The sensitivities of emissions reductions for the mitigation of UK PM$_{2.5}$

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Abstract. The reduction of ambient concentrations of fine particulate matter (PM$_{2.5}$) is a key objective for air pollution control policies in the UK and elsewhere. Long-term exposure to PM$_{2.5}$ has been identified as a major contributor to adverse human health effects in epidemiological studies and underpins ambient PM$_{2.5}$ legislation. As a range of emission sources and atmospheric chemistry transport processes contribute to PM$_{2.5}$ concentrations, atmospheric chemistry transport models are an essential tool to assess emissions control effectiveness. The EMEP4UK atmospheric chemistry transport model was used to investigate the impact of reductions in UK anthropogenic emissions of primary PM$_{2.5}$, NH$_3$, NO$_x$, SO$_x$ or non-methane VOC on surface concentrations of PM$_{2.5}$ in the UK for a recent year (2010) and for a future current legislation emission (CLE) scenario (2030). In general, the sensitivity to UK mitigation is rather small. A 30 % reduction in UK emissions of any one of the above components yields (for the 2010 simulation) a maximum reduction in PM$_{2.5}$ in any given location of $\sim 0.6\,\mu g\,m^{-3}$ (equivalent to ~ 6 % of the modelled PM$_{2.5}$). On average across the UK, the sensitivity of PM$_{2.5}$ concentrations to a 30 % reduction in UK emissions of individual contributing components, for both the 2010 and 2030 CLE baselines, increases in the order NMVOC, NO$_x$, SO$_x$, NH$_3$ and primary PM$_{2.5}$; however there are strong spatial differences in the PM$_{2.5}$ sensitivities across the UK. Consequently, the sensitivity of PM$_{2.5}$ to individual component emissions reductions varies between area and population weighting. Reductions in NH$_3$ have the greatest effect on area-weighted PM$_{2.5}$. A full UK population weighting places greater emphasis on reductions of primary PM$_{2.5}$ emissions, which is simulated to be the most effective single-component control on PM$_{2.5}$ for the 2030 scenario. An important conclusion is that weighting corresponding to the average exposure indicator metric (using data from the 45 model grids containing a monitor whose measurements are used to calculate the UK AEI) further increases the emphasis on the effectiveness of primary PM$_{2.5}$ emissions reductions (and of NO$_x$ emissions reductions) relative to the effectiveness of NH$_3$ emissions reductions. Reductions in primary PM$_{2.5}$ have the largest impact on the AEI in both 2010 and the 2030 CLE scenario. The summation of the modelled reductions to the UK PM$_{2.5}$ AEI from 30 % reductions in UK emissions of primary PM$_{2.5}$, NH$_3$, SO$_x$, NO$_x$ and VOC totals 1.17 and 0.82 $\mu g\,m^{-3}$ for the 2010 and 2030 CLE simulations, respectively (not accounting for non-linearity).

1 Introduction

Atmospheric particulate matter (PM) has a range of adverse impacts including on climate change through radiative forcing (IPCC, 2013) and on human health (WHO, 2006, 2013). The global health burden from exposure to ground-level ambient fine particulate matter (as characterized by the PM$_{2.5}$ metric) is substantial. The Global Burden of Disease project attributed 3.2 million premature deaths and 76 million disability-adjusted life years to exposure to ambient PM$_{2.5}$ concentrations prevailing in 2005 (Lim et al., 2012).

Exposure to ambient PM$_{2.5}$ remains a major health issue in Europe. The European Environment Agency report that for the period 2010–2012, 10–14 % of the urban population in the EU28 countries were exposed to ambient concentrations of PM$_{2.5}$ exceeding the EU annual-average PM$_{2.5}$ reference value of 25 µg m^{-3}, but 91–93 % were exposed to concentrations exceeding the WHO annual-average PM$_{2.5}$ air quality guideline of 10 µg m^{-3} (EEA, 2014).

European Commission (EC) legislation for PM$_{2.5}$ includes an obligation on individual member states to reduce exposure to PM$_{2.5}$ in areas of population by a prescribed percentage between 2010 and 2020. The exposure to PM$_{2.5}$ is quantified through the average exposure indicator (AEI) which is the average of the annual PM$_{2.5}$ measured across designated urban background and suburban sites spread over cities and large towns (averaged over the 3-year periods spanning 2010 and 2020). The AEI is therefore a quasi-indicator of population-weighted PM$_{2.5}$. For the UK, the calculation of the AEI uses data from 45 sites (Brookes et al., 2012) and the required reduction by 2020 is 15 % from its 2010 value of 13 µg m^{-3} (Defra, 2012).

While standards focus on PM$_{2.5}$ mass concentrations, meeting these standards is complicated by the considerable chemical heterogeneity, which arises because ambient PM$_{2.5}$ comprises both primary PM emissions and secondary inorganic and organic components formed within the atmosphere from gaseous precursor emissions, specifically NH$_3$, NO$_x$ (NO and NO$_2$), SO$_2$, and a wide range of non-methane volatile organic compounds (VOC) (USEPA, 2009; AQEG, 2015). Meteorological conditions also control PM$_{2.5}$ concentrations through their influences on dispersion, chemistry, and deposition.

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

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

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

2 Methods

2.1 Model description and set-up

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

The EMEP4UK model meteorological driver is the Weather Research and Forecast (WRF) model version 3.1.1 (www.wrf-model.org). The EMEP4UK and WRF model horizontal resolution is 50 km × 50 km for the extended European domain and 5 km × 5 km for the inner domain as illustrated in Fig. 1. The EMEP4UK model uses a nested approach, the European domain concentrations are used as boundary condition for the UK domain. The boundary conditions at the edge of the European domain are prescribed concentrations in terms of latitude and adjusted for each year. For ozone, 3-D fields for the whole domain are specified from climatological ozone-sonde data sets, modified monthly against clean-air surface observations as described in Simpson et al. (2012).

The default EMEP MSC-W chemical scheme was used for the present study, as it has been extensively validated at the European scale (Simpson et al., 2012, www.emep.int). The scheme has 72 species and 137 reactions, and full details are given in Simpson et al. (2012). The gas/aerosol partitioning is the model for aerosols reacting system (MARS) formulation (Simpson et al., 2012). In the model version used here, PM$_{2.5}$ is the sum of the fine (PM$_{2.5}$) fraction of ammonium (NH$_4^+$), sulphate (SO$_4^{2-}$), nitrate (NO$_3^-$), elemental carbon (EC), organic matter (OM), sea salt (SS), mineral dust, and
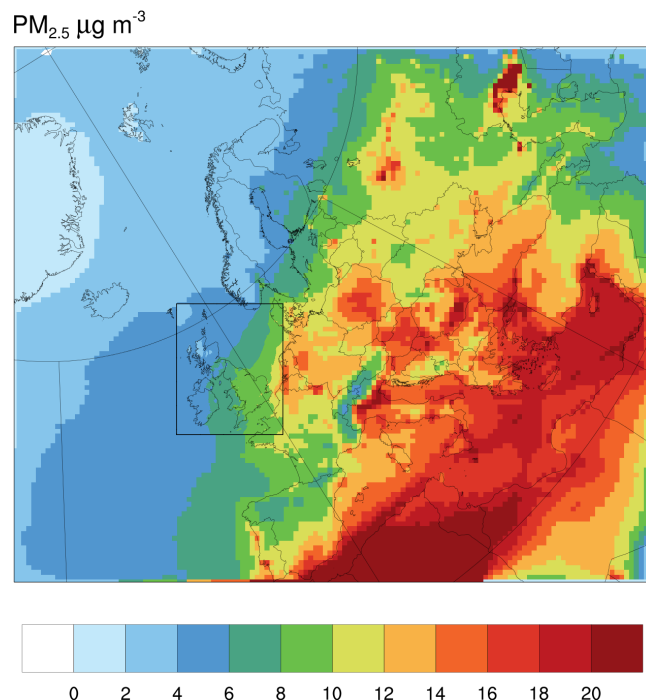
PM$_{2.5}$ µg m^{-3}

Figure 1. 2010 EMEP4UK annual-average surface concentrations of PM$_{2.5}$ (µg m^{-3}) at 50 km × 50 km horizontal resolution for the European model domain, and at 5 km × 5 km horizontal resolution for the nested Great Britain and Ireland domain (black box).

27 % of the coarse nitrate. PM$_{10}$ is the sum of PM$_{2.5}$ plus the coarse (PM$_{2.5-10}$) fraction of EC, OM, NO$_3^-$, SS, and dust.

Whilst fine nitrate production is modelled using a thermodynamic model (MARS), the formation of coarse nitrate from nitric acid (HNO$_3$) uses a parameterized approach that seeks to capture the HNO$_3$ reaction with sea salt and crustal material. The conversion rate of HNO$_3$ to coarse nitrate depends on relative humidity, as described by Simpson et al. (2012), but is not explicitly linked to the surface area of the existing coarse aerosol. Both nitrate generation mechanisms compete for the same HNO$_3$, and whilst this constrains the total amount of nitrate produced, it is acknowledged that the resulting split into fine and coarse nitrate is somewhat uncertain as discussed in Aas et al. (2012). A more explicit aerosol scheme is under development for the model.

Anthropogenic emissions of NO$_x$, NH$_3$, SO$_2$, primary PM$_{2.5}$, primary PM$_{coarse}$, CO, and non-methane VOC for the UK are derived from the National Atmospheric Emission Inventory (NAEI, http://naei.defra.gov.uk) at 1 km × 1 km resolution and aggregated to 5 km × 5 km resolution. For the European domain, the model uses the EMEP 50 km × 50 km resolution emission estimates provided by the Centre for Emission Inventories and Projections (CEIP, http://www.ceip.at/). Shipping emissions estimates, for the inner domain, are derived from the ENTEC (now Amec Foster Wheeler)

emissions estimate (ENTEC, 2010). Natural emissions of isoprene and DMS are as described in Simpson et al. (2012).

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

2.2 Model experiments

A base run and a set of 5 sensitivity experiments were carried out for emissions and meteorology for 2010. The experiments applied 30 % reductions to UK anthropogenic emissions from all sectors for each of the following pollutants individually: primary PM$_{2.5}$, NH$_3$, NO$_x$, SO$_x$ and NMVOC. This 30 % perturbation was applied to land-based emissions only; shipping emissions (both domestic and international) were left unchanged.

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

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

As well as maps of annual-average surface PM$_{2.5}$ concentrations the following three summary statistics for UK PM$_{2.5}$ were calculated: (i) the area-weighted average, i.e. the average of all 5 km × 5 km model grids over the UK; (ii) the population-weighted average, i.e. the 5 km × 5 km gridded estimates of PM$_{2.5}$ surface concentrations re-projected onto the British National Grid and multiplied by population es-

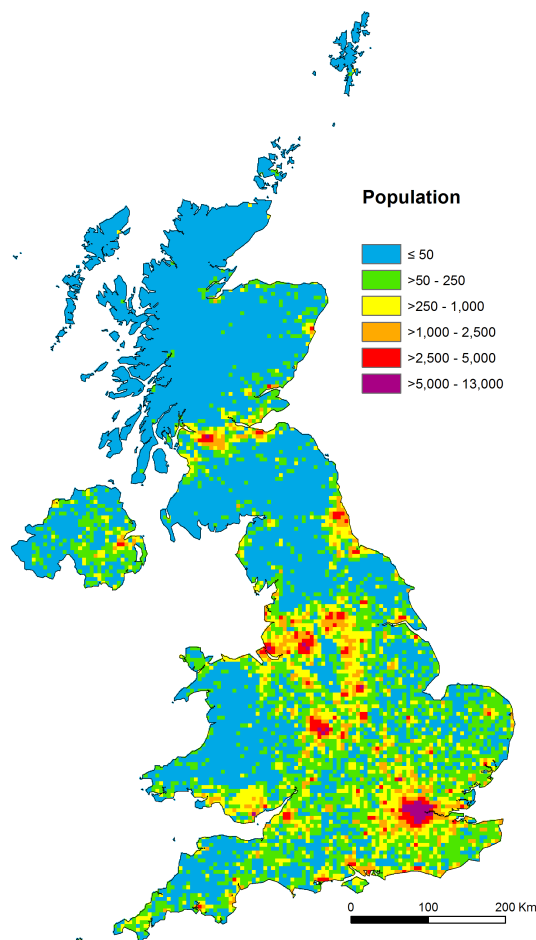**Figure 2.** Gridded UK population density based on the UK census at the 5 km × 5 km grid spatial resolution. Units are population km^{-2}.

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

timates at the same spatial resolution (derived from the UK census, http://census.edina.ac.uk/) (Fig. 2) and divided by the sum of the UK population; (iii) a value analogous to the average exposure indicator (AEI), calculated as the average of the concentrations for the 45 model grids containing a PM$_{2.5}$ monitor whose measurements are used to define the UK's 2010 AEI value (Brookes et al., 2012).

3 Results

Example comparisons between EMEP4UK-modelled surface concentrations of PM$_{2.5}$ components and total measured PM$_{2.5}$ are shown in Fig. 3 for three UK national network monitoring sites: Edinburgh St. Leonards, an urban background site in the north of the UK; London North Kensington, an urban background site in central London in the south-east of the UK; and Harwell, a rural background site in central England. Monthly averages of the hourly measured and modelled data are presented. Model simulations follow
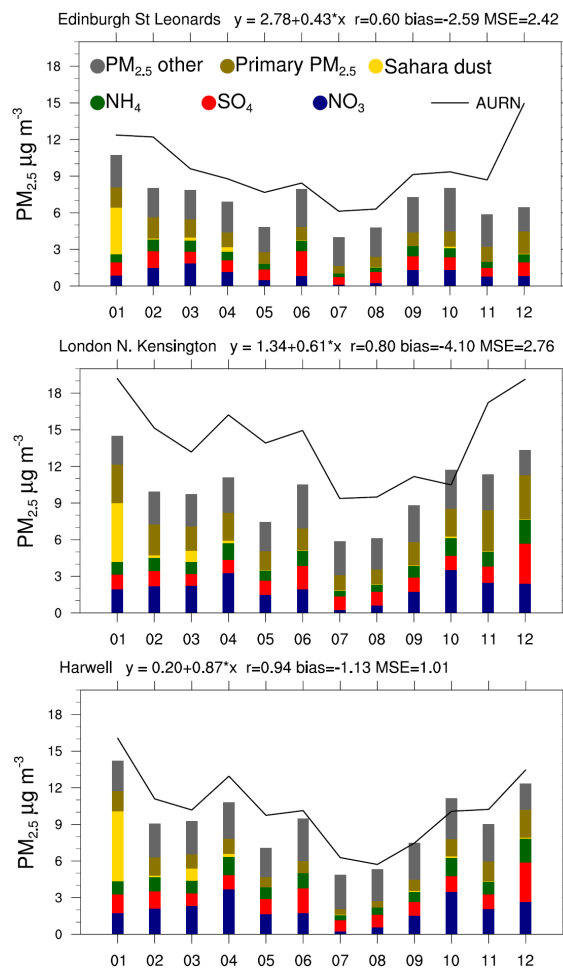
the observational time trends well. The model simulations of the SIA components SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$ have previously been individually evaluated by Vieno et al. (2014) against 10 years of speciated observations made at ∼ 30 sites across the UK in the AGANET network (Conolly et al., 2011). The four UK sites included in Vieno et al. (2014) showed good agreement between the monthly averaged EMEP4UK simulation and the observed NO$_3^-$ and SO$_4^{2-}$, with a bias range of 0.28 to −0.62 and 0.8 to −0.27 µg m^{-3}, respectively. The EMEP4UK model was also evaluated against observations and other models in a UK model inter-comparison organized by the UK Department for Environment, Food & Rural Affairs (Defra) (Carslaw, 2011a, b). The persistent negative bias in the sum of the modelled PM$_{2.5}$ against observation in
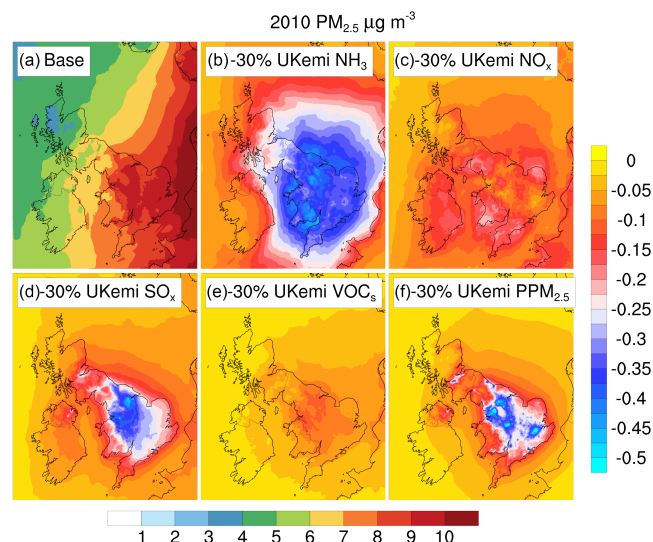
Figure 4. Model simulations of the impact of 30 % UK emissions reductions on annual-average surface concentration of PM$_{2.5}$. Panel **(a)** 2010 base-case scenario, no emissions reduction (bottom colour scale); remaining panels, the change in annual-average PM$_{2.5}$ for 30 % UK emissions reductions in **(b)** NH$_3$, **(c)** NO$_x$, **(d)** SO$_x$, **(e)** VOC, and **(f)** primary PM$_{2.5}$ (right colour scale). All units are μg m^{-3}.

Figure 5. The impact of 30 % UK terrestrial emissions reductions in primary PM$_{2.5}$, NH$_3$, SO$_x$, NO$_x$, and VOC (individually) on three measures of UK-average surface concentrations of PM$_{2.5}$: area weighted; population weighted; and the average for the 45 model grids containing the monitors used to calculate the UK PM$_{2.5}$ average exposure indicator (AEI). Data are shown for simulations for 2010, and for 2030 under a CLE emission scenario (using 2010 meteorology).

Fig. 3 is consistent with the absence of re-suspended dust in the model configuration used here, and possibly also reflects a difference in the treatment of particle-bound water in model and measurement. The omission of re-suspended dust does not impact on the investigations here of the sensitivities of PM$_{2.5}$ concentrations to anthropogenic emissions reductions; however it is acknowledged that since particle-bound water is related to the mass of secondary inorganic components its omission will have some impact on the sensitivity of PM$_{2.5}$ to inorganic precursor gas emissions reductions. Different measurement techniques and conditions incorporate different proportions of the ambient PM$_{2.5}$ water content. We focus here on changes to the dry mass concentrations of surface PM$_{2.5}$ derived from changes in the emissions of primary PM$_{2.5}$ and secondary PM$_{2.5}$ precursor gases. (It is also noted that values of relative reductions in modelled PM$_{2.5}$ will be slightly higher than if expressed relative to measured PM$_{2.5}$ at that location). Some model underestimation may also derive from dilution of primary PM$_{2.5}$ emissions into the 5 km grid of the model compared with the primary emissions more local to an urban background monitor.

The simulated "baseline" 2010 annual-average surface concentrations for PM$_{2.5}$ at 50 km horizontal resolution for the EMEP4UK European domain and for the nested 5 km horizontal resolution Great Britain and Ireland domain are shown in Fig. 1. The UK 2010 annual-average surface concentrations of PM$_{2.5}$ are generally lower compared with neighbouring continental countries such as France, the
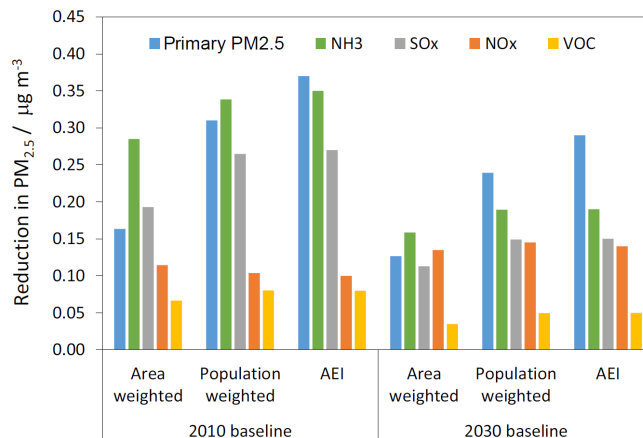
Netherlands, and Germany. The influence of emissions originating from continental Europe is revealed by the gradient of decreasing PM$_{2.5}$ concentrations away from the continent. An analysis presented in AQEG (2015) also using the EMEP4UK model showed that UK emissions contribute around 55 % of the total PM$_{2.5}$ in the UK. This limits the extent to which long-term average concentrations can be reduced by UK action alone.

Figure 4 shows maps of the impacts on 2010 surface PM$_{2.5}$ for 30 % reductions in UK terrestrial emissions of each of NH$_3$, NO$_x$, SO$_x$, VOC, and primary PM$_{2.5}$. The effect of these emissions reductions on the three measures of UK-average surface concentrations of PM$_{2.5}$ are illustrated in Fig. 5, based on the data given in Table 1. The principal observations from the two figures are that PM$_{2.5}$ levels in the UK do not show strong responses to UK-only reductions in emissions of individual components and/or precursors of PM$_{2.5}$, and that the responses are highly geographically variable. The maximum reduction in PM$_{2.5}$ concentrations (at a 5 km grid resolution) reaches $\sim 0.6\,\mu$g m^{-3} (~ 6 % of the modelled components) in response to a 30 % reduction in UK emissions of individual components, and in most locations the reductions in PM$_{2.5}$ concentrations are considerably smaller. This again indicates the influence on PM$_{2.5}$ in the UK (on an annual average basis) from emissions outside of the UK. In the case of the formation of SIA components, it also reflects the non-linearity in the precursor oxidation chemistry and gas-particle phase partitioning that occurs between emission location and receptor location (Harrison et al., 2013; Vieno et al., 2014).

Table 1. EMEP4UK-modelled estimates of the impact of 30 % UK terrestrial emissions reductions on three measures of UK-average surface concentrations of PM$_{2.5}$ (μg m^{-3}): (i) the average of the model grids containing the 45 monitors used to calculate the UK PM$_{2.5}$ average exposure indicator (AEI), (ii) the population-weighted average, and (iii) the area-weighted (i.e. geographical) average, for 2010, and for 2030 under a CLE emission scenario (using 2010 meteorology). For context, the modelled reductions in the baselines between 2010 and 2030 CLE for the three measures of UK-average PM$_{2.5}$ are 2.42, 2.24, and 1.70 μg m^{-3}, respectively.

Emissions reduced	AEI		Population-weighted		Area-weighted	
	2010	2030 CLE	2010	2030 CLE	2010	2030 CLE
Primary PM$_{2.5}$	0.37	0.29	0.31	0.24	0.16	0.13
NH$_3$	0.35	0.19	0.34	0.19	0.28	0.16
SO$_x$	0.27	0.15	0.26	0.15	0.19	0.11
NO$_x$	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
Total	1.17	0.82	1.10	0.77	0.82	0.57

Figures 4 and 5 show that, on average across the UK, the sensitivity of PM$_{2.5}$ concentrations to a 30 % reduction in UK emissions of individual contributing components increases in the order VOC, NO$_x$, SO$_x$, primary PM$_{2.5}$, and NH$_3$. The exact order varies slightly with the UK-average measure used (Fig. 5). This is due to differences in the spatial patterns of the PM$_{2.5}$ reductions shown in Fig. 4 in relation to the distribution of UK population shown in Fig. 2.

The 30 % reductions in UK VOC emissions gives maximum reductions of $\sim 0.15 \mu$g m^{-3} (1.5 %) in PM$_{2.5}$ concentrations in central and northern England and central Scotland (Fig. 4e). The 30 % reductions in UK NO$_x$ emissions yield around 0.2 μg m^{-3} (3 %) reductions in PM$_{2.5}$ over some rural areas (Fig. 4c), and generally a maximum of 0.15 μg m^{-3} (1.5 %) reductions in PM$_{2.5}$ over other rural areas. An important observation is that reductions of PM$_{2.5}$ over urban centres are smaller (no more than 0.15 μg m^{-3}) than in rural areas for these reductions in NO$_x$ emissions. The 30 % reductions in UK SO$_x$ emissions yield up to ~ 0.45–0.5 μg m^{-3} (5 %) reductions in PM$_{2.5}$ in the Trent valley and up to around 0.3–0.35 μg m^{-3} (3 %) reductions in PM$_{2.5}$ over large areas of central and northern England and central Scotland (Fig. 4d). The locations with greatest sensitivities to the 30 % NO$_x$ emissions reductions (Fig. 4c) are generally those with the lowest sensitivities to SO$_x$ emissions reductions (Fig. 4d). As with the NO$_x$ emissions reductions, the reductions in PM$_{2.5}$ concentrations for reductions in SO$_x$ emissions is not, in general, associated with the major urban areas, except where these also have major SO$_x$ sources in the vicinity (e.g. Trent Valley, West Midlands, Cheshire). This is primarily caused by the spatial distribution of major sources of SO$_x$ emissions. As ~ 80 % of UK SO$_x$ 2010 emissions originate from large point sources (power plants, industrial facilities), which are not located in the heart of urban areas, associated emission reductions have the most profound effects in rural areas. However, the greater sensitivity to SO$_x$ close to large point sources (e.g. coal-fired power plants) may in part be an artefact due to the model assumption that 5 % of SO$_x$

emissions are directly in the form of SO$_4^{2-}$, which may no longer be appropriate for these sources or for models running at relatively high horizontal spatial resolution. The SO$_x$ and NO$_x$ gases compete in their reaction with NH$_3$ to form particulate ammonium sulphate ((NH$_4$)$_2$SO$_4$) or ammonium nitrate (NH$_4$NO$_3$). The larger sensitivity of PM$_{2.5}$ formation to NH$_3$ emissions reductions indicates that NH$_3$ is the limiting species; whilst the greater sensitivity to SO$_x$ than to NO$_x$ emissions reductions reflects that the reaction between NH$_3$ and SO$_x$ is fast and essentially irreversible compared with the equilibrium reactions between gaseous NH$_3$ and NO$_x$ species and NH$_4$NO$_3$.

The largest reductions in PM$_{2.5}$ (when weighted towards areas of greatest population) derive from 30 % reductions in UK NH$_3$ and primary PM$_{2.5}$ emissions (Fig. 4b and f), up to 0.45 μg m^{-3} for NH$_3$ reductions and greater for primary PM$_{2.5}$ reductions (up to ~ 6 % of modelled PM$_{2.5}$ in both cases). There is a distinct inverse geographic relationship in the PM$_{2.5}$ sensitivity to reductions of these two components. The reductions in NH$_3$ emissions give greatest PM$_{2.5}$ decreases in agricultural areas, whereas the reductions in primary PM$_{2.5}$ give greatest decreases in the large conurbations and other areas of high population density. The difference in geographical patterns is highlighted more clearly in Fig. 6a which shows the data in Fig. 4b minus the data in Fig. 4f. Blue colours in Fig. 6a indicate where reductions in PM$_{2.5}$ from a 30 % reduction in NH$_3$ emissions exceed the reductions in PM$_{2.5}$ from a 30 % reduction in primary PM$_{2.5}$ emissions, and vice-versa for red colours. White colours indicate comparable reductions in PM$_{2.5}$ via primary PM$_{2.5}$ or NH$_3$ emissions reductions. The geographical pattern in PM$_{2.5}$ sensitivity reflects the geographical pattern of the emission sources and the fact that, because of the short atmospheric lifetime of NH$_3$, UK emissions of NH$_3$ also generally have short-range influence.

Figure 7 shows the map of annual-average surface concentration of PM$_{2.5}$ estimated for the 2030 CLE emissions projections, and of the difference between the PM$_{2.5}$ concen-
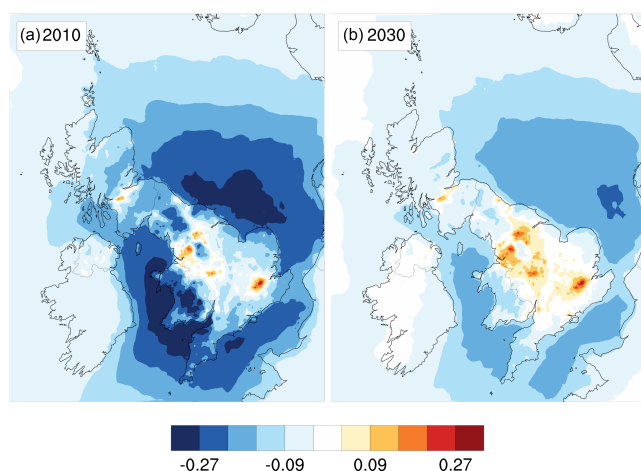
Figure 6. The difference between changes in simulated annual-average PM$_{2.5}$ (µg m^{-3}) for 30 % reductions in UK NH$_3$ emissions reduction and for 30 % reductions in UK primary PM$_{2.5}$ emissions reduction: **(a)** for the year 2010 (i.e. the data in Fig. 4b minus the data in Fig. 4f); and **(b)** for the year 2030 (i.e. the data in Figure 8b minus the data in Fig. 8f). Blue colours indicate where reductions in PM$_{2.5}$ for 30 % reduction in NH$_3$ emissions exceed the reductions in PM$_{2.5}$ for 30 % reduction in primary PM$_{2.5}$ emissions, and vice versa for the red colours. The same meteorological year 2010 was used.

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

trations in 2030 and 2010. Surface concentrations of PM$_{2.5}$ over the UK are simulated to reduce by up to 2.8 µg m^{-3} between 2010 and the 2030 CLE emissions scenario used. The UK-wide reductions in PM$_{2.5}$ between 2010 and 2030 CLE are 1.70, 2.24, and 2.42 µg m^{-3} for the area-weighted, population-weighted and AEI summary measures, respectively. The impacts on surface PM$_{2.5}$ in 2030 of additional 30 % reductions applied to UK-only terrestrial emissions of each of NH$_3$, NO$_x$, SO$_x$, VOC, and primary PM$_{2.5}$ individually are shown in Fig. 8. Figure 5 illustrates the quantitative effect of these further emissions reductions against the 2030 CLE scenario on the three summary measures of UK-average surface concentrations of PM$_{2.5}$.

The maps in Fig. 8 show qualitatively very similar findings to their equivalent maps in Fig. 4. In 2030, UK PM$_{2.5}$ is projected to remain more sensitive to reductions in UK emissions of NH$_3$ and primary PM$_{2.5}$ than to reductions in UK SO$_x$ and NO$_x$; and, from a population-weighted perspective, to be relatively more sensitive to further primary PM$_{2.5}$ and NH$_3$ emissions reductions, particularly to primary PM$_{2.5}$ emissions reductions, than was the case for the 2010 simulations (Fig. 5). For the 2030 simulations, additional 30 % reductions in UK primary PM$_{2.5}$ or NH$_3$ emissions yield reductions in PM$_{2.5}$ of up to 0.5 or 0.25 µg m^{-3}, respectively (Fig. 8), whilst in 2010 additional 30 % reductions in primary PM$_{2.5}$ or NH$_3$ emissions yield reductions in PM$_{2.5}$ of up to 0.6 or 0.45 µg m^{-3}, respectively (Fig. 4). The 2030 results again emphasize a geographic pattern of greatest sensitivity
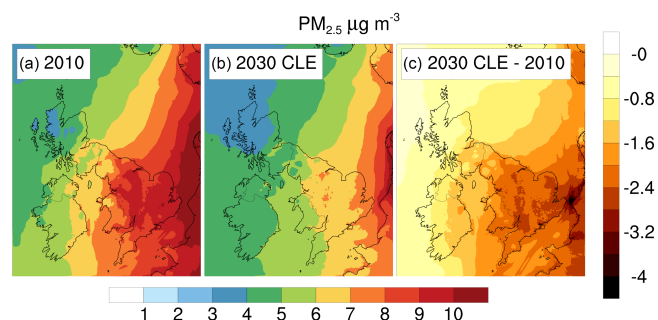
of PM$_{2.5}$ to reductions in the areas of high population density. Figure 6b plots the difference in response to the NH$_3$ and primary PM$_{2.5}$ emissions reductions in 2030, analogous to the plot in Fig. 6a for the 2010 sensitivities. Figure 6b clearly emphasizes that for this projection for 2030, UK PM$_{2.5}$ is relatively even more sensitive to further reductions in UK primary PM$_{2.5}$ emissions compared with further reductions in UK NH$_3$ emissions, particularly in populated areas, than is the case for 2010; albeit that the additional absolute reductions in PM$_{2.5}$ for a given percentage of emissions reductions is smaller in 2030 than in 2010 (Fig. 5) because of the general decline in emissions across Europe during this period for this scenario.

4 Discussion

Simulations were undertaken for both 2010 and a 2030 scenario to investigate whether conclusions on effectiveness of potential UK mitigation differ between the two time points. It is recognized that reductions in emissions of primary PM$_{2.5}$ and precursor gases from many anthropogenic sources are already anticipated going forward under current legislation, so it is important to know, for a future policy perspective, the anticipated sensitivities of UK PM$_{2.5}$ to additional UK emission reductions in the future.

The simulations for both 2010 and 2030 CLE show that if the focus is on the reduction of spatially averaged PM$_{2.5}$ concentrations then the most effective UK control, via an individual component, is achieved through reduction of UK emissions of NH$_3$, as shown in Fig. 5. However, the conclusion is different when considering population-weighted PM$_{2.5}$ reductions for the mitigation of human health effects. For a full population weighting across all 5 km × 5 km model grids, reductions in UK primary PM$_{2.5}$ emissions are almost as effective as reductions in UK NH$_3$ emissions for the 2010 simulations, but primary PM$_{2.5}$ emissions reductions are simulated to be the most effective additional control in the
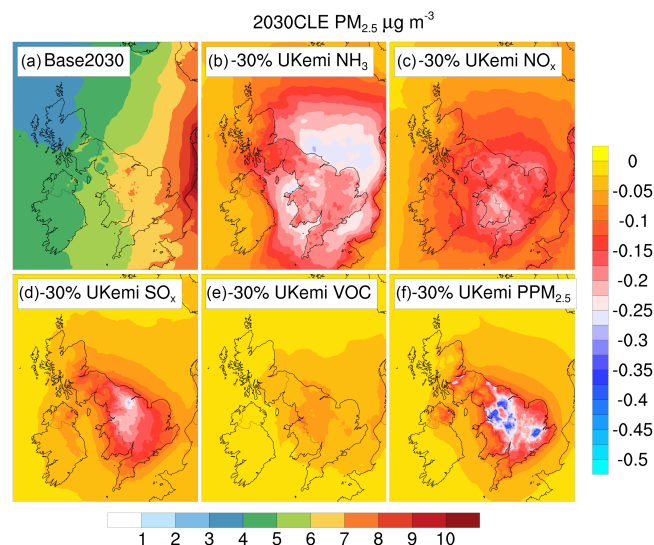
2030CLE PM$_{2.5}$ µg m^{-3}

Figure 8. Model simulations of impact of 30 % UK emissions reductions on annual-average surface concentration of PM$_{2.5}$ for a future scenario (with 2010 meteorology). Panel (**a**), 2030 CLE scenario, no emissions reduction (bottom colour scale); remaining panels, the change in annual-average PM$_{2.5}$ for 30 % UK emissions reductions in (**b**) NH$_3$, (**c**) NO$_x$, (**d**) SO$_x$, (**e**) VOC, and (**f**) primary PM$_{2.5}$ (right colour scale). All units are µg m^{-3}.

2030 CLE future (Fig. 5). Emphasis on population weighting also increases the sensitivities of PM$_{2.5}$ to reductions in NO$_x$ emissions in both 2010 and 2030 CLE because a major source of NO$_x$ is road traffic whose emissions are associated with where the population live. On the other hand, the sensitivity of PM$_{2.5}$ to further reductions in UK SO$_x$ emissions is markedly lower in 2030 than in 2010 because of the large reductions in SO$_x$ emissions already implemented under the CLE scenario. It is also recognized that reductions in NO$_x$ and VOC emissions have the potential to deliver health benefits separately from their contribution to reduction in PM$_{2.5}$ through reductions in population exposure to surface NO$_2$ and O$_3$.

An important observation is that the effectiveness of emissions reductions on PM$_{2.5}$ using a population weighting for the quantification differs between evaluation via full nationwide gridded population-weighting or via use of data only at the locations used to derive the AEI. Quantification through the AEI puts greater emphasis on the effectiveness of primary PM$_{2.5}$ emissions reduction, and on NO$_x$ emissions reductions, (Fig. 5) because the monitor locations contributing to the AEI are sited in the largest cities and towns where emissions of primary PM$_{2.5}$ and NO$_x$ are prevalent. Based on the AEI, control of primary PM$_{2.5}$ is the most effective individual component in 2010 as well as in 2030 CLE. These observations are pertinent given that the AEI is the air quality metric for PM$_{2.5}$.

Analyses from the EUCAARI study in Kulmala et al. (2011) and a more recent European study in Megaritis et al. (2013) both suggest that reducing NH$_3$ emissions is the most effective way to reduce PM$_{2.5}$ under present-day conditions. Whilst the current study also emphasizes the sensitivity of PM$_{2.5}$ to NH$_3$ emissions reductions, it also emphasizes that, for the UK, a sensitivity to primary PM$_{2.5}$ emissions reductions is at least as great as for NH$_3$ when considering population-weighting of PM$_{2.5}$ concentrations, both currently and for a future CLE scenario. In fact the sensitivity to primary PM$_{2.5}$ emissions may be underestimated by the simulations because of dilution of primary PM$_{2.5}$ emissions into the 5 km × 5 km grid resolution of the model. It has been calculated that a 1 : 1 relationship between UK primary PM$_{2.5}$ emissions reductions and the reduction in the primary PM$_{2.5}$ component of the UK 2010 AEI would lead to a reduction in the 2010 AEI of 0.8 µg m^{-3} (AQEG, 2015), compared with the 0.37 µg m^{-3} derived from the model simulations in this work (Table 1). Even so, the total impact of 30 % reductions in UK emissions of all the components and/or precursors listed in Table 1 on the 2010 baseline, is only of comparable magnitude (1.2 µg m^{-3}) to the 15 % (or 1.3 µg m^{-3}) reduction required in the UK AEI by 2020. However, reductions in these emissions from outside the UK will also contribute to reducing the UK PM$_{2.5}$ AEI. Conversely, reductions of emissions in the UK will also yield benefits for surface PM$_{2.5}$ concentrations elsewhere in Europe. The country-to-country source-receptor matrices developed by EMEP MSC-W at the 50 km resolution indicate that reductions in the UK of the same primary and precursor species considered in this work would (for 2011 emissions) lead to reductions in PM$_{2.5}$ in neighbouring countries up to about one-third the magnitude of the PM$_{2.5}$ reductions in the UK (Fagerli et al., 2014). Reductions of emissions in the UK would also lead to other benefits outside the UK on, for example, NO$_2$ and O$_3$ exposure and on N and S deposition.

Although the model used in this study is widely applied across Europe for air quality policy development (Fagerli et al., 2014), the data presented here are from simulations from a single model. The model simulations of the effect of inorganic precursor gases on the secondary inorganic PM$_{2.5}$ are dependent on accurate representation of the relevant chemistry and phase partitioning. It is possible that the SIA representation in the EMEP4UK model may underestimate the nitrate in the PM$_{2.5}$ size fraction, and hence downplay somewhat the sensitivity of PM$_{2.5}$ to NO$_x$ emissions reductions. In addition, not explicitly calculating the uptake of HNO$_3$ by mineral dust may reduce the NO$_3^-$ changes due to NO$_x$ emissions reduction. However, the EMEP4UK particle sulphate, nitrate and ammonium concentrations all compare well with the multi-year time series of measurements of these components at ∼ 30 sites across the UK in the Acid Gas and Aerosol Network (AGANet) and National Ammonia Monitoring Network (NAMN) (Vieno et al., 2014). Variation in particle-bound water may also impact on the exact PM$_{2.5}$ mass sen-

sitivities associated with inorganic precursor gas emissions reductions.

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

The interpretation of the modelling results has been undertaken from the perspective that reduction in all anthropogenically derived components of PM$_{2.5}$ is equally important. This remains the current position for the EU legislation that sets limits and targets for concentrations of PM$_{2.5}$ (Heal et al., 2012); i.e. no consideration is given to the potential different toxicity to human health of different components of PM$_{2.5}$. The UK Committee on the Medical Effects of Air Pollutants has also recently concluded that reductions in concentrations of both primary and secondary particles are likely to benefit public health (COMEAP, 2015). Nevertheless, although not conclusive, there is evidence that traffic-related sources of PM, or combustion sources more generally, are particularly associated with adverse health outcomes (Grahame and Schlesinger, 2007, 2010; Janssen et al., 2011; Stanek et al., 2011; WHO, 2013; Grahame et al., 2014). The possibility that primary PM$_{2.5}$ is more toxic per unit mass than secondary PM$_{2.5}$, places greater emphasis on the finding from this work on the effectiveness of reductions in emissions of primary PM$_{2.5}$. Interpretation of the modelling results has also not considered the relative costs or feasibilities of implementing further reductions in the emissions of the individual precursors and components investigated.

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

5 Conclusions

The sensitivity of annual-average surface concentrations of PM$_{2.5}$ across the UK to reductions in UK terrestrial anthropogenic emissions in primary PM$_{2.5}$, NH$_3$, NO$_x$, SO$_x$, and non-methane VOC was investigated using the EMEP4UK atmospheric chemistry transport model for 2010 and for a 2030 current legislation scenario that includes projected pan-European emission changes. In general, the sensitivity of modelled concentrations to UK-only mitigation is rather small. A 30 % reduction in UK emissions of any one of the above listed PM components yields (for the 2010 simulation) a maximum reduction in PM$_{2.5}$ concentrations in any given location of ∼ 0.6 µg m^{-3} (equivalent to ∼ 6 % of the

total modelled PM$_{2.5}$ mass concentration). On average across the UK, the sensitivity of PM$_{2.5}$ concentrations to a 30 % reduction in UK emissions of individual contributing components, for both the 2010 and 2030 CLE baselines, increases in the order NMVOC, NO$_x$, SO$_x$, NH$_3$, and primary PM$_{2.5}$, but there are strong spatial differences in the PM$_{2.5}$ sensitivities across the UK. Consequently, the sensitivity of PM$_{2.5}$ to individual component emissions reductions varies between area and population weighting. Reductions in NH$_3$ have the greatest area-weighted effect on PM$_{2.5}$. A full UK population weighting places greater emphasis on reductions of primary PM$_{2.5}$ emissions, which is simulated to be the most effective single-component control on PM$_{2.5}$ for the 2030 scenario. An important observation is that weighting corresponding to the average exposure indicator metric (using data from the 45 model grids containing a monitor whose measurements are used to calculate the UK AEI) further increases the emphasis on the effectiveness of primary PM$_{2.5}$ emissions reductions (and of NO$_x$ emissions reductions) relative to the effectiveness of NH$_3$ emissions reductions. Reductions in primary PM$_{2.5}$ has the largest impact on the AEI in 2010 as well as the 2030 CLE scenario. The summation of the reductions to the UK PM$_{2.5}$ AEI of the 30 % reductions in UK emissions of primary PM$_{2.5}$ and of NH$_3$, SO$_x$, NO$_x$ and VOC totals ∼ 1.2 and ∼ 0.8 µg m^{-3} with respect to the 2010 and 2030 CLE baselines, respectively (not accounting for non-linearity).

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