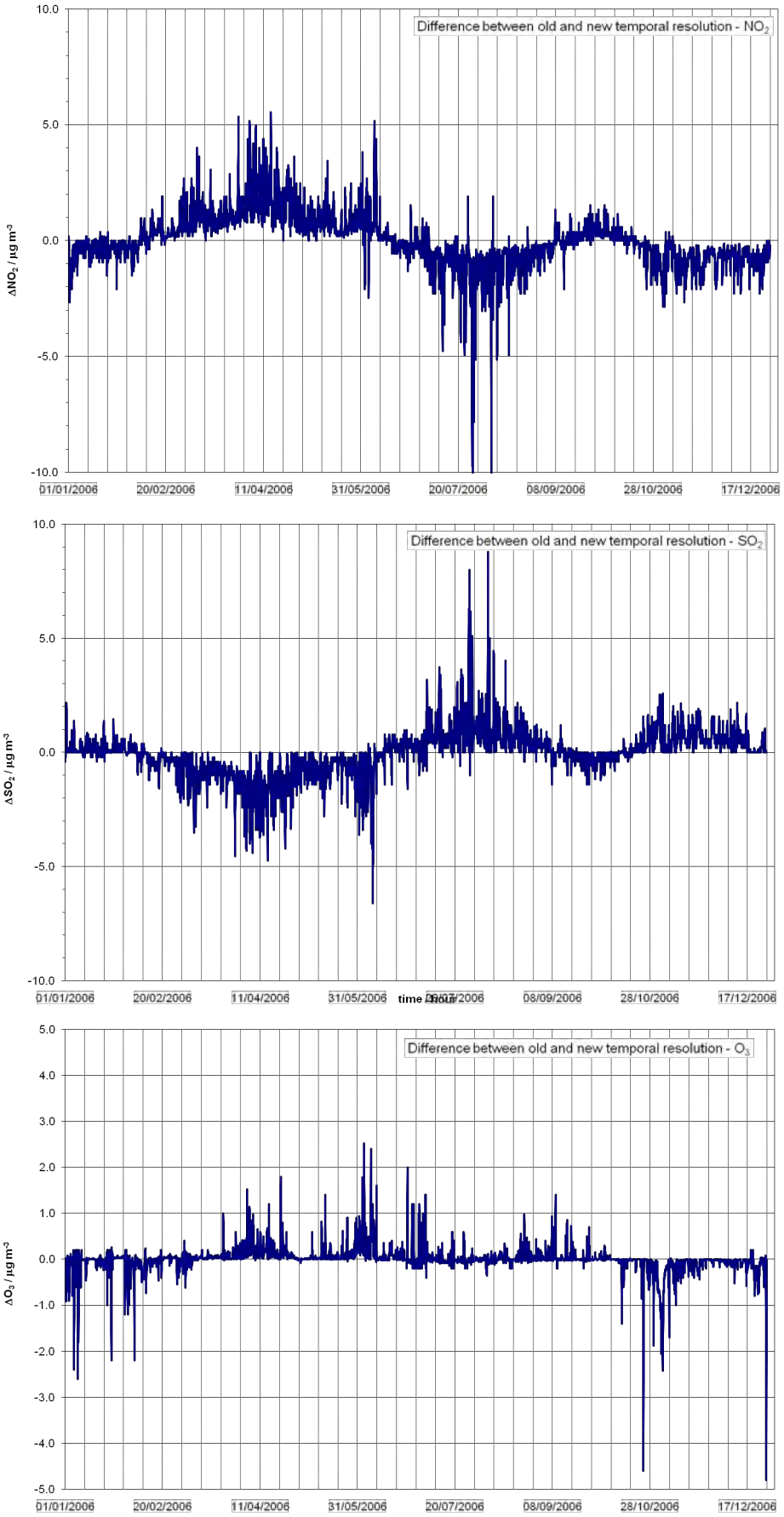

Fig. 14. Comparison of EMEP4UK model runs for the year 2006 using old and improved temporal resolutions for emission input data (calculated as *concentration*[new temp. resolution] – *concentration*[old temp. resolution])

8 Summary and conclusions

To begin with, it is vital to state that the results discussed here need to be views as early stage results, as the focus of this first part of the study was on revisiting the methodology and data availability to improve the temporal resolution of emission input data and to conduct modelling tests to quantify the effect of the changes applied.

Regarding the development of atmospheric dispersion models in the recent past, it is somewhat surprising that comparatively little emphasis has been put on the improvement of how emission input data are processed and introduced into the models. Progress has mainly been made in the case of spatial resolution, with grid resolutions of models going down to 50×50 km^2 on a regional scale and 10×10, 5×5 or even 1×1 km^2 for national or local scale modelling. Where source sectors are concerned, dispersion models often do not realise the full potential of the sectoral detail provided by available emission inventories.

The temporal resolution of emissions, where it has been taken into account at all, is often based on rather crude diurnal or seasonal factors. This introduces potential issues in particular for models applied on a high spatial resolution, where the proximity to the sources and the small scale chemical transformation and transport can have a substantial effect on modelled concentrations and depositions.

The case discussed in this paper, applying the EMEP4UK atmospheric dispersion model for the United Kingdom first with its built-in temporal profiles (which are outdated and coarse) and then with improved temporal factors for selected source sectors (power generation, residential and commercial combustion and road transport) gives a first indication. The discussion of these early results needs to take into account limitations of the data and results displayed. For instance, only weekly and monthly temporal resolution has yet been applied, as the EMEP4UK model (owing to the model structure of its origin, the EMEP Unified Model) is not able to deal with a full hourly resolution at this stage. This necessary adaptation is currently being implemented and will be tested using highly resolved datasets for road transport emissions. In addition to that, the data used to derive temporal profiles are equally prone to uncertainties, as they are based on statistical figures collected for purposes different than generating highly resolved emission datasets. Hence, they often only serve as proxy datasets giving a means to distribute annual total emissions into a reasonable pattern, while not allowing for a fully flexible bottom-up calculation of temporally explicit emission data using emission factors and activity data as a function of time. Finally, the approach taken for deriving the temporal profiles in this study does not directly take meteorological effects into account. As it has been indicated in the discussions above, temperature substantially affects the consumption of fossil fuels in power generation and space heating. However, temperature and other factors indirectly affect emissions for instance from agriculture, due to the temperature dependence of both activities (manure distribution only above certain temperature tresholds) and emission factors (volatilisation of ammonia from manure application). While it is undisputed that these factors have a significant impact on the temporal resolution of emissions, accurate and highly resolved datasets on e.g. manure spreading activities or other relevant activities are not generally available.

The indications emerging from the results described above provide a clear guidance for a further, more thorough analysis of the model performance and as a next step, a set of new temporal factors for a number of years will be tested, including an hourly resolution of road transport emissions. This analysis will be published within the next few months and – hopefully – provide a strong incentive to support the development of up-to-date and sector/source specific temporal profiles for atmospheric dispersion modelling.

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