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

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

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## Summary

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 climate change between of 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.

## 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 (OUJ, 2008) contains a 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 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

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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 (2010) suggests that local sources tend to contribute less than half of the total predicted $PM_{2.5}$ concentration, and often less than 10%, even at roadside receptors. Urban sources give rise to an annual average urban background enhancement of around 3 to 6 µg $m^{-3}$ and secondary $PM_{2.5}$ contributes around 4 to 6 µg $m^{-3}$. SNIFFER recommended that modelling studies are carried out to establish the contributions of UK and other EU emissions of precursor gases to annual mean $PM_{2.5}$ concentrations, and to determine how these contributions will respond to changes in emissions, in order to guide the development of the most cost-effective control programme. This study is a step to fulfilling this need. The SNIFFER report discusses the 'average exposure indicator', which requires reductions in the annual average $PM_{2.5}$ concentration of about 1.5 to 2 µg $m^{-3}$ between 2010 and 2020[3].

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 background sites. Given the ratios of $PM_{2.5}$ to $PM_{10}$ identified for different

[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. These 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, or a failure in the application of new technology on road vehicles.

[3] The 'average exposure indicator' representing the 3 year average exposure at urban background monitoring sites 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%.

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 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 annual National Ambient Air Quality Standard in the USA for $PM_{2.5}$ is 15 μg $m^{-3}$ (Hogrefe *et al*., 2011). These authors used CMAQ to investigate differences depending on the biogenic emissions in the $PM_{2.5}$ concentrations under a NOx emission control scenario in eastern North America.

There is evidence (Jannsen *et al*., 2011) that black carbon particles could be the 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 regional concentrations using another metric. However one should correct the calculated regional 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 a 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 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 make 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.

| 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

These ratios were used to scale UK major industrial emissions to obtain representative emissions for 2020. The influence of changes of order 30% or more in the main air pollution emissions are assessed in the model. 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 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 are 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 the 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 and 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.

| 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.

Rather than set an uncertainty bound on predictions it was assumed that the CMAQ model under-predicts at regional background sites, because the contribution from local sources, within the 6km long grid squares containing monitoring sites, is not included. It was assumed that the regional estimate was approximately a 20% under-prediction of the regional 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 met the indicative limit value concentration of about 20 μg $m^{-3}$. A precautionary estimate of the regional 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. The SNIFFER (2010) estimate of the rural background concentration in southern England was about 10 μg $m^{-3}$, in 2010, and the additional urban background in central London was estimated to be about 5 μg $m^{-3}$. Later in this paper the road contribution is included as a separate item in the $PM_{2.5}$ budget. Other sources within grid squares, which make a local contribution, arise from small stationary sources, but are not included in the calculation. They are not as widely distributed as traffic sources, affect fewer grid squares and should be subject to local air quality management control.

## 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.

[4] In 2011 there were 67 AURN (Automatic and Rural Monitoring Network) sites at which $PM_{2.5}$ was measured but most would not be regarded as rural 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.

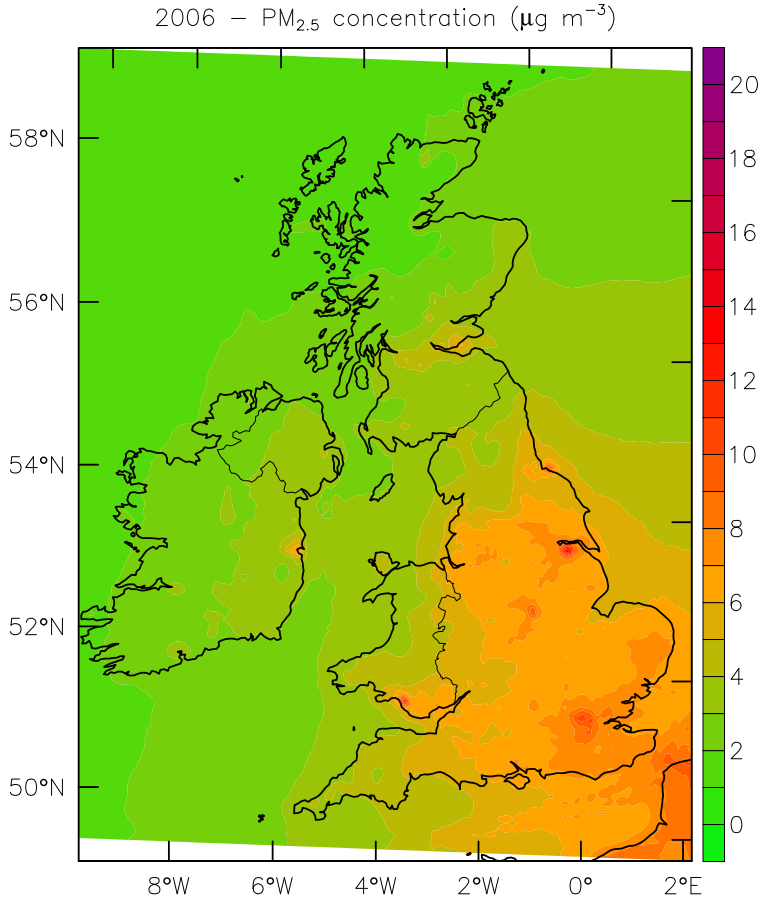


Fig. 1 Annual average regional $PM_{2.5}$ concentrations (µg $m^{-3}$) in 2006 from all sources excluding local contributions

The regional contribution to annual average $PM_{2.5}$ concentrations in 2006 from the CMAQ model is well below the indicative limit value of 20 µg $m^{-3}$. It is clearly below the equivalent AQEG (2012) limit values of 15 µg $m^{-3}$, in northern Britain, and 20 µg $m^{-3}$ in the south east. In some other parts of Europe the regional concentration in 2006 is higher, approaching but not exceeding the indicative limit value of 20 µg $m^{-3}$. The local road (and small stationary source) contribution 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 further understanding, the contribution from major stationary sources was estimated by rerunning the full model but excluding all emissions from all major industrial sources regulated in the UK. This then gives the regional footprint of the $PM_{2.5}$, over distances exceeding 6km, arising from major industrial sources regulated in the UK.

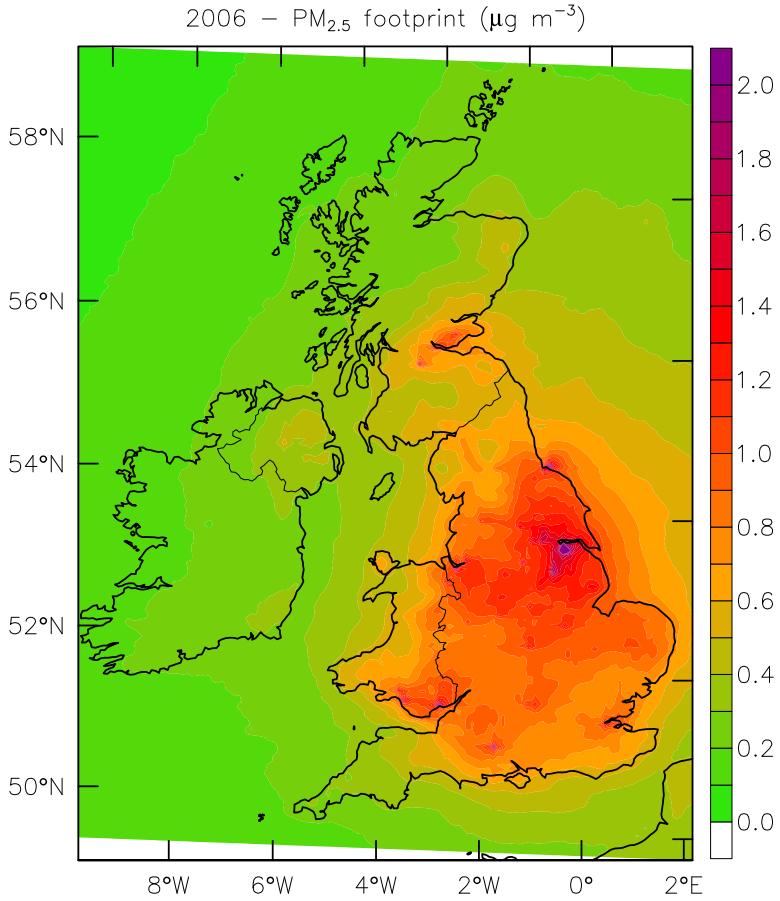


Fig. 2 Annual average regional $PM_{2.5}$ concentrations (µg $m^{-3}$) from major industrial source regulated in the UK in 2006

The highest concentrations of $PM_{2.5}$ 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 amount to 1 to 2 µg $m^{-3}$ in the Midlands. As a percentage, the major industrial sources regulated in the UK make up between 10 and 20 % of the regional $PM_{2.5}$ over England in 2006. 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.

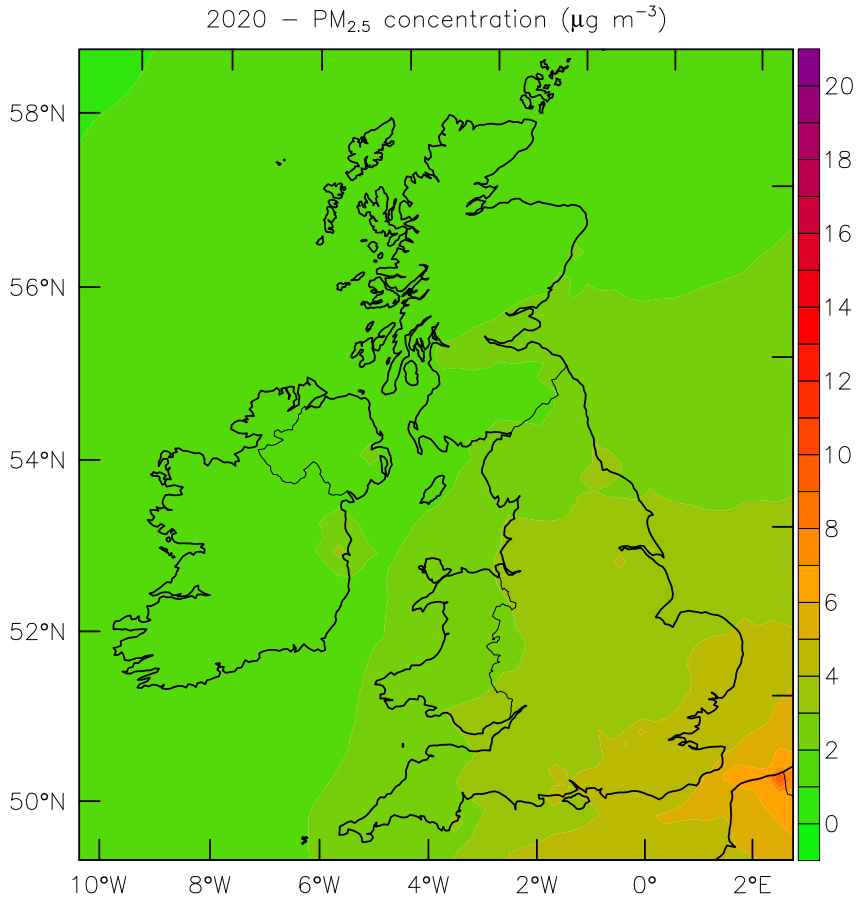


Fig. 3 Annual average regional $PM_{2.5}$ concentrations (µg $m^{-3}$) in 2020 from all sources using 2006 meteorology

Fig.3 shows the annual average regional $PM_{2.5}$ concentrations (in µg $m^{-3}$) from all sources in the 2020 emissions projection, excluding local contributions and using 2006 meteorology. This shows a significant reduction in the regional contribution compared with 2006 with levels less than 6 µg $m^{-3}$ in most of the country and only reach 6 µg $m^{-3}$ in the south east of the country. This is a reduction of about 40%. Concentrations are a mixture of contributions from sources at various distances subject to a variety of processes. From this single calculation, the sectors with the main reductions *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 5-15 % 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 production of secondary particulate matter from emissions of $SO_2$, NOx and PM. In 2006 the total source from major regulated stationary sources in the UK amounted to about 453 kt $yr^{-1}$ $SO_2$, 441 kt $yr^{-1}$ NOx, 17 kt $yr^{-1}$ $PM_{10}$, 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 $yr^{-1}$ $PM_{10}$, 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 by 2020 to about 40% of its 2006 level. The reduction in $PM_{2.5}$ concentrations of about 50% in the region of the sources is approximately of the same order. Thus 'potential source strength' is a useful indicator of the possible 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 different 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.

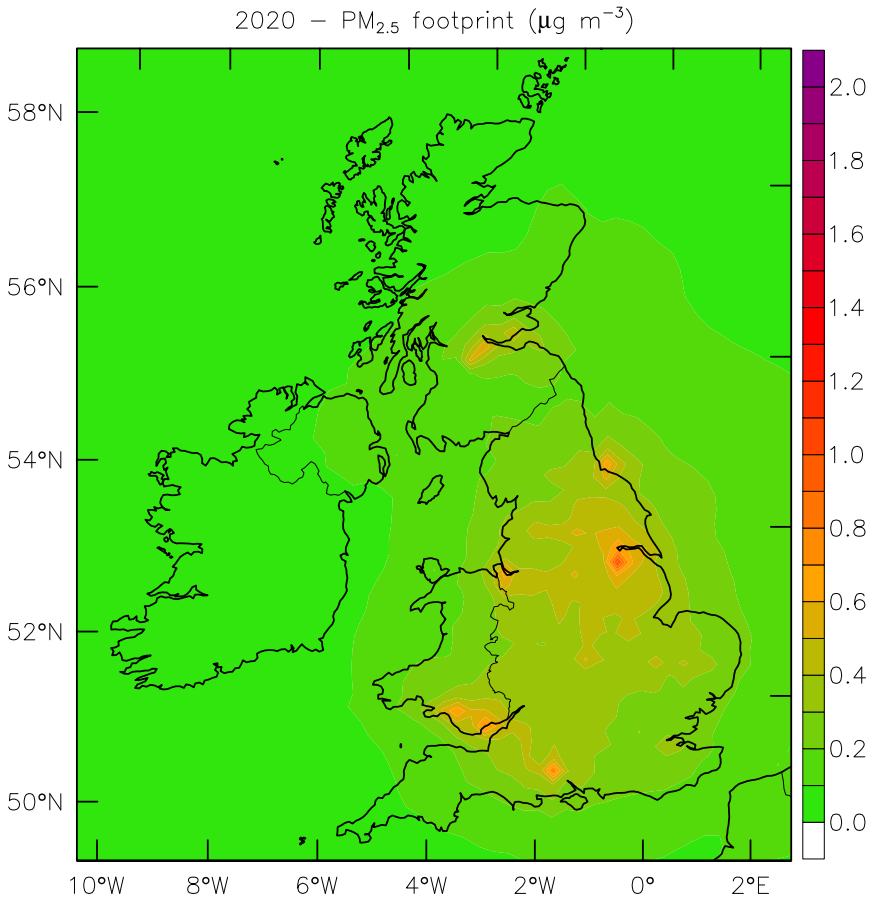

Fig. 4 Annual average regional $PM_{2.5}$ concentrations (μg $m^{-3}$) from emissions from major industrial sources regulated in the UK in 2020 using 2006 meteorology, the 'industrial footprint' in 2020

## 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 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 emissions 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 mean. The reduction in the population weighted mean is about 40% because of emission reductions between 2006 and 2020. 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. An estimate the health benefit of removing all major industrial emissions regulated in the UK, if such a policy were possible, can also be made. 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 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 urban background observations, is not equivalent to the CMAQ estimate of regional background concentrations in 2006. However the reduction from the CMAQ estimate of a nearly 40% reduction between 2006 and 2020 appears large enough to satisfy the 'average exposure indicator' reduction target, given that emission reductions will progressively have an effect throughout the period 2006 to 2020.

## 6 The local contribution to $PM_{2.5}$

The CMAQ estimate of the population weighted $PM_{2.5}$ 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 the concentrations decline rapidly with distance from the road. However it is valuable to know the possible local 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 specific locations in major urban areas, such as London.

We have estimated the local 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 2). The persistent negative bias (underestimate) in the CMAQ calculations also provides a rough estimate of the local contribution.

| | | LDV | | | HDV | |
|---|---|---|---|---|---|---|
| Year | 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 2 Emission factors (g $PM_{2.5}$/km) of light duty vehicles and heavy duty vehicles in current and future years

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 road 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 background concentration in central London is 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 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 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 future trends in emission are subject to uncertainty and policy should be flexible to adjust for the actual future trends in emission when they occur. 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 standards. At hot spots where traffic flows are higher than at typical roads, or near small stationary sources where building

[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, tyre and road abrasion for recent years. Fleet projections giving vehicle type according to Euro class in future years, as well as primary NO2 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 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.

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 regional 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.

## 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 makes up 10 to 20% of the regional annual average $PM_{2.5}$ concentration across most of the UK.

The impact of 2020 emission, using 2006 meteorology, is a reduction of 2 to 3 μg $m^{-3}$ in the regional annual average $PM_{2.5}$ concentrations over England, 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 the Midlands of the country where there is the main cluster of industrial emissions. This contribution makes up 5 to 15 % of the regional annual average $PM_{2.5}$ concentrations 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 $PM_{2.5}$ 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 appears therefore 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

[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,

$m^{-3}$ 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. If, for example, one or other of the primary particulate components was the most harmful species or particle number was the appropriate health metric, then the local primary contribution would be more important than the regional contribution and the emphasis on modelling regional transport using CMAQ would be less important. In contrast if secondary organics are found to be the most harmful species, then a full treatment of the atmospheric chemistry within a chemical transport model would be the appropriate assessment tool. An air quality standard also depends on the choice of year by which a specified concentration should be achieved. 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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