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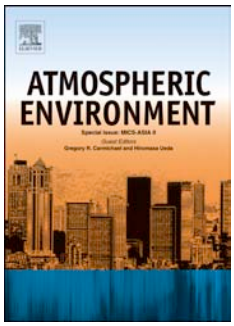

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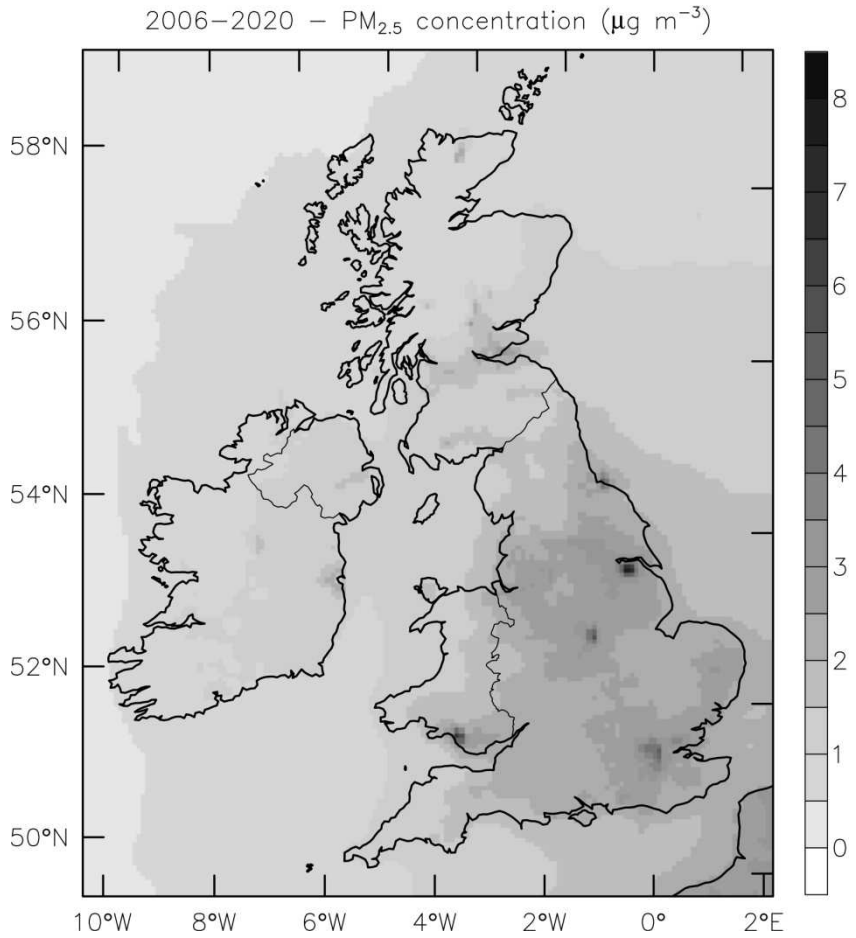

Reduction in the annual average $PM_{2.5}$ concentration between 2006 and 2020 (excluding the contribution from sources within the grid square containing the receptor)

Application of chemical transport model CMAQ to policy decisions regarding PM2.5 in the UK

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Abstract

This paper shows how the advanced chemical transport model CMAQ can be used to estimate future levels of $PM_{2.5}$ in the UK, the key air pollutant in terms of human health effects, but one which is largely made up from the formation of secondary particulate in the atmosphere. By adding the primary particulate contribution from typical urban roads and including a margin for error, it is concluded that the current indicative limit value for $PM_{2.5}$ will largely be met in 2020 assuming 2006 meteorological conditions. Contributions to annual average regional $PM_{2.5}$ concentration from wild fires in Europe in 2006 and from possible climate change between 2006 and 2020 are shown to be small compared with the change in $PM_{2.5}$ concentration arising from changes in emissions between 2006 and 2020. The contribution from emissions from major industrial sources regulated in the UK is estimated from additional CMAQ calculations. The potential source strength of these emissions is a useful indicator of the linearity of the response of the atmosphere to changes in emissions. Uncertainties in the modelling of regional and local sources are taken into account based on previous evaluations of the models. Future actual trends in emissions mean that exceedences of limit values may arise, and these and further research into $PM_{2.5}$ health effects will need to be part of the future strategy to manage $PM_{2.5}$ concentrations.

Keywords

Regional air quality; CMAQ; limit values; emissions inventory; industrial footprint; local traffic pollution

1 Introduction

This paper describes an application of the complex air quality model CMAQ to assess when the UK is likely to meet air quality limit values for $PM_{2.5}$. The Ambient Air Quality Directive (OJEU, 2008) contains an annual mean limit value for $PM_{2.5}$ of 25 μg m^{-3}. Although $PM_{2.5}$ (defined as particulate matter that passes through a size-selective inlet with a 50% efficiency cut-off at 2.5 μm) is thought to be the species of greatest concern to human health, this is not the strictest air quality standard in the Directive. If we assume that the requirement to meet a daily average concentration for PM_{10} of 50 μg m^{-3}, for no more than 35 days in the year, is equivalent to a long-term average of 31.5 μg m^{-3} for PM_{10}, the PM_{10} limit value is equivalent to a $PM_{2.5}$ concentration of about 20 μg m^{-3} $PM_{2.5}$ as an annual average. The purpose of this paper is to determine when such a standard might be reached, given current legislation regarding emission controls. The paper draws on evaluation studies (Fisher, 2013, Fisher *et*

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al., 2013, Hayman *et al*. 2013a, b, c and d) which concluded that there was no formal procedure for evaluating a model and then approving its use. Instead the conclusion was that one should use as good a model as was currently available. The CMAQ model (Chemel *et al*., 2011) is one of the most up to date and sophisticated chemical transport models and is used here. A simple 'local' correction is applied to account for sources within grid squares not treated explicitly within the model. There are other, wider subjective judgments to be made when deciding which model to use, such as whether to employ the same team to do assessments as performed the evaluation studies. The possibility of mistakes being made may then be reduced. However with complex models such considerations cannot be rigorously applied.

Other recent studies have addressed issues relating to the contribution from various source sectors to $PM_{2.5}$. Yim and Barrett (2012) suggest that about 1/6 of the $PM_{2.5}$ concentration is attributable to industrial sources for the year 2005 based on similar CMAQ modelling and suggest that 40% of the $PM_{2.5}$ originates from outside the UK. The authors did not extend their calculations to a future year, nor can one be sure that the modelling was done in exactly the same way as in this paper. The SNIFFER (2010) report references a study by Derwent *et al.* (2009) who examined the modelled concentrations resulting from a 30% reduction in emissions of SO_2, NOx, NH_3, VOC and CO. It was concluded that $PM_{2.5}$ concentrations in rural southern UK are likely to be influenced strongly by reductions in SO_2, NOx and NH_3 emissions in a complex and interlinked manner. The largest reduction in $PM_{2.5}$ was derived from a reduction in NH_3 but such a large decrease is unlikely to occur[2].

The present study examines realistic changes in national emissions between 2006 and 2020 and is therefore directly relevant to policy. The SNIFFER report discusses the 'average exposure indicator', which requires reductions in the annual average $PM_{2.5}$ concentration between 2010 and 2020[3]. AQEG (2012) and Harrison *et al.* (2013) also estimate $PM_{2.5}$ concentrations in 2020 but not by using a fully comprehensive chemical transport model.

AQEG (2012) determines the appropriate equivalent $PM_{2.5}$ limit value in the following way. The 24-hour limit value for PM_{10} of no more than 35 days >50 μg m^{-3} is taken to be equivalent to an annual mean PM_{10} concentration of 31.5 μg m^{-3}. The $PM_{2.5}/PM_{10}$ ratio shown in AQEG (2012) is about 0.7 at urban sites not close to roads. Given the ratios of $PM_{2.5}$ to PM_{10} identified for different parts of the UK, the indicative PM_{10} annual mean limit value can be equated to an annual mean $PM_{2.5}$ value which ranges from 17 μg m^{-3} in Scotland to 24 μg m^{-3} in south east England and about 20 μg m^{-3} in the rest of the country (AQEG, 2012). This paper does not

[2] The parties to the UNECE Air Pollution Convention agreed on 4 May 2012 to a new emission reduction commitment for the main air pollutants in Europe (revision of the Gothenburg Protocol). The revised Protocol requires an overall emissions reduction in the EU of 59% for SO_2, 42% for NOx, 6% for NH_3 and 28% for NMVOC between 2005 and 2020, and for the first time a limit on primary $PM_{2.5}$ emissions involving a reduction of 22% between 2005 and 2020. For the UK the reductions are 59% for SO_2, 55% for NOx, 8% for NH_3, 32% for NMVOC and 30% for $PM_{2.5}$. The reductions in emissions from the large stationary sources which the Environment Agency regulate are approximately similar fractions, and are broadly based on the assumption that all processes will operate with new technology. The revision of the Gothenburg Protocol is implemented in practice by regulations on sources, through the Integrated Pollution Prevention and Control Directive, undergoing replacement by the Industrial Emissions Directive (OJEU, 2010), and Euro standards on motor vehicles. Other Directives, such as the Large Combustion Plant Directive (OJEU, 2001), require emissions standards consistent with the revised Protocol. Emission reductions are expected to be achieved through the gradual introduction of new technology standards, unless there is an unexpected increase in emissions from small unregulated sources, through wood burning for example, or a failure in the application of new technology to road vehicles.

[3] The 'average exposure indicator' representing the 3 year average exposure at urban monitoring sites, not close to roads, is related to the population weighted annual mean $PM_{2.5}$ concentration in urban areas of the UK. The UK target is for a 15% reduction between 2010 and 2020, while the EU target is either 10% or 15%.

consider other particle metrics, such as the concentration of black carbon particles. The EU limit value for $PM_{2.5}$ is 25 µg m^{-3} by 2015, with a stage 2 indicative value of 20 µg m^{-3} by 2020. The National Ambient Air Quality Standard in the USA for $PM_{2.5}$ is an annual mean of 15 µg m^{-3} averaged over 3 years (Hogrefe *et al*., 2011). These authors used CMAQ to investigate differences in the $PM_{2.5}$ concentrations depending on the biogenic emissions under a NOx emission control scenario in eastern North America.

There is evidence (Jannsen *et al*., 2011) that black carbon particles could be the most important indicator of the health risk of particles. This would be significant in situations where particulate concentrations are dominated by primary road traffic sources, whereas the calculations presented in this paper refer to regional $PM_{2.5}$ with a large secondary component. It is assumed that the levels in the Ambient Air Quality Directive determine the health benefit of emission reductions, and therefore it would not be appropriate to evaluate calculated concentrations using another metric. However one should correct the calculated concentrations by estimating roadside concentrations of $PM_{2.5}$.

2 The CMAQ Model Setup

The Community Multiscale Air Quality (CMAQ) modelling system is a comprehensive modelling system developed by the US Environmental Protection Agency (USEPA). CMAQ is an Eulerian photochemical air quality model in which complex interactions between atmospheric pollutants on urban, regional and hemispheric scales are treated in a consistent framework. It is designed for assessing the impact of multiple pollutants including tropospheric ozone and other oxidants, aerosols and acid deposition.

In this application of CMAQ an outer domain over Europe with a grid resolution of 18km was used, with an inner domain over the UK with a grid resolution of 6km. So over the UK primary roadside and urban emissions within about 3 km of a monitoring site will not be included. The inner domain covers the British Isles. The outer domain stretches from about 33 degrees north 20 degrees west to about 70 degrees north 32 degrees east.

The European emission inventory for 2006 is based on the TNO (http://www.tno.nl/) inventory, which consists of anthropogenic emissions from ten Selected Nomenclature for Air Pollution (SNAP) source sectors and international shipping. Biogenic gas emissions were included in the CMAQ model from the Model of Emissions of Gases and Aerosols from Nature (MEGAN) system at the same resolution as the anthropogenic emissions. Biomass burning emissions from wild fires for 2006 were based on daily fire estimates from the Moderate Resolution Imaging Spectroradiometer (MODIS) fire radiative power product (Sofiev *et al*., 2009).

A 2020 European emissions scenario was considered based on the MEGAPOLI (Baklanov *et al*., 2010) project results (Theloke *et al.* 2010), which makes use of the:
(1) Integrated MARKAL-EFOM System (TIMES) Pan-European (TIMES PanEU) energy system model for the energy related sectors;
(2) Greenhouse gas – Air pollution Interactions and Synergies (GAINS) model and other assumptions for the non-energy related sectors; and
(3) a reduction of the greenhouse gas emissions by 30% by 2020 compared to 1990.

Major industrial sources are defined as those with annual emissions of SO_2 greater than 500t yr^{-1} and/or annual emissions of NO_x greater than 500t yr^{-1} and/or annual emissions of PM_{10} greater than 200t yr^{-1}. 2006 is the base line year for the calculations. All major industrial emissions regulated in the UK in 2006 were adjusted to enable 2020 concentrations to be modelled. Information on likely trends in regulation between 2006 and 2020 is available, but

the activity of each industrial sector cannot be reliably forecast. A site by site 2020 emissions estimate was therefore not feasible. Two industrial estimates for 2020 are available in the public domain from the AEA Atmospheric Emissions team (Wagner *et al.*, 2009) from which the mean was adopted.

The ratio of the emissions of SO_2, NOx, PM_{10} and NMVOC between 2006 and 2020 are estimated for major industrial releases and for the national total emissions. These are shown in Table 1.

[Table 1 here]

These ratios were used to scale UK major industrial emissions to obtain representative emissions for 2020. Ammonia is a special case and no correction is made. Since the ratio SO_2:NH_3 should be lower in 2020 than 2006, the rate of production of $PM_{2.5}$ for a given fixed source could be greater in 2020 compared to 2006. Based on results of the EMEP chemical transport model, a comparison (Fisher, 2012) of the radial footprints of annual average $PM_{2.5}$ concentration for different past years and for different individual countries, UK and Germany with different source strengths, does not indicate strong differences in behaviour as a function of distance from the centre of the country. The tentative conclusion is that chemical transport models have not demonstrated that the annual average $PM_{2.5}$ concentration has a very strong non-linear dependence on the change in emissions. However further studies investigating greater changes in background atmospheric composition are needed to understand the relationship more fully.

The estimated ratios in emissions between 2006 and 2020 in Table 1 are in reasonable agreement with previous projections by Vincent and Abbott (2008). There are some differences with the MEGAPOLI projections for the UK energy related sectors, but when differences arise the most pessimistic case is adopted.

The CMAQ system setup involves the Weather Research and Forecasting (WRF) meso-scale model, embedded within the ECMWF regional model for the meteorological fields. The initial and lateral meteorological boundary conditions of the outer domain were derived from the European Centre for Medium-range Weather Forecasts (ECMWF) gridded analyses available every 6 h with a horizontal resolution of 0.5 degrees on operational pressure levels up to 50 hPa for vertically distributed data, and at surface and soil levels for surface and deep-soil data. A grid nudging technique was employed for the outer domain every 6 h in order to constrain the model towards the analyses. The Met Office have run their global HADGEM2_ES model (Collins *et al*., 2011) and obtained regional meteorological fields for 2020 applying the RCP8.5 emissions scenario, which may be regarded as a pessimistic scenario. The Met Office 2020 meteorological fields used to develop an alternative 2020 $PM_{2.5}$ climate change projection are thought to be representative of the decade. The difference between the 2006 and 2020 meteorological fields is one of many possibilities which could occur, but the approach illustrates a straightforward methodology which further large-scale computing could develop further.

The CMAQ modelling system requires hourly emissions data of primary pollutants. SMOKE has been developed for this purpose and can be adapted to process annual emissions data (from point, line and area sources) into temporally-resolved, spatially-distributed and speciated emissions files ready for the chemical transport model.

For CMAQ version 4.7, adopted in this study, the CB05 chemical mechanism was used. The CB05 mechanism treats the formation of secondary organic aerosols. The tri-modal approach to aerosol size distribution was used in order to model particulate matter. The species modelled include sulphate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), sodium (Na^+), chloride

(Cl^-), water (H_2O) and organics from precursors of anthropogenic and biogenic origin but only the total PM_{10} and $PM_{2.5}$ concentrations are considered in this paper. Each mode (Aitken, accumulation and coarse) is subject to both wet and dry deposition. Documentation on CMAQ is available from the official CMAQ website (http://www.cmaq-model.org) [accessed 21 May 2013]. Chemical boundary conditions were obtained from runs of the global model Geos-Chem.

3 Model Evaluation and Uncertainty

The Defra Model Evaluation Protocol (Derwent *et al*. 2010) sets the following criteria for the acceptance of a model. The predictions of a model should be accepted if the percentage of model predictions within a factor of two (*FAC2*) of the observations is greater than 50 per cent and the magnitude of the normalised mean bias (*NMB*) is less than 0.2.

The normalised mean bias (*NMB*) is defined as:

$$NMB = \frac{\sum_{i=1}^{N} M_i - O_i}{\sum_{i=1}^{N} O_i} \qquad (1)$$

where N is the number of observations, M_i are the calculated values, O_i are the observed values. *NMB* should satisfy $-0.2 \le NMB \le 0.2$.

A limited evaluation of CMAQ predictions of $PM_{2.5}$ over the UK for the year 2005 was undertaken by Yim and Barrett (2012). Only three AURN monitoring sites, where $PM_{2.5}$ was measured, were operating in 2005: Harwell (rural), Stoke (rural), Bloomsbury (urban background) and a fourth roadside site at Marylebone. The *NMB* was -0.23 at Harwell -0.09 at Stoke and -0.27 at Bloomsbury.

The multi-model AQMEII study (Solazzo *et al.* 2012) compared $PM_{2.5}$ at many sites in North America and Europe in 2006. It included some CMAQ calculations for Europe by the authors of this paper. These generally showed significant under-prediction with a *NMB* of about -0.4. However the CMAQ predictions reported in this paper of individual components of particulate matter at Harwell for the year 2006, such as inorganic species (SO_4, NO_3 and NH_4), and elemental carbon and organic carbon, do not show the same systematic under-prediction. The implication is that the model does not include all components and sources of $PM_{2.5}$. One would anticipate better model performance for industrial sources for which the primary and secondary particulate species source terms are better known.[4] The evaluations undertaken in previous studies of CMAQ (see Table 2) are taken as the justification for CMAQ's use in this paper.

[Table 2 here]

It was assumed that the estimate from the CMAQ model, excluding the contribution from sources within the grid square containing the receptor, is a 20% under-prediction of the concentration, based on previous regional model evaluations, and therefore a margin of safety of 20% was assumed, in order to be confident that total concentrations meet the indicative limit

[4] In 2011 there were 67 AURN (Automatic and Rural Monitoring Network) sites at which $PM_{2.5}$ was measured but most are influenced by nearby sourcesl and therefore of limited value in evaluating the performance of CMAQ (http://uk-air.defra.gov.uk/data/exceedence [accessed 21 May 2013]). These sites mainly used equipment giving the total concentration of particulate matter, not its components.

value concentration of about 20 μg m^{-3}. A precautionary estimate of the annual average $PM_{2.5}$ concentration in 2006 (see Fig. 1) varies between 7 (=6x6/5) μg m^{-3} in the north of the UK to 12 (=6x10/5) μg m^{-3} in south-east England. Later in this paper the road contribution is included as a separate item in the $PM_{2.5}$ budget. Other sources within grid squares, such as from small stationary sources, are excluded from the calculation. Small stationary sources are not as widely distributed as traffic sources, affect fewer grid squares and should be subject to local air quality management control.

[Fig.1 here]

4 Annual average $PM_{2.5}$ concentrations over the UK in past and future years

2006 was a year when forest fires were prevalent in parts of Europe. Runs of CMAQ, including and not including the forest fire contribution, suggested that the impact of forest fires on annual average $PM_{2.5}$ concentration over the UK was small, of order 3% or less. The contribution of wild fires can therefore be discounted.

The contribution to annual average $PM_{2.5}$ concentrations in 2006 from the CMAQ model excluding sub-grid scale sources is well below the indicative limit value of 20 μg m^{-3}. In some other parts of Europe the concentration in 2006 is higher, approaching but not exceeding the indicative limit value of 20 μg m^{-3}. The sub-grid scale road (and small stationary source) contributions are not included in these estimates.

The observed annual average $PM_{2.5}$ concentration consists of contributions from a mixture of primary and secondary sources over very short and very long travel distances from their point of emission. In order to improve understanding, the contribution from major stationary sources was estimated by rerunning the full model excluding all emissions from major industrial sources regulated in the UK. This then gives the footprint of the $PM_{2.5}$, over distances exceeding 6km, arising from major industrial sources regulated in the UK.

[Fig. 2 here]

The highest concentrations of $PM_{2.5}$ from major industrial sources lie close to the cluster of major industrial sources in the Midlands. The concentration declines with distance from the Midlands mainly as a consequence of the spread in air mass trajectories (Fisher *et al*., 2011). The highest concentrations in the Midlands amount to 1 to 2 μg m^{-3}. As a percentage, the major industrial sources regulated in the UK make up between 10 and 20% of the $PM_{2.5}$ over England in 2006 excluding the contribution from sources within the grid square containing the receptor. Using the simpler semi-empirical TRACK-ADMS model, Vincent and Abbott (2008) estimated that the contribution of major regulated sources was 10% of the average PM_{10} concentrations in the UK in 2005.

[Fig. 3 here]

Fig. 3 shows the annual average $PM_{2.5}$ concentrations (in μg m^{-3}) from all sources excluding the contribution from sources within the grid square containing the receptor in the 2020 emissions projection, using 2006 meteorology. This shows a significant reduction in concentrations compared with 2006, with levels less than 6 μg m^{-3} over most of the country, only reaching 6 μg m^{-3} in the south east of the country. This is a reduction of about 40%. The difference between Fig. 1 and Fig. 3, the reduction in the annual mean $PM_{2.5}$ concentration, is above 2 μg m^{-3} over much of England and about 1 μg m^{-3} over areas more remote from major population centres. The reduction is shown in the graphical abstract to this paper. From this

single calculation, the sector with the main reduction *e.g*. possibly Europe, UK road transport or UK industrial emissions, cannot be identified.

A further calculation was made to determine the contribution of emissions from major industrial sources regulated in the UK in 2020, using 2006 meteorology, the so-called industrial footprint in 2020 (see Fig. 4). The maximum concentration from major sources regulated in the UK amounts to 0.5 to 1 μg m^{-3} in the Midlands, near to the main cluster of major industrial sources. As a fraction of the total annual average $PM_{2.5}$ concentration, the industrial footprint makes up 5 to 15% of $PM_{2.5}$ concentrations across most of the UK in 2020. This constitutes a reduction of about 50% in the major industrial source contribution in the region close to the main cluster of major industrial sources. The main source contributing to the industrial footprint is the production of secondary particulate matter from emissions of SO_2 and NOx. In 2006, the total emissions from major regulated stationary sources in the UK amounted to about 453 kt yr^{-1} SO_2, 441 kt yr^{-1} NOx, 17 kt yr^{-1} PM_{10} and 6 kt yr^{-1} $PM_{2.5}$ giving a total of about 900 kt per annum of 'potential' $PM_{2.5}$ emissions (SO_2+NOx+$PM_{2.5}$) if all these primary emissions were converted to $PM_{2.5}$. The equivalent industrial footprint source strength in 2020 is 168 kt yr^{-1} SO_2, 197 kt yr^{-1} NOx, 7 kt yr^{-1} PM_{10} and 5 kt yr^{-1} $PM_{2.5}$ giving an indicative 'potential' $PM_{2.5}$ annual emission (SO_2+NOx+$PM_{2.5}$) of about 370kt. Thus the total primary emission from regulated major industry expressed as the total of the SO_2 + NOx + $PM_{2.5}$ emissions would have reduced to about 40% of its 2006 level by 2020. The reduction in annual mean $PM_{2.5}$ concentrations of about 50% in the region of the sources is approximately of the same order. Thus 'potential source strength' is a possible, useful indicator of the response of the atmosphere to changes in emissions.

The change in the total $PM_{2.5}$ concentration between 2006 and 2020 is not a useful indicator because the change comes about from a mixture of contributions to $PM_{2.5}$ concentration over the UK. The response will be variable, depending on whether the response is from distant primary emissions in Europe, mainly from sources in the south east, or from regional industrial UK sources centred in the Midlands, or from urban UK emissions, spread around the country. No single indicator is representative of all these responses, which depend on different geographical distribution changes between 2006 and 2020.

Chemical transport models are designed to inform on the possible non-linear relationship between the change in emissions and the resulting change in concentration of $PM_{2.5}$. One could imagine that if the atmosphere became relatively more reactive, secondary $PM_{2.5}$ could be formed more quickly near to locations where it was emitted. Lifetimes can be estimated from the footprints of a specified source at a single location (Fisher *et al*., 2011). This is not possible here because the major industrial sources are distributed around the country. However the change in the primary emissions of major industrial sources is roughly in proportion to the change in the $PM_{2.5}$ concentration, near the cluster of major industrial sources, suggesting no evidence of very large non-linearity. The change in emissions between 2006 and 2020 may not be large enough to provide evidence of non-linearity and more testing of responses to changes in emission should be undertaken.

[Fig. 4 here]

5 Climate change and population exposure

Because of the availability of an alternative meteorological scenario in 2020 from results of the Met Office's HadGEM model, an estimate can be made of the change in annual mean $PM_{2.5}$ concentrations in 2020 as a possible consequence of climate change. It turns out that the effect of the chosen climate change scenario is to reduce annual average $PM_{2.5}$ concentrations

in 2020 by of order 10% over the country, representing reductions of 0.2 to 0.5 μg m^{-3} in England as a result of a different climate. This is much less than the reduction in regional concentration from emission changes between 2006 and 2020. The treatment of climate change did not include interactions between climate and pollution, such as the effect particle concentrations arising from emission changes might have on radiation and cloud formation, cloud duration and thickness. The version of CMAQ used, v4.7, is not a fully coupled model calculation. As the climate effect appears small, this is a preliminary indication that it may not be worthwhile running the CMAQ model for a range of alternative 2020 $PM_{2.5}$ climate change projections.

An alternative way of estimating the benefit of emission reductions between 2006 and 2020 is to determine the population weighted annual mean $PM_{2.5}$ concentration. The reduction in the population weighted annual mean is about 40%. The additional fractional contribution of climate change in 2020 is estimated to be about 5%. Some slight non-proportional differences between 2006 and 2020 may occur because of changes in population. The fractional contribution of major industrial sources to the population weighted annual mean $PM_{2.5}$ concentration in 2006 is about 14%.

The percentage change in population weighted mean $PM_{2.5}$ concentrations between 2006 and 2020 can be compared with the EU Directive 'average exposure indicator' target value. For a 15% reduction in average exposure indicator, SNIFFER (2010) estimated that a reduction in the annual mean $PM_{2.5}$ of 1.5 μg m^{-3} over England, and a 2 μg m^{-3} over inner London between 2010 and 2020, is required. The SNIFFER estimate of $PM_{2.5}$ concentrations in 2010, based on observations at urban sites not at the roadside, is not strictly equivalent to the CMAQ estimate. However the reduction from the CMAQ estimate of a nearly 40% reduction in the population weighted annual mean $PM_{2.5}$ concentration between 2006 and 2020 appears large enough to satisfy the 'average exposure indicator' reduction target.

6 The local contribution to $PM_{2.5}$

The CMAQ estimate of the population weighted annual mean $PM_{2.5}$ concentration reduction does not include the roadside contribution to $PM_{2.5}$ concentrations. This is likely to be a small fraction of the population exposure as traffic concentrations decline rapidly with distance from the road. However it is valuable to know the possible contributions from sources within a grid square to check whether the indicative annual average limit value of 20 μg m^{-3} is likely to be exceeded at some locations in major urban areas, such as London.

We have estimated the concentration of primary $PM_{2.5}$ near to roads using the GRAM model (Fisher and Sokhi, 2000). This requires future road vehicle emission factors[5] (see Table 3).

[5] The road traffic Emissions Factors Toolkit released by the LAQM Support Helpdesk http://laqm.defra.gov.uk/review-and-assessment/tools/emissions.html#eft [accessed 21 May 2013] utilises revised NOx emissions factors and vehicle fleet information. NOx emissions factors previously based on DFT/TRL https://www.gov.uk/government/publications/road-vehicle-emission-factors-2009 [accessed 3 May 2013] functions have been replaced by factors from COPERT 4 v8.1. This reference gives standard emission factors under urban, rural and motorway conditions for various types of vehicles according to Euro category. These emission factors were published in May 2011 by the European Environment Agency and are widely used for the purpose of calculating emissions from road traffic in Europe. The COPERT model is available to download from http://www.emisia.com/copert/ [accessed 21 May 2013]. Fleet weighted road transport emission factors based on the COPERT model are also available from the National Atmospheric Emission Inventory web site http://naei.defra.gov.uk/data/ef-transport [accessed 21 May 2013] and include emission from cold starts, brake and tyre wear, and road abrasion for recent years. Fleet projections giving vehicle type according to Euro class in future years, as well as primary NO_2 emissions, are also provided according to type of road (urban, rural and motorway). The simple addition of the roadside concentration to the regional concentrations from the CMAQ model would not be appropriate for calculating roadside NO_2 because of the non-linear chemistry involving ozone

The persistent negative bias (underestimate) in the CMAQ calculations may also provide a rough estimate of the sub-grid square contribution.

[Table 3 here]

Assuming representative flows of 25,000, 50,000 and 100,000 vehicles per day for typical urban and rural roads and a motorway, and a margin of error of +20%, the roadside concentrations at distances of 5 to 100m from the road centre line have been calculated using the model GRAM. The typical urban road is likely to be a street canyon.

The urban concentrations in central London are taken to be 12 μg m^{-3}, 7.2 μg m^{-3} and 7.2 μg m^{-3} in 2010, 2020 and 2030, from the 2005 and 2020 CMAQ results in this paper, including a 20% margin of error. Estimates of the local primary $PM_{2.5}$ concentration at a typical roadside location in London, 5m from an urban road with the daily traffic flow of 25,000 in a street canyon are 5.3 μg m^{-3}, 4.6 μg m^{-3} and 4.4 μg m^{-3} in 2010, 2020 and 2030. The small extra improvement beyond 2020 arises partly because road emissions are then dominated by non-exhaust emissions. Future Euro standards will have little effect on these emissions. In addition in urban areas, where most people start their journeys, emissions during cold starts are an important factor. The total $PM_{2.5}$ roadside concentrations in 2006, 2020 and 2030 are estimated to be 17.3 μg m^{-3}, 11.8 μg m^{-3} and 11.6 μg m^{-3} near a typical road, suggesting widespread compliance with the target limit value. This does not definitely confirm that $PM_{2.5}$ will not be an issue at some locations, but it appears not to be a general issue accepting approved emissions standards. Near small stationary sources where building downwash may occur, local air quality management should be considered the tool for addressing exceedences.

This practical estimate of future $PM_{2.5}$ concentrations over the UK combines a simple assessment of road traffic emissions with the contribution from a complex model. The estimate does not contain estimates near to every kind of road. Exceptional cases, such as an urban road with very heavy traffic, could lead to higher concentrations but these cases should be treated using local air quality management action plans. The local air quality management system can also be used as the process for checking that future emissions follow expected trends. Actual trends may not follow the expected trend because (1) technological measures addressing vehicle emissions do not perform as expected, (2) trends in other types of emissions do not occur as forecast[6], or (3) unanticipated future trends in the way mobility, power and heating services are provided. Policy should be flexible to adjust for the actual future trends in emission when they occur.

7 Conclusions

From calculations using the CMAQ model it was concluded that in 2006 wild fires make a minor contribution to annual mean $PM_{2.5}$ concentrations in the UK. The contribution of emissions from major industrial sources regulated in the UK, in 2006, amounts to 1 to 2 μg m^{-3} in the Midlands. This sector makes up 10 to 20% of the annual average $PM_{2.5}$ concentration across most of the UK, excluding the contribution from sources within the grid square containing the receptor.

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and primary NO_2 emissions, but for $PM_{2.5}$ for which roadside concentrations are mainly primary emissions, either from exhaust (cold starts) and non-exhaust emissions (tyre and brake wear, road abrasion), such an approach is acceptable.

[6] Biomass burning in small installations in urban areas has been promoted in recent years as a way of addressing greenhouse gas emissions, but this has the disadvantage of increasing primary particulate emissions,

The impact of 2020 emissions, using 2006 meteorology, is a reduction of above 2 μg m^{-3} in the annual average $PM_{2.5}$ concentrations over England excluding the sub-grid square contribution, a reduction of 20 to 40% across most of the UK. The reduction in the population weighted mean because of emission reductions between 2006 and 2020 is estimated to be 40%.

The contribution of emissions from major industrial sources regulated in the UK to regional annual average $PM_{2.5}$ concentrations in 2020, using 2006 meteorology, amounts to 0.5 to 1 μg m^{-3} in central regions of the country near the main cluster of industrial emissions. This contribution makes up 5 to 15% of the annual average $PM_{2.5}$ concentrations excluding the sub-grid square contribution, across most of the UK. The potential source strength of regulated major industrial emissions expressed as the total of SO_2 + NOx + $PM_{2.5}$ emissions would have reduced to about 40% of their 2006 level in 2020. The reduction in annual mean $PM_{2.5}$ concentrations of about 50% in the Midlands is approximately of the same order. Thus 'potential source strength' is a possible, useful indicator of the response of the atmosphere to changes in emissions.

Applying meteorological conditions in 2020 representing a possible climate change scenario, with a 2020 emissions scenario, suggests reductions of 0.2 to 0.5 μg m^{-3} in the regional annual average $PM_{2.5}$ concentrations over England, equivalent to a reduction of 2 to 5% across most of the UK compared to $PM_{2.5}$ concentrations under 2006 meteorological conditions and a 2020 emissions scenario. The 'climate change scenario' reduction is between 5 to 10 times smaller than the reduction brought about by emissions changes between 2006 and 2020.

Estimates of the local primary $PM_{2.5}$ concentration at a typical roadside location in London have been added to the regional $PM_{2.5}$ concentration to test whether compliance with the indicative $PM_{2.5}$ limit value of 20 μg m^{-3} in 2020 will be achieved. The total $PM_{2.5}$ concentrations in 2006, 2020 and 2030, including a margin of error, are 17.3 μg m^{-3} , 11.8 μg m^{-3} and 11.6 μg m^{-3} suggesting widespread compliance with the target limit value. This does not confirm that $PM_{2.5}$ will not be an issue at some locations, but it appears not to be a general issue accepting current air quality standards and likely trends in emissions. At hot spots, where traffic flows are higher than at a typical urban road, local air quality management should be considered the tool for addressing $PM_{2.5}$ exceedences. The introduction of new emission technology standards is the tool for addressing air quality considerations on a national basis.

There is uncertainty associated with the air quality standard used in this paper. We have adopted the approach, implicit in the Ambient Air Quality Directive, that all components of $PM_{2.5}$ have the same health disbenefit and the aim should be to reach and maintain the strictest standard implied by the values stated in the Directive. If further research suggests that certain components of $PM_{2.5}$ are the active components causing harm and $PM_{2.5}$ acts as the surrogate for these components in the underlying epidemiological studies, then the conclusions may be different. Moreover understanding the observed trends in annual average concentrations is a necessary step to ensure compliance, and modelling can only suggest a strategy towards ensuring future compliance. Unexpected changes in emissions and unexplained observational trends compared with model predictions may still arise.

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Figure Captions

Fig. 1 Annual average regional $PM_{2.5}$ concentrations (μg m^{-3}) in 2006 from all sources (excluding the contribution from sources within the grid square containing the receptor)

Fig. 2 Annual average regional $PM_{2.5}$ concentrations (μg m^{-3}) from major industrial source regulated in the UK in 2006

Fig. 3 Annual average regional $PM_{2.5}$ concentrations (μg m^{-3}) in 2020 from all sources using 2006 meteorology (excluding the contribution from sources within the grid square containing the receptor)

Fig. 4 Annual average regional $PM_{2.5}$ concentrations (μg m^{-3}) from emissions from major industrial sources regulated in the UK in 2020 using 2006 meteorology (the 'industrial footprint' in 2020)

Pollutant	Ratio 2020:2006 major industry emissions	Ratio 2020:2006 national total UK emissions
SO_2	0.37	0.47
NOx	0.45	0.52
PM_{10}	0.42	0.72
$PM_{2.5}$	0.65	0.68
NMVOC	0.87	0.72

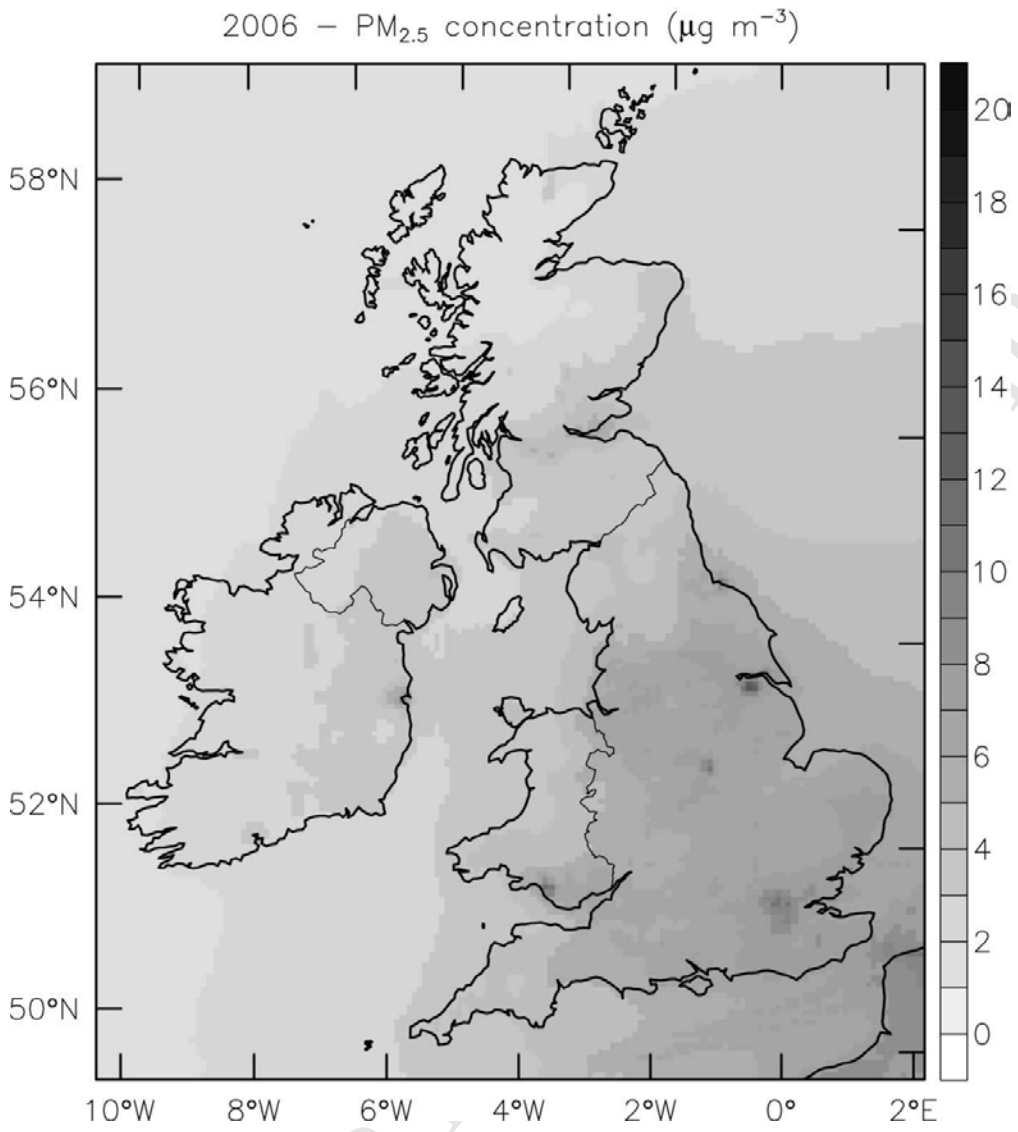
Table 1 Estimated ratio of the major industrial emissions and national total emissions of SO_2, NOx, PM_{10} and NMVOC between 2006 and 2020

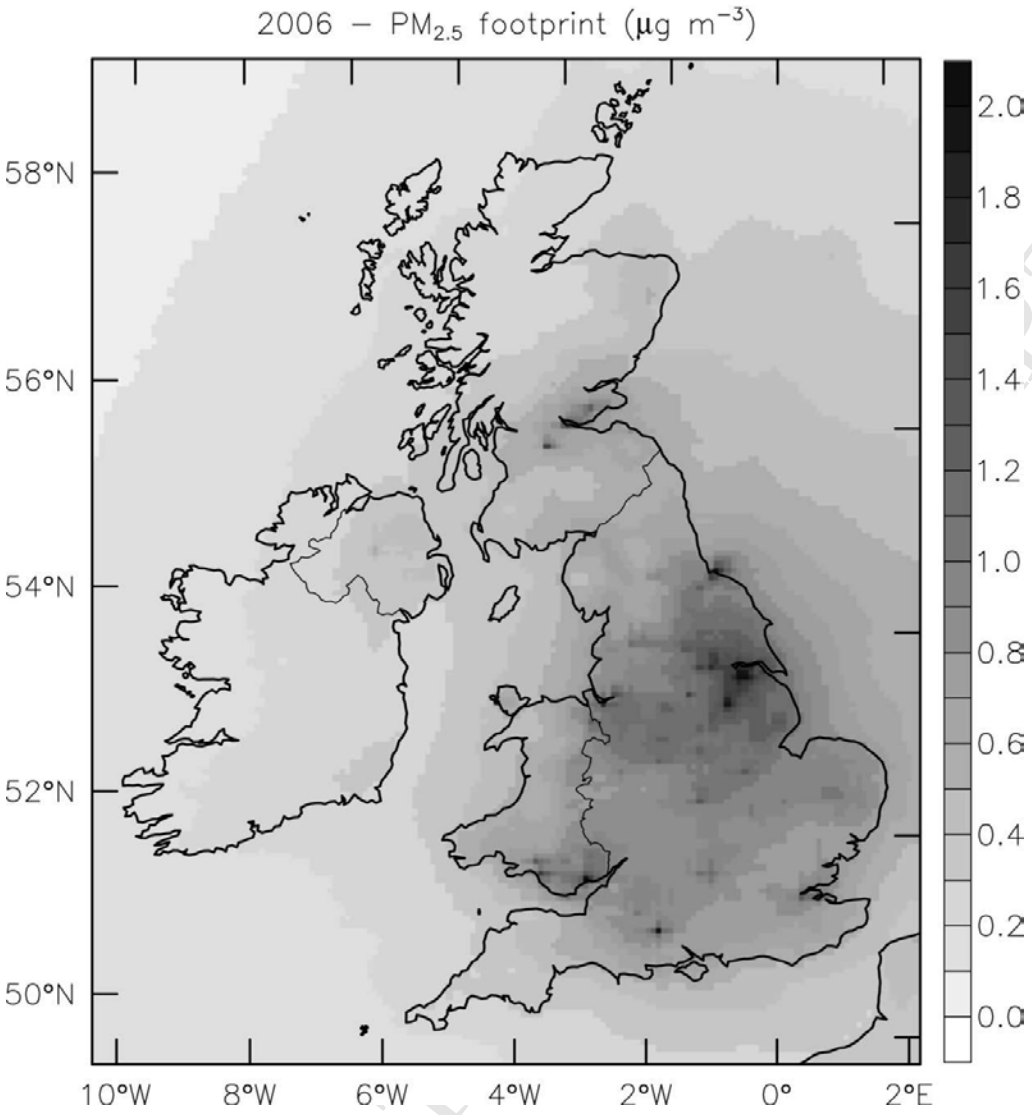
Model metric PM_{10} for 2003	CMAQ v4.6	CMAQ v4.7	TRACK-ADMS
FAC2 (%)	88.2	100	100
*r (*correlation coefficient)	0.09	0.0	0.45
NMB	-0.33	-0.09	-0.20
Single power station contribution (%)	0.34	0.28	0.28

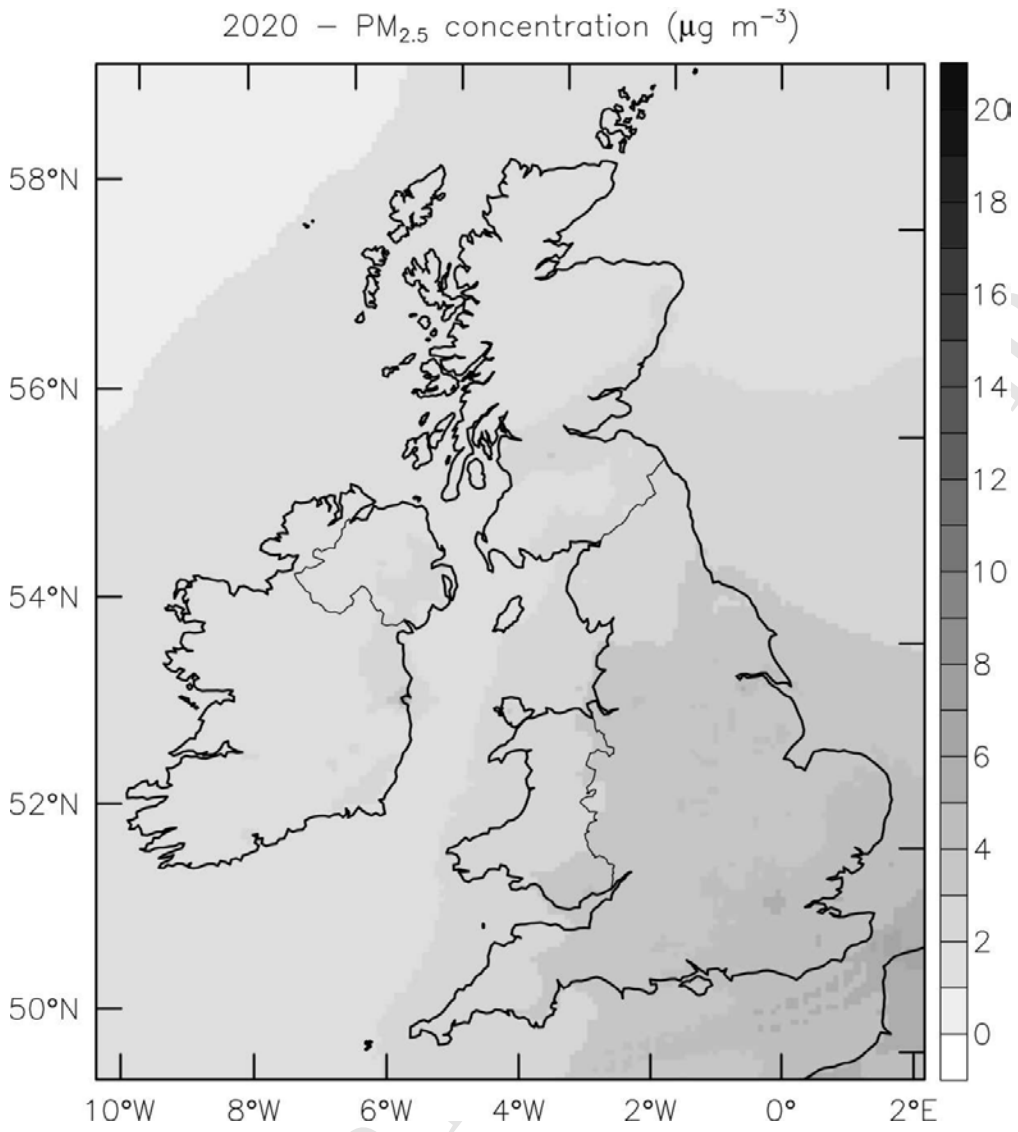
Table 2 Comparison of performance in predicting annual average PM_{10} concentration in 2003 at 40 urban background and rural background sites in the AURN network in the UK for two versions of the advanced model CMAQ (Chemel *et al.* 2011) and a simpler, semi-empirical model TRACK-ADMS (Vincent and Abbott, 2008).

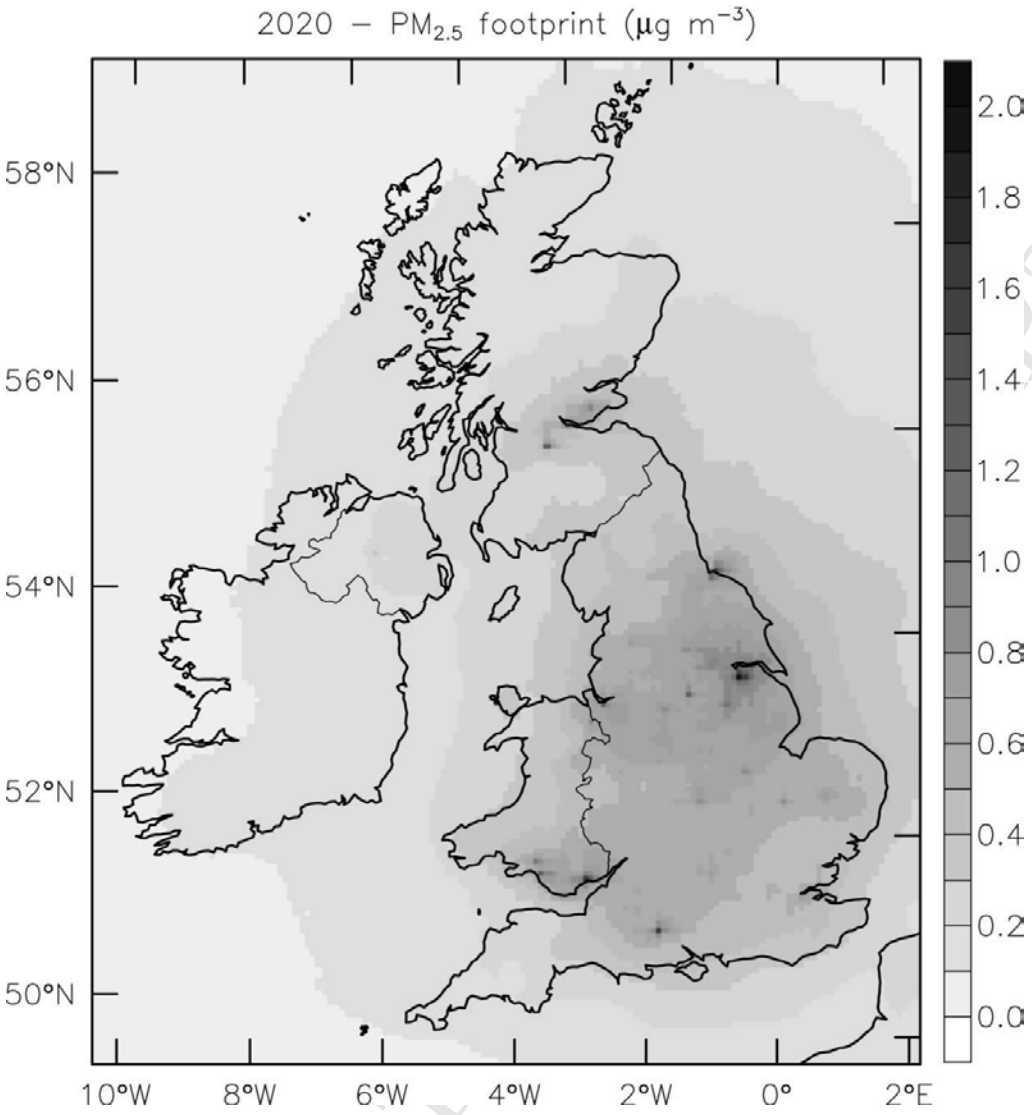
Year	LDV			HDV		
	Urban	Rural	Motorway	Urban	Rural	Motorway
2010	0.38	0.37	0.47	5.02	4.71	5.23
2020	0.28	0.21	0.42	1.47	0.76	0.77
2030	0.14	0.15	0.23	0.45	0.45	0.53

Table 3 Emission factors (g $PM_{2.5}$/km) for light duty vehicles (LDV) and heavy duty vehicles (HDV) in current and future years for typical roads









- Model CMAQ evaluates UK regional PM2.5 concentration for 2006 and 2020.
- A correction for local traffic sources within a major city is made.
- PM2.5 complies with implied air quality standards.
- CMAQ model can be applied to answer policy questions.
- Industrial source footprint contribution is less than 15% of PM2.5.