

# 1 The sensitivities of emissions reductions for the mitigation of UK

## 2 PM<sub>2.5</sub>

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### 12 13 ABSTRACT

14  
15 The reduction of ambient concentrations of fine particulate matter (PM<sub>2.5</sub>) is a key objective for  
16 air pollution control policies in the UK and elsewhere. Long-term exposure to PM<sub>2.5</sub> has been  
17 identified as a major contributor to adverse human health effects in epidemiological studies and  
18 underpins ambient PM<sub>2.5</sub> legislation. As a range of emission sources and atmospheric chemistry  
19 transport processes contribute to PM<sub>2.5</sub> concentrations, atmospheric chemistry transport models  
20 are an essential tool to assess emissions control effectiveness. The EMEP4UK atmospheric  
21 chemistry transport model was used to investigate the impact of reductions in UK  
22 anthropogenic emissions of primary PM<sub>2.5</sub>, NH<sub>3</sub>, NO<sub>x</sub>, SO<sub>x</sub> or non-methane VOC on surface  
23 concentrations of PM<sub>2.5</sub> in the UK for a recent year (2010) and for a future current legislation  
24 emission (CLE) scenario (2030). In general, the sensitivity to UK mitigation is rather small. A  
25 30% reduction in UK emissions of any one of the above components yields (for the 2010  
26 simulation) a maximum reduction in PM<sub>2.5</sub> in any given location of ~0.6 µg m<sup>-3</sup> (equivalent to  
27 ~6% of the modelled PM<sub>2.5</sub>). On average across the UK, the sensitivity of PM<sub>2.5</sub> concentrations  
28 to a 30% reduction in UK emissions of individual contributing components, for both the 2010  
29 and 2030 CLE baselines, increases in the order NMVOC, NO<sub>x</sub>, SO<sub>x</sub>, NH<sub>3</sub> and primary PM<sub>2.5</sub>;  
30 however there are strong spatial differences in the PM<sub>2.5</sub> sensitivities across the UK.  
31 Consequently, the sensitivity of PM<sub>2.5</sub> to individual component emissions reductions varies  
32 between area and population weighting. Reductions in NH<sub>3</sub> have the greatest effect on area-

33 weighted PM<sub>2.5</sub>. A full UK population weighting places greater emphasis on reductions of  
34 primary PM<sub>2.5</sub> emissions, which is simulated to be the most effective single-component control  
35 on PM<sub>2.5</sub> for the 2030 scenario. An important conclusion is that weighting corresponding to the  
36 Average Exposure Indicator metric (using data from the 45 model grids containing a monitor  
37 whose measurements are used to calculate the UK AEI) further increases the emphasis on the  
38 effectiveness of primary PM<sub>2.5</sub> emissions reductions (and of NO<sub>x</sub> emissions reductions) relative  
39 to the effectiveness of NH<sub>3</sub> emissions reductions. Reductions in primary PM<sub>2.5</sub> have the largest  
40 impact on the AEI in both 2010 and the 2030 CLE scenario. The summation of the modelled  
41 reductions to the UK PM<sub>2.5</sub> AEI from 30% reductions in UK emissions of primary PM<sub>2.5</sub>, NH<sub>3</sub>,  
42 SO<sub>x</sub>, NO<sub>x</sub> and VOC totals 1.17 µg m<sup>-3</sup> and 0.82 µg m<sup>-3</sup> for the 2010 and 2030 CLE simulations,  
43 respectively (not accounting for non-linearity).

44

## 45 **1 Introduction**

46

47 Atmospheric particulate matter (PM) has a range of adverse impacts including on climate  
48 change through radiative forcing (IPCC, 2013) and on human health (WHO, 2006, 2013). The  
49 global health burden from exposure to ground-level ambient fine particulate matter (as  
50 characterised by the PM<sub>2.5</sub> metric) is substantial. The Global Burden of Disease project  
51 attributed 3.2 million premature deaths and 76 million disability-adjusted life years to exposure  
52 to ambient PM<sub>2.5</sub> concentrations prevailing in 2005 (Lim et al., 2012). Exposure to ambient  
53 PM<sub>2.5</sub> remains a major health issue in Europe. The European Environment Agency report that  
54 for the period 2010-2012, 10-14% of the urban population in the EU28 countries was exposed  
55 to ambient concentrations of PM<sub>2.5</sub> exceeding the EU annual-average PM<sub>2.5</sub> reference value of  
56 25 µg m<sup>-3</sup>, but 91-93% were exposed to concentrations exceeding the WHO annual-average  
57 PM<sub>2.5</sub> air quality guideline of 10 µg m<sup>-3</sup> (EEA, 2014).

58

59 European Commission (EC) legislation for PM<sub>2.5</sub> includes an obligation on individual member  
60 states to reduce exposure to PM<sub>2.5</sub> in areas of population by a prescribed percentage between  
61 2010 and 2020. The exposure to PM<sub>2.5</sub> is quantified through the Average Exposure Indicator  
62 (AEI) which is the average of the annual PM<sub>2.5</sub> measured across designated urban background  
63 and suburban sites spread over cities and large towns (averaged over the 3-year periods  
64 spanning 2010 and 2020). The AEI is therefore a quasi-indicator of population-weighted PM<sub>2.5</sub>.

65 For the UK, the calculation of the AEI uses data from 45 sites (Brookes et al., 2012) and the  
66 required reduction by 2020 is 15% from its 2010 value of  $13 \mu\text{g m}^{-3}$  (Defra, 2012).

67  
68 While standards focus on  $\text{PM}_{2.5}$  mass concentrations, meeting these standards is complicated  
69 by the considerable chemical heterogeneity, which arises because ambient  $\text{PM}_{2.5}$  comprises  
70 both primary PM emissions and secondary inorganic and organic components formed within  
71 the atmosphere from gaseous precursor emissions, specifically  $\text{NH}_3$ ,  $\text{NO}_x$  ( $\text{NO}$  &  $\text{NO}_2$ ),  $\text{SO}_2$   
72 and a wide range of non-methane volatile organic compounds (VOC) (USEPA, 2009; AQEG,  
73 2012). Meteorological conditions also control  $\text{PM}_{2.5}$  concentrations through their influences on  
74 dispersion, chemistry and deposition.

75  
76 European legislation sets current and future caps on anthropogenic emissions of primary and  
77 secondary-precursor components of  $\text{PM}_{2.5}$  at national level and from individual sources (Heal  
78 et al., 2012). Although it is well-known that much of the ambient  $\text{PM}_{2.5}$  in the UK derives from  
79 trans-boundary emissions and transport into the UK (Vieno et al., 2014;AQEG, 2015), a  
80 pertinent policy question to address is: what additional surface  $\text{PM}_{2.5}$  reductions could the UK  
81 unilaterally achieve, at least in principle? In other words, what are the sensitivities of UK  $\text{PM}_{2.5}$   
82 to UK reductions in emissions of relevant components?

83  
84 This is the motivation for the work presented here, which investigates the impact of reductions  
85 from UK anthropogenic sources of emissions of primary  $\text{PM}_{2.5}$  and of precursors of secondary  
86  $\text{PM}_{2.5}$  on surface  $\text{PM}_{2.5}$  concentrations across the whole UK. To adequately simulate the UK  
87 national domain requires the use of a regional atmospheric chemistry transport model (ACTM),  
88 in this study the EMEP4UK Eulerian ACTM (Vieno et al., 2009;Vieno et al., 2010;Vieno et al.,  
89 2014). Recognising that reductions in UK and rest-of-Europe emissions are already projected  
90 under current legislation, this work compares the present-day sensitivity of UK emissions  
91 reductions on UK  $\text{PM}_{2.5}$  with a future time point (2030) to examine the effectiveness of potential  
92 options in the future. It is recognised that climate change may also have some influence on  
93 future  $\text{PM}_{2.5}$  concentrations in the UK; however the focus is here on UK precursor emission  
94 sensitivity and many studies have concluded that on the 2030 timescale air pollutant  
95 concentrations will be much more strongly influenced by changes in precursor emissions than  
96 by changes in climate (e.g. Langner et al. (2012);Coleman et al. (2013);Colette et al. (2013)).

97 Throughout, the focus is on annual average  $PM_{2.5}$ , since this is the metric within the AEI, which  
98 in turn is driven by the evidence from epidemiological studies that demonstrate associations  
99 between adverse health outcomes and long-term (annual average) concentrations of  $PM_{2.5}$   
100 (COMEAP, 2010;WHO, 2013). It is also recognised that, whilst the focus here is on reduction  
101 in concentrations of  $PM_{2.5}$  from the perspective of its impact on human health, the reduction of  
102 anthropogenic emissions in general will also have other benefits including on human health, on  
103 N and S deposition, and on ozone formation.

104

## 105 **2 Methods**

106

### 107 **2.1 Model description and set-up**

108 The EMEP4UK model used here is a regional ACTM based on version rv4.4 ([www.emep.int](http://www.emep.int))  
109 of the EMEP MSC-W model which is described in Simpson et al. (2012). A detailed description  
110 of the EMEP4UK model is given in Vieno et al. (2010), and Vieno et al. (2014).

111

112 The EMEP4UK model meteorological driver is the Weather Research and Forecast (WRF)  
113 model version 3.1.1 ([www.wrf-model.org](http://www.wrf-model.org)). The EMEP4UK and WRF model horizontal  
114 resolution is  $50\text{ km} \times 50\text{ km}$  for the extended European domain and  $5\text{ km} \times 5\text{ km}$  for the inner  
115 domain as illustrated in Figure 1. The EMEP4UK model uses a nested approach, the European  
116 domain concentrations are used as boundary condition for the UK domain. The boundary  
117 conditions at the edge of the European domain are prescribed concentrations in terms of latitude  
118 and adjusted for each year. For ozone, 3-D fields for the whole domain are specified from  
119 climatological ozone-sonde data-sets, modified monthly against clean-air surface observations  
120 as described in Simpson et al. (2012).

121

122 The default EMEP MSC-W chemical scheme was used for the present study, as it has been  
123 extensively validated at the European scale (Simpson et al. (2012), [www.emep.int](http://www.emep.int)). The scheme  
124 has 72 species and 137 reactions, and full details are given in Simpson et al. (2012). The  
125 gas/aerosol partitioning is the model for aerosols reacting system (MARS) formulation  
126 (Simpson et al., 2012). In the model version used here,  $PM_{2.5}$  is the sum of the fine ( $PM_{2.5}$ )  
127 fraction of: ammonium ( $NH_4^+$ ), sulphate ( $SO_4^{2-}$ ), nitrate ( $NO_3^-$ ), elemental carbon (EC), organic

128 matter (OM), sea salt (SS), mineral dust, and 27% of the coarse nitrate.  $PM_{10}$  is the sum of  
129  $PM_{2.5}$  plus the coarse ( $PM_{2.5-10}$ ) fraction of EC, OM,  $NO_3^-$ , SS, and dust.

130 Whilst fine nitrate production is modelled using a thermodynamic model (MARS), the  
131 formation of coarse nitrate from nitric acid ( $HNO_3$ ) uses a parameterised approach that seeks  
132 to capture the  $HNO_3$  reaction with sea salt and crustal material. The conversion rate of  $HNO_3$   
133 to coarse nitrate depends on relative humidity, as described by Simpson et al. (2012), but is not  
134 explicitly linked to the surface area of the existing coarse aerosol. Both nitrate generation  
135 mechanisms compete for the same  $HNO_3$ , and whilst this constrains the total amount of nitrate  
136 produced, it is acknowledged that the resulting split into fine and coarse nitrate is somewhat  
137 uncertain as discussed in Aas et al. (2012). A more explicit aerosol scheme is under  
138 development for the model.

139

140 Anthropogenic emissions of  $NO_x$ ,  $NH_3$ ,  $SO_2$ , primary  $PM_{2.5}$ , primary  $PM_{coarse}$ , CO and non-  
141 methane VOC for the UK are derived from the National Atmospheric Emission Inventory  
142 (NAEI, <http://naei.defra.gov.uk>) at  $1\text{ km} \times 1\text{ km}$  resolution and aggregated to  $5\text{ km} \times 5\text{ km}$   
143 resolution. For the European domain, the model uses the EMEP  $50\text{ km} \times 50\text{ km}$  resolution  
144 emission estimates provided by the Centre for Emission Inventories and Projections (CEIP,  
145 <http://www.ceip.at/>). Shipping emissions estimates, for the inner domain, are derived from the  
146 ENTEC (now Amec Foster Wheeler) emissions estimate (ENTEC, 2010). Natural emissions of  
147 isoprene and DMS are as described in [Simpson et al. \(2012\)](#).

148

149 The EMEP MSC-W model from which the EMEP4UK model is derived is used widely in  
150 support of European air quality science and policy development and the performances of both  
151 have been extensively evaluated (Carslaw, 2011b;Schulz et al., 2013;Simpson et al.,  
152 2014;Schaap et al., 2015).

153

## 154 **2.2 Model experiments**

155 A base run and a set of 5 sensitivity experiments were carried out for emissions and meteorology  
156 for 2010. The experiments applied 30% reductions to UK anthropogenic emissions from all  
157 sectors for each of the following pollutants individually: primary  $PM_{2.5}$ ,  $NH_3$ ,  $NO_x$ ,  $SO_x$  and  
158 NMVOC. This 30% perturbation was applied to land-based emissions only; shipping emissions  
159 (both domestic and international) were left unchanged.

160

161 Model runs were repeated for a 2030 future emissions scenario to investigate sensitivities of  
162 UK PM<sub>2.5</sub> to UK emissions reductions further along the pathway of current legislation (CLE)  
163 emissions. The 2030 CLE emissions used in the model runs were based on the 2030 IIASA  
164 CLE projection (IIASA, 2012) for Europe and the *Updated Energy Projections* (UEP, version  
165 45) for the UK. The UEPs are developed and regularly updated by analysing and projecting  
166 future energy use and are based on assumptions of future economic growth, fossil fuel prices,  
167 UK population development and other key variables. A set of projections is based on a range  
168 of assumptions to represent the uncertainty in making such projections into the future. For this  
169 manuscript, the mid-range estimates were used. For a full description of the UEPs and the  
170 methodology for their compilation, see DECC (2015). Emissions from shipping were 2020  
171 emissions estimate provided by ENTEC (now Amec Foster Wheeler) (ENTEC, 2010).

172

173 No change in the spatial distribution of emissions was made. Whilst there will likely be some  
174 changes in the spatial distribution of emissions, such changes are not easily predicted for a  
175 future scenario, and may be anticipated to be smaller than the changes in absolute amounts of  
176 emissions. The boundary and initial conditions for ozone and particles outside the European  
177 domain were left unchanged to the year 2010, as was the meteorology. The use of the same  
178 meteorology isolates the sensitivity of surface PM<sub>2.5</sub> to emissions reductions at some future date  
179 from the effects on surface PM<sub>2.5</sub> due to differences in meteorology.

180

181 As well as maps of annual-average surface PM<sub>2.5</sub> concentrations the following three summary  
182 statistics for UK PM<sub>2.5</sub> were calculated: (i) the area-weighted average, i.e. the average of all 5  
183 km × 5 km model grids over the UK; (ii) the population-weighted average, i.e. the 5 km × 5  
184 km gridded estimates of PM<sub>2.5</sub> surface concentrations re-projected onto the British National  
185 Grid and multiplied by population estimates at the same spatial resolution (derived from the  
186 UK census, <http://census.edina.ac.uk/>) (Figure 2) and divided by the sum of the UK population;  
187 (iii) a value analogous to the Average Exposure Indicator (AEI), calculated as the average of  
188 the concentrations for the 45 model grids containing a PM<sub>2.5</sub> monitor whose measurements are  
189 used to define the UK's 2010 AEI value (Brookes et al., 2012).

190

### 191 3 Results

192

193 Example comparisons between EMEP4UK-modelled surface concentrations of PM<sub>2.5</sub>  
194 components and total measured PM<sub>2.5</sub> are shown in Figure 3 for three UK national network  
195 monitoring sites: Edinburgh St. Leonards, an urban background site in the north of the UK;  
196 London North Kensington, an urban background site in central London in the south-east of the  
197 UK; and Harwell, a rural background site in central England. Monthly averages of the hourly  
198 measured and modelled data are presented. Model simulations follow the observational time  
199 trends well. The model simulations of the SIA components SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> have  
200 previously been individually evaluated by Vieno et al. (2014) against 10 years of speciated  
201 observations made at ~30 sites across the UK in the AGANET network (Conolly et al., 2011).  
202 The four UK sites included in Vieno et al. (2014) showed good agreement between the **monthly**  
203 **averaged** EMEP4UK simulation and the observed NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>, with a bias range of 0.28 to  
204 -0.62 and 0.8 to -0.27 µg m<sup>-3</sup>, respectively. The EMEP4UK model was also evaluated against  
205 observations and other models in a UK model inter-comparison organised by the UK  
206 Department for Environment, Food & Rural Affairs (Defra) (Carslaw, 2011b, a). The persistent  
207 negative bias in the sum of the modelled PM<sub>2.5</sub> against observation in Figure 3 is consistent  
208 with the absence of re-suspended dust in the model configuration used here, and possibly also  
209 reflects a difference in the treatment of particle-bound water in model and measurement. The  
210 omission of re-suspended dust does not impact on the investigations here of the sensitivities of  
211 PM<sub>2.5</sub> concentrations to anthropogenic emissions reductions; however it is acknowledged that  
212 since particle-bound water is related to the mass of secondary inorganic components its  
213 omission will have some impact on the sensitivity of PM<sub>2.5</sub> to inorganic precursor gas emissions  
214 reductions. Different measurement techniques and conditions incorporate different proportions  
215 of the ambient PM<sub>2.5</sub> water content. **We focus here on changes to the dry mass concentrations**  
216 **of surface PM<sub>2.5</sub> derived from changes in the emissions of primary PM<sub>2.5</sub> and in secondary PM<sub>2.5</sub>**  
217 **precursor gases.** (It is also noted that values of relative reductions in modelled PM<sub>2.5</sub> will be  
218 slightly higher than if expressed relative to measured PM<sub>2.5</sub> at that location.) Some model  
219 underestimation may also derive from dilution of primary PM<sub>2.5</sub> emissions into the 5 km grid  
220 of the model compared with the primary emissions more local to an urban background monitor.  
221 The simulated 'baseline' 2010 annual-average surface concentrations for PM<sub>2.5</sub> at 50 km  
222 horizontal resolution for the EMEP4UK European domain and for the nested 5 km horizontal

223 resolution British Isles domain are shown in Figure 1. The UK 2010 annual-average surface  
224 concentrations of PM<sub>2.5</sub> are generally lower compared with neighbouring continental countries  
225 such as France, the Netherlands and Germany. The influence of emissions originating from  
226 continental Europe is revealed by the gradient of decreasing PM<sub>2.5</sub> concentrations away from  
227 the continent. An analysis presented in AQEG (2015) also using the EMEP4UK model showed  
228 that UK emissions contribute around 55% of the total PM<sub>2.5</sub> in the UK. This limits the extent to  
229 which long-term average concentrations can be reduced by UK action alone.

230

231 Figure 4 shows maps of the impacts on 2010 surface PM<sub>2.5</sub> for 30% reductions in UK terrestrial  
232 emissions of each of NH<sub>3</sub>, NO<sub>x</sub>, SO<sub>x</sub>, VOC and primary PM<sub>2.5</sub>. The effect of these emissions  
233 reductions on the three measures of UK-average surface concentrations of PM<sub>2.5</sub> are illustrated  
234 in Figure 5, based on the data given in Table 1. The principal observations from the two figures  
235 are that PM<sub>2.5</sub> levels in the UK do not show strong responses to UK-only reductions in emissions  
236 of individual components/precursors of PM<sub>2.5</sub>, and that the responses are highly geographically  
237 variable. The maximum reduction in PM<sub>2.5</sub> concentrations (at a 5 km grid resolution) reaches  
238 ~0.6 µg m<sup>-3</sup> (~6% of the modelled components) in response to a 30% reduction in UK emissions  
239 of individual components, and in most locations the reductions in PM<sub>2.5</sub> concentrations are  
240 considerably smaller. This again indicates the influence on PM<sub>2.5</sub> in the UK (on an annual  
241 average basis) from emissions outside of the UK. In the case of the formation of SIA  
242 components, it also reflects the non-linearity in the precursor oxidation chemistry and gas-  
243 particle phase partitioning that occurs between emission location and receptor location  
244 (Harrison et al., 2013; Vieno et al., 2014).

245

246 Figure 4 and Figure 5 show that, on average across the UK, the sensitivity of PM<sub>2.5</sub>  
247 concentrations to a 30% reduction in UK emissions of individual contributing components  
248 increases in the order VOC, NO<sub>x</sub>, SO<sub>x</sub>, primary PM<sub>2.5</sub> and NH<sub>3</sub>. The exact order varies slightly  
249 with the UK-average measure used (Figure 5). This is due to differences in the spatial patterns  
250 of the PM<sub>2.5</sub> reductions shown in Figure 4 in relation to the distribution of UK population shown  
251 in Figure 2.

252

253 The 30% reductions in UK VOC emissions gives maximum reductions of ~0.15 µg m<sup>-3</sup> (1.5%)  
254 in PM<sub>2.5</sub> concentrations in central and northern England and central Scotland (Figure 4e). The



255 30% reductions in UK NO<sub>x</sub> emissions yield around 0.2 µg m<sup>-3</sup> (3%) reductions in PM<sub>2.5</sub> over  
256 some rural areas (Figure 4c), and generally a maximum of 0.15 µg m<sup>-3</sup> (1.5%) reductions in  
257 PM<sub>2.5</sub> over other rural areas. An important observation is that reductions of PM<sub>2.5</sub> over urban  
258 centres are smaller (no more than 0.15 µg m<sup>-3</sup>) than in rural areas for these reductions in NO<sub>x</sub>  
259 emissions. The 30% reductions in UK SO<sub>x</sub> emissions yield up to ~0.45-0.5 µg m<sup>-3</sup> (5%)  
260 reductions in PM<sub>2.5</sub> in the Trent valley and up to around 0.3-0.35 µg m<sup>-3</sup> (3%) reductions in  
261 PM<sub>2.5</sub> over large areas of central and northern England and central Scotland (Figure 4d). The  
262 locations with greatest sensitivities to the 30% NO<sub>x</sub> emissions reductions (Figure 4c) are  
263 generally those with the lowest sensitivities to SO<sub>x</sub> emissions reductions (Figure 4d). As with  
264 the NO<sub>x</sub> emissions reductions, the reductions in PM<sub>2.5</sub> concentrations for reductions in SO<sub>x</sub>  
265 emissions is not, in general, associated with the major urban areas, except where these also have  
266 major SO<sub>x</sub> sources in the vicinity (e.g. Trent Valley, West Midlands, Cheshire). This is  
267 primarily caused by the spatial distribution of major sources of SO<sub>x</sub> emissions. As ~80% of UK  
268 SO<sub>x</sub> 2010 emissions originate from large point sources (power plants, industrial facilities),  
269 which are not located in the heart of urban areas, associated emission reductions have the most  
270 profound effects in rural areas. However, the greater sensitivity to SO<sub>x</sub> close to large point  
271 sources (e.g. coal-fired power plants) may in part be an artefact due to the model assumption  
272 that 5% of SO<sub>x</sub> emissions are directly in the form of SO<sub>4</sub><sup>2-</sup>, which may no longer be appropriate  
273 for these sources or for models running at relatively high horizontal spatial resolution. The SO<sub>x</sub>  
274 and NO<sub>x</sub> gases compete in their reaction with NH<sub>3</sub> to form particulate ammonium sulphate  
275 ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>) or ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>). The larger sensitivity of PM<sub>2.5</sub> formation to NH<sub>3</sub>  
276 emissions reductions indicates that NH<sub>3</sub> is the limiting species; whilst the greater sensitivity to  
277 SO<sub>x</sub> than to NO<sub>x</sub> emissions reductions reflects that the reaction between NH<sub>3</sub> and SO<sub>x</sub> is fast  
278 and essentially irreversible compared with the equilibrium reactions between gaseous NH<sub>3</sub> and  
279 NO<sub>x</sub> species and NH<sub>4</sub>NO<sub>3</sub>.

280

281 The largest reductions in PM<sub>2.5</sub> (when weighted towards areas of greatest population) derive  
282 from 30% reductions in UK NH<sub>3</sub> and primary PM<sub>2.5</sub> emissions (Figure 4b and Figure 4f), up to  
283 0.45 µg m<sup>-3</sup> for NH<sub>3</sub> reductions and greater for primary PM<sub>2.5</sub> reductions (up to ~6% of  
284 modelled PM<sub>2.5</sub> in both cases). There is a distinct inverse geographic relationship in the PM<sub>2.5</sub>  
285 sensitivity to reductions of these two components. The reductions in NH<sub>3</sub> emissions give  
286 greatest PM<sub>2.5</sub> decreases in agricultural areas, whereas the reductions in primary PM<sub>2.5</sub> give

287 greatest decreases in the large conurbations and other areas of high population density. The  
288 difference in geographical patterns is highlighted more clearly in Figure 6a which shows the  
289 data in Figure 4b minus the data in Figure 4f. Blue colours in Figure 6a indicate where  
290 reductions in PM<sub>2.5</sub> from a 30% reduction in NH<sub>3</sub> emissions exceed the reductions in PM<sub>2.5</sub>  
291 from a 30% reduction in primary PM<sub>2.5</sub> emissions, and vice-versa for red colours. White colours  
292 indicate comparable reductions in PM<sub>2.5</sub> via primary PM<sub>2.5</sub> or NH<sub>3</sub> emissions reductions. The  
293 geographical pattern in PM<sub>2.5</sub> sensitivity reflects the geographical pattern of the emission  
294 sources and the fact that, because of the short atmospheric lifetime of NH<sub>3</sub>, UK emissions of  
295 NH<sub>3</sub> also generally have short-range influence.

296  
297 Figure 7 shows the map of annual-average surface concentration of PM<sub>2.5</sub> estimated for the  
298 2030 CLE emissions projections, and of the difference between the PM<sub>2.5</sub> concentrations in  
299 2030 and 2010. Surface concentrations of PM<sub>2.5</sub> over the UK are simulated to reduce by up to  
300 2.8 µg m<sup>-3</sup> between 2010 and the 2030 CLE emissions scenario used. The UK-wide reductions  
301 in PM<sub>2.5</sub> between 2010 and 2030 CLE are 1.70, 2.24 and 2.42 µg m<sup>-3</sup> for the area-weighted,  
302 population-weighted and AEI summary measures, respectively. The impacts on surface PM<sub>2.5</sub>  
303 in 2030 of additional 30% reductions applied to UK-only terrestrial emissions of each of NH<sub>3</sub>,  
304 NO<sub>x</sub>, SO<sub>x</sub>, VOC and primary PM<sub>2.5</sub> individually are shown in Figure 8. Figure 5 illustrates the  
305 quantitative effect of these further emissions reductions against the 2030 CLE scenario on the  
306 three summary measures of UK-average surface concentrations of PM<sub>2.5</sub>.

307  
308 The maps in Figure 8 show qualitatively very similar findings to their equivalent maps in Figure  
309 4. In 2030, UK PM<sub>2.5</sub> is projected to remain more sensitive to reductions in UK emissions of  
310 NH<sub>3</sub> and primary PM<sub>2.5</sub> than to reductions in UK SO<sub>x</sub> and NO<sub>x</sub>; and, from a population-  
311 weighted perspective, to be relatively more sensitive to further primary PM<sub>2.5</sub> and NH<sub>3</sub>  
312 emissions reductions, particularly to primary PM<sub>2.5</sub> emissions reductions, than was the case for  
313 the 2010 simulations (Figure 5). For the 2030 simulations, additional 30% reductions in UK  
314 primary PM<sub>2.5</sub> or NH<sub>3</sub> emissions yield reductions in PM<sub>2.5</sub> of up to 0.5 µg m<sup>-3</sup> or 0.25 µg m<sup>-3</sup>,  
315 respectively (Figure 8), whilst in 2010 additional 30% reductions in primary PM<sub>2.5</sub> or NH<sub>3</sub>  
316 emissions yield reductions in PM<sub>2.5</sub> of up to 0.6 µg m<sup>-3</sup> or 0.45 µg m<sup>-3</sup>, respectively (Figure 4).  
317 The 2030 results again emphasise a geographic pattern of greatest sensitivity of PM<sub>2.5</sub> to  
318 reductions in the areas of high population density. Figure 6b plots the difference in response to

319 the NH<sub>3</sub> and primary PM<sub>2.5</sub> emissions reductions in 2030, analogous to the plot in Figure 6a for  
320 the 2010 sensitivities. Figure 6b clearly emphasises that for this projection for 2030, UK PM<sub>2.5</sub>  
321 is relatively even more sensitive to further reductions in UK primary PM<sub>2.5</sub> emissions compared  
322 with further reductions in UK NH<sub>3</sub> emissions, particularly in populated areas, than is the case  
323 for 2010; albeit that the additional absolute reductions in PM<sub>2.5</sub> for a given percentage of  
324 emissions reductions is smaller in 2030 than in 2010 (Figure 5) because of the general decline  
325 in emissions across Europe during this period for this scenario.

326

#### 327 **4 Discussion**

328

329 Simulations were undertaken for both 2010 and a 2030 scenario to investigate whether  
330 conclusions on effectiveness of potential UK mitigation differ between the two time points. It  
331 is recognised that reductions in emissions of primary PM<sub>2.5</sub> and precursor gases from many  
332 anthropogenic sources are already anticipated going forward under current legislation, so it is  
333 important to know, for a future policy perspective, the anticipated sensitivities of UK PM<sub>2.5</sub> to  
334 additional UK emission reductions in the future.

335

336 The simulations for both 2010 and 2030 CLE show that if the focus is on the reduction of  
337 spatially-averaged PM<sub>2.5</sub> concentrations then the most effective UK control, via an individual  
338 component, is achieved through reduction of UK emissions of NH<sub>3</sub>, as shown in Figure 5.  
339 However, the conclusion is different when considering population-weighted PM<sub>2.5</sub> reductions  
340 for the mitigation of human health effects. For a full population weighting across all 5 km × 5  
341 km model grids, reductions in UK primary PM<sub>2.5</sub> emissions are almost as effective as reductions  
342 in UK NH<sub>3</sub> emissions for the 2010 simulations, but primary PM<sub>2.5</sub> emissions reductions are  
343 simulated to be the most effective additional control in the 2030 CLE future (Figure 5).  
344 Emphasis on population weighting also increases the sensitivities of PM<sub>2.5</sub> to reductions in NO<sub>x</sub>  
345 emissions in both 2010 and 2030 CLE because a major source of NO<sub>x</sub> is road traffic whose  
346 emissions are associated with where population live. On the other hand, the sensitivity of PM<sub>2.5</sub>  
347 to further reductions in UK SO<sub>x</sub> emissions is markedly lower in 2030 than in 2010 because of  
348 the large reductions in SO<sub>x</sub> emissions already implemented under the CLE scenario. It is also  
349 recognised that reductions in NO<sub>x</sub> and VOC emissions have the potential to deliver health

350 benefits separately from their contribution to reduction in  $PM_{2.5}$  through reductions in  
351 population exposure to surface  $NO_2$  and  $O_3$ .

352

353 An important observation is that the effectiveness of emissions reductions on  $PM_{2.5}$  using a  
354 population weighting for the quantification differs between evaluation via full nation-wide  
355 gridded population-weighting or via use of data only at the locations used to derive the AEI.  
356 Quantification through the AEI puts greater emphasis on the effectiveness of primary  $PM_{2.5}$   
357 emissions reduction, and on  $NO_x$  emissions reductions, (Figure 5) because the monitor locations  
358 contributing to the AEI are sited in the largest cities and towns where emissions of primary  
359  $PM_{2.5}$  and  $NO_x$  are prevalent. Based on the AEI, control of primary  $PM_{2.5}$  is the most effective  
360 individual component in 2010 as well as in 2030 CLE. These observations are pertinent given  
361 that the AEI is the air quality metric for  $PM_{2.5}$ .

362

363 Analyses from the EUCAARI study in Kulmala et al. (2011) and a more recent European study  
364 in Megaritis et al. (2013) both suggest that reducing  $NH_3$  emissions is the most effective way  
365 to reduce  $PM_{2.5}$  under present-day conditions. Whilst the current study also emphasises the  
366 sensitivity of  $PM_{2.5}$  to  $NH_3$  emissions reductions, it also emphasises that, for the UK, a  
367 sensitivity to primary  $PM_{2.5}$  emissions reductions is at least as great as for  $NH_3$  when  
368 considering population-weighting of  $PM_{2.5}$  concentrations, both currently and for a future CLE  
369 scenario. In fact the sensitivity to primary  $PM_{2.5}$  emissions may be underestimated by the  
370 simulations because of dilution of primary  $PM_{2.5}$  emissions into the  $5\text{ km} \times 5\text{ km}$  grid resolution  
371 of the model. It has been calculated that a 1:1 relationship between UK primary  $PM_{2.5}$  emissions  
372 reductions and the reduction in the primary  $PM_{2.5}$  component of the UK 2010 AEI would lead  
373 to a reduction in the 2010 AEI of  $0.8\ \mu\text{g m}^{-3}$  (AQEG, 2015), compared with the  $0.37\ \mu\text{g m}^{-3}$   
374 derived from the model simulations in this work (Table 1). Even so, the total impact of 30%  
375 reductions in UK emissions of all the components/precursors listed in Table 1 on the 2010  
376 baseline, is only of comparable magnitude ( $1.2\ \mu\text{g m}^{-3}$ ) to the 15% (or  $1.3\ \mu\text{g m}^{-3}$ ) reduction  
377 required in the UK AEI by 2020. However, reductions in these emissions from outside the UK  
378 will also contribute to reducing the UK  $PM_{2.5}$  AEI. Conversely, reductions of emissions in the  
379 UK will also yield benefits for surface  $PM_{2.5}$  concentrations elsewhere in Europe. The country-  
380 to-country source-receptor matrices developed by EMEP MSC-W at the 50 km resolution  
381 indicate that reductions in the UK of the same primary and precursor species considered in this

382 work would (for 2011 emissions) lead to reductions in PM<sub>2.5</sub> in neighbouring countries up to  
383 about one-third the magnitude of the PM<sub>2.5</sub> reductions in the UK (Fagerli et al., 2014).  
384 Reductions of emissions in the UK would also lead to other benefits outside the UK on, for  
385 example, NO<sub>2</sub> and O<sub>3</sub> exposure and on N and S deposition.

386

387 Although the model used in this study is widely applied across Europe for air quality policy  
388 development (Fagerli et al., 2014), the data presented here are from simulations from a single  
389 model. The model simulations of the effect of inorganic precursor gases on the secondary  
390 inorganic PM<sub>2.5</sub> are dependent on accurate representation of the relevant chemistry and phase  
391 partitioning. It is possible that the SIA representation in the EMEP4UK model may  
392 underestimate the nitrate in the PM<sub>2.5</sub> size fraction, and hence downplay somewhat the  
393 sensitivity of PM<sub>2.5</sub> to NO<sub>x</sub> emissions reductions. In addition, not explicitly calculating the  
394 uptake of HNO<sub>3</sub> by mineral dust may reduce the NO<sub>3</sub><sup>-</sup> changes due to NO<sub>x</sub> emissions reduction.  
395 However, the EMEP4UK particle sulphate, nitrate and ammonium concentrations all compare  
396 well with the multi-year time series of measurements of these components at ~30 sites across  
397 the UK in the Acid Gas and Aerosol Network (AGANet) and National Ammonia Monitoring  
398 Network (NAMN) (Vieno et al., 2014). Variation in particle-bound water may also impact on  
399 the exact PM<sub>2.5</sub> mass sensitivities associated with inorganic precursor gas emissions reductions.

400

401 Inter-annual variability in meteorology may also have an influence, in particular in determining  
402 the balance in any year between PM<sub>2.5</sub> in the UK derived from UK emissions and that derived  
403 from emissions outside the UK (Vieno et al., 2014). However, whilst the precise quantitative  
404 sensitivities of annual average PM<sub>2.5</sub> to emissions reductions will be subject to inter-annual  
405 meteorological variability, it is anticipated that the broad findings of this study will hold.

406

407 The interpretation of the modelling results has been undertaken from the perspective that  
408 reduction in all anthropogenically-derived components of PM<sub>2.5</sub> is equally important. This  
409 remains the current position for the EU legislation that sets limits and targets for concentrations  
410 of PM<sub>2.5</sub> (Heal et al., 2012); i.e. no consideration is given to the potential different toxicity to  
411 human health of different components of PM<sub>2.5</sub>. The UK Committee on the Medical Effects of  
412 Air Pollutants has also recently concluded that reductions in concentrations of both primary and  
413 secondary particles are likely to benefit public health (COMEAP, 2015). Nevertheless, although

414 not conclusive, there is evidence that traffic-related sources of PM, or combustion sources more  
415 generally, are particularly associated with adverse health outcomes (Grahame and Schlesinger,  
416 2007, 2010;Janssen et al., 2011;Stanek et al., 2011;WHO, 2013;Grahame et al., 2014). The  
417 possibility that primary PM<sub>2.5</sub> is more toxic per unit mass than secondary PM<sub>2.5</sub>, places greater  
418 emphasis on the finding from this work on the effectiveness of reductions in emissions of  
419 primary PM<sub>2.5</sub>. Interpretation of the modelling results has also not considered the relative costs  
420 or feasibilities of implementing further reductions in the emissions of the individual precursors  
421 and components investigated.

422

423 Finally, it should be remembered that measures taken in the UK to reduce concentrations of  
424 ambient PM<sub>2.5</sub> and of precursor gases, both within and outside of populated areas, will have  
425 multiple co-benefits on human health, N and S deposition, ozone formation and radiative  
426 forcing, not just in the UK but elsewhere.

427

## 428 **5 Conclusions**

429

430 The sensitivity of annual-average surface concentrations of PM<sub>2.5</sub> across the UK to reductions  
431 in UK terrestrial anthropogenic emissions in primary PM<sub>2.5</sub>, NH<sub>3</sub>, NO<sub>x</sub>, SO<sub>x</sub> and non-methane  
432 VOC was investigated using the EMEP4UK atmospheric chemistry transport model for 2010  
433 and for a 2030 current legislation scenario that includes projected pan-European emission  
434 changes. In general, the sensitivity of modelled concentrations to UK-only mitigation is rather  
435 small. A 30% reduction in UK emissions of any one of the above listed PM components yields  
436 (for the 2010 simulation) a maximum reduction in PM<sub>2.5</sub> concentrations in any given location  
437 of ~0.6 µg m<sup>-3</sup> (equivalent to ~6% of the total modelled PM<sub>2.5</sub> mass concentration). On average  
438 across the UK, the sensitivity of PM<sub>2.5</sub> concentrations to a 30% reduction in UK emissions of  
439 individual contributing components, for both the 2010 and 2030 CLE baselines, increases in  
440 the order NMVOC, NO<sub>x</sub>, SO<sub>x</sub>, NH<sub>3</sub> and primary PM<sub>2.5</sub>, but there are strong spatial differences  
441 in the PM<sub>2.5</sub> sensitivities across the UK. Consequently, the sensitivity of PM<sub>2.5</sub> to individual  
442 component emissions reductions varies between area and population weighting. Reductions in  
443 NH<sub>3</sub> have the greatest area-weighted effect on PM<sub>2.5</sub>. A full UK population weighting places  
444 greater emphasis on reductions of primary PM<sub>2.5</sub> emissions, which is simulated to be the most  
445 effective single-component control on PM<sub>2.5</sub> for the 2030 scenario. An important observation

446 is that weighting corresponding to the Average Exposure Indicator metric (using data from the  
447 45 model grids containing a monitor whose measurements are used to calculate the UK AEI)  
448 further increases the emphasis on the effectiveness of primary PM<sub>2.5</sub> emissions reductions (and  
449 of NO<sub>x</sub> emissions reductions) relative to the effectiveness of NH<sub>3</sub> emissions reductions.  
450 Reductions in primary PM<sub>2.5</sub> has the largest impact on the AEI in 2010 as well as the 2030 CLE  
451 scenario. The summation of the reductions to the UK PM<sub>2.5</sub> AEI of the 30% reductions in UK  
452 emissions of primary PM<sub>2.5</sub> and of NH<sub>3</sub>, SO<sub>x</sub>, NO<sub>x</sub> and VOC totals ~1.2 µg m<sup>-3</sup> and ~0.8 µg m<sup>-3</sup>  
453 <sup>3</sup> with respect to the 2010 and 2030 CLE baselines, respectively (not accounting for non-  
454 linearity).

455

456

457

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464

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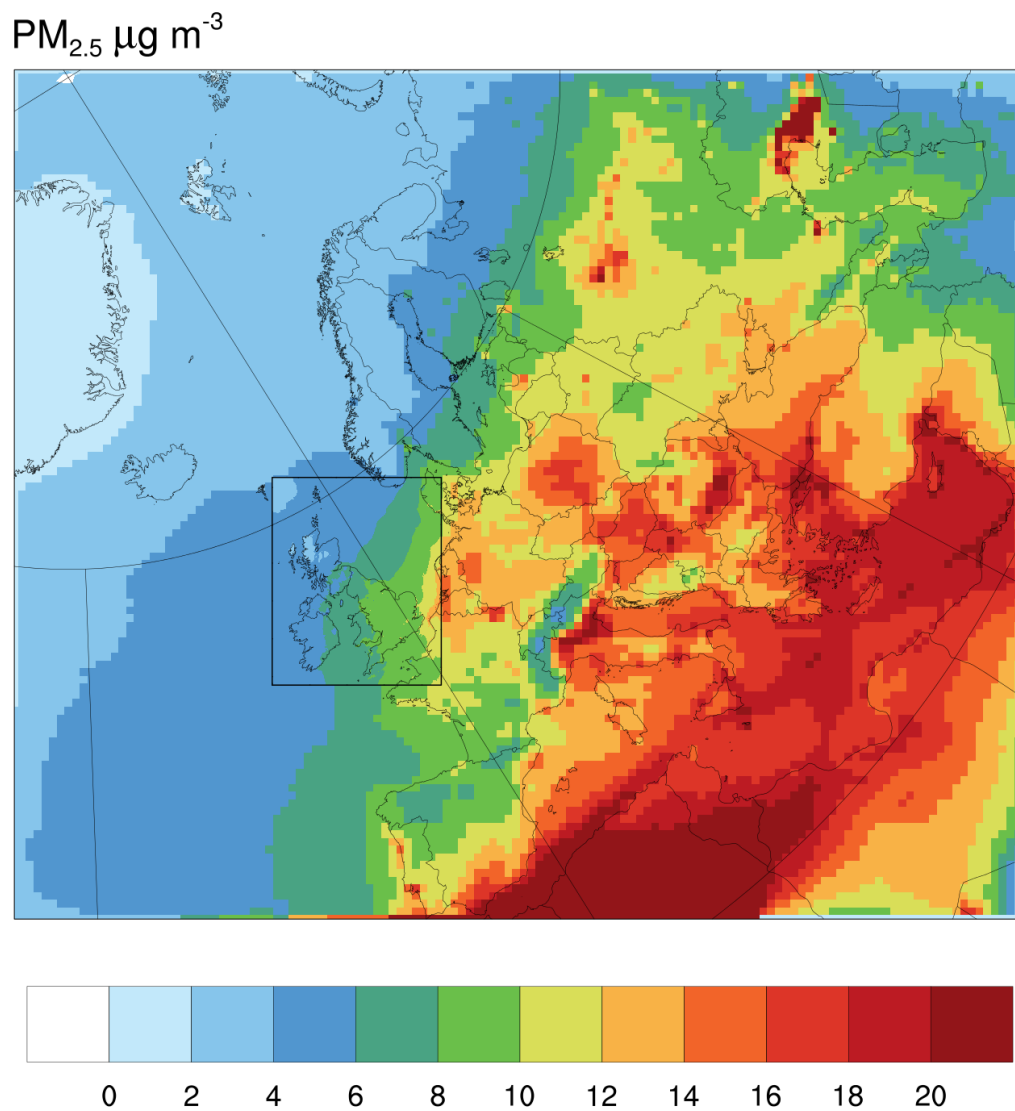
654 **Table:**

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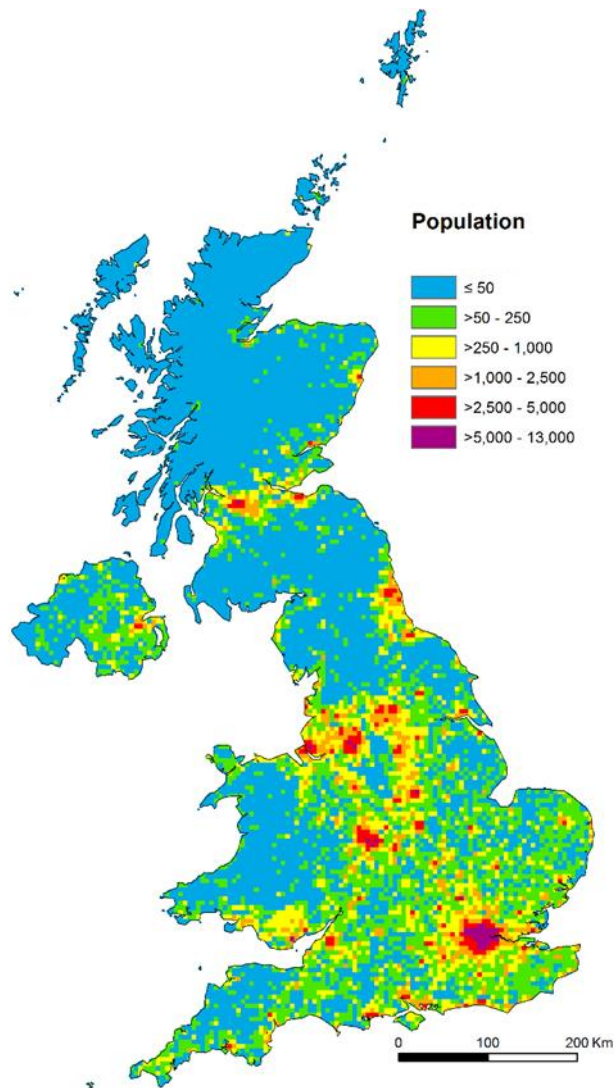
657 Table 1: EMEP4UK-modelled estimates of the impact of 30% UK terrestrial emissions  
658 reductions on three measures of UK-average surface concentrations of PM<sub>2.5</sub> (µg m<sup>-3</sup>): (i) the  
659 average of the model grids containing the 45 monitors used to calculate the UK PM<sub>2.5</sub> Average  
660 Exposure Indicator (AEI), (ii) the population-weighted average, and (iii) the area-weighted (i.e.  
661 geographical) average, for 2010, and for 2030 under a CLE emission scenario (using 2010  
662 meteorology). For context, the modelled reductions in the baselines between 2010 and 2030  
663 CLE for the three measures of UK-average PM<sub>2.5</sub> are 2.42, 2.24, and 1.70 µg m<sup>-3</sup>, respectively.  
664

Emissions reduced	AEI		Population-weighted		Area-weighted	
	2010	2030 CLE	2010	2030 CLE	2010	2030 CLE
Primary PM <sub>2.5</sub>	0.37	0.29	0.31	0.24	0.16	0.13
NH <sub>3</sub>	0.35	0.19	0.34	0.19	0.28	0.16
SO <sub>x</sub>	0.27	0.15	0.26	0.15	0.19	0.11
NO <sub>x</sub>	0.10	0.14	0.10	0.15	0.11	0.13
VOC	0.08	0.05	0.08	0.05	0.07	0.03
<b>Total</b>	<b>1.17</b>	<b>0.82</b>	<b>1.10</b>	<b>0.77</b>	<b>0.82</b>	<b>0.57</b>



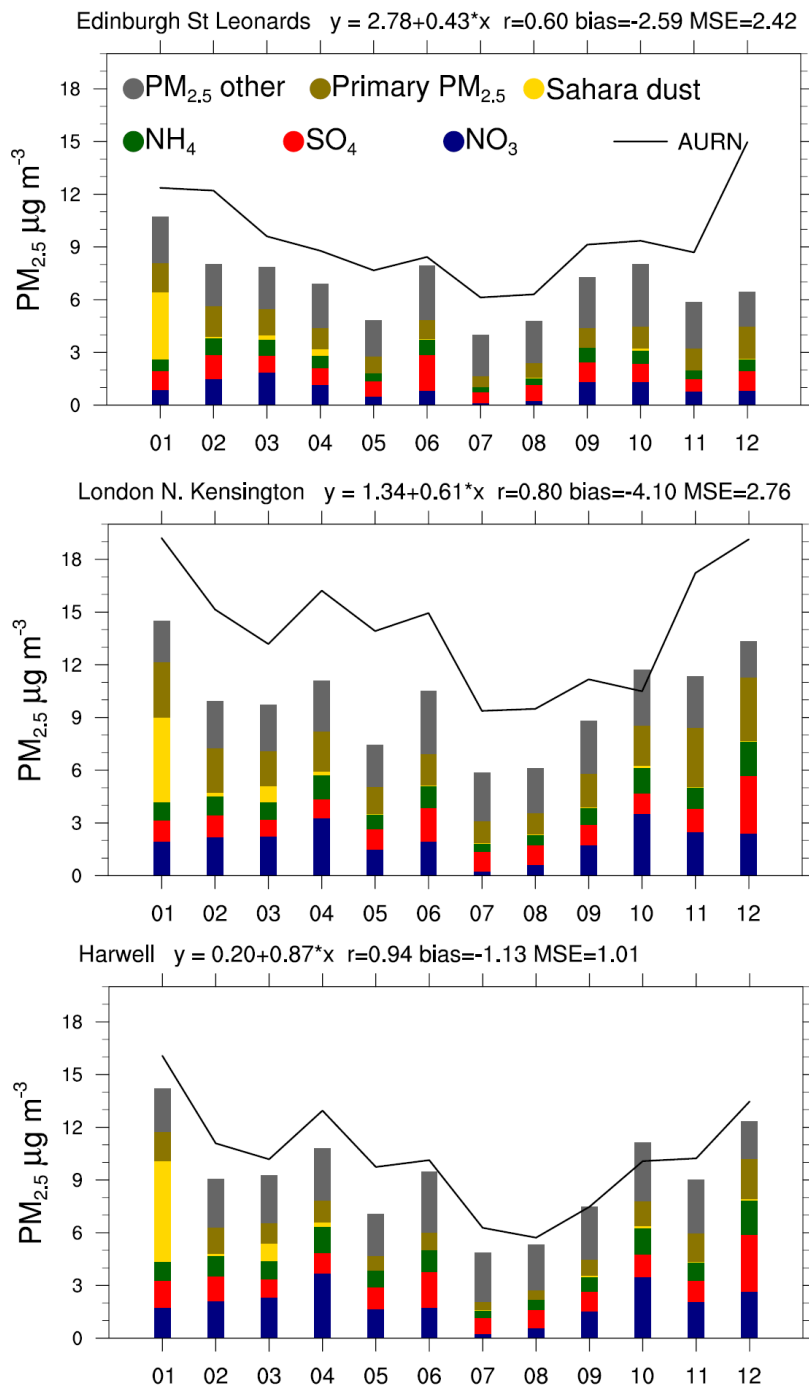
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666 Figure 1: 2010 EMEP4UK annual-average surface concentrations of PM<sub>2.5</sub> ( $\mu\text{g m}^{-3}$ ) at 50 km ×  
667 50 km horizontal resolution for the European model domain, and at 5 km × 5 km horizontal  
668 resolution for the nested British Isles domain (black box).



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670 Figure 2: Gridded UK population density based on the UK census at the 5 km × 5 km grid  
 671 spatial resolution. Units are population km<sup>-2</sup>.

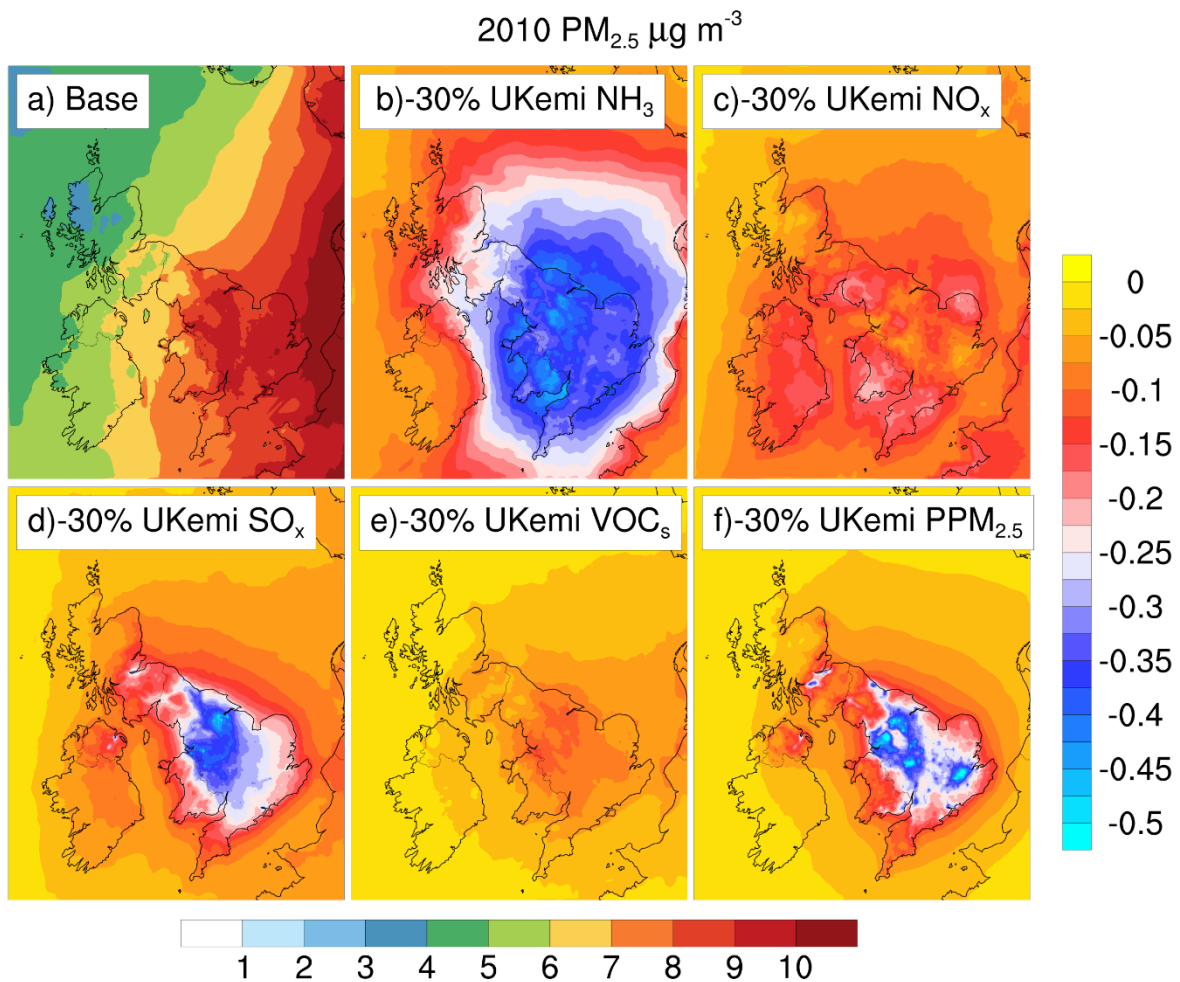


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673 Figure 3: 2010 monthly-averaged EMEP4UK simulated PM<sub>2.5</sub> components and total PM<sub>2.5</sub>  
 674 observations by TEOM-FDMS at the Edinburgh St. Leonards, London North Kensington and  
 675 Harwell UK national network (AURN) monitoring sites. Both the modelled and observed data  
 676 are averaged from hourly values. The linear regression between the **monthly averaged**  
 677 observation and model is also shown at the top of each panel, along with the correlation  
 678 coefficient, r, bias and mean square error.



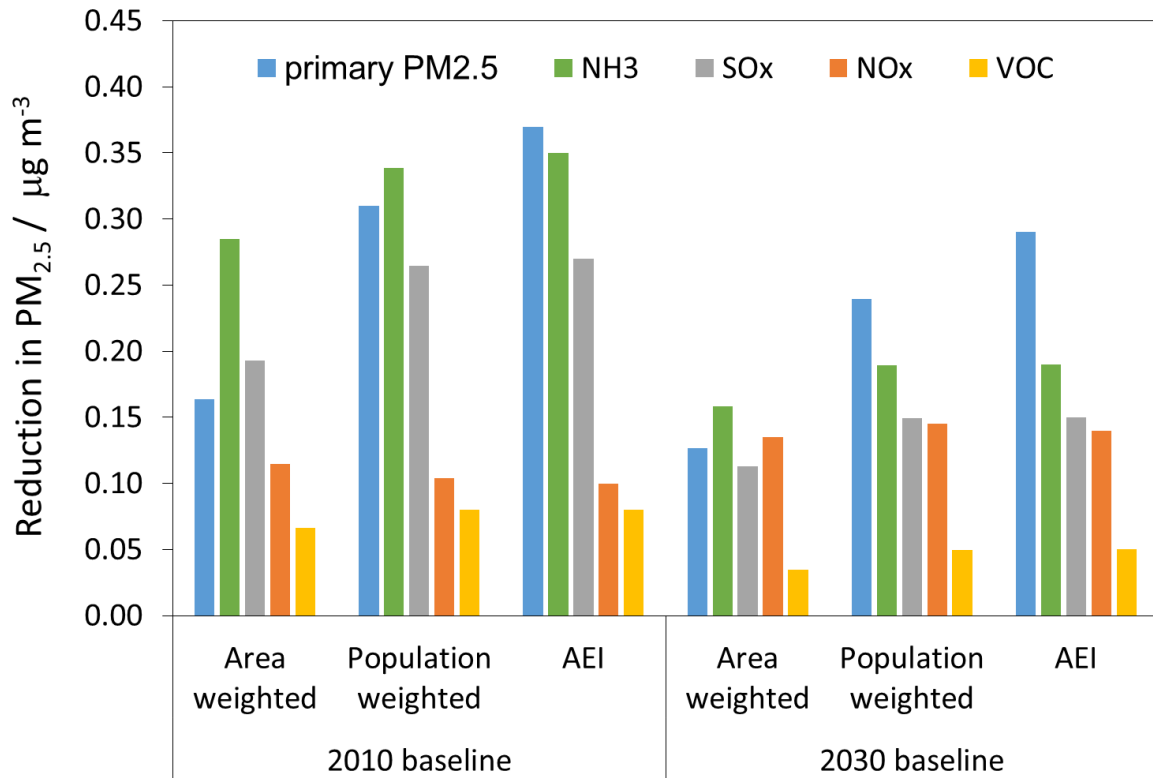
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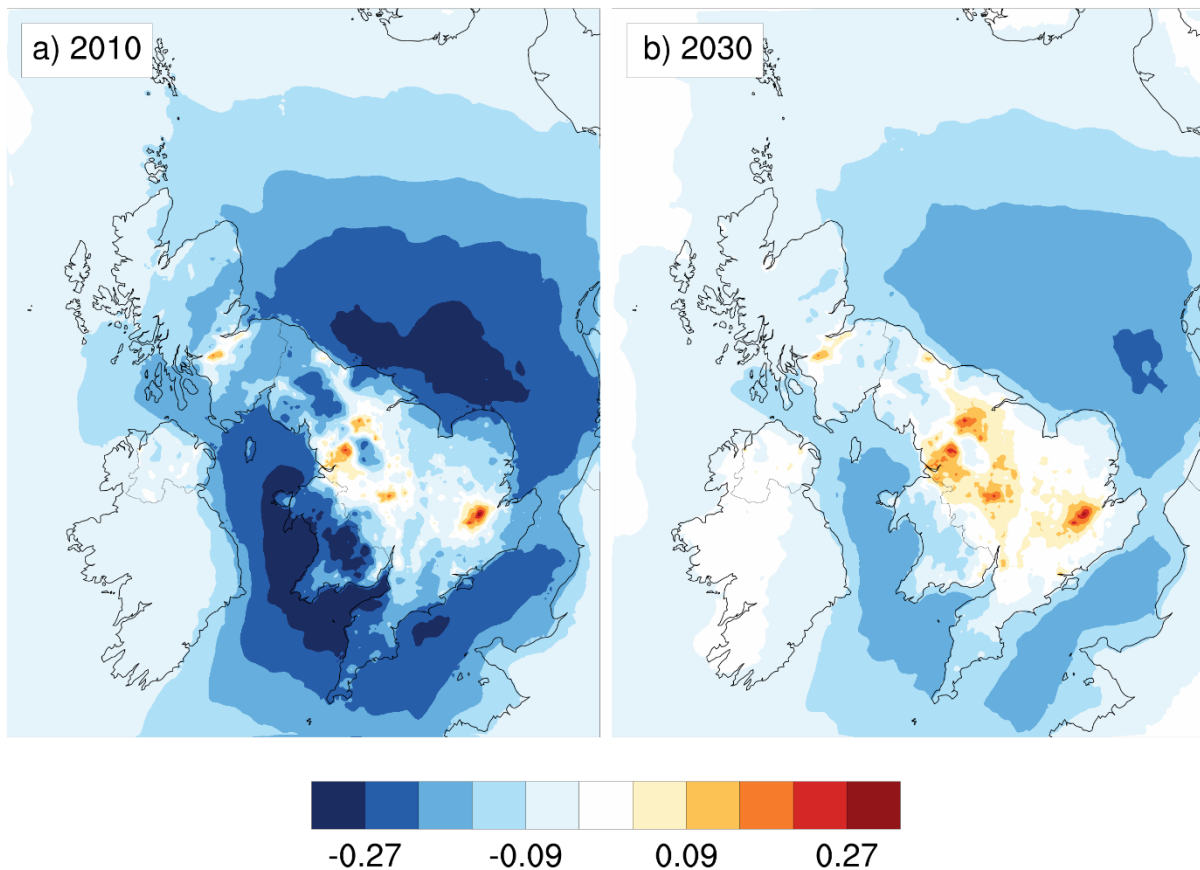
Figure 4: Model simulations of the impact of 30% UK emissions reductions on annual-average surface concentration of PM<sub>2.5</sub>. Panel (a) 2010 base-case scenario, no emissions reduction (bottom colour scale); remaining panels, the change in annual-average PM<sub>2.5</sub> for 30% UK emissions reductions in (b) NH<sub>3</sub>, (c) NO<sub>x</sub>, (d) SO<sub>x</sub>, (e) VOC, and (f) primary PM<sub>2.5</sub> (right colour scale). All units are  $\mu\text{g m}^{-3}$ .

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Figure 5: The impact of 30% UK terrestrial emissions reductions in primary PM<sub>2.5</sub>, NH<sub>3</sub>, SO<sub>x</sub>, NO<sub>x</sub>, and VOC (individually) on three measures of UK-average surface concentrations of PM<sub>2.5</sub>: area weighted; population weighted; and the average for the 45 model grids containing the monitors used to calculate the UK PM<sub>2.5</sub> Average Exposure Indicator (AEI). Data are shown for simulations for 2010, and for 2030 under a CLE emission scenario (using 2010 meteorology).

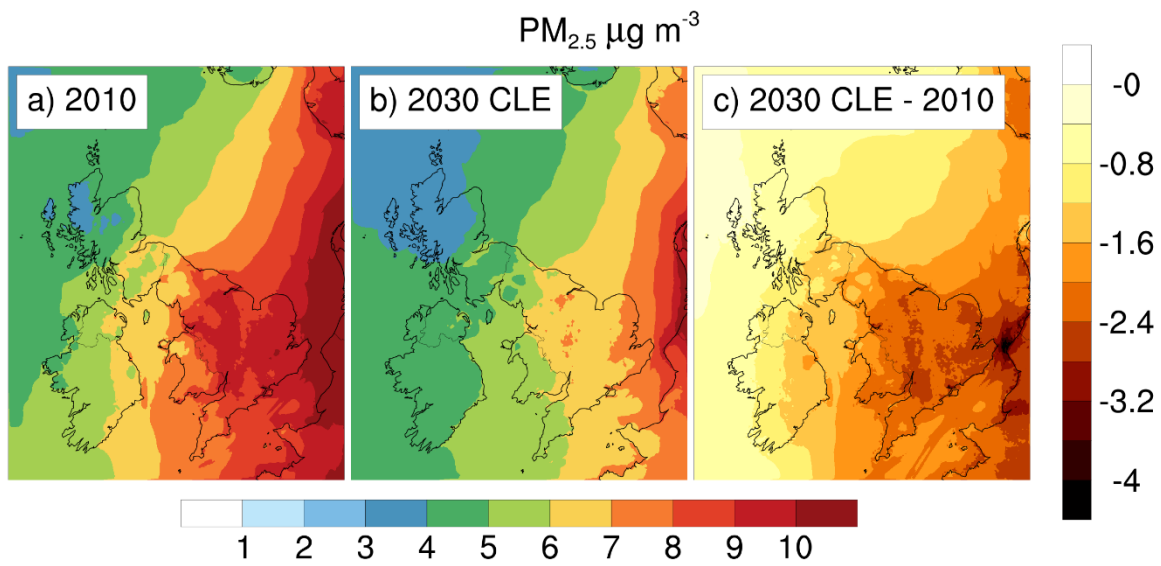


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703 Figure 6: The difference between changes in simulated annual-average PM<sub>2.5</sub> (µg m<sup>-3</sup>) for 30%  
 704 reductions in UK NH<sub>3</sub> emissions reduction and for 30% reductions in UK primary PM<sub>2.5</sub>  
 705 emissions reduction: (a) for the year 2010 (i.e. the data in Figure 4b minus the data in Figure  
 706 4f); and (b) for the year 2030 (i.e. the data in Figure 8b minus the data in Figure 8f). Blue  
 707 colours indicate where reductions in PM<sub>2.5</sub> for 30% reduction in NH<sub>3</sub> emissions exceed the  
 708 reductions in PM<sub>2.5</sub> for 30% reduction in primary PM<sub>2.5</sub> emissions, and vice versa for the red  
 709 colours. The same meteorological year 2010 was used.

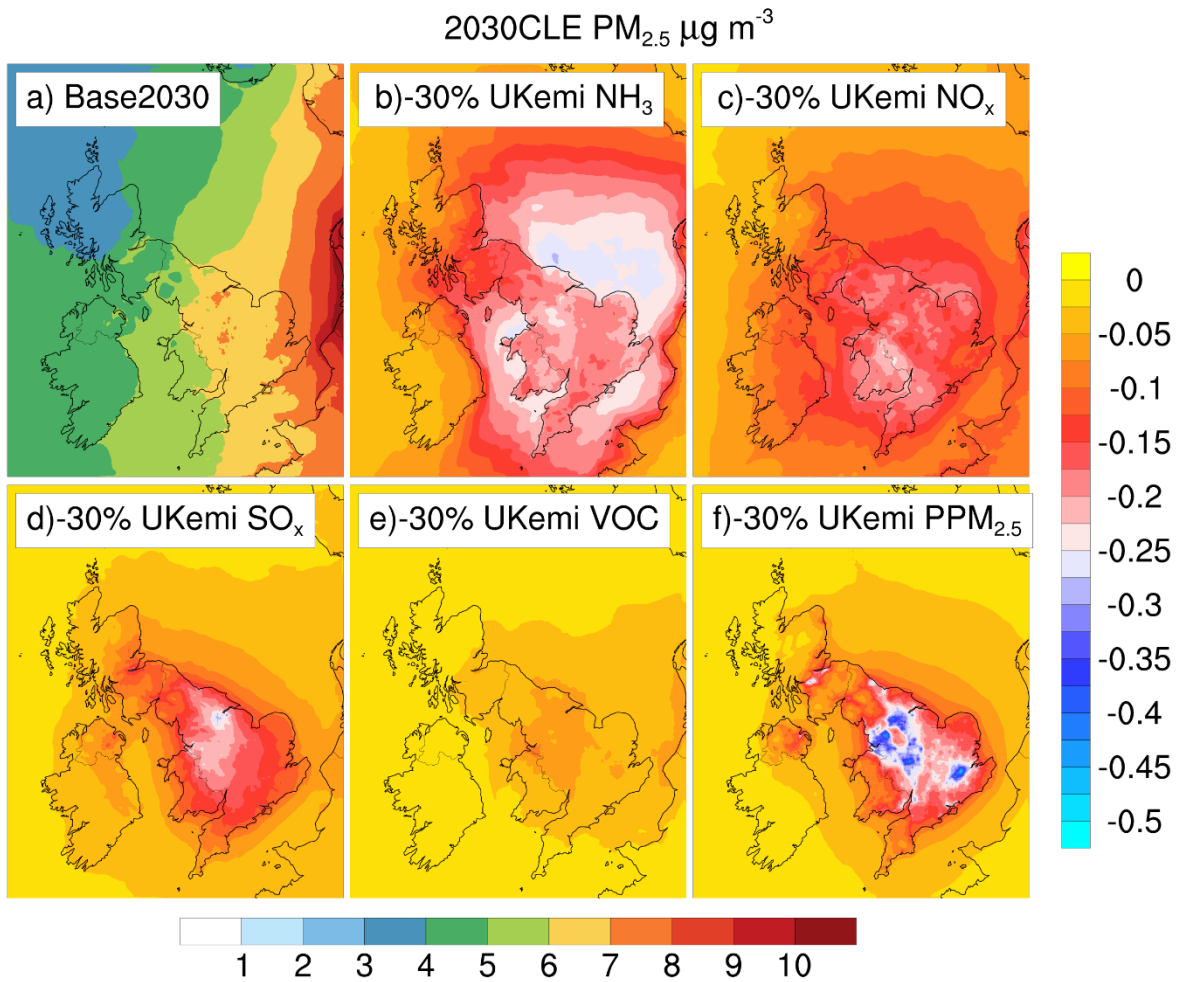
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713 Figure 7: EMEP4UK annual-average surface concentration of  $\text{PM}_{2.5}$  ( $\mu\text{g m}^{-3}$ ) for a) 2010  
 714 emissions, and b) 2030 CLE emissions projection (bottom colour scale), and c) the difference  
 715 2030 CLE – 2010 CLE (right colour scale). The same meteorological year 2010 was used.



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718 Figure 8: Model simulations of impact of 30% UK emissions reductions on annual-average  
 719 surface concentration of PM<sub>2.5</sub> for a future scenario (with 2010 meteorology). Panel (a), 2030  
 720 CLE scenario, no emissions reduction (bottom colour scale); remaining panels, the change in  
 721 annual-average PM<sub>2.5</sub> for 30% UK emissions reductions in (b) NH<sub>3</sub>, (c) NO<sub>x</sub>, (d) SO<sub>x</sub>, (e) VOC,  
 722 and (f) primary PM<sub>2.5</sub> (right colour scale). All units are  $\mu\text{g m}^{-3}$ .