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Application of chemical transport model CMAQ to policy decisions regarding PM_{2.5} 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 $\mu\text{g m}^{-3}$. 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 PM₁₀ of 50 $\mu\text{g m}^{-3}$, for no more than 35 days in the year, is equivalent to a long-term average of 31.5 $\mu\text{g m}^{-3}$ for PM₁₀, the PM₁₀ limit value is equivalent to a PM_{2.5} concentration of about 20 $\mu\text{g m}^{-3}$ 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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50 local correction is applied to account for sources within grid squares not treated explicitly within
51 the model. There are other, wider subjective judgments to be made when deciding which
52 model to use, such as whether to employ the same team to do assessments as performed the
53 evaluation studies. The possibility of mistakes being made may then be reduced. However with
54 complex models such considerations cannot be rigorously applied.

55
56 Other recent studies have addressed issues relating to the contribution from various source
57 sectors to PM_{2.5}. Yim and Barrett (2012) suggest that about 1/6 of the PM_{2.5} concentration is
58 attributable to industrial sources for the year 2005 based on similar CMAQ modelling and
59 suggest that 40% of the PM_{2.5} originates from outside the UK. The authors did not extend their
60 calculations to a future year, nor can one be sure that the modelling was done in exactly the
61 same way as in this paper. The SNIFFER (2010) report references a study by Derwent *et al.*
62 (2009) who examined the modelled concentrations resulting from a 30% reduction in emissions
63 of SO₂, NO_x, NH₃, VOC and CO. It was concluded that PM_{2.5} concentrations in rural southern
64 UK are likely to be influenced strongly by reductions in SO₂, NO_x and NH₃ emissions in a
65 complex and interlinked manner. The largest reduction in PM_{2.5} was derived from a reduction in
66 NH₃ but such a large decrease is unlikely to occur².

67
68 The present study examines realistic changes in national emissions between 2006 and 2020
69 and is therefore directly relevant to policy. The SNIFFER report (2010) suggests that local
70 sources tend to contribute less than half of the total predicted PM_{2.5} concentration, and often
71 less than 10%, even at roadside receptors. Urban sources give rise to an annual average
72 urban background enhancement of around 3 to 6 µg m⁻³ and secondary PM_{2.5} contributes
73 around 4 to 6 µg m⁻³. SNIFFER recommended that modelling studies are carried out to
74 establish the contributions of UK and other EU emissions of precursor gases to annual mean
75 PM_{2.5} concentrations, and to determine how these contributions will respond to changes in
76 emissions, in order to guide the development of the most cost-effective control programme.
77 This study is a step to fulfilling this need. The SNIFFER report discusses the 'average
78 exposure indicator', which requires reductions in the annual average PM_{2.5} concentration of
79 about 1.5 to 2 µg m⁻³ between 2010 and 2020³.

80
81 AQEG (2012) determines the appropriate equivalent PM_{2.5} limit value in the following way. The
82 24-hour limit value for PM₁₀ of no more than 35 days >50 µg m⁻³ is taken to be equivalent to an
83 annual mean PM₁₀ concentration of 31.5 µg m⁻³. The PM_{2.5}/PM₁₀ ratio shown in AQEG (2012)
84 is about 0.7 at urban background sites. Given the ratios of PM_{2.5} to PM₁₀ 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 SO₂, 42% for NO_x, 6% for NH₃ 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 SO₂, 55% for NO_x, 8% for NH₃, 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%.

85 parts of the UK, the indicative PM₁₀ annual mean limit value can be equated to an annual
86 mean PM_{2.5} value which ranges from 17 µg m⁻³ in Scotland to 24 µg m⁻³ in south east England
87 and about 20 µg m⁻³ in the rest of the country (AQEG, 2012). This paper does not consider
88 other particle metrics, such as the concentration of black carbon particles. The EU limit value
89 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
90 annual National Ambient Air Quality Standard in the USA for PM_{2.5} is 15 µg m⁻³ (Hogrefe *et*
91 *al.*, 2011). These authors used CMAQ to investigate differences depending on the biogenic
92 emissions in the PM_{2.5} concentrations under a NO_x emission control scenario in eastern North
93 America.

94
95 There is evidence (Jannsen *et al.*, 2011) that black carbon particles could be the important
96 indicator of the health risk of particles. This would be significant in situations where particulate
97 concentrations are dominated by primary road traffic sources, whereas the calculations
98 presented in this paper refer to regional PM_{2.5} with a large secondary component. It is
99 assumed that the levels in the Ambient Air Quality Directive determine the health benefit of
100 emission reductions, and therefore it would not be appropriate to evaluate calculated regional
101 concentrations using another metric. However one should correct the calculated regional
102 concentrations by estimating roadside concentrations of PM_{2.5}.

103 104 **2 The CMAQ Model Setup**

105
106 The Community Multiscale Air Quality (CMAQ) modelling system is a comprehensive modelling
107 system developed by the US Environmental Protection Agency (USEPA). CMAQ is a Eulerian
108 photochemical air quality model in which complex interactions between atmospheric pollutants
109 on urban, regional and hemispheric scales are treated in a consistent framework. It is designed
110 for assessing the impact of multiple pollutants including tropospheric ozone and other oxidants,
111 aerosols and acid deposition.

112
113 In this application of CMAQ an outer domain over Europe with a grid resolution 18km was
114 used, with an inner domain over the UK with a grid resolution of 6km. So over the UK primary
115 roadside and urban emissions within about 3 km of a monitoring site will not be included. The
116 inner domain covers the British Isles. The outer domain stretches from about 33 degrees north
117 20 degrees west to about 70 degrees north 32 degrees east.

118
119 The European emission inventory for 2006 is based on the TNO (<http://www.tno.nl/>) inventory,
120 which consists of anthropogenic emissions from ten Selected Nomenclature for Air Pollution
121 (SNAP) source sectors and international shipping. Biogenic gas emissions were included in the
122 CMAQ model from the Model of Emissions of Gases and Aerosols from Nature (MEGAN)
123 system at the same resolution as the anthropogenic emissions. Biomass burning emissions
124 from wild fires for 2006 were based on daily fire estimates from the Moderate Resolution
125 Imaging Spectroradiometer (MODIS) fire radiative power product (Sofiev *et al.*, 2009).

126
127 A 2020 European emissions scenario was considered based on the MEGAPOLI (Baklanov *et*
128 *al.*, 2010) project results (Theloke *et al.* 2010), which make use of the:
129 (1) Integrated MARKAL-EFOM System (TIMES) Pan-European (TIMES PanEU) energy
130 system model for the energy related sectors;
131 (2) Greenhouse gas – Air pollution Interactions and Synergies (GAINS) model and other
132 assumptions for the non-energy related sectors; and
133 (3) a reduction of the greenhouse gas emissions by 30% by 2020 compared to 1990.

134
135 Major industrial sources are defined as those with annual emissions of SO₂ greater than 500t
136 yr⁻¹ and/or annual emissions of NO_x greater than 500t yr⁻¹ and/or annual emissions of PM₁₀

137 greater than 200t yr⁻¹. 2006 is the base line year for the calculations. All major industrial
 138 emissions regulated in the UK in 2006 were adjusted to enable 2020 concentrations to be
 139 modelled. Information on likely trends in regulation between 2006 and 2020 is available, but
 140 the activity of each industrial sector cannot be reliably forecast. A site by site 2020 emissions
 141 estimate was therefore not feasible. Two industrial estimates for 2020 are available in the
 142 public domain from the AEA Atmospheric Emissions team (Wagner *et al.*, 2009) from which the
 143 mean was adopted.

144
 145 The ratio of the emissions of SO₂, NO_x, PM₁₀ and NMVOC between 2006 and 2020 are
 146 estimated for major industrial releases and for the national total emissions. These are shown in
 147 Table 1.
 148

Pollutant	Ratio 2020:2006 major industry emissions	Ratio 2020:2006 national total UK emissions
SO ₂	0.37	0.47
NO _x	0.45	0.52
PM ₁₀	0.42	0.72
PM _{2.5}	0.65	0.68
NMVOC	0.87	0.72

149
 150 Table 1 Estimated ratio of the major industrial emissions and national total emissions of SO₂,
 151 NO_x, PM₁₀ and NMVOC between 2006 and 2020
 152

153 These ratios were used to scale UK major industrial emissions to obtain representative
 154 emissions for 2020. The influence of changes of order 30% or more in the main air pollution
 155 emissions are assessed in the model. Ammonia is a special case and no correction is made.
 156 Since the ratio SO₂:NH₃ should be lower in 2020 than 2006, the rate of production of PM_{2.5} for
 157 a given fixed source could be greater in 2020 compared to 2006. Based on results of the
 158 EMEP chemical transport model, a comparison (Fisher, 2012) of the radial footprints of annual
 159 average PM_{2.5} concentration for different past years and different individual countries, UK and
 160 Germany with different source strengths, does not indicate strong differences in behaviour as a
 161 function of distance from the centre of the country. The tentative conclusion is that chemical
 162 transport models have not demonstrated that the annual average PM_{2.5} concentration has a
 163 very strong non-linear dependence on the change in emissions. However further studies
 164 investigating greater changes in background atmospheric composition are needed to
 165 understand the relationship more fully.
 166

167 The estimated ratios in emissions between 2006 and 2020 in Table 1 are in reasonable
 168 agreement with previous projections by Vincent and Abbott (2008). There are some differences
 169 with the MEGAPOLI projections for the UK energy related sectors, but when differences arise
 170 the most pessimistic case is adopted.
 171

172 The CMAQ system setup involves the Weather Research and Forecasting (WRF) meso-scale
 173 model, embedded within the ECMWF regional model for the meteorological fields. The initial
 174 and lateral meteorological boundary conditions of the outer domain were derived from the
 175 European Centre for Medium-range Weather Forecasts (ECMWF) gridded analyses available
 176 every 6 h with a horizontal resolution of 0.5 degrees on operational pressure levels up to 50
 177 hPa for vertically distributed data, and at surface and soil levels for surface and deep-soil data.
 178 A grid nudging technique was employed for the outer domain every 6 h in order to constrain
 179 the model towards the analyses. The Met Office have run their global HADGEM2_ES model
 180 (Collins *et al.*, 2011) and obtained regional meteorological fields for 2020 applying the RCP8.5

181 emissions scenario, which may be regarded as a pessimistic scenario. The Met Office 2020
182 meteorological fields used to develop an alternative 2020 PM_{2.5} climate change projection are
183 thought to be representative of the decade. The difference between the 2006 and 2020
184 meteorological fields are one of many possibilities which could occur, but the approach
185 illustrates a straightforward methodology which further large-scale computing could develop
186 further.

187
188 The CMAQ modelling system requires hourly emissions data of primary pollutants. SMOKE
189 has been developed for this purpose and can be adapted to process annual emissions data
190 (from point, line and area sources) into temporally-resolved, spatially-distributed and speciated
191 emissions files ready for the chemical transport model.

192
193 For CMAQ version 4.7, adopted in this study, the CB05 chemical mechanism was used. The
194 CB05 mechanism treats the formation of secondary organic aerosols. The tri-modal approach
195 to aerosol size distribution was used in order to model particulate matter. The species
196 modelled include sulphate (SO₄²⁻), nitrate (NO₃⁻), ammonium (NH₄⁺), sodium (Na⁺), chloride
197 (Cl⁻), water (H₂O) and organics from precursors of anthropogenic and biogenic origin but only
198 the total PM₁₀ and PM_{2.5} concentrations are considered in this paper. Each mode (Aitken,
199 accumulation and coarse) is subject to both wet and dry deposition. Documentation on CMAQ
200 is available from the official CMAQ website (<http://www.cmaq-model.org>) [accessed 21 May
201 2013]. Chemical boundary conditions were obtained from runs of the global model Geos-
202 Chem.

203 204 **3 Model Evaluation and Uncertainty**

205
206 The Defra Model Evaluation Protocol (Derwent *et al.* 2010) sets the following criteria for the
207 acceptance of a model. The predictions of the model should be accepted if the percentage of
208 model predictions within a factor of two (*FAC2*) of the observations is greater than 50 per cent
209 and the magnitude of the normalised mean bias (*NMB*) is less than 0.2.

210
211 The normalised mean bias (*NMB*) is defined as:

$$212 \quad NMB = \frac{\sum_{i=1}^N M_i - O_i}{\sum_{i=1}^N O_i} \quad (1)$$

213
214 where *N* is the number of observations, *M_i* are the calculated values, *O_i* are the observed
215 values. *NMB* should satisfy $-0.2 \leq NMB \leq 0.2$.

216
217 A limited evaluation of CMAQ predictions of PM_{2.5} over the UK for the year 2005 was
218 undertaken by Yim and Barrett (2012). Only three AURN monitoring sites, where PM_{2.5} was
219 measured, were operating in 2005: Harwell (rural), Stoke (rural), Bloomsbury (urban
220 background) and a fourth roadside site at Marylebone. The *NMB* was -0.23 at Harwell -0.09 at
221 Stoke and -0.27 at Bloomsbury.

222
223 The multi-model AQMEII study (Solazzo *et al.* 2012) compared PM_{2.5} at many sites in North
224 America and Europe in 2006 and included some CMAQ calculations for Europe by the authors
225 of this paper. These generally showed significant under-prediction with a *NMB* of about -0.4.
226 However the CMAQ predictions reported in this paper of individual components of particulate
227 matter at Harwell for the year 2006, such as inorganic species (SO₄, NO₃ and NH₄), and
228 elemental carbon and organic carbon, do not show the same systematic under-prediction. The
229

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
<i>FAC2</i> (%)	88.2	100	100
<i>r</i> (correlation coefficient)	0.09	0.0	0.45
<i>NMB</i>	-0.33	-0.09	-0.20
Single power station contribution (%)	0.34	0.28	0.28

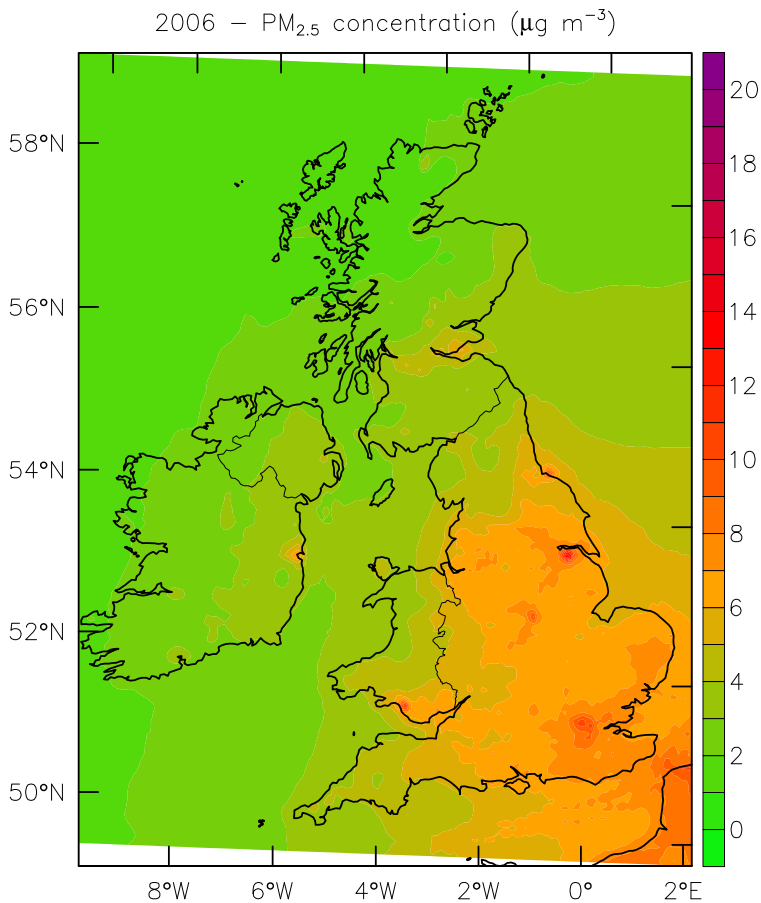
Table 2 Comparison of performance in predicting annual average PM₁₀ concentration in 2003 at 40 urban background and rural background sites in the AURN network in the UK for two versions of the advanced model CMAQ (Chemel *et al.* 2011) and a simpler, semi-empirical model TRACK-ADMS.

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.

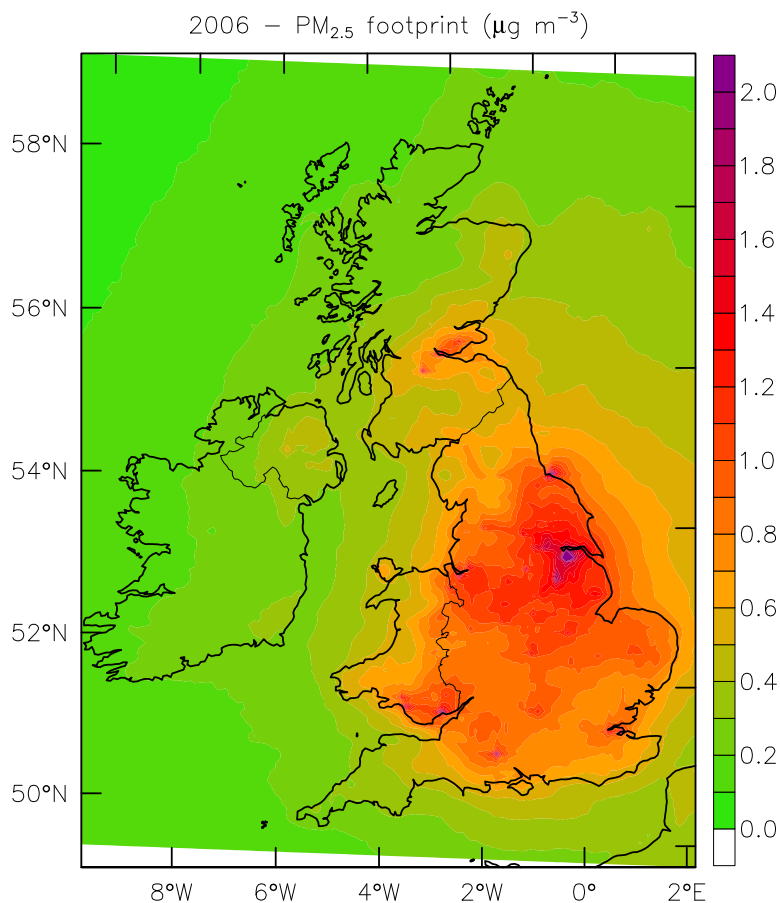
⁴ In 2011 there were 67 AURN (Automatic and Rural Monitoring Network) sites at which PM_{2.5} was measured but most would not be regarded as rural and therefore of limited value in evaluating the performance of CMAQ (<http://uk-air.defra.gov.uk/data/exceedence> [accessed 21 May 2013]). These sites mainly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



264
265 Fig. 1 Annual average regional PM_{2.5} concentrations ($\mu\text{g m}^{-3}$) in 2006 from all sources
266 excluding local contributions

267
268 The regional contribution to annual average PM_{2.5} concentrations in 2006 from the CMAQ
269 model is well below the indicative limit value of $20 \mu\text{g m}^{-3}$. It is clearly below the equivalent
270 AQEG (2012) limit values of $15 \mu\text{g m}^{-3}$, in northern Britain, and $20 \mu\text{g m}^{-3}$ in the south east. In
271 some other parts of Europe the regional concentration in 2006 is higher, approaching but not
272 exceeding the indicative limit value of $20 \mu\text{g m}^{-3}$. The local road (and small stationary source)
273 contribution are not included in these estimates.

274
275 The observed annual average PM_{2.5} concentration consists of contributions from a mixture of
276 primary and secondary sources over very short and very long travel distances from their point
277 of emission. In order to further understanding, the contribution from major stationary sources
278 was estimated by rerunning the full model but excluding all emissions from all major industrial
279 sources regulated in the UK. This then gives the regional footprint of the PM_{2.5}, over distances
280 exceeding 6km, arising from major industrial sources regulated in the UK.
281



282
 283 Fig. 2 Annual average regional PM_{2.5} concentrations (µg m⁻³) from major industrial source
 284 regulated in the UK in 2006

285
 286 The highest concentrations of PM_{2.5} lie close to the cluster of major industrial sources in the
 287 Midlands. The concentration declines with distance from the Midlands mainly as a
 288 consequence of the spread in air mass trajectories (Fisher *et al.*, 2011). The highest
 289 concentrations amount to 1 to 2 µg m⁻³ in the Midlands. As a percentage, the major industrial
 290 sources regulated in the UK make up between 10 and 20 % of the regional PM_{2.5} over England
 291 in 2006. Using the simpler semi-empirical TRACK-ADMS model, Vincent and Abbott (2008)
 292 estimated that the contribution of major regulated sources was 10% of the average PM₁₀
 293 concentrations in the UK in 2005.
 294

2020 – PM_{2.5} concentration ($\mu\text{g m}^{-3}$)

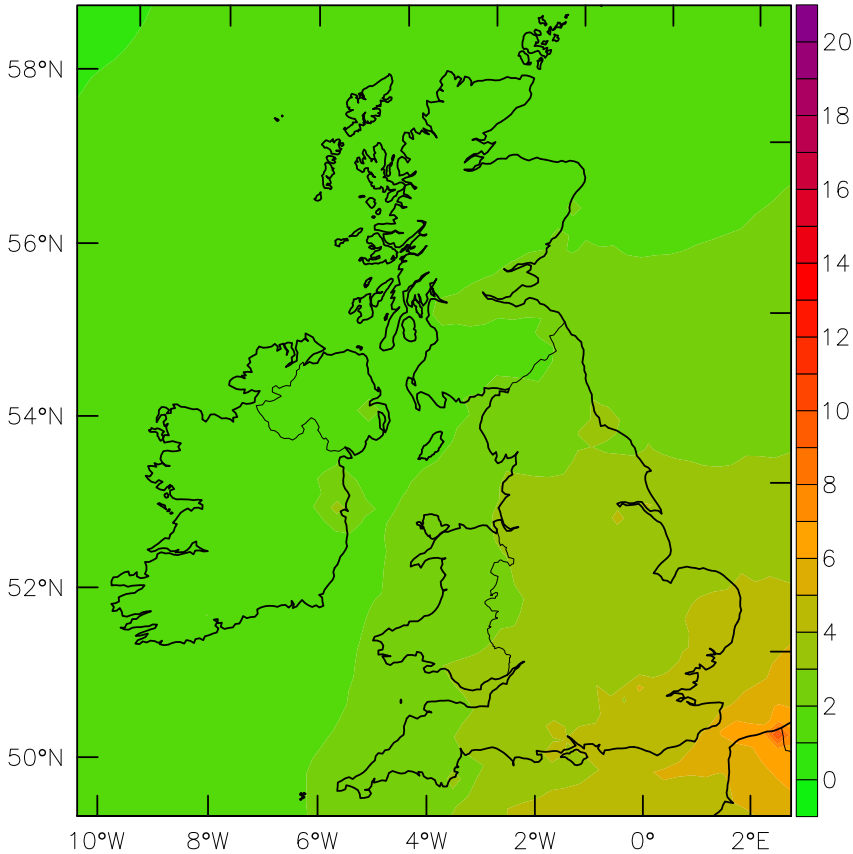


Fig. 3 Annual average regional PM_{2.5} concentrations ($\mu\text{g m}^{-3}$) in 2020 from all sources using 2006 meteorology

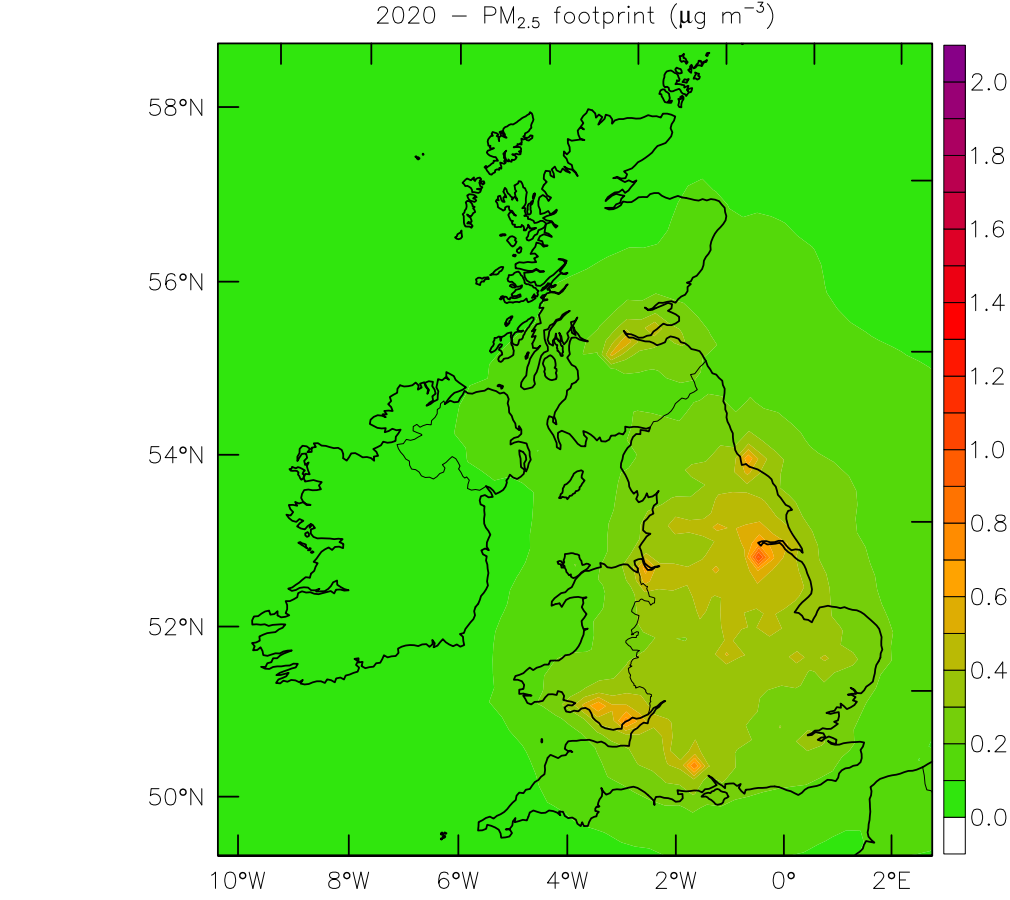
Fig.3 shows the annual average regional PM_{2.5} concentrations (in $\mu\text{g m}^{-3}$) from all sources in the 2020 emissions projection, excluding local contributions and using 2006 meteorology. This shows a significant reduction in the regional contribution compared with 2006 with levels less than $6 \mu\text{g m}^{-3}$ in most of the country and only reach $6 \mu\text{g m}^{-3}$ 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 $1 \mu\text{g m}^{-3}$ 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 SO₂, NO_x and PM. In 2006 the total source from major regulated stationary sources in the UK amounted to about 453 kt yr^{-1} SO₂, 441 kt yr^{-1} NO_x, 17 kt yr^{-1} PM₁₀, 6 kt yr^{-1} PM_{2.5} giving a total of about 900 kt per annum of 'potential' PM_{2.5} emissions (SO₂+NO_x+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} SO₂, 197 kt yr^{-1} NO_x, 7 kt yr^{-1} PM₁₀, 5 kt yr^{-1} PM_{2.5} giving an indicative 'potential' PM_{2.5} annual emission (SO₂+NO_x+PM_{2.5})

322 of about 370kt. Thus the total primary emission from regulated major industry expressed as the
323 total of the SO₂ + NO_x + PM_{2.5} emissions would have reduced by 2020 to about 40% of its
324 2006 level. The reduction in PM_{2.5} concentrations of about 50% in the region of the sources is
325 approximately of the same order. Thus 'potential source strength' is a useful indicator of the
326 possible response of the atmosphere to changes in emissions.
327

328 The change in the total PM_{2.5} concentration between 2006 and 2020 is not a useful indicator
329 because the change comes about from a mixture of contributions to PM_{2.5} concentration over
330 the UK. The response will be variable, depending on whether the response is from distant
331 primary emissions in Europe, mainly from sources in the south east, or from regional industrial
332 UK sources, centred in the Midlands, or from urban UK emissions, spread around the country.
333 No single indicator is representative of all these different responses, which depend on different
334 geographical distribution changes between 2006 and 2020.
335

336 Chemical transport models are designed to inform on the possible non-linear relationship
337 between the change in emissions and the resulting change in concentration of PM_{2.5}. One
338 could imagine that if the atmosphere became relatively more reactive, secondary PM_{2.5} could
339 be formed more quickly near to locations where it was emitted. Lifetimes can be estimated
340 from the footprints of a specified source at a single location (Fisher *et al.*, 2011). This is not
341 possible here because the major industrial sources are distributed around the country.
342 However the change in the primary emissions of major industrial sources is roughly in
343 proportion to the change in the PM_{2.5} concentration, near the cluster of major industrial sources
344 suggesting no evidence of very large non-linearity. The change in emissions between 2006
345 and 2020 may not be large enough to provide evidence of non-linearity and more testing of
346 responses to changes in emission should be undertaken.
347



348
349

350 Fig. 4 Annual average regional PM_{2.5} concentrations ($\mu\text{g m}^{-3}$) from emissions from major
351 industrial sources regulated in the UK in 2020 using 2006 meteorology, the 'industrial footprint'
352 in 2020
353

354 **5 Climate change and population exposure**

355
356 Because of the availability of an alternative meteorological scenario in 2020 from results of the
357 Met Office's HadGEM model, an estimate can be made of the change in 2020 as a possible
358 consequence of climate change. It turns out that the effect of the chosen climate change
359 scenario is to reduce annual average PM_{2.5} concentrations in 2020 by of order 10% over the
360 country, representing reductions of 0.2 to 0.5 $\mu\text{g m}^{-3}$ in England as a result of a different
361 climate. This is much less than the reduction in regional concentration from emission changes
362 between 2006 and 2020. The treatment of climate change did not include interactions between
363 climate and pollution, such as the effect particle concentrations arising from emissions changes
364 might have on radiation and cloud formation, cloud duration and thickness. The version of
365 CMAQ used, v4.7, is not a fully coupled model calculation. As the climate effect appears small,
366 this is a preliminary indication that it may not be worthwhile running the CMAQ model for a
367 range of alternative 2020 PM_{2.5} climate change projections.
368

369 An alternative way of estimating the benefit of emission reductions between 2006 and 2020 is
370 to determine the population weighted mean. The reduction in the population weighted mean is
371 about 40% because of emission reductions between 2006 and 2020. The additional fractional
372 contribution of climate change in 2020 is estimated to be about 5%. Some slight non-
373 proportional differences between 2006 and 2020 may occur because of changes in population.
374 An estimate the health benefit of removing all major industrial emissions regulated in the UK, if
375 such a policy were possible, can also be made. The fractional contribution of major industrial
376 sources to the population weighted annual mean PM_{2.5} concentration in 2006 is about 14%.
377

378 The percentage change in population weighted concentrations between 2006 and 2020 can be
379 compared with the EU Directive 'average exposure indicator' target value. For a 15%
380 reduction in average exposure indicator, SNIFFER (2010) estimated that a reduction in the
381 annual mean PM_{2.5} of 1.5 $\mu\text{g m}^{-3}$ over England, and a 2 $\mu\text{g m}^{-3}$ over inner London between
382 2010 and 2020, is required. The SNIFFER estimate of PM_{2.5} concentrations in 2010, based on
383 urban background observations, is not equivalent to the CMAQ estimate of regional
384 background concentrations in 2006. However the reduction from the CMAQ estimate of a
385 nearly 40% reduction between 2006 and 2020 appears large enough to satisfy the 'average
386 exposure indicator' reduction target, given that emission reductions will progressively have an
387 effect throughout the period 2006 to 2020.
388

389 **6 The local contribution to PM_{2.5}**

390
391 The CMAQ estimate of the population weighted PM_{2.5} concentration reduction does not include
392 the roadside contribution to PM_{2.5} concentrations. This is likely to be a small fraction of the
393 population exposure as the concentrations decline rapidly with distance from the road.
394 However it is valuable to know the possible local contributions from sources within a grid
395 square to check whether the indicative annual average limit value of 20 $\mu\text{g m}^{-3}$ is likely to be
396 exceeded at specific locations in major urban areas, such as London.
397

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 factors⁵ (see table 2). The persistent negative bias (underestimate) in the CMAQ calculations also provides a rough estimate of the local contribution.

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 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 NO₂ because of the non-linear chemistry involving ozone and primary NO₂ 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.

427 downwash may occur, local air quality management should be considered the tool for
428 addressing exceedences.

429
430 This practical estimate of future PM_{2.5} concentrations over the UK combines a simple
431 assessment of road traffic emissions with the regional contribution from a complex model. The
432 estimate does not contain estimates near to every kind of road. Exceptional cases, such as an
433 urban road with very heavy traffic, could lead to higher concentrations but these cases should
434 be treated using local air quality management action plans. The local air quality management
435 system can also be used as the process for checking that future emissions follow expected
436 trends. Actual trends may not follow the expected trend because (1) technological measures
437 addressing vehicle emissions do not perform as expected, (2) trends in other types of
438 emissions do not occur as forecast⁶, or (3) unanticipated future trends in the way mobility,
439 power and heating services are provided.

440 441 **7 Conclusions**

442
443 From calculations using the CMAQ model it was concluded that in 2006 wild fires make a
444 minor contribution to annual mean PM_{2.5} concentrations in the UK. The contribution of
445 emissions from major industrial sources regulated in the UK, in 2006, amounts to 1 to 2 µg m⁻³
446 in the Midlands. This makes up 10 to 20% of the regional annual average PM_{2.5} concentration
447 across most of the UK.

448
449 The impact of 2020 emission, using 2006 meteorology, is a reduction of 2 to 3 µg m⁻³ in the
450 regional annual average PM_{2.5} concentrations over England, a reduction of 20 to 40% across
451 most of the UK. The reduction in the population weighted mean because of emission
452 reductions between 2006 and 2020 is estimated to be 40%.

453
454 The contribution of emissions from major industrial sources regulated in the UK to regional
455 annual average PM_{2.5} concentrations in 2020, using 2006 meteorology, amounts to 0.5 to 1 µg
456 m⁻³ in the Midlands of the country where there is the main cluster of industrial emissions. This
457 contribution makes up 5 to 15 % of the regional annual average PM_{2.5} concentrations across
458 most of the UK. The potential source strength of regulated major industrial emissions
459 expressed as the total of SO₂ + NO_x + PM_{2.5} emissions would have reduced to about 40% of
460 their 2006 level in 2020. The reduction in PM_{2.5} concentrations of about 50% in the Midlands is
461 approximately of the same order. Thus 'potential source strength' is a possible, useful indicator
462 of the response of the atmosphere to changes in emissions.

463
464 Applying meteorological conditions in 2020 representing a possible climate change scenario,
465 with a 2020 emissions scenario, suggests reductions of 0.2 to 0.5 µg m⁻³ in the regional annual
466 average PM_{2.5} concentrations over England, equivalent to a reduction of 2 to 5 % across most
467 of the UK compared to PM_{2.5} concentrations under 2006 meteorological conditions and a 2020
468 emissions scenario. The 'climate change scenario' reduction appears therefore between 5 to
469 10 times smaller than the reduction brought about by emissions changes between 2006 and
470 2020.

471
472 Estimates of the local primary PM_{2.5} concentration at a typical roadside location in London
473 have been added to the regional PM_{2.5} concentration to test whether compliance with the
474 indicative PM_{2.5} limit value of 20 µg m⁻³ in 2020 will be achieved. The total PM_{2.5} concentrations
475 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,

476 m⁻³ suggesting widespread compliance with the target limit value. This does not confirm that
477 PM_{2.5} will not be an issue at some locations, but it appears not to be a general issue accepting
478 current air quality standards and likely trends in emissions. At hot spots, where traffic flows are
479 higher than at a typical urban road, local air quality management should be considered the tool
480 for addressing PM_{2.5} exceedences. The introduction of new emission technology standards is
481 the tool for addressing air quality considerations on a national basis.
482

483 There is uncertainty associated with the air quality standard used in this paper. We have
484 adopted the approach, implicit in the Ambient Air Quality Directive, that all components of
485 PM_{2.5} have the same health disbenefit and the aim should be to reach and maintain the
486 strictest standard implied by the values stated in the Directive. If further research suggests that
487 certain components of PM_{2.5} are the active components causing harm and PM_{2.5} acts as the
488 surrogate for these components in the underlying epidemiological studies, then the conclusions
489 may be different. If, for example, one or other of the primary particulate components was the
490 most harmful species or particle number was the appropriate health metric, then the local
491 primary contribution would be more important than the regional contribution and the emphasis
492 on modelling regional transport using CMAQ would be less important. In contrast if secondary
493 organics are found to be the most harmful species, then a full treatment of the atmospheric
494 chemistry within a chemical transport model would be the appropriate assessment tool. An air
495 quality standard also depends on the choice of year by which a specified concentration should
496 be achieved. Understanding the observed trends in annual average concentrations is a
497 necessary step to ensure compliance, and modelling can only suggest a strategy towards
498 ensuring future compliance. Unexpected changes in emissions and unexplained observational
499 trends compared with model predictions may still arise.
500

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507

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