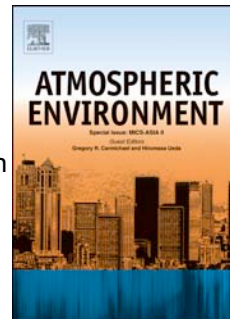


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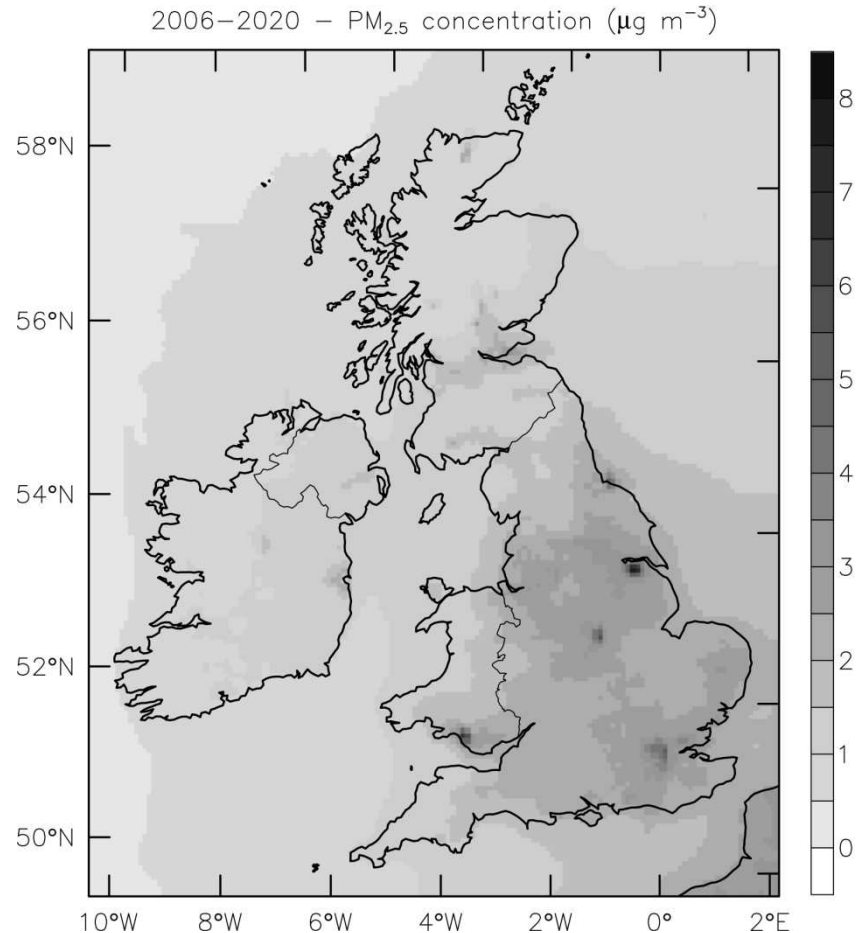
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Reduction in the annual average PM<sub>2.5</sub> concentration between 2006 and 2020 (excluding the contribution from sources within the grid square containing the receptor)

# Application of chemical transport model CMAQ to policy decisions regarding PM<sub>2.5</sub> 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<sub>2.5</sub> 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<sub>2.5</sub> will largely be met in 2020 assuming 2006 meteorological conditions. Contributions to annual average regional PM<sub>2.5</sub> 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<sub>2.5</sub> 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<sub>2.5</sub> health effects will need to be part of the future strategy to manage PM<sub>2.5</sub> 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<sub>2.5</sub>. The Ambient Air Quality Directive (OJEU, 2008) contains an annual mean limit value for PM<sub>2.5</sub> of 25 µg m<sup>-3</sup>. Although PM<sub>2.5</sub> (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<sub>10</sub> of 50 µg m<sup>-3</sup>, for no more than 35 days in the year, is equivalent to a long-term average of 31.5 µg m<sup>-3</sup> for PM<sub>10</sub>, the PM<sub>10</sub> limit value is equivalent to a PM<sub>2.5</sub> concentration of about 20 µg m<sup>-3</sup> PM<sub>2.5</sub> 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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51 *al.*, 2013, Hayman *et al.* 2013a, b, c and d) which concluded that there was no formal  
52 procedure for evaluating a model and then approving its use. Instead the conclusion was that  
53 one should use as good a model as was currently available. The CMAQ model (Chemel *et al.*,  
54 2011) is one of the most up to date and sophisticated chemical transport models and is used  
55 here. A simple 'local' correction is applied to account for sources within grid squares not  
56 treated explicitly within the model. There are other, wider subjective judgments to be made  
57 when deciding which model to use, such as whether to employ the same team to do  
58 assessments as performed the evaluation studies. The possibility of mistakes being made may  
59 then be reduced. However with complex models such considerations cannot be rigorously  
60 applied.

61  
62 Other recent studies have addressed issues relating to the contribution from various source  
63 sectors to PM<sub>2.5</sub>. Yim and Barrett (2012) suggest that about 1/6 of the PM<sub>2.5</sub> concentration is  
64 attributable to industrial sources for the year 2005 based on similar CMAQ modelling and  
65 suggest that 40% of the PM<sub>2.5</sub> originates from outside the UK. The authors did not extend their  
66 calculations to a future year, nor can one be sure that the modelling was done in exactly the  
67 same way as in this paper. The SNIFFER (2010) report references a study by Derwent *et al.*  
68 (2009) who examined the modelled concentrations resulting from a 30% reduction in emissions  
69 of SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub>, VOC and CO. It was concluded that PM<sub>2.5</sub> concentrations in rural southern  
70 UK are likely to be influenced strongly by reductions in SO<sub>2</sub>, NO<sub>x</sub> and NH<sub>3</sub> emissions in a  
71 complex and interlinked manner. The largest reduction in PM<sub>2.5</sub> was derived from a reduction in  
72 NH<sub>3</sub> but such a large decrease is unlikely to occur<sup>2</sup>.

73  
74 The present study examines realistic changes in national emissions between 2006 and 2020  
75 and is therefore directly relevant to policy. The SNIFFER report discusses the 'average  
76 exposure indicator', which requires reductions in the annual average PM<sub>2.5</sub> concentration  
77 between 2010 and 2020<sup>3</sup>. AQEG (2012) and Harrison *et al.* (2013) also estimate PM<sub>2.5</sub>  
78 concentrations in 2020 but not by using a fully comprehensive chemical transport model.

79  
80 AQEG (2012) determines the appropriate equivalent PM<sub>2.5</sub> limit value in the following way. The  
81 24-hour limit value for PM<sub>10</sub> of no more than 35 days >50 µg m<sup>-3</sup> is taken to be equivalent to an  
82 annual mean PM<sub>10</sub> concentration of 31.5 µg m<sup>-3</sup>. The PM<sub>2.5</sub>/PM<sub>10</sub> ratio shown in AQEG (2012)  
83 is about 0.7 at urban sites not close to roads. Given the ratios of PM<sub>2.5</sub> to PM<sub>10</sub> identified for  
84 different parts of the UK, the indicative PM<sub>10</sub> annual mean limit value can be equated to an  
85 annual mean PM<sub>2.5</sub> value which ranges from 17 µg m<sup>-3</sup> in Scotland to 24 µg m<sup>-3</sup> in south east  
86 England and about 20 µg m<sup>-3</sup> in the rest of the country (AQEG, 2012). This paper does not

---

<sup>2</sup> 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<sub>2</sub>, 42% for NO<sub>x</sub>, 6% for NH<sub>3</sub> and 28% for NMVOC between 2005 and 2020, and for the first time a limit on primary PM<sub>2.5</sub> emissions involving a reduction of 22% between 2005 and 2020. For the UK the reductions are 59% for SO<sub>2</sub>, 55% for NO<sub>x</sub>, 8% for NH<sub>3</sub>, 32% for NMVOC and 30% for PM<sub>2.5</sub>. 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.

<sup>3</sup> 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<sub>2.5</sub> 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%.

87 consider other particle metrics, such as the concentration of black carbon particles. The EU  
88 limit value for PM<sub>2.5</sub> is 25 µg m<sup>-3</sup> by 2015, with a stage 2 indicative value of 20 µg m<sup>-3</sup> by 2020.  
89 The National Ambient Air Quality Standard in the USA for PM<sub>2.5</sub> is an annual mean of 15 µg m<sup>-3</sup>  
90 <sup>3</sup> averaged over 3 years (Hogrefe *et al.*, 2011). These authors used CMAQ to investigate  
91 differences in the PM<sub>2.5</sub> concentrations depending on the biogenic emissions under a NO<sub>x</sub>  
92 emission control scenario in eastern North America.

93  
94 There is evidence (Jannsen *et al.*, 2011) that black carbon particles could be the most  
95 important indicator of the health risk of particles. This would be significant in situations where  
96 particulate concentrations are dominated by primary road traffic sources, whereas the  
97 calculations presented in this paper refer to regional PM<sub>2.5</sub> with a large secondary component.  
98 It is assumed that the levels in the Ambient Air Quality Directive determine the health benefit of  
99 emission reductions, and therefore it would not be appropriate to evaluate calculated  
100 concentrations using another metric. However one should correct the calculated concentrations  
101 by estimating roadside concentrations of PM<sub>2.5</sub>.

102

## 103 **2 The CMAQ Model Setup**

104

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

111

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

117

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

125

126 A 2020 European emissions scenario was considered based on the MEGAPOLI (Baklanov *et al.*  
127 *et al.*, 2010) project results (Theloke *et al.* 2010), which makes use of the:

128 (1) Integrated MARKAL-EFOM System (TIMES) Pan-European (TIMES PanEU) energy  
129 system model for the energy related sectors;

130 (2) Greenhouse gas – Air pollution Interactions and Synergies (GAINS) model and other  
131 assumptions for the non-energy related sectors; and

132 (3) a reduction of the greenhouse gas emissions by 30% by 2020 compared to 1990.

133

134 Major industrial sources are defined as those with annual emissions of SO<sub>2</sub> greater than 500t  
135 yr<sup>-1</sup> and/or annual emissions of NO<sub>x</sub> greater than 500t yr<sup>-1</sup> and/or annual emissions of PM<sub>10</sub>  
136 greater than 200t yr<sup>-1</sup>. 2006 is the base line year for the calculations. All major industrial  
137 emissions regulated in the UK in 2006 were adjusted to enable 2020 concentrations to be  
138 modelled. Information on likely trends in regulation between 2006 and 2020 is available, but

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

143  
144 The ratio of the emissions of SO<sub>2</sub>, NO<sub>x</sub>, PM<sub>10</sub> and NMVOC between 2006 and 2020 are  
145 estimated for major industrial releases and for the national total emissions. These are shown in  
146 Table 1.

147  
148 [Table 1 here]

149  
150 These ratios were used to scale UK major industrial emissions to obtain representative  
151 emissions for 2020. Ammonia is a special case and no correction is made. Since the ratio  
152 SO<sub>2</sub>:NH<sub>3</sub> should be lower in 2020 than 2006, the rate of production of PM<sub>2.5</sub> for a given fixed  
153 source could be greater in 2020 compared to 2006. Based on results of the EMEP chemical  
154 transport model, a comparison (Fisher, 2012) of the radial footprints of annual average PM<sub>2.5</sub>  
155 concentration for different past years and for different individual countries, UK and Germany  
156 with different source strengths, does not indicate strong differences in behaviour as a function  
157 of distance from the centre of the country. The tentative conclusion is that chemical transport  
158 models have not demonstrated that the annual average PM<sub>2.5</sub> concentration has a very strong  
159 non-linear dependence on the change in emissions. However further studies investigating  
160 greater changes in background atmospheric composition are needed to understand the  
161 relationship more fully.

162  
163 The estimated ratios in emissions between 2006 and 2020 in Table 1 are in reasonable  
164 agreement with previous projections by Vincent and Abbott (2008). There are some differences  
165 with the MEGAPOLI projections for the UK energy related sectors, but when differences arise  
166 the most pessimistic case is adopted.

167  
168 The CMAQ system setup involves the Weather Research and Forecasting (WRF) meso-scale  
169 model, embedded within the ECMWF regional model for the meteorological fields. The initial  
170 and lateral meteorological boundary conditions of the outer domain were derived from the  
171 European Centre for Medium-range Weather Forecasts (ECMWF) gridded analyses available  
172 every 6 h with a horizontal resolution of 0.5 degrees on operational pressure levels up to 50  
173 hPa for vertically distributed data, and at surface and soil levels for surface and deep-soil data.  
174 A grid nudging technique was employed for the outer domain every 6 h in order to constrain  
175 the model towards the analyses. The Met Office have run their global HADGEM2\_ES model  
176 (Collins *et al.*, 2011) and obtained regional meteorological fields for 2020 applying the RCP8.5  
177 emissions scenario, which may be regarded as a pessimistic scenario. The Met Office 2020  
178 meteorological fields used to develop an alternative 2020 PM<sub>2.5</sub> climate change projection are  
179 thought to be representative of the decade. The difference between the 2006 and 2020  
180 meteorological fields is one of many possibilities which could occur, but the approach illustrates  
181 a straightforward methodology which further large-scale computing could develop further.

182  
183 The CMAQ modelling system requires hourly emissions data of primary pollutants. SMOKE  
184 has been developed for this purpose and can be adapted to process annual emissions data  
185 (from point, line and area sources) into temporally-resolved, spatially-distributed and speciated  
186 emissions files ready for the chemical transport model.

187  
188 For CMAQ version 4.7, adopted in this study, the CB05 chemical mechanism was used. The  
189 CB05 mechanism treats the formation of secondary organic aerosols. The tri-modal approach  
190 to aerosol size distribution was used in order to model particulate matter. The species  
191 modelled include sulphate (SO<sub>4</sub><sup>2-</sup>), nitrate (NO<sub>3</sub><sup>-</sup>), ammonium (NH<sub>4</sub><sup>+</sup>), sodium (Na<sup>+</sup>), chloride

192 (Cl<sup>-</sup>), water (H<sub>2</sub>O) and organics from precursors of anthropogenic and biogenic origin but only  
 193 the total PM<sub>10</sub> and PM<sub>2.5</sub> concentrations are considered in this paper. Each mode (Aitken,  
 194 accumulation and coarse) is subject to both wet and dry deposition. Documentation on CMAQ  
 195 is available from the official CMAQ website (<http://www.cmaq-model.org>) [accessed 21 May  
 196 2013]. Chemical boundary conditions were obtained from runs of the global model Geos-  
 197 Chem.

198

### 199 **3 Model Evaluation and Uncertainty**

200

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

205

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

207

$$208 \quad NMB = \frac{\sum_{i=1}^N M_i - O_i}{\sum_{i=1}^N O_i} \quad (1)$$

209

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

212

213 A limited evaluation of CMAQ predictions of PM<sub>2.5</sub> over the UK for the year 2005 was  
 214 undertaken by Yim and Barrett (2012). Only three AURN monitoring sites, where PM<sub>2.5</sub> was  
 215 measured, were operating in 2005: Harwell (rural), Stoke (rural), Bloomsbury (urban  
 216 background) and a fourth roadside site at Marylebone. The NMB was -0.23 at Harwell -0.09 at  
 217 Stoke and -0.27 at Bloomsbury.

218

219 The multi-model AQMEII study (Solazzo *et al.* 2012) compared PM<sub>2.5</sub> at many sites in North  
 220 America and Europe in 2006. It included some CMAQ calculations for Europe by the authors of  
 221 this paper. These generally showed significant under-prediction with a NMB of about -0.4.  
 222 However the CMAQ predictions reported in this paper of individual components of particulate  
 223 matter at Harwell for the year 2006, such as inorganic species (SO<sub>4</sub>, NO<sub>3</sub> and NH<sub>4</sub>), and  
 224 elemental carbon and organic carbon, do not show the same systematic under-prediction. The  
 225 implication is that the model does not include all components and sources of PM<sub>2.5</sub>. One would  
 226 anticipate better model performance for industrial sources for which the primary and secondary  
 227 particulate species source terms are better known.<sup>4</sup> The evaluations undertaken in previous  
 228 studies of CMAQ (see Table 2) are taken as the justification for CMAQ's use in this paper.

229

230 [Table 2 here]

231

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

<sup>4</sup> In 2011 there were 67 AURN (Automatic and Rural Monitoring Network) sites at which PM<sub>2.5</sub> was measured but most are influenced by nearby sources 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.

236 value concentration of about  $20 \mu\text{g m}^{-3}$ . A precautionary estimate of the annual average  $\text{PM}_{2.5}$   
237 concentration in 2006 (see Fig. 1) varies between  $7 (=6 \times 6/5) \mu\text{g m}^{-3}$  in the north of the UK to  $12$   
238  $(=6 \times 10/5) \mu\text{g m}^{-3}$  in south-east England. Later in this paper the road contribution is included as  
239 a separate item in the  $\text{PM}_{2.5}$  budget. Other sources within grid squares, such as from small  
240 stationary sources, are excluded from the calculation. Small stationary sources are not as  
241 widely distributed as traffic sources, affect fewer grid squares and should be subject to local air  
242 quality management control.

243

244 [Fig.1 here]

245

#### 246 **4 Annual average $\text{PM}_{2.5}$ concentrations over the UK in past and future years**

247 2006 was a year when forest fires were prevalent in parts of Europe. Runs of CMAQ, including  
248 and not including the forest fire contribution, suggested that the impact of forest fires on annual  
249 average  $\text{PM}_{2.5}$  concentration over the UK was small, of order 3% or less. The contribution of  
250 wild fires can therefore be discounted.

251

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

257

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

264

265 [Fig. 2 here]

266

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

276

277 [Fig. 3 here]

278

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



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

289  
290 A further calculation was made to determine the contribution of emissions from major industrial  
291 sources regulated in the UK in 2020, using 2006 meteorology, the so-called industrial footprint  
292 in 2020 (see Fig. 4). The maximum concentration from major sources regulated in the UK  
293 amounts to 0.5 to 1  $\mu\text{g m}^{-3}$  in the Midlands, near to the main cluster of major industrial sources.  
294 As a fraction of the total annual average  $\text{PM}_{2.5}$  concentration, the industrial footprint makes up  
295 5 to 15% of  $\text{PM}_{2.5}$  concentrations across most of the UK in 2020. This constitutes a reduction of  
296 about 50% in the major industrial source contribution in the region close to the main cluster of  
297 major industrial sources. The main source contributing to the industrial footprint is the  
298 production of secondary particulate matter from emissions of  $\text{SO}_2$  and  $\text{NO}_x$ . In 2006, the total  
299 emissions from major regulated stationary sources in the UK amounted to about 453  $\text{kt yr}^{-1}$   
300  $\text{SO}_2$ , 441  $\text{kt yr}^{-1}$   $\text{NO}_x$ , 17  $\text{kt yr}^{-1}$   $\text{PM}_{10}$  and 6  $\text{kt yr}^{-1}$   $\text{PM}_{2.5}$  giving a total of about 900  $\text{kt}$  per  
301 annum of 'potential'  $\text{PM}_{2.5}$  emissions ( $\text{SO}_2 + \text{NO}_x + \text{PM}_{2.5}$ ) if all these primary emissions were  
302 converted to  $\text{PM}_{2.5}$ . The equivalent industrial footprint source strength in 2020 is 168  $\text{kt yr}^{-1}$   
303  $\text{SO}_2$ , 197  $\text{kt yr}^{-1}$   $\text{NO}_x$ , 7  $\text{kt yr}^{-1}$   $\text{PM}_{10}$  and 5  $\text{kt yr}^{-1}$   $\text{PM}_{2.5}$  giving an indicative 'potential'  $\text{PM}_{2.5}$   
304 annual emission ( $\text{SO}_2 + \text{NO}_x + \text{PM}_{2.5}$ ) of about 370  $\text{kt}$ . Thus the total primary emission from  
305 regulated major industry expressed as the total of the  $\text{SO}_2 + \text{NO}_x + \text{PM}_{2.5}$  emissions would  
306 have reduced to about 40% of its 2006 level by 2020. The reduction in annual mean  $\text{PM}_{2.5}$   
307 concentrations of about 50% in the region of the sources is approximately of the same order.  
308 Thus 'potential source strength' is a possible, useful indicator of the response of the  
309 atmosphere to changes in emissions.

310  
311 The change in the total  $\text{PM}_{2.5}$  concentration between 2006 and 2020 is not a useful indicator  
312 because the change comes about from a mixture of contributions to  $\text{PM}_{2.5}$  concentration over  
313 the UK. The response will be variable, depending on whether the response is from distant  
314 primary emissions in Europe, mainly from sources in the south east, or from regional industrial  
315 UK sources centred in the Midlands, or from urban UK emissions, spread around the country.  
316 No single indicator is representative of all these responses, which depend on different  
317 geographical distribution changes between 2006 and 2020.

318  
319 Chemical transport models are designed to inform on the possible non-linear relationship  
320 between the change in emissions and the resulting change in concentration of  $\text{PM}_{2.5}$ . One  
321 could imagine that if the atmosphere became relatively more reactive, secondary  $\text{PM}_{2.5}$  could  
322 be formed more quickly near to locations where it was emitted. Lifetimes can be estimated  
323 from the footprints of a specified source at a single location (Fisher *et al.*, 2011). This is not  
324 possible here because the major industrial sources are distributed around the country.  
325 However the change in the primary emissions of major industrial sources is roughly in  
326 proportion to the change in the  $\text{PM}_{2.5}$  concentration, near the cluster of major industrial  
327 sources, suggesting no evidence of very large non-linearity. The change in emissions between  
328 2006 and 2020 may not be large enough to provide evidence of non-linearity and more testing  
329 of responses to changes in emission should be undertaken.

330  
331 [Fig. 4 here]

## 332 333 **5 Climate change and population exposure**

334  
335 Because of the availability of an alternative meteorological scenario in 2020 from results of the  
336 Met Office's HadGEM model, an estimate can be made of the change in annual mean  $\text{PM}_{2.5}$   
337 concentrations in 2020 as a possible consequence of climate change. It turns out that the  
338 effect of the chosen climate change scenario is to reduce annual average  $\text{PM}_{2.5}$  concentrations

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

348  
349 An alternative way of estimating the benefit of emission reductions between 2006 and 2020 is  
350 to determine the population weighted annual mean  $\text{PM}_{2.5}$  concentration. The reduction in the  
351 population weighted annual mean is about 40%. The additional fractional contribution of  
352 climate change in 2020 is estimated to be about 5%. Some slight non-proportional differences  
353 between 2006 and 2020 may occur because of changes in population. The fractional  
354 contribution of major industrial sources to the population weighted annual mean  $\text{PM}_{2.5}$   
355 concentration in 2006 is about 14%.

356  
357 The percentage change in population weighted mean  $\text{PM}_{2.5}$  concentrations between 2006 and  
358 2020 can be compared with the EU Directive 'average exposure indicator' target value. For a  
359 15% reduction in average exposure indicator, SNIFFER (2010) estimated that a reduction in  
360 the annual mean  $\text{PM}_{2.5}$  of 1.5  $\mu\text{g m}^{-3}$  over England, and a 2  $\mu\text{g m}^{-3}$  over inner London between  
361 2010 and 2020, is required. The SNIFFER estimate of  $\text{PM}_{2.5}$  concentrations in 2010, based on  
362 observations at urban sites not at the roadside, is not strictly equivalent to the CMAQ estimate.  
363 However the reduction from the CMAQ estimate of a nearly 40% reduction in the population  
364 weighted annual mean  $\text{PM}_{2.5}$  concentration between 2006 and 2020 appears large enough to  
365 satisfy the 'average exposure indicator' reduction target.

## 366 367 **6 The local contribution to $\text{PM}_{2.5}$**

368  
369 The CMAQ estimate of the population weighted annual mean  $\text{PM}_{2.5}$  concentration reduction  
370 does not include the roadside contribution to  $\text{PM}_{2.5}$  concentrations. This is likely to be a small  
371 fraction of the population exposure as traffic concentrations decline rapidly with distance from  
372 the road. However it is valuable to know the possible contributions from sources within a grid  
373 square to check whether the indicative annual average limit value of 20  $\mu\text{g m}^{-3}$  is likely to be  
374 exceeded at some locations in major urban areas, such as London.

375  
376 We have estimated the concentration of primary  $\text{PM}_{2.5}$  near to roads using the GRAM model  
377 (Fisher and Sokhi, 2000). This requires future road vehicle emission factors<sup>5</sup> (see Table 3).

---

<sup>5</sup> 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<sub>2</sub> 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<sub>2</sub> because of the non-linear chemistry involving ozone

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

380  
381 [Table 3 here]  
382  
383 Assuming representative flows of 25,000, 50,000 and 100,000 vehicles per day for typical  
384 urban and rural roads and a motorway, and a margin of error of +20%, the roadside  
385 concentrations at distances of 5 to 100m from the road centre line have been calculated using  
386 the model GRAM. The typical urban road is likely to be a street canyon.

387  
388 The urban concentrations in central London are taken to be  $12 \mu\text{g m}^{-3}$ ,  $7.2 \mu\text{g m}^{-3}$  and  $7.2 \mu\text{g m}^{-3}$   
389 <sup>3</sup> in 2010, 2020 and 2030, from the 2005 and 2020 CMAQ results in this paper, including a  
390 20% margin of error. Estimates of the local primary  $\text{PM}_{2.5}$  concentration at a typical roadside  
391 location in London, 5m from an urban road with the daily traffic flow of 25,000 in a street  
392 canyon are  $5.3 \mu\text{g m}^{-3}$ ,  $4.6 \mu\text{g m}^{-3}$  and  $4.4 \mu\text{g m}^{-3}$  in 2010, 2020 and 2030. The small extra  
393 improvement beyond 2020 arises partly because road emissions are then dominated by non-  
394 exhaust emissions. Future Euro standards will have little effect on these emissions. In addition  
395 in urban areas, where most people start their journeys, emissions during cold starts are an  
396 important factor. The total  $\text{PM}_{2.5}$  roadside concentrations in 2006, 2020 and 2030 are  
397 estimated to be  $17.3 \mu\text{g m}^{-3}$ ,  $11.8 \mu\text{g m}^{-3}$  and  $11.6 \mu\text{g m}^{-3}$  near a typical road, suggesting  
398 widespread compliance with the target limit value. This does not definitely confirm that  $\text{PM}_{2.5}$   
399 will not be an issue at some locations, but it appears not to be a general issue accepting  
400 approved emissions standards. Near small stationary sources where building downwash may  
401 occur, local air quality management should be considered the tool for addressing  
402 exceedences.

403  
404 This practical estimate of future  $\text{PM}_{2.5}$  concentrations over the UK combines a simple  
405 assessment of road traffic emissions with the contribution from a complex model. The estimate  
406 does not contain estimates near to every kind of road. Exceptional cases, such as an urban  
407 road with very heavy traffic, could lead to higher concentrations but these cases should be  
408 treated using local air quality management action plans. The local air quality management  
409 system can also be used as the process for checking that future emissions follow expected  
410 trends. Actual trends may not follow the expected trend because (1) technological measures  
411 addressing vehicle emissions do not perform as expected, (2) trends in other types of  
412 emissions do not occur as forecast<sup>6</sup>, or (3) unanticipated future trends in the way mobility,  
413 power and heating services are provided. Policy should be flexible to adjust for the actual  
414 future trends in emission when they occur.

## 415 416 **7 Conclusions**

417  
418 From calculations using the CMAQ model it was concluded that in 2006 wild fires make a  
419 minor contribution to annual mean  $\text{PM}_{2.5}$  concentrations in the UK. The contribution of  
420 emissions from major industrial sources regulated in the UK, in 2006, amounts to 1 to  $2 \mu\text{g m}^{-3}$   
421 in the Midlands. This sector makes up 10 to 20% of the annual average  $\text{PM}_{2.5}$  concentration  
422 across most of the UK, excluding the contribution from sources within the grid square  
423 containing the receptor.

---

and primary  $\text{NO}_2$  emissions, but for  $\text{PM}_{2.5}$  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.

<sup>6</sup> 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,

424  
425 The impact of 2020 emissions, using 2006 meteorology, is a reduction of above  $2 \mu\text{g m}^{-3}$  in the  
426 annual average  $\text{PM}_{2.5}$  concentrations over England excluding the sub-grid square contribution,  
427 a reduction of 20 to 40% across most of the UK. The reduction in the population weighted  
428 mean because of emission reductions between 2006 and 2020 is estimated to be 40%.

429  
430 The contribution of emissions from major industrial sources regulated in the UK to regional  
431 annual average  $\text{PM}_{2.5}$  concentrations in 2020, using 2006 meteorology, amounts to  $0.5$  to  $1 \mu\text{g}$   
432  $\text{m}^{-3}$  in central regions of the country near the main cluster of industrial emissions. This  
433 contribution makes up 5 to 15% of the annual average  $\text{PM}_{2.5}$  concentrations excluding the sub-  
434 grid square contribution, across most of the UK. The potential source strength of regulated  
435 major industrial emissions expressed as the total of  $\text{SO}_2 + \text{NO}_x + \text{PM}_{2.5}$  emissions would have  
436 reduced to about 40% of their 2006 level in 2020. The reduction in annual mean  $\text{PM}_{2.5}$   
437 concentrations of about 50% in the Midlands is approximately of the same order. Thus  
438 'potential source strength' is a possible, useful indicator of the response of the atmosphere to  
439 changes in emissions.

440  
441 Applying meteorological conditions in 2020 representing a possible climate change scenario,  
442 with a 2020 emissions scenario, suggests reductions of  $0.2$  to  $0.5 \mu\text{g m}^{-3}$  in the regional annual  
443 average  $\text{PM}_{2.5}$  concentrations over England, equivalent to a reduction of 2 to 5% across most  
444 of the UK compared to  $\text{PM}_{2.5}$  concentrations under 2006 meteorological conditions and a 2020  
445 emissions scenario. The 'climate change scenario' reduction is between 5 to 10 times smaller  
446 than the reduction brought about by emissions changes between 2006 and 2020.

447  
448 Estimates of the local primary  $\text{PM}_{2.5}$  concentration at a typical roadside location in London  
449 have been added to the regional  $\text{PM}_{2.5}$  concentration to test whether compliance with the  
450 indicative  $\text{PM}_{2.5}$  limit value of  $20 \mu\text{g m}^{-3}$  in 2020 will be achieved. The total  $\text{PM}_{2.5}$  concentrations  
451 in 2006, 2020 and 2030, including a margin of error, are  $17.3 \mu\text{g m}^{-3}$ ,  $11.8 \mu\text{g m}^{-3}$  and  $11.6 \mu\text{g}$   
452  $\text{m}^{-3}$  suggesting widespread compliance with the target limit value. This does not confirm that  
453  $\text{PM}_{2.5}$  will not be an issue at some locations, but it appears not to be a general issue accepting  
454 current air quality standards and likely trends in emissions. At hot spots, where traffic flows are  
455 higher than at a typical urban road, local air quality management should be considered the tool  
456 for addressing  $\text{PM}_{2.5}$  exceedences. The introduction of new emission technology standards is  
457 the tool for addressing air quality considerations on a national basis.

458  
459 There is uncertainty associated with the air quality standard used in this paper. We have  
460 adopted the approach, implicit in the Ambient Air Quality Directive, that all components of  
461  $\text{PM}_{2.5}$  have the same health disbenefit and the aim should be to reach and maintain the  
462 strictest standard implied by the values stated in the Directive. If further research suggests that  
463 certain components of  $\text{PM}_{2.5}$  are the active components causing harm and  $\text{PM}_{2.5}$  acts as the  
464 surrogate for these components in the underlying epidemiological studies, then the conclusions  
465 may be different. Moreover understanding the observed trends in annual average  
466 concentrations is a necessary step to ensure compliance, and modelling can only suggest a  
467 strategy towards ensuring future compliance. Unexpected changes in emissions and  
468 unexplained observational trends compared with model predictions may still arise.

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474 Results of this work may not necessarily reflect the views of the Environment Agency and no  
475 official endorsement should be inferred.

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626

1 **Figure Captions**

2

3 Fig. 1 Annual average regional PM<sub>2.5</sub> concentrations ( $\mu\text{g m}^{-3}$ ) in 2006 from all  
4 sources (excluding the contribution from sources within the grid square containing  
5 the receptor)

6

7 Fig. 2 Annual average regional PM<sub>2.5</sub> concentrations ( $\mu\text{g m}^{-3}$ ) from major industrial  
8 source regulated in the UK in 2006

9

10 Fig. 3 Annual average regional PM<sub>2.5</sub> concentrations ( $\mu\text{g m}^{-3}$ ) in 2020 from all  
11 sources using 2006 meteorology (excluding the contribution from sources within the  
12 grid square containing the receptor)

13

14 Fig. 4 Annual average regional PM<sub>2.5</sub> concentrations ( $\mu\text{g m}^{-3}$ ) from emissions from  
15 major industrial sources regulated in the UK in 2020 using 2006 meteorology (the  
16 'industrial footprint' in 2020)

17



Pollutant	Ratio 2020:2006 major industry emissions	Ratio 2020:2006 national total UK emissions
SO <sub>2</sub>	0.37	0.47
NO <sub>x</sub>	0.45	0.52
PM <sub>10</sub>	0.42	0.72
PM <sub>2.5</sub>	0.65	0.68
NMVOC	0.87	0.72

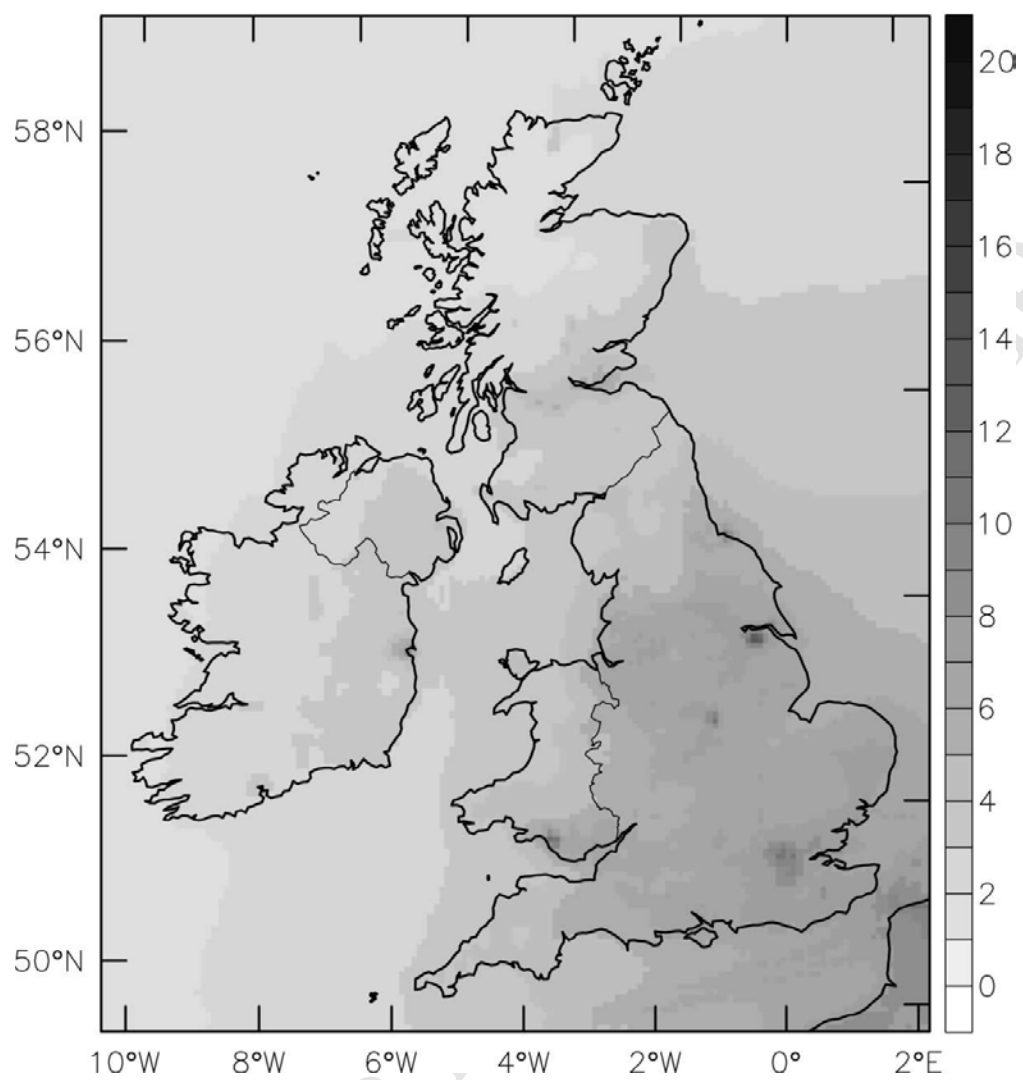
Table 1 Estimated ratio of the major industrial emissions and national total emissions of SO<sub>2</sub>, NO<sub>x</sub>, PM<sub>10</sub> and NMVOC between 2006 and 2020

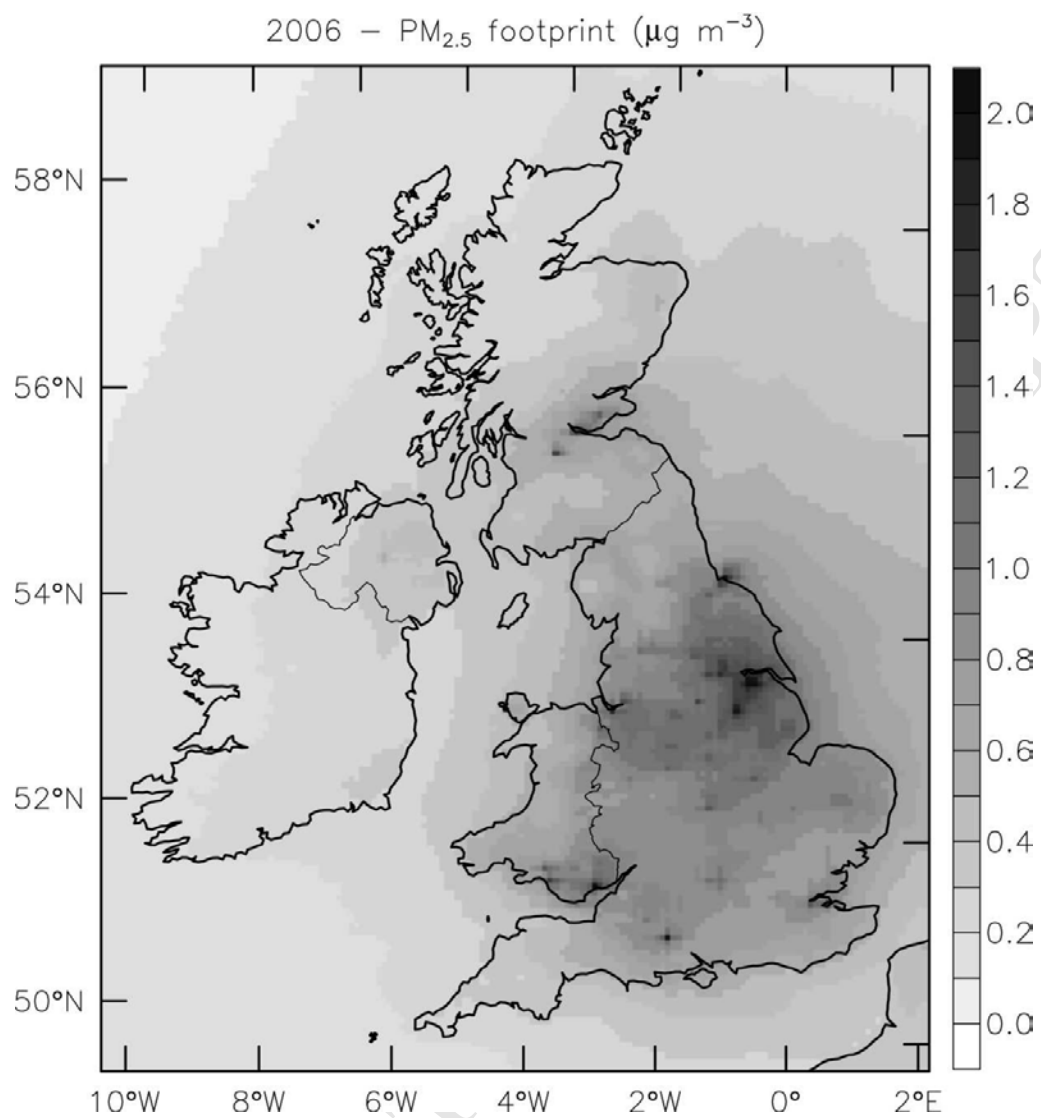
Model metric PM <sub>10</sub> for 2003	CMAQ v4.6	CMAQ v4.7	TRACK-ADMS
<i>FAC2</i> (%)	88.2	100	100
<i>r</i> (correlation coefficient)	0.09	0.0	0.45
<i>NMB</i>	-0.33	-0.09	-0.20
Single power station contribution (%)	0.34	0.28	0.28

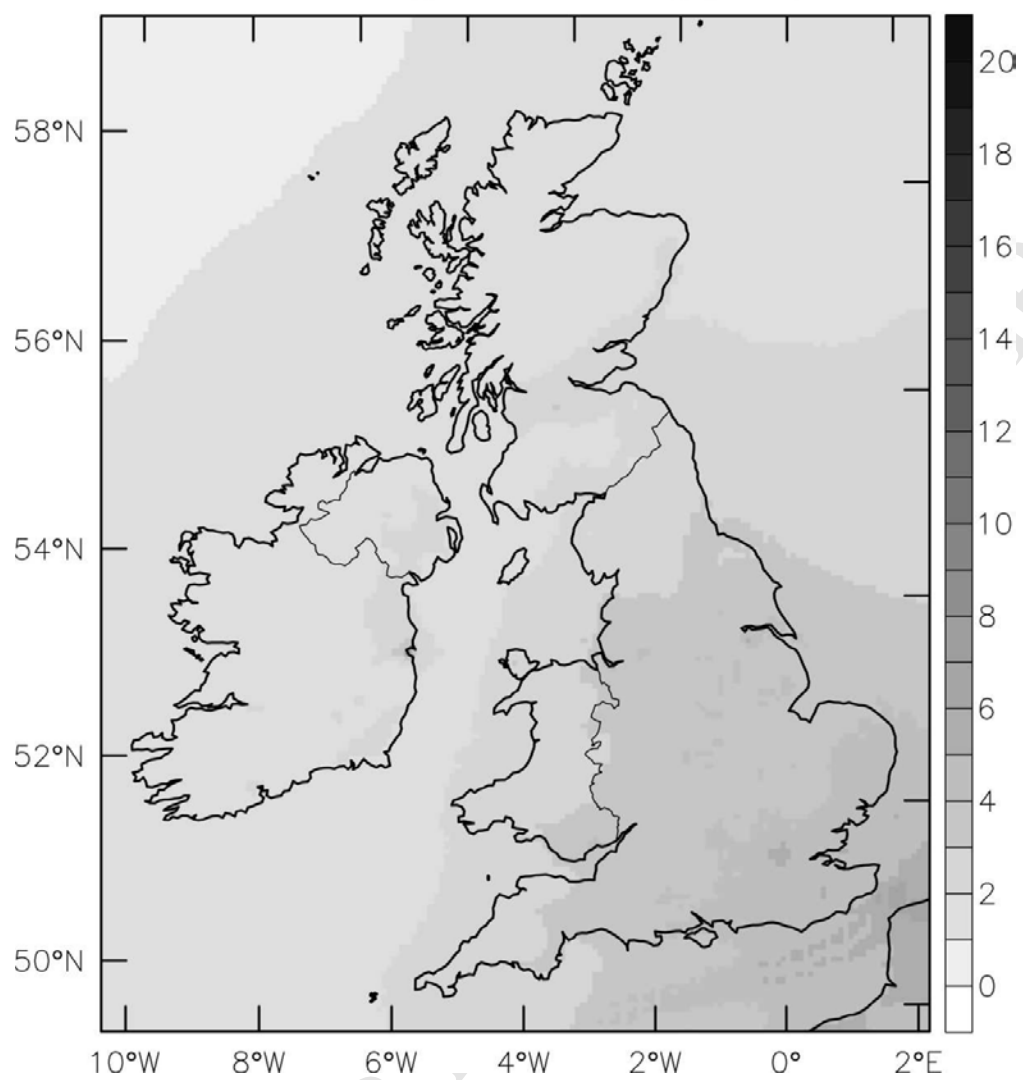
Table 2 Comparison of performance in predicting annual average PM<sub>10</sub> concentration in 2003 at 40 urban background and rural background sites in the AURN network in the UK for two versions of the advanced model CMAQ (Chemel *et al.* 2011) and a simpler, semi-empirical model TRACK-ADMS (Vincent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

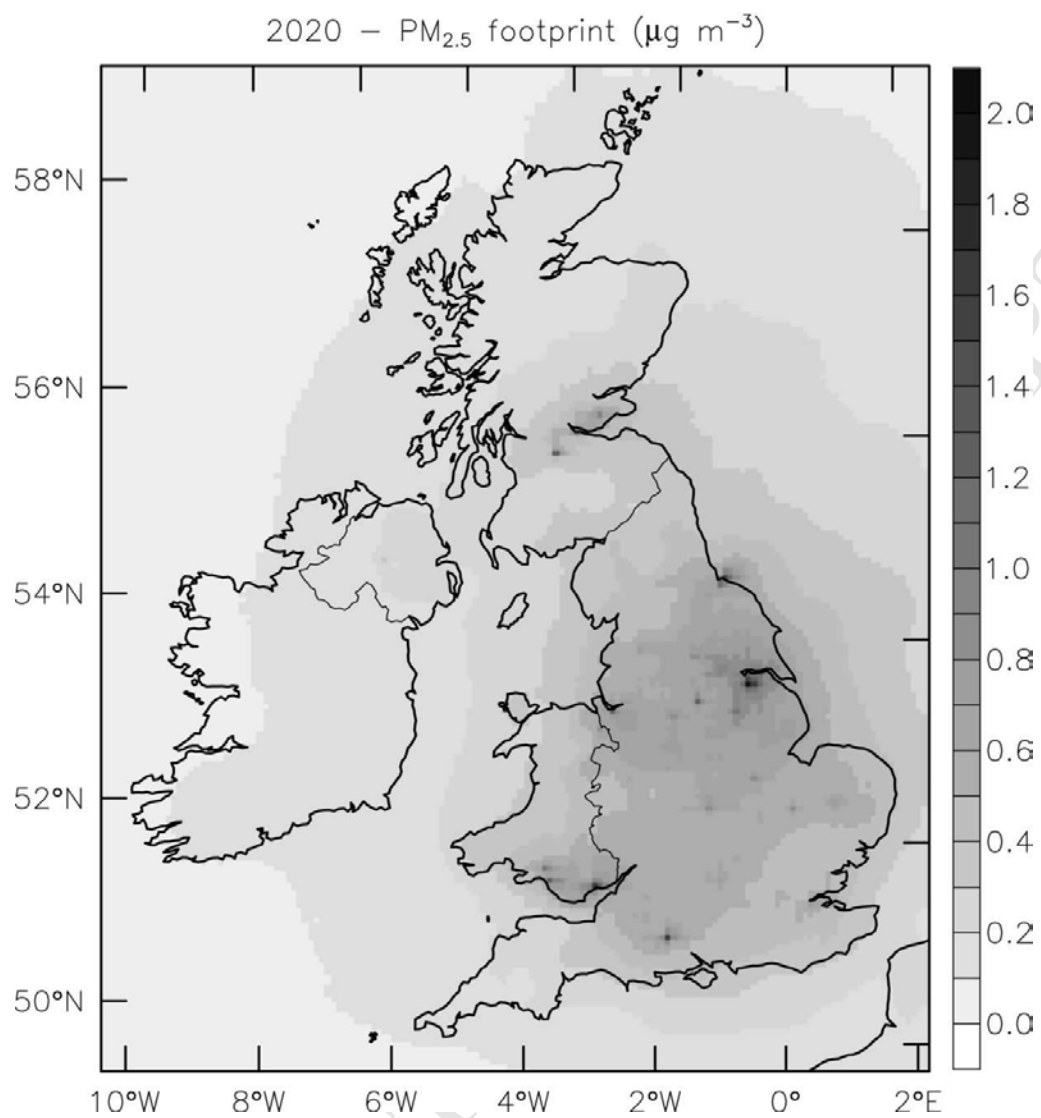
Year	LDV			HDV		
	Urban	Rural	Motorway	Urban	Rural	Motorway
2010	0.38	0.37	0.47	5.02	4.71	5.23
2020	0.28	0.21	0.42	1.47	0.76	0.77
2030	0.14	0.15	0.23	0.45	0.45	0.53

Table 3 Emission factors (g PM<sub>2.5</sub>/km) for light duty vehicles (LDV) and heavy duty vehicles (HDV) in current and future years for typical roads

2006 – PM<sub>2.5</sub> concentration ( $\mu\text{g m}^{-3}$ )



2020 – PM<sub>2.5</sub> concentration ( $\mu\text{g m}^{-3}$ )



- Model CMAQ evaluates UK regional PM<sub>2.5</sub> concentration for 2006 and 2020.
- A correction for local traffic sources within a major city is made.
- PM<sub>2.5</sub> complies with implied air quality standards.
- CMAQ model can be applied to answer policy questions.
- Industrial source footprint contribution is less than 15% of PM<sub>2.5</sub>.