Future air pollution related health burdens associated with RCP emission changes in the UK

Future air pollution related health burdens associated with RCP emission changes in the UK

Sara Fenech[a,d,1], Ruth M. Doherty[a], Fiona M. O'Connor[b], Clare Heaviside[c], Helen L. Macintyre[d,e], Sotiris Vardoulakis[f], Paul Agnew[g], Lucy S. Neal[g]

[a]School of GeoSciences, University of Edinburgh, Edinburgh, UK

[b]Met Office, Hadley Centre, FitzRoy Road, Exeter, UK

[c]Institute for Environmental Design and Engineering, University College London, Central House, 14 Woburn Place, London, WC1H 0NN, UK

[d]Centre for Radiation, Chemical and Environmental Hazards, Public Health England, Chilton, UK

[e]School of Geography, Earth and Environmental Sciences, University of Birmingham, Edgbaston, Birmingham, UK

[f]National Centre for Epidemiology and Population Health, Research School of Population Health, Australian National University, Canberra, Australia

[g]Met Office, FitzRoy Road, Exeter, UK

Correspondence to: Sara Fenech (sara.fenech@um.edu.mt)

[1]Present address: Department of Geosciences, University of Malta, Malta

Abstract. Three Intergovernmental Panel on Climate Change (IPCC) Representative Concentration Pathways (RCPs) are used to simulate future ozone (O_3), nitrogen dioxide (NO_2), and fine particulate matter ($PM_{2.5}$) in the United Kingdom (UK) for the 2050s relative to the 2000s with an air quality model (AQUM) at a 12 km horizontal resolution. The present-day and future attributable fractions (AF) of mortality associated with long-term exposure to annual mean O_3, NO_2 and $PM_{2.5}$ have accordingly been estimated for the first time for regions across England, Scotland and Wales.

Across the three RCPs (RCP2.6, RCP6.0 and RCP8.5), simulated annual mean of the daily maximum 8-hr mean (MDA8) O_3 concentrations increase compared to present-day, likely due to decreases in NO_x (nitrogen oxides) emissions, leading to less titration of O_3 by NO. Annual mean NO_2 and $PM_{2.5}$ concentrations decrease under all RCPs for the 2050s, mostly driven by decreases in NO_x and sulphur dioxide (SO_2) emissions, respectively.

The AF of mortality associated with long-term exposure to annual mean MDA8 O_3 is estimated to increase in the future across all the regions and for all RCPs. Reductions in NO_2 and $PM_{2.5}$ concentrations lead to reductions in the AF estimated for future periods under all RCPs, for both pollutants. Total mortality burdens are also highly sensitive to future population projections. Accounting for population projections exacerbates differences in total UK-wide MDA8 O_3-health burdens between present-day and future by up to a factor of ~3 but diminishes differences in NO_2-health burdens. For $PM_{2.5}$, accounting for future population projections results in additional UK-wide deaths brought forward compared to present-day under RCP2.6 and RCP6.0, even though the simulated $PM_{2.5}$ concentrations for the 2050s are estimated to decrease. Thus, these results highlight the sensitivity of future health burdens in the UK to future trends in atmospheric emissions over the UK as well as future population projections.

Keywords: RCPs, health impacts, ozone, nitrogen dioxide, particulate matter, population projections, climate change

1 Introduction

Long-term exposure to ambient air pollution has been linked with adverse health impacts in a number of studies (e.g. Burnett et al., 2014; 2018; Jerrett et al., 2009; Krewski, 2009; Turner et al., 2015). Risk estimates for mortality associated with long-term exposure to ambient particulate matter less than 2.5 µm in diameter ($PM_{2.5}$) are well established (e.g. Hoek et al., 2013; Krewski, 2009). Several studies have quantified exposure-response relationships (often called concentration-response functions, or CRFs) associated with long-term exposure to ozone (O_3) (Jerrett et al., 2009; Turner et al., 2015) and nitrogen dioxide (NO_2) (COMEAP, 2018; Crouse et al., 2015; Faustini et al., 2014; Hoek et al., 2013). However, for both O_3 and NO_2, long-term effects are still emerging, with most studies to date performed over North America.

Future air pollution levels (and related health burdens) can be influenced by a number of factors including changes in emissions of air pollutants (both natural and anthropogenic) and climate variability. In this study we focus on the emissions-driven impacts on air pollutants and the corresponding health burdens in the UK. To estimate future ambient air quality, several future pathways are employed utilising the Intergovernmental Panel on Climate Change (IPCC) Representative Concentration Pathways (RCPs) (van Vuuren et al., 2011a). These are future pathways of global greenhouse gas (GHG) and air pollutants and their precursor emissions as used in the Coupled Model Intercomparison Project Phase 5 (CMIP5) project. They consist of four pathways that lead to radiative forcing levels from the combined effects of greenhouse gases and aerosols of 2.6 W m^{-2} (van Vuuren et al., 2011b), 4.5 W m^{-2} (Thomson et al., 2011), 6 W m^{-2} (Masui et al., 2011), and 8.5 W m^{-2} (Riahi et al., 2011) by 2100. For all RCPs, large decreases in emissions of particulate matter (PM) and precursors of O_3, especially nitrogen oxides (NO_x), carbon monoxide (CO), sulphur dioxide (SO_2), black carbon (BC) and organic carbon (OC), are projected globally following the assumption of more stringent air pollution control measures over time (van Vuuren et al., 2011a). An exception is ammonia (NH_3) which globally increases in nearly all pathways, and methane (CH_4) which increases especially for RCP8.5. However, these are global trends which may vary both spatially and temporally. Note that the assumption of more stringent air pollution measures across the globe is a caveat of the RCPs as these may not represent the true uncertainty in emissions pathways. In addition, the RCPs are developed independently and are governed by different assumptions about social, economic and political development.

Several studies have analysed the impact of emission changes on future pollutant concentrations both globally and regionally. For Europe, several studies suggest O_3 reductions due to decreases in O_3 precursors under different future emission scenarios (e.g. Hedegaard et al., 2013; Wild et al., 2012). However, increases in O_3 concentrations have also been observed especially under RCP8.5 due to increases in CH_4 concentrations (Wild et al., 2012) and in regions with high NO_x levels (e.g. the Benelux region and the south of the UK) (Hedegaard et al., 2013; Colette et al., 2012). In a multi-model study, Im et al. (2018) suggest an average increase in O_3

concentrations (by up to ~6 %) for Europe in response to a 20 % reduction of anthropogenic emissions; this study also suggests a larger contribution of non-local sources for O_3 concentrations in Europe.

Future emission changes both globall and in Europe, are likely to lead to lower $PM_{2.5}$ concentrations due to reduced primary emissions as well as changes in secondary inorganic aerosol (Chemel et al., 2014; Hedegaard et al., 2013; Im et al., 2018; Vieno et al., 2016). Sulphate and nitrate aerosols compete for ammonium, such that reductions in SO_2 emissions (which lead to reductions in sulphate concentrations) and increases in NH_3 emissions could increase nitrate aerosol levels even though NOx emissions decrease (Pye et al., 2009).

Most recent studies focusing on future health burdens associated with long-term exposure to O_3 and/or $PM_{2.5}$ concentrations under the RCPs typically analyse the combined impacts of changes in emissions and changes in the climate. Following RCP4.5, the estimated $PM_{2.5}$ and O_3-related global mortality burden is 1.1±0.5 and 0.2±0.1 million avoided deaths per year in 2050, respectively (West et al., 2013). Future health burdens associated with long-term exposure to air pollution do not solely depend on projected future air pollutant concentrations but also depend on changes in mortality rates and demographic or population changes, as well as underlying rates of diseases. Despite projected future reductions in pollutant concentrations, Silva et al. (2016) suggest increases in premature mortality when accounting for increases in total population size in 2050. West et al. (2013) also suggest that globally and in Europe, the population growth and baseline mortality rates can dominate changes in the health impacts resulting from the RCPs particularly in 2100.

Some studies have focused on future health impacts in the UK following different emission scenarios. Using three IPCC Special Report on Emissions Scenarios (SRES; Nakicenovic et al., 2000) and including population projections for 2030, Heal et al. (2013) estimate that the O_3-health burden increases over the UK by 16-28 % compared to that in 2003. A recent modelling study examined health impacts due to air pollutant concentration changes in 2035 and 2050 for three UK-specific energy-related scenarios (Williams et al., 2018). In all three scenarios, NO_2 concentrations decrease in 2050 due to reductions in NOx emissions. The subsequent reduction in long-term NO_2- related mortality following the baseline scenario in 2050 is estimated to be ~6.5 million life-years gained compared to 2011 (Williams et al., 2018). $PM_{2.5}$ concentrations across the UK also decrease for this baseline scenario leading to ~17.8 million life-years gained in 2050 compared to 2011 (Williams et al., 2018). Using the AQUM (Air Quality in the Unified Model; Savage et al., 2013) as used in our study, Pannullo et al. (2017) find reductions in NO_2 concentrations for three RCPs across the UK in the 2050 lead to reductions in respiratory hospital admissions associated with exposure to NO_2 by 1.7 % (RCP2.6), 1.4 % (RCP6.0) and 2.4 % (RCP8.5) (Pannullo et al., 2017).

As discussed above, several global and regional studies have analysed the impact of emission changes on future air quality. However, to date such health impact assessment studies conducted using the RCPs comprehensively for three pollutants O_3, NO_2 and $PM_{2.5}$ over the UK are limited and generally do not account for future population projections. The focus of this study is on UK primary and precursor emission changes for the 2050 based on three RCPs: RCP2.6, RCP6.0 and RCP8.5 and the subsequent impacts of these changes on ambient levels of O_3, NO_2 and $PM_{2.5}$. We also assess the health effects associated with long-term exposure to each pollutant under each RCP, and the sensitivity of estimated future health burdens to two population scenarios.

The study is organised as follows: The impact of changes in future UK emissions on simulated annual mean MDA8 O_3, NO_2 and $PM_{2.5}$ concentrations are discussed in Section 3. The corresponding future health burdens associated with long-term exposure to all three pollutants are described in Section 4 together with the sensitivity of estimated mortality burdens associated with each pollutant to future population projections. Finally, summary and conclusions are presented in Section 5.

2 Methods

2.1 Air Quality in the Unified Model – AQUM

In this study we use air pollutant concentrations for both present-day and future which are derived from configurations of a limited-area model system, AQUM (Air Quality in the Unified Model), based on the UK Met Office Unified Model (Brown et al., 2012). AQUM has a horizontal resolution of 0.11° × 0.11° (~12 km; Savage et al., 2013) with a domain covering the UK and parts of the western European region. The model has 38 vertical levels from the surface up to 39 km (with the lowest model level at 20 m). The model includes an interactive aerosol scheme CLASSIC (Coupled Large-scale Aerosol Simulator for Studies in Climate; Bellouin et al., 2013, 2011; Jones et al., 2001) which simulates ammonium sulphate and nitrate, fossil-fuel organic carbon (OC), mineral dust, soot and biomass burning (BB) aerosol interactively. Biogenic secondary organic aerosols are prescribed from a climatology (Bellouin et al., 2011) and sea salt is calculated over sea points only and does not contribute to PM concentrations over land. Within AQUM, sulphate and nitric acid compete for available ammonium to form ammonium nitrate and ammonium sulphate aerosols (Bellouin et al., 2011). Gas-phase chemistry is simulated within AQUM by a tropospheric configuration of the United Kingdom Chemistry and Aerosol (UKCA) model

(Morgenstern et al., 2009; O'Connor et al., 2014). The chemistry scheme used is the Regional Air Quality (RAQ) chemistry scheme, which has 58 chemical species, 116 gas phase reactions and 23 photolysis reactions. Photolysis rates are calculated with the on-line photolysis scheme Fast-JX (Neu et al., 2007). Lateral boundary conditions for chemistry and aerosols are derived from the GEMS (Global and regional Earth-system Monitoring using Satellite and in-situ data) and MACC (Monitoring Atmospheric Composition and Climate) global reanalysis fields (Flemming et al., 2009) whilst meteorology is obtained from the UK Met Office Unified Model (MetUM) global forecast fields. Further details on AQUM, including evaluation, can be found in Savage et al. (2013). We use the same AQUM configuration as described in Fenech et al. (2018).

2.2 Model Set-up

In this study we conduct a total of four simulations using AQUM: one to derive air pollutant concentrations for the 2000s ('present-day') and another three simulations to derive air pollutant concentrations following three different RCPs for the 2050s ('future'). For all these simulations the same meteorology was employed – that of the year 2006 (Fenech et al. 2018). For the present day simulation, anthropogenic and biomass burning emissions are taken from decadal mean emissions centred on the year 2000 (Lamarque et al., 2011, 2010). These historical emissions were originally created for the 5th Climate Model Intercomparison Program (CMIP5) in support of the IPCC Fifth Assessment Report (AR5) and are used as a starting point for all RCPs. For the future simulations, anthropogenic and biomass burning emissions are obtained from decadal mean emissions centred on 2050 following three IPCC RCPs: RCP2.6, RCP6.0 and RCP8.5. Biogenic emissions of isoprene for all simulations are diagnosed from simulations with a fully coupled nested configuration of the MetUM (described in Neal et al., 2017) and then prescribed in AQUM with a diurnal cycle imposed. Therefore, although the climate is unchanged between the 2000s and the 2050s, the prescribed biogenic isoprene emissions have responded to changes in CO_2 and temperature in the 2050s compared to the 2000s following Pacifico et al. (2011). Emission changes are discussed in more detail in Section 2.2.1.

The lateral boundary conditions used for future simulations are kept the same as for the present-day. For all simulations, feedbacks of aerosols and greenhouse gases on the radiation scheme are excluded, thus ensuring the climate is unchanged between the present-day and future simulations. Hence the projected changes in air pollutant concentrations show the influence of UK anthropogenic, biomass burning and biogenic emission changes only. Greenhouse gas (GHG) such as CH_4, carbon dioxide (CO_2) and nitrous oxide (N_2O) have prescribed concentrations for each simulation due to their long lifetime.

All simulations are conducted for 18 months with the first 6 months discarded as spin-up. Hourly pollutant concentrations taken from the lowest model vertical level (20 m) are then extracted, from which the annual mean MDA8 O_3, NO_2 and $PM_{2.5}$ concentrations are calculated.

2.2.1 Present-day and future UK emissions

Emissions totals and percentage differences over the UK between the 2000s and 2050s for key O_3 and $PM_{2.5}$ primary and precursor species under each pathway are shown in Fig. 1. Most of the emissions over the UK decrease in the future compared to present-day, with the greatest reductions occurring for RCP8.5 (Fig.1). For example, nitrogen oxides (NOx) emissions fall from ~ 1.3 Tg (NO) yr^{-1} in 2000s to ~ 0.5 Tg (NO) yr^{-1} for RCP2.6 and RCP6.0 and ~ 0.3 Tg (NO) yr^{-1} for RCP8.5 in 2050s, corresponding to reductions of 60 %, 58 % and 73 % (Fig.1a). Spatial distributions of NOx emissions for present-day and for differences in future emissions compared to present-day are illustrated in Fig.2. For all RCPs, the largest reductions in NOx emissions occur in regions having high present-day emissions (e.g. central and south east England) (Fig. 2). Emissions following RCP8.5 exhibit the largest reductions compared to present-day while the smallest reductions are for RCP6.0.

Emission changes between the 2000s and 2050s for CO, OC and BC follow a similar pattern to that of NOx with the highest reductions occurring for RCP8.5 and lower reductions for RCP2.6 and RCP6.0 (Fig. 1 b-d). SO_2 emissions also decrease in the future in all RCPs. However, reductions in each RCP follow a different pattern, with the highest reductions occurring for RCP2.6 (-92 %) and RCP8.5 (-94 %) and the lowest reductions occurring for RCP6.0 (-43 %; Fig. 1e). Ammonia (NH_3) emissions and methane (CH_4) concentrations increase over the UK for certain future pathways (Fig. 1f and g). Ammonia emissions total ~ 0.35 Tg (NH) yr^{-1} in 2000s and increase for all RCPs, with the highest increase occurring for RCP6.0 (29 %) followed by RCP2.6 (16 %) and RCP8.5 (7 %) (Fig. 1f). As mentioned in the previous section, CH_4 concentrations are prescribed for all four simulations due to its long lifetime relative to the simulation period. CH_4 concentrations over the UK are ~1200 $\mu g\ m^{-3}$ for present-day and decrease by 16 % in 2050s following RCP2.6 (Fig. 1g). In contrast CH_4 concentrations increase following RCP6.0 (8 %) and RCP8.5 in 2050s (54 %; Fig. 1g).

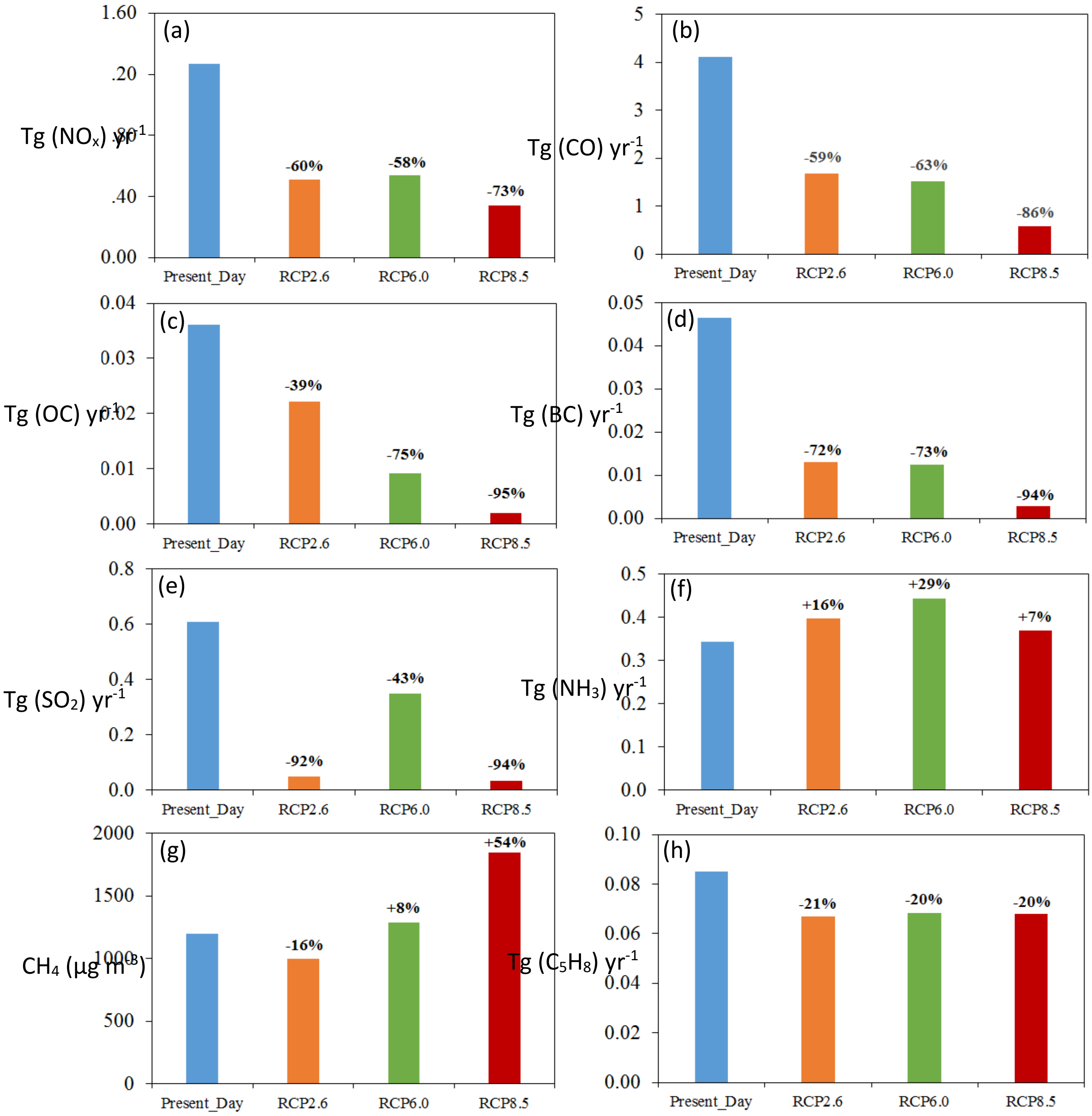

Figure 1: Emissions totals and percentage differences over the UK (land only) between 2000s and 2050s for the key O_3 and $PM_{2.5}$ primary and precursor species: (a) nitrogen oxides (NO_x) (b) carbon monoxide (CO), (c) fossil fuel organic carbon (OC), (d) fossil fuel black carbon (BC), (e) sulphur dioxide (SO_2), (f) ammonia (NH_3), (g) methane (CH_4) (concentrations) and (h) isoprene (C_5H_8) for present-day (blue), RCP2.6 (orange), RCP6.0 (green) and RCP8.5 (red). Percentage differences between future and present-day are shown above each pathway.

Isoprene (C_5H_8) emissions used in this study are obtained from a model simulation in which biogenic VOC emissions are calculated interactively responding to changes in carbon dioxide (CO_2) and temperature (Pacifico et al., 2011). These are then prescribed in the UK domain within AQUM. C_5H_8 emissions decrease from ~0.09 Tg (C_5H_8) yr^{-1} for present-day to ~0.07 Tg (C_5H_8) yr^{-1} under all RCPs (~20 % reduction; Fig 1h). This suggests that the main driver for reductions in isoprene emissions for all future pathways is CO_2 inhibition of isoprene (high levels of CO_2 suppressing leaf isoprene production) (Arneth et al., 2007) which offset the temperature-driven emission increases, as found by other studies (e.g. Pacifico et al., 2012; Squire et al., 2015) .

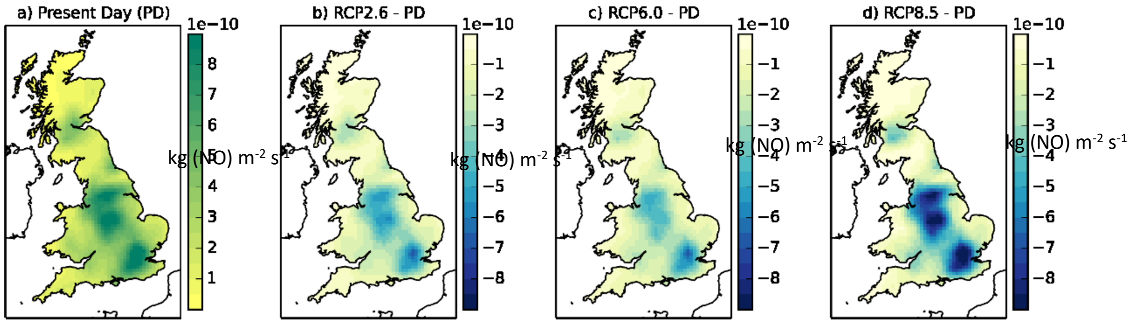

Figure 2: Total NOx emissions for (a) present-day (PD – 2000s) and (b-d) differences in NOx emissions between present-day and future (2050s) under three RCPs.

2.3 Heath impact assessment

Estimated health burdens attributable to long-term exposure to annual mean MDA8 O_3, NO_2 and $PM_{2.5}$ concentrations are calculated as follows for each of the nine Government Office Regions (GOR) for England, and for Scotland and Wales (Fig. 3):

$$M_r = BM_r \times AF_r \tag{1}$$

where

$$AF_r = \frac{RR_r - 1}{RR_r} \tag{2}$$

and

$$RR_r = \exp(CRF \times x_r) \tag{3}$$

and

$$x_r = \frac{\sum_{j \in region} (x_j \times p_j)}{\sum_{j \in region} p_j} \quad (4)$$

In equation 1, M_r is the mortality associated with long-term exposure to MDA8 O_3, NO_2 or $PM_{2.5}$ for each region, r (Fig. 3), BM_r is the total regional annual respiratory mortality for O_3-related health estimates and all-cause (excluding external) mortality for NO_2 and $PM_{2.5}$-related health estimates for 2006 (henceforth referred to as baseline mortality) and AF_r is the attributable fraction associated with long-term exposure to annual mean MDA8 O_3 or NO_2 or $PM_{2.5}$ concentrations that is calculated for each region using equation 2. Baseline mortality data for 2006 was obtained from the Office for National Statistics for England and Wales (ons.gov.uk) and from the National Records of Scotland (nrscotland.gov.uk) of which only ages above 30 years were considered. In equations 2 and 3, RR is the relative risk associated with long-term exposure to annual mean air pollutant levels. In equation 3, *CRF* is the concentration-response function coefficient and x_{ir} is the regional annual population-weighted pollutant concentration. The *CRF* used in this study for long-term exposure to annual mean MDA8 O_3 concentrations is taken from Turner et al (2015) and for long-term exposure to annual mean NO_2 and $PM_{2.5}$ the *CRF* used is from COMEAP (2015) and WHO (2013), respectively. The CRF recommended by Turner et al. (2015) for the effects of long-term O_3 exposure on respiratory mortality is 1.06 (95 % Confidence Interval (CI) = 1.04,1.08) per 10 $\mu g\ m^{-3}$ increase in annual mean MDA8 O_3 concentrations with a threshold of 53.4 $\mu g\ m^{-3}$ (Turner et al. 2015). For long-term NO_2 exposure on all-cause (natural) mortality COMEAP (2015) suggest a coefficient of 1.025 (95 % CI = 1.01,1.04) per 10 $\mu g\ m^{-3}$ increase in annual mean NO_2 concentrations (with no threshold) while for long-term $PM_{2.5}$ exposure on all-cause (natural) mortality, we use a coefficient of 1.062 (95 % CI = 1.040,1.083) per 10 $\mu g\ m^{-3}$ increase in annual average concentrations (with no threshold), which is based on a meta-analysis of cohort studies by Hoek et al. (2013). For all three pollutants, the health estimates in this study are for people aged 30 years and above. The uncertainty in AF estimates, presented in the figures of the supplement section of this manuscript as error bars, represents the 95% CI, as taken from the literature and described above, associated with the uncertainties in the concentration-response coefficients used only. Burdens for the effects of each pollutant are calculated separately following recommendations from the studies from which the coefficients are drawn, though it should be noted that NO_2 effects could to some extent overlap with effects of long-term exposure to $PM_{2.5}$ as the two pollutants are strongly correlated (COMEAP, 2018).

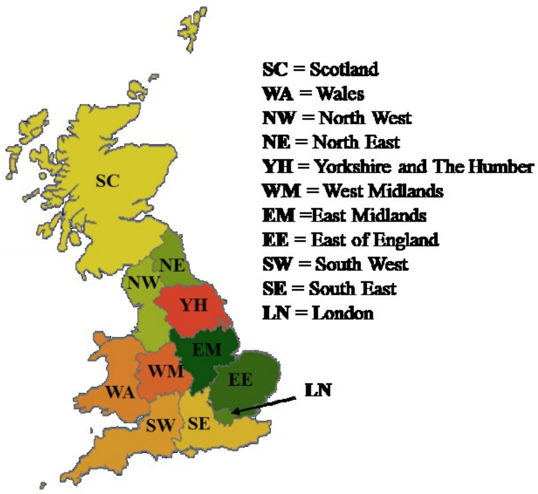

Figure 3: Government Office Regions (GOR) for England, Scotland and Wales used in this study (adapted from Fenech et al. 2019).

The population-weighted pollutant concentrations x_r are calculated by first counting the total residential gridded population data for people aged 30 years and over (p, at a resolution of 5 km (GWPv3), obtained from the Socioeconomic Data and Applications Centre (SEDAC) (sedac.ciesin.columbia.edu) within each model grid cell, j (equation 4). This population total (p_j) is then multiplied by the annual pollutant concentration within each grid cell x_j, summed over every grid cell within the region r, and divided by the total population of the region. To quantify differences in health burdens between present-day and future we simply subtract the present-day estimate from the future one (ie Mort $_{\text{Future}}$ – Mort $_{\text{PD}}$).

Future mortality estimates do not solely depend on future pollutant concentrations but also on future population projections and mortality rates. In this study we analyse how changes in future population projections may influence estimated future health burdens. Each RCP is associated with a future population projection. However, to isolate the effect of changes in air pollutant concentrations across the RCPs we choose to use a single population projection. This is done by using the future population projections from the Shared Socioeconomic Pathways (SSP) gridded at a resolution of 17 km (Jones and O'Neill, 2017). The SSPs are a set of five different socioeconomic development narratives that describe plausible pathways for the evolution of society over the next century and are intended to provide a range of pathways that can be combined with the RCPs (Riahi et al., 2017). In conjunction with each RCP the population projection following SSP1, or "Sustainability" storyline and SSP5, or "Fossil-fuelled Development" storyline are applied. These two SSPs represent the upper (SSP5) and lower (SSP1) limits of projected population totals for the UK for 2050s.

Based on SSP1 and SSP5, the total UK population is estimated to reach 74.7 million and 85.3 million, respectively, in 2050s (a factor of 1.2 and 1.4 greater than present-day totals used in this study). The UK Office of National Statistics (ONS) projects a total UK population of 72.9 million in mid-2041 based on 2016 data (ONS, 2017), while the Directorate-General of the European Commission (Eurostat) estimates suggest that at the start of 2040 the total UK population will be 75 million (Eurostat, 2017). The baseline mortality rates and demographics for the future estimates are kept at present-day rates.

3 Results and Discussion

3.1 The emissions driven impact on O_3 and NO_2 concentrations in the UK

Simulated annual mean MDA8 O_3 concentrations in 2000s are on average ~ 77 µg m^{-3} across the UK, ranging from ~ 62 µg m^{-3} in central England (West and East Midlands regions) to more than 80 µg m^{-3} in the south west of England, Wales, and Scotland (Fig 4a). O_3 concentrations increase under all RCPs (2050s) compared to present-day (2000s) and are on average between ~4 µg m^{-3} and ~9 µg m^{-3} higher depending on the RCP (Fig. 4b-d). For all three RCPs the largest increases for 2050s occur in regions where present-day O_3 concentrations are low (Fig. 4b-d). The largest increases in O_3 concentrations occur for RCP8.5 where concentrations are more than 12 µg m^{-3} (~40 %) and up to 18 µg m^{-3} (~50 %) higher across much of the central and southern regions of England (Fig. 4d). For RCP2.6 and RCP6.0 increases in O_3 concentrations are about 6 µg m^{-3} (~25 %) compared to present-day in Central England with smaller increases elsewhere in the UK (Fig. 4b and c).

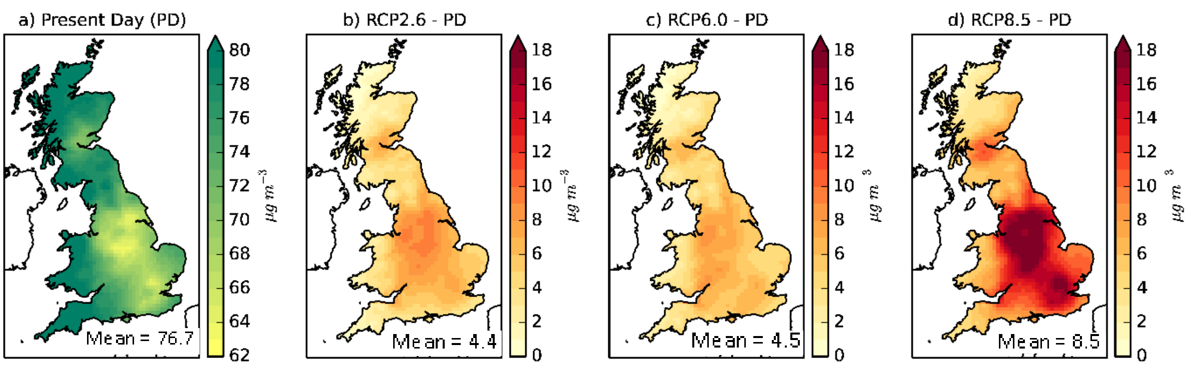

Figure 4: Simulated annual mean daily maximum 8-hr running mean (MDA8) O_3 concentrations for (a) present-day (PD, 2000s), (b-d) differences in simulated annual mean MDA8 O_3 concentrations between present-day (2000s) and future (2050s) calculated as MDA8 $O_{3\ future}$ – MDA8 $O_{3\ present\text{-}day}$ for each future pathway. Mean concentrations (µg m^{-3}) are shown at the bottom of each panel.

Changes in the abundance of O_3 concentrations can occur via changes in precursor emissions. NO_X emissions play a key role in determining surface O_3 concentrations. For all three RCP simulations, UK NO_X emissions decrease in 2050s (by up to 73 %; Fig. 1 and 2) with the largest decreases occurring in regions having high present-day NO_x emissions (Fig. 2), for example central and south eastern England (Fig. 2b-d). This results in substantial decreases in annual mean NO_2 concentrations across the UK (Fig. 5b-d). Present-day annual mean NO_2 concentrations averaged across the UK are 11.2 μg m^{-3} and range from ~3 μg m^{-3} in most of Scotland to ~ 27 μg m^{-3} in Central and South East England (Fig. 5a), with the spatial distribution being inversely to that of O_3 concentrations (Fig. 4a). Annual-mean NO_2 concentrations reduce in the future, with decreases between 6 μg m^{-3} (~-25 %) and 12 μg m^{-3} (~-50 %) in most of central England for RCP2.6 and RCP6.0 (Fig. 5b and c) and higher than 15 μg m^{-3} (~-75%) for RCP8.5 (Fig. 5d). Regions showing low present-day O_3 concentrations and high NO_2 concentrations (Fig. 4a and 5a), also exhibit the greatest increases in O_3 concentrations and the largest decreases in NO_2 concentrations in the 2050s relative to 2000s (Fig. 4 b-d and Fig. 5 b-d). Reductions in NO_2 concentrations are highest for RCP8.5 (Fig. 5d) followed by RCP 2.6 (Fig. 5b) and RCP6.0 (Fig. 5c), consistent with reductions in NO_X emissions (Fig 1 and 2). The higher O_3 concentrations in the future are likely due to the titration effect of NO on O_3. Thus, UK NO_X emission reductions between present-day and future increase O_3 concentrations, suggesting a NO_x saturated environment across all the UK, but particularly in highly polluted regions, where estimated NO_X emission reductions are highest.

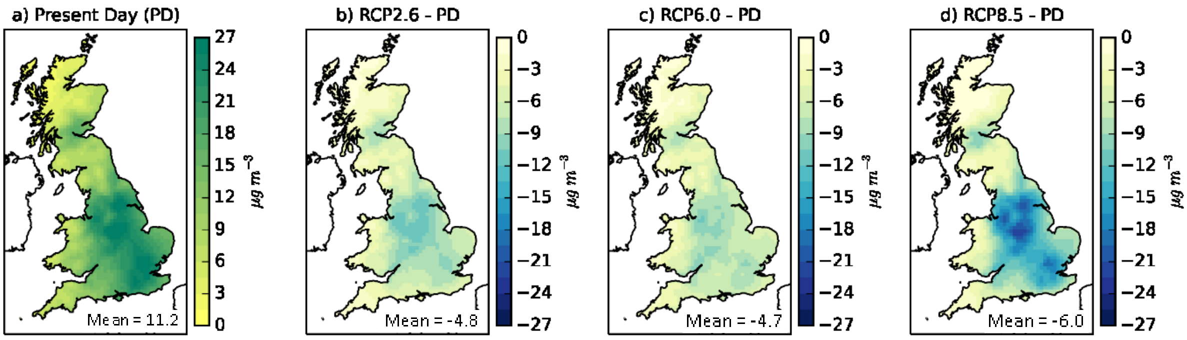

Figure 5: Annual mean NO_2 concentrations for (a) present-day (PD – 2000s) and (b-d) differences in NO_2 concentrations between present-day and future (2050s) under three RCPs. Mean concentrations (μg m^{-3}) are shown at the bottom of each panel.

While annual mean CH_4 concentrations decrease for RCP2.6 (-16 %) across the UK, they increase for RCP6.0 (+8 %) and RCP8.5 (+ 54 %) (Fig. 1). This increase in CH_4 concentrations for RCP6.0 and RCP8.5 between present-day and future may contribute to increases in O_3 concentrations for these two pathways (Fig. 4c and d) but fails to explain increases in annual mean MDA8 O_3 concentrations for RCP2.6 (Fig. 4b). Nonetheless, the spatial patterns in O_3 differences between present-day and future (Fig. 4) are primarily driven by changes in NO_x emissions (Fig. 5) as discussed above.

These findings are consistent with previous studies over Europe. A number of global and Europe-wide studies have shown a strong impact of NO_x emission reductions and higher CH_4 levels leading to higher O_3 concentrations in the northern European region. Hedegaard et al. (2013) find the chemical environment for O_3 production to differ between North-west Europe and elsewhere in Europe. In the Benelux regions and surrounding countries (including the UK), O_3 concentrations increased under RCP4.5 between 1990 and 2090 because of NO_x decreases. However, this NOx- saturated environment was not consistent across the UK as found in this study. Elsewhere in Europe, NO_x emissions reductions under RCP 4.5 were found to lead to lower O_3 concentrations (~20 %) between 1990 and 2090 (NO_x limited; Hedegaard et al. 2013). In another regional European study with different future emission scenarios (Global Energy Assessment scenarios) but which also feature NO_x emission reductions, annual mean O_3 was found to increase across most of north west Europe and the UK in particular central and south east England (Colette et al. 2012), also highlighting the North West Europe and the UK as a NO_x saturated environment.

Similar to the work presented in this study, a UK-focused report evaluating nine regional models (including AQUM) suggests that reducing the total anthropogenic NO_x and VOC emissions by 30 % for 2006 across the UK, led to an increase in annual mean simulated O_3 concentrations ranging between 1.2 and 6.1 µg m^{-3} for all models (Defra, 2013). A similar increase in annual mean O_3 concentrations over the UK was also found for a 30 % reduction in anthropogenic NO_x and VOC emissions across the UK and Europe. Thus, this study suggests that increases in O_3 concentrations are more likely driven by reductions in the UK NOx and VOC rather than changes in emissions outside the UK.

3.2 The emissions driven impact on $PM_{2.5}$ concentrations in the UK

Present-day simulated annual mean $PM_{2.5}$ concentrations averaged across the UK are ~9 µg m^{-3} and range from ~ 4 µg m^{-3} in Scotland to ~ 12 µg m^{-3} in eastern England (Fig. 6a). For all RCPs, annual mean $PM_{2.5}$ concentrations decrease compared to present-day, with a notable north-south gradient (Fig. 6 b-d). Across the UK, the largest reductions in $PM_{2.5}$ concentrations in the 2050s relative to 2000s occur under RCP8.5 (Fig. 6d). For RCP8.5 and RCP2.6, annual mean $PM_{2.5}$ concentrations are between ~ 1 µg m^{-3} (~-15 %) in Scotland to ~ 4.5 µg m^{-3} (~-50 %) in central and eastern England lower compared to present-day (Fig. 6 b and d). In 2050s, reductions in simulated

$PM_{2.5}$ concentrations under RCP6.0 relative to 2000s are smaller than the other two future pathways with differences ranging from ~ -0.5 µg m^{-3}(~-10 %) in Scotland to ~ -2 µg m^{-3}(~ -20 %) in eastern England (Fig. 6c).

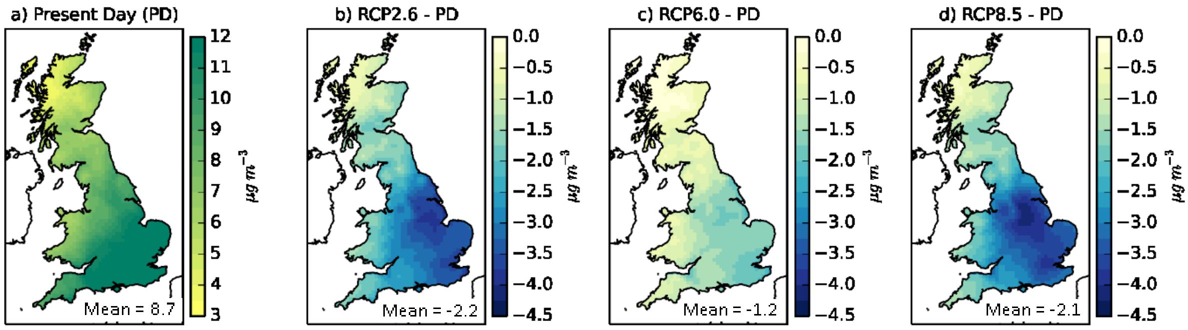

Figure 6: Simulated annual mean $PM_{2.5}$ concentrations for (a) present-day (PD – 2000s), (b-d) differences in simulated annual mean $PM_{2.5}$ concentrations between present-day and future (2050s) calculated as $PM_{2.5\ future}$ – $PM_{2.5\ present\text{-}day}$ for each future pathway. Mean concentrations (µg m^{-3}) are shown at the bottom of each panel.

In this study, for both present-day and future, the two major components contributing to total simulated annual mean $PM_{2.5}$ concentrations are ammonium sulphate and, to a lesser extent, ammonium nitrate (Fig. 7). For present-day, ammonium nitrate constitutes 35 % of total $PM_{2.5}$ concentrations while ammonium sulphate constitutes 45 % (Fig. 7a). For all RCPs, this distribution is modified compared to present-day, with ammonium nitrate increasingly becoming the more dominant constituent of $PM_{2.5}$ concentrations especially under RCP2.6 and RCP8.5 (Fig. 7 b-d).

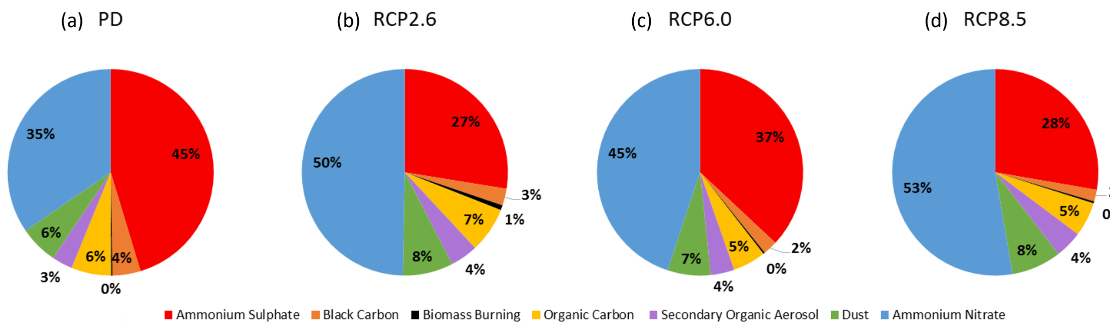

Figure 7: The individual components that add up to the total $PM_{2.5}$ concentration as simulated by the AQUM model for (a) present-day (2000s) and (b-d) future pathways following RCP2.6, RCP6.0 and RCP8.5.

The magnitude and spatial distribution of differences in ammonium sulphate concentrations between present-day and future for all three RCPs is similar to that of simulated total $PM_{2.5}$ concentrations with differences ranging from ~ -1 μg m^{-3} in Scotland to ~ -4.5 μg m^{-3} in central and eastern England (Fig. S1 of the supplement to this article).

In contrast, simulated ammonium nitrate concentrations over the UK are higher by ~0.25 μg m^{-3} to ~1 μg m^{-3} for all three RCPs compared to present-day (Fig. S2). For all future RCPs, UK SO_2 emissions decrease (Fig. 1) leading to a reduction in sulphate aerosol in the future. This reduction in SO_2 emissions in conjunction with increases in ammonia emissions under all three RCPs results in an overall increase in the simulated nitrate aerosols even though NO_x emissions in the UK are projected to decrease in the future.

These decreases in $PM_{2.5}$ concentrations in 2050s following the RCPs compared to 2000s are consistent with those of Hedegaard et al. (2013), suggesting that changes in anthropogenic emissions of SO_x and BC following RCP4.5 lead to a decrease in $PM_{2.5}$ concentrations across Europe. Under a European emission reduction scenario for 2020 based on the MEGAPOLI project, Chemel et al. (2014) find annual average $PM_{2.5}$ concentrations reductions > 2 μg m^{-3} over England due to reductions in SO_2, NOx and NMVOCs. Vieno et al. (2016) also suggest that under current legislation (CLE) emissions for 2030, surface annual-average $PM_{2.5}$ concentrations over the UK reduce by up to 2.8 μg m^{-3} compared to 2010. Furthermore, these findings on the impacts of SO_2 emission reductions on the ammonium sulphate and ammonium nitrate balance are similar to those reported in previous studies (e.g. Pye et al., 2009).

4 Future health burdens due to changes in UK emissions

Using the simulated annual mean MDA8 O_3, NO_2 and $PM_{2.5}$ concentrations discussed in Section 3 we first calculated the population-weighted pollutant concentrations, then estimated the regional attributable fraction (AF) of mortality, and mortality burdens (Section 4.1) associated with long-term exposure to each pollutant for present-day (2000s) and all three future pathways (2050s) following the method described in Section 2.3.

We estimate that across all UK regions the percentage (AF) of respiratory mortality associated with long-term exposure to annual mean MDA8 O_3 is higher for all three RCPs in 2050s compared to present-day estimates (refer to Fig. S3 in the Supplement to this manuscript). In contrast, for all RCPs the AF of mortality associated with long-term exposure to annual mean NO_2 and $PM_{2.5}$ concentrations is lower for 2050s relative to 2000s (Fig. S4 and S5, respectively). These differences are driven by differences in the respective population-weighted pollutant concentrations; annual mean MDA8 O_3 concentrations are higher for future pathways and annual mean NO_2 and $PM_{2.5}$ concentrations are lower.

The uncertainty estimates based on the 95 % confidence interval of all three CRFs used are relatively large such that the confidence range between present-day and all three future pathways overlap for most regions (Fig. S3-5). There are exceptions for specific regions following RCP8.5 (Fig. S3a-5a). This suggests a significant difference in the AF of mortality between present-day and RCP8.5. The large uncertainty is further reflected in the confidence intervals of differences between present-day and each future pathways (Fig. S3b-5b). For all regions under all three RCPs, the limit of the 95 % confidence interval of the difference is close to zero and in some regions includes both positive and negative AF values thus suggesting that AF differences are not significant (Fig. S3b-5b), although this is not the case for most of the regions under RCP8.5 (Fig. S3b-5b).

4.1 Health impacts associated with long-term exposure to annual mean MDA8 O_3

The estimated respiratory mortality burdens associated with long-term exposure to annual mean MDA8 O_3 are shown in Fig. 8a. The estimated total UK-wide present-day (2000s) mortality burden is 7,705 attributable deaths. Under all RCPs, regions which have high population totals, such as the North West England region, have high estimated mortality burdens in 2050s (Fig. 8a). For example, under RCP8.5, the North West and South East regions are the regions with the highest estimated mortality burden in 2050s (Fig. 8a). Under all three RCPs, the future total estimated mortality burden in 2050s is higher than present-day (positive differences, Fig. 8b), as a result of higher total AFs of respiratory mortality (Fig S3b). For RCP2.6 and RCP6.0 there are 2,710 and 2,529 additional attributable deaths respectively, and 5,396 additional attributable deaths for RCP8.5 (Fig. 8b).

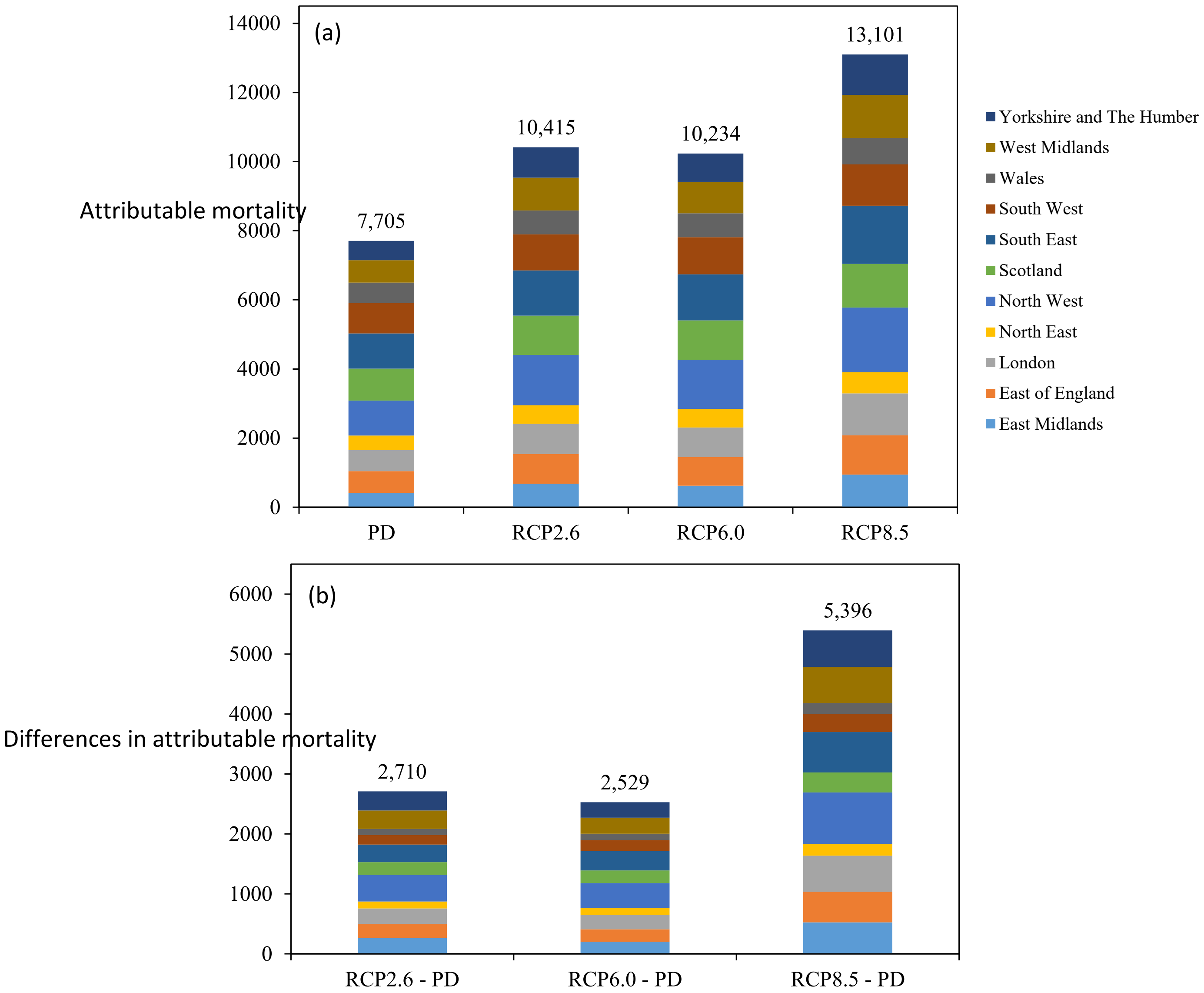

Figure 8: (a) UK annual respiratory attributable deaths associated with long-term exposure to annual mean MDA8 O_3 for present-day (PD – 2000s), and all three RCPs for 2050s. (b) Differences in UK annual attributable deaths between present-day and future under RCP2.6, RCP6.0 and RCP8.5. Colours indicate the annual attributbale deaths for each region in England and Scotland and Wales. N.B. no population projections are included in these future health burdens.

4.2 Health impacts associated with long-term exposure to annual mean NO_2

The present-day NO_2-related UK mortality burden is 25,278 attributable deaths (Fig. 9a), with regions having high present-day population (e.g. South East region) resulting in the highest total attributable mortality. Estimated mortality burdens for 2050s range from 9,496 to 15,860 attributable deaths for RCP8.5 and RCP6.0, respectively (Fig 9a). For all RCPs, the estimated mortality burdens in 2050s are lower compared to present-day; differences in mortality burdens range between 15,782 and 9,418 avoided attributable deaths for RCP6.0 and RCP8.5, respectively (Fig 9b). These reductions in estimated mortality burdens are driven by reductions in annual mean NO_2 concentrations. It should be noted that NO_2 effects could to some extent overlap with effects of long-term exposure to $PM_{2.5}$ as it is difficult to attribute health effects independently due to the typically strong correlation between these two pollutants (COMEAP, 2018).

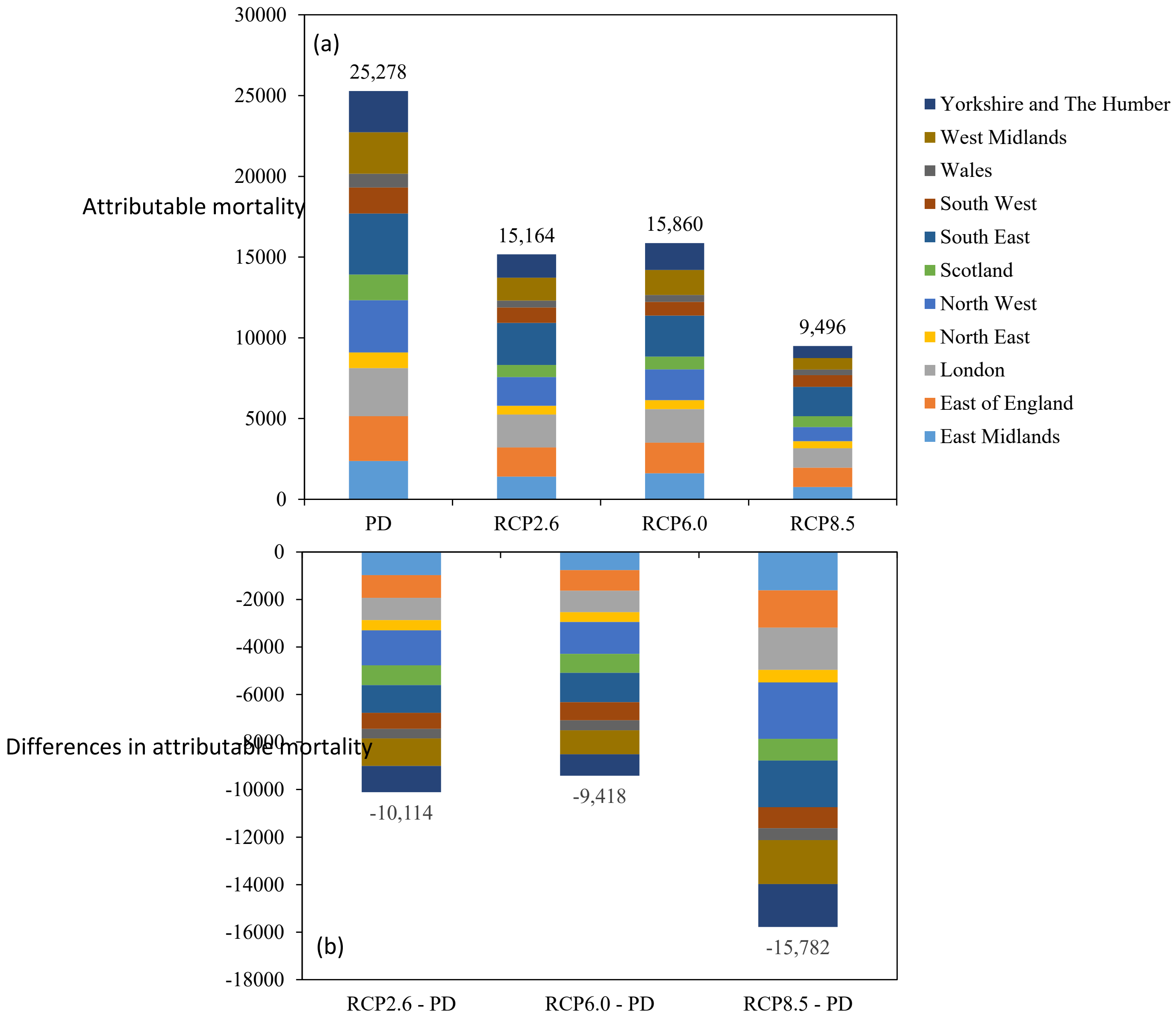

Figure 9: (a) UK annual attributable deaths associated with long-term exposrue to annual mean NO_2 for present-day (PD – 2000s), and all three RCPs in 2050s. (b) Differences in UK annual attributable deaths between present-day and future under RCP2.6, RCP6.0 and RCP8.5. Colours indicate the annual attributable deaths for each region in England and Scotland and Wales. N.B. no population projections are included in these future health burdens.

4.3 Health impacts associated with long-term exposure to annual mean $PM_{2.5}$

For present-day, we estimate that 32,996 attributable deaths in the UK are associated with long-term exposure to annual mean $PM_{2.5}$ concentrations (Fig 10a). The total UK mortality burdens for each of the future emission pathways are estimated at 24,034, 28,475 and 23,514 attributable deaths over the UK under RCP2.6, RCP6.0 and RCP8.5 in 2050s, respectively (Fig 10a). For all pathways, the estimated mortality burdens are highest in regions having a combination of a high PM2.5 concentration (Fig 10a) and a high population. For example, this

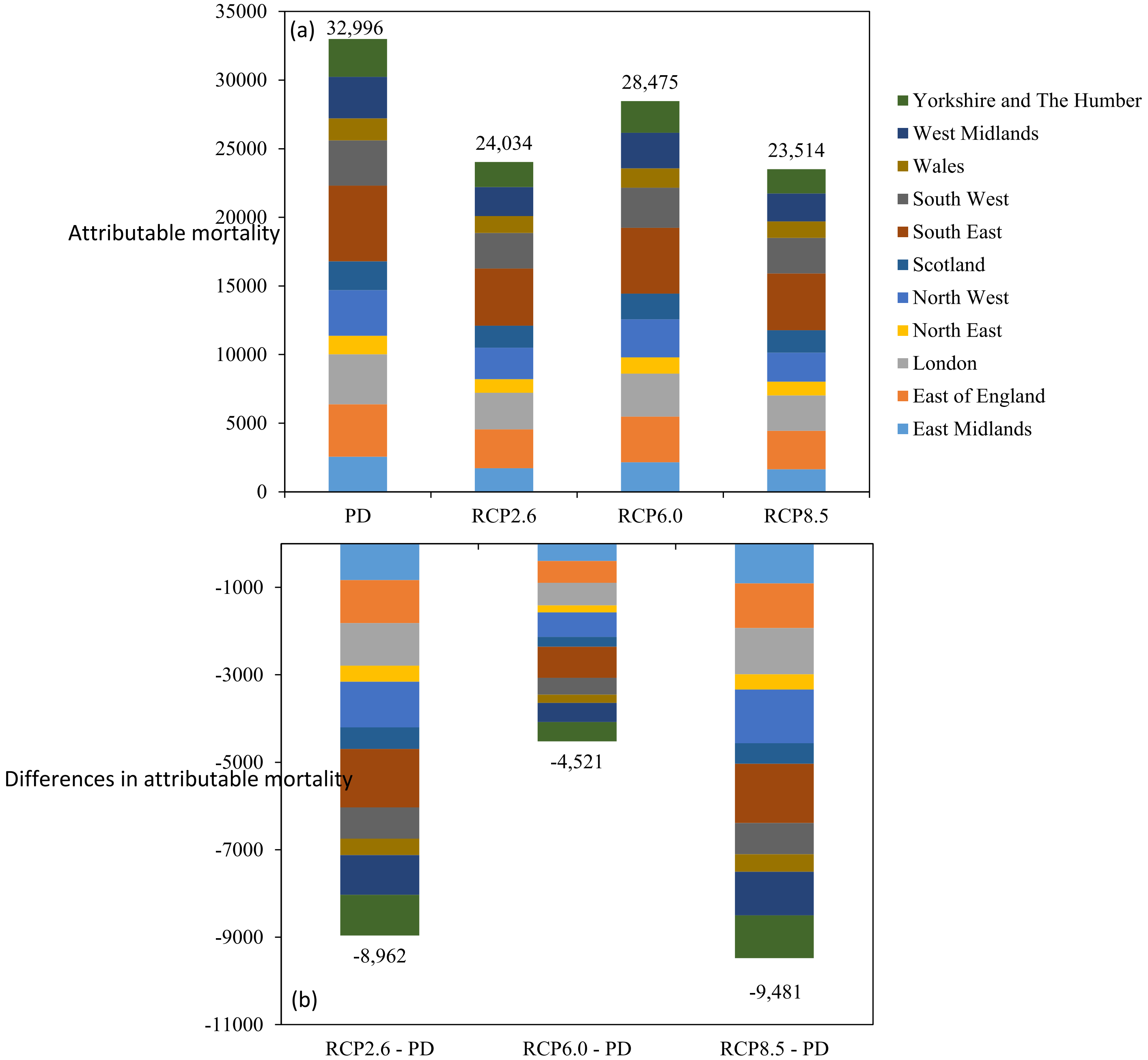

Figure 10: (a) UK annual attributable deaths associated with long-term exposure to annual mean $PM_{2.5}$ for present-day (PD – 2000s), and all three RCPs for 2050s. (b) Differences in UK annual attributbale deaths between present-day and future under RCP2.6, RCP6.0 and RCP8.5. Colours indicate the annual attributable deaths for each region in England and Scotland and Wales. N.B. no population ptojections are included in these future health burdens.

occurs in the South East, London and East of England regions (Fig 10a). As with differences in the AF of mortality (Fig. S5b), the estimated mortality burden for all RCPs in 2050s are lower compared to present-day (Fig 10b). The largest decreases in estimated attributable deaths associated with $PM_{2.5}$ occur under RCP2.6 and RCP 8.5 (8,962 and 9,481 avoided attributable deaths, respectively) while the smallest decreases occur under RCP6.0 (4,521 avoided attributable deaths).

4.4 Comparison of estimated future health impacts to literature

Models used to simulate future air pollutant concentrations generally have a coarse (~50km) horizontal resolution and studies typically only quantify impacts at a global or European scale, thus a direct comparison of results in this section with other studies is quantitatively limited. Instead we compare the direction of changes under different pathways.

Using a similar model set-up to this paper but including climate change effects, Pannullo et al. (2017) estimate the respiratory hospital admissions attributable to NO_2 in 2050s and find reductions of 1.7 %, 1.4 % and 2.4 % in hospital admissions under RCP2.6, RCP6.0 and RCP8.5, respectively in England (relative to average present-day respiratory hospital admissions per year across England – 613,052). This is to some extent similar to findings in this study where reductions in NO_2 concentrations under all RCPs in 2050s lead to reductions in corresponding mortality burdens across the UK compared to present-day. Williams et al. (2018) also estimated the health benefits for the UK following emission reductions under three energy consumption scenarios for 2050s, suggesting a decrease in annual mean NO_2 and $PM_{2.5}$ concentrations and their respective mortality burdens as a result of emission reductions in 2050s. In a broader European context, O_3-related mortality burdens are generally found to reduce for different RCPs in 2050s, with a slight increase for RCP8.5 as a result of increases in methane concentrations (e.g. Silva et al., 2016; West et al., 2013), not found in this study likely due to the finer model resolution of 12 km. $PM_{2.5}$ mortality burdens are generally found to decrease in the future as a result of reductions in primary and precursor emissions (e.g. Pozzer et al., 2017; Silva et al., 2016).

Overall, the findings in this study compare well with previous UK-based studies based on future emissions reductions, in terms of directions of change in future NO_2 and $PM_{2.5}$-related mortality burdens. Differences in the direction of change between the future O_3-related health burdens in this study and those found in different European studies (West et al. 2013; Silva et al. 2016) may be due to the coarser model resolutions used in these studies which may influence how the non-linear chemistry of NO_X and O_3 are captured (an increase vs. a decrease in O_3 related health burdens, respectively).

4.5 Sensitivity of estimated regional mortality burdens to future population projections

In this section we assess the sensitivity of future mortality burdens presented in Sections 4.1 - 4.3 to future population projections based on two shared socio-economic pathways (SSPs): SSP1 and SSP5 (a detailed description of changes in individual regions as well as results for SSP5 can be found in Section S3 of the Supplement to this manuscript). Under these pathways the population of the UK increases by a factor of 1.2 and

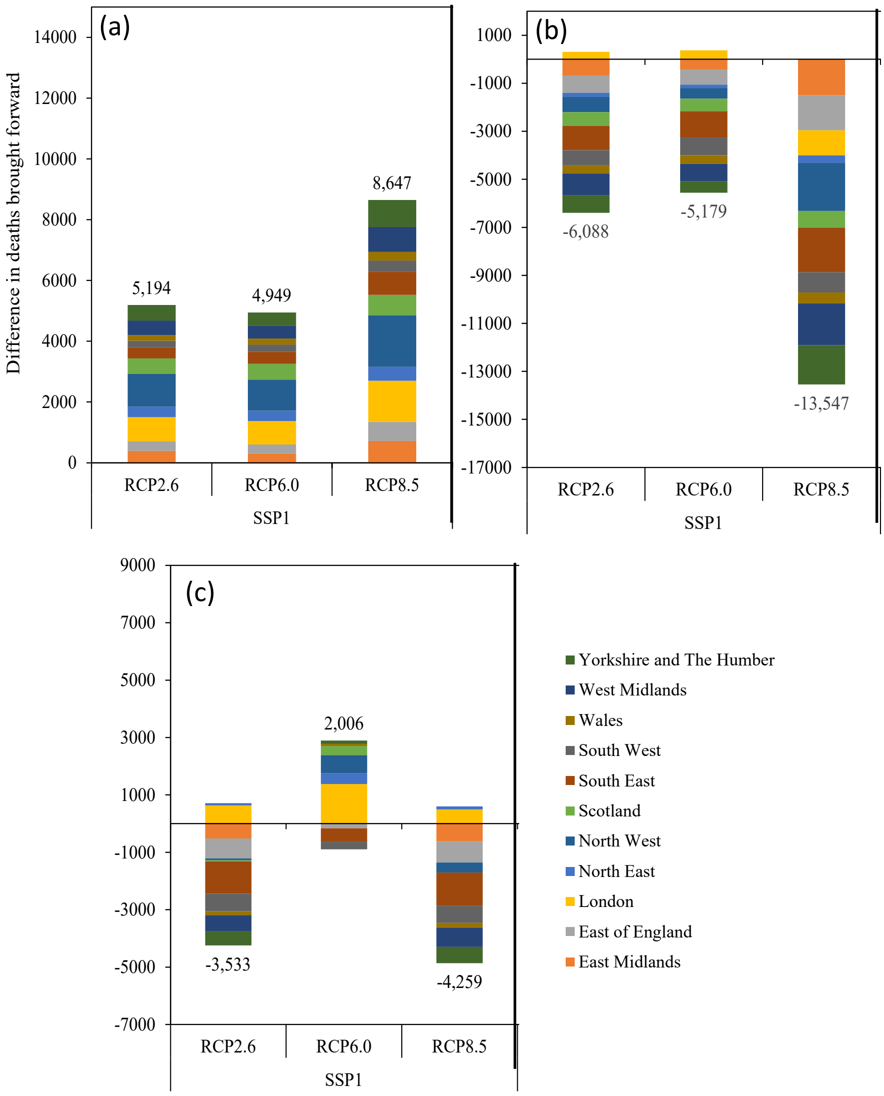

Figure 11: Differences in UK annual attributable deaths associated with long-term exposure to annual mean (a) MDA8 O_3 (b) NO_2 and (c) $PM_{2.5}$ between present-day (PD) for the 2000s pathway and the future under RCP2.6, RCP6.0 and RCP8.5 emission pathways including future population projections following SSP1. Colours indicate differences in annual attributable deaths for each region in England and Scotland and Wales.

1.4 from present-day, respectively (Section 2.3). Regional annual baseline mortality rates used to calculate future mortality burdens are assumed to be the same as for present-day.

Including the projected population pathways when estimating future mortality burdens based on the SSP1 and SSP5, results in higher future mortality burdens especially in densely populated regions such as London (Fig. 11a and Fig. S6a). For O_3, whose levels are estimated to increase in the future, the difference in mortality burdens between present-day and future is amplified when incorporating future population pathways (up to a factor of ~3). However, for NO_2 and $PM_{2.5}$, pollutants that are estimated to decrease under future UK emissions pathways, future increases in population may result in a change in sign as well as the magnitude for the total differences in UK mortality in the future (Fig. 11b-c and Fig. S6b-c). This result is broadly consistent with other studies. For example, Silva et al (2016) suggest that a larger population in 2050s magnifies the impact of changes in $PM_{2.5}$ concentrations and find the future global mortality burden of air pollution for $PM_{2.5}$ can exceed the current burden, even where $PM_{2.5}$ concentrations decrease.

5 Conclusions

The influence of changes in UK emissions following three RCPs; RCP2.6, RCP6.0 and RCP8.5 on simulated air pollutant concentrations of O_3, NO_2 and $PM_{2.5}$ over the UK for the 2050s relative to the 2000s, has been quantified for the first time using an air quality model (AQUM) at 12 km horizontal resolution. The corresponding changes in attributable fraction (AF) of mortality and mortality burdens associated with long-term exposure to annual mean MDA8 O_3, NO_2 and $PM_{2.5}$ between present-day and future pathways are estimated for different regions of the UK. In addition, the sensitivity of these health impact estimates to future population projections is also analysed.

Under all the three RCPs, O_3 concentrations increase across the entire UK due to substantial decreases in NOx emissions (ranging between -58 % and -73 %) resulting in less titration of O_3 by NO. Projected increases in CH_4 concentrations for the UK under RCP6.0 and RCP8.5 may also contribute to increases in O_3 concentrations. In contrast, under all RCPs, simulated annual mean NO_2 concentrations decrease across the UK. Annual mean $PM_{2.5}$ concentrations also decrease in 2050s for all RCPs, driven by large reductions in precursor emissions however, the $PM_{2.5}$ composition is noted to change for future pathways with ammonium nitrate increasingly becoming the more dominant constituent of $PM_{2.5}$. The lateral boundary conditions used in this study for all three RCPs are constant to the present-day run therefore, the impact of changes in regional and global emissions outside the UK are not accounted for. Results concerning the impact of changes in European emissions for the 2050s on

UK air pollution are variable. This typically depends on the models used as well as the source of precursors for the pollutant being studied. For example, for PM as well as gaseous pollutants such as NO_2, CO and SO_2, changes in local emissions are more likely to be dominant however, a larger impact from non-local sources may be expected for, for example O_3.

For all regions and under all three RCPs, the corresponding total respiratory mortality burden associated with long-term exposure to MDA8 O_3 between the RCPs is estimated to increase by 2,529 (RCP6.0) to 5,396 (RCP8.5) additional attributable deaths in the 2050s relative to the 2000s. Differences between present-day and future NO_2-related mortality burdens range between 9,418 and 15,782 avoided attributable deaths while differences in $PM_{2.5}$ related mortality burdens range from 4,521 to 9,481 avoided attributable deaths for RCP6.0 and RCP8.5, respectively. Even though the $PM_{2.5}$ concentrations are suggested to decrease for all three RCPs, we note that the $PM_{2.5}$ composition changes. Evidence on the toxicity and chemical composition of $PM_{2.5}$ is very limited. Thus, while our results suggest reductions in $PM_{2.5}$-related mortality for the 2050s under all three RCPs, we cannot estimate the difference in toxicity due to a change in $PM_{2.5}$ composition between present-day and future estimates.

Results show that future mortality burdens are sensitive to future population pathways for all pollutants. We note that, the mortality burdens presented in this study may be overestimate as the effect of exposure to NO_2 and $PM_{2.5}$ may not be independent as these pollutants are typically strongly correlated. In terms of uncertainty, the 95 % CI representing uncertainties associated with the CRF is quantified for the AF of mortality due to long-term exposure to O_3, NO_2 and $PM_{2.5}$. Results suggest that the uncertainty in the CRF has a substantial impact on the estimates of the AF of mortality for each UK region in terms of their magnitude and direction of change, except for four UK regions where significant differences in AF estimates are found between present-day and following RCP 8.5 in 2050s. We also note the large uncertainty in future emission scenarios and their drivers, for example, the recent changes in emissions across the globe driven by the limited movement of people following the unprecedented spread of the COVID-19 virus. In addition, although uncertainties in population projections are considered, uncertainties associated with the population demographics, e.g. an ageing population, are not considered. Changes in future baseline mortality rates are also neglected. These may in turn have a significant impact on the overall results as baseline mortality rates are expected to increase for an ageing and larger population total which will result in higher estimated mortality burdens. Furthermore, the estimates presented in this study are highly dependent on changes in local policies.

Nonetheless results in this study highlight the sensitivity of annual mean MDA8 O_3, NO_2 and $PM_{2.5}$ concentrations to reducing UK NO_x emissions which in turn drive changes in estimates of UK and regional mortality burdens associated with long-term exposure to these pollutants under three RCPs for the 2050s compared to the 2000s. In addition, mortality burdens are also shown to be sensitivity to future population pathways.

Data availability

The following data have been used for this study:

AQUM model results courtesy of UK Met Office facilities, Baseline mortality data for 2006 was obtained from the Office for National Statistics for England and Wales (ons.gov.uk) and from the National Records of Scotland (nrscotland.gov.uk).

Author Contribution

SF, RD and FO designed the experiments and SF carried them out. PA and LN developed the model code and SF performed the simulations. CH, HM and SV helped with the requisition of health-related data and advised on health impact assessment methodology. SF prepared the manuscript with contributions from all co-authors.

Competing Interests

The authors declare that they have no conflict of interest.

Acknowledgements

Sara Fenech's PhD was funded by Public Health England (PHE). The development of the United Kingdom Chemistry and Aerosol (UKCA) Model and Fiona M. O'Connor is supported by the Joint UK BEIS/Defra Met Office Hadley Centre Climate Programme (GA01101). Clare Heaviside is supported by a NERC fellowship (NE/R01440X/1). The research was part funded by the National Institute for Health Research Health Protection Research Unit (NIHR HPRU) in Environmental Change and Health at the London School of Hygiene and Tropical Medicine in partnership with Public Health England (PHE), and in collaboration with the University of Exeter, University College London, and the Met Office. The views expressed are those of the author(s) and not necessarily those of the NHS, the NIHR, the Department of Health or Public Health England.

References

Arneth, A., Miller, P.A., Scholze, M., Hickler, T., Schurgers, G., Smith, B., Prentice, I.C., 2007. CO2 inhibition of global terrestrial isoprene emissions: Potential implications for atmospheric chemistry. Geophys. Res. Lett. 34, 1–5.

Bellouin, N., Mann, G.W., Woodhouse, M.T., Johnson, C., Carslaw, K.S., Dalvi, M., 2013. Impact of the modal aerosol scheme GLOMAP-mode on aerosol forcing in the hadley centre global environmental model. Atmos. Chem. Phys. 13, 3027–3044.

Bellouin, N., Rae, J., Jones, A., Johnson, C., Haywood, J., Boucher, O., 2011. Aerosol forcing in the Climate Model Intercomparison Project (CMIP5) simulations by HadGEM2-ES and the role of ammonium nitrate. J. Geophys. Res. Atmos. 116, 1–25.

Brown, A., Milton, S., Cullen, M., Golding, B., Mitchell, J., Shelly, A., 2012. Unified modeling and prediction of weather and climate: A 25-year journey. Bull. Am. Meteorol. Soc. 93, 1865–1877.

Burnett, R., Chen, H., Fann, N., Hubbell, B., Pope, C.A., Frostad, J., Lim, S.S., Kan, H., Walker, K.D., Thurston, G.D., Hayes, R.B., Lim, C.C., Turner, M.C., Jerrett, M., Krewski, D., Gapstur, S.M., Diver, W.R., Ostro, B., Goldberg, D., Crouse, D.L., Martin, R. V, Peters, P., Pinault, L., Tjepkema, M., Donkelaar, A. Van, Villeneuve, P.J., Miller, A.B., Yin, P., Zhou, M., Wang, L., Janssen, N.A.H., Marra, M., Atkinson, R.W., Tsang, H., Quoc, T., Cannon, J.B., Allen, R.T., Hart, J.E., Laden, F., Cesaroni, G., Forastiere, F., Weinmayr, G., Jaensch, A., Nagel, G., Concin, H., Spadaro, J. V, 2018. Global estimates of mortality associated with long- term exposure to outdoor fine particulate matter. PNAS 115.

Burnett, R.T., Arden Pope, C., Ezzati, M., Olives, C., Lim, S.S., Mehta, S., Shin, H.H., Singh, G., Hubbell, B., Brauer, M., Ross Anderson, H., Smith, K.R., Balmes, J.R., Bruce, N.G., Kan, H., Laden, F., Prüss-Ustün, A., Turner, M.C., Gapstur, S.M., Diver, W.R., Cohen, A., 2014. An integrated risk function for estimating the global burden of disease attributable to ambient fine particulate matter exposure. Environ. Health Perspect. 122, 397–403.

Chemel, C., Fisher, B.E.A., Kong, X., Francis, X. V., Sokhi, R.S., Good, N., Collins, W.J., Folberth, G.A., 2014. Application of chemical transport model CMAQ to policy decisions regarding PM2.5 in the UK. Atmos. Environ. 82, 410–417.

Colette, A., Granier, C., Hodnebrog, Jakobs, H., Maurizi, A., Nyiri, A., Rao, S., Amann, M., Bessagnet, B., D'Angiola, A., Gauss, M., Heyes, C., Klimont, Z., Meleux, F., Memmesheimer, M., Mieville, A., Rouïl, L., Russo, F., Schucht, S., Simpson, D., Stordal, F., Tampieri, F., Vrac, M., 2012. Future air quality in Europe: A multi-model assessment of projected exposure to ozone. Atmos. Chem. Phys. 12, 10613–10630.

COMEAP, 2015. Interim Statement on Quantifying the Association of Long-Term Average Concentrations of Nitrogen Dioxide and Mortality.

COMEAP, 2018. Associations of long-term average concentrations of nitrogen dioxide with mortality.

Crouse, D.L., Peters, P.A., Hystad, P., Brook, J.R., van Donkelaar, A., Martin, R. V., Villeneuve, P.J., Jerrett, M., Goldberg, M.S., Arden Pope, C., Brauer, M., Brook, R.D., Robichaud, A., Menard, R., Burnett, R.T., 2015. Ambient PM2.5, O3, and NO2exposures and associations with mortality over 16 years of follow-up in the canadian census health and environment cohort (CanCHEC). Environ. Health Perspect. 123, 1180–1186.

Defra, 2013. Defra Phase 2 urban model evaluation, King's College London.

Eurostat, 2017. Population Projections [WWW Document]. Eurostat, Stat. Off. Eur. Union. URL http://ec.europa.eu/eurostat/web/population-demography-migration-projections/population-projections-data (accessed 7.4.18).

Faustini, A., Rapp, R., Forastiere, F., 2014. Nitrogen dioxide and mortality: review and meta-analysis of long-term studies. Eur. Respir. J. 3, 744–753.

Flemming, J., Inness, a., Flentje, H., Huijnen, V., Moinat, P., Schultz, M.G., Stein, O., 2009. Coupling global chemistry transport models to ECMWF's integrated forecast system. Geosci. Model Dev. Discuss. 2, 763–795.

Heal, M.R., Heaviside, C., Doherty, R.M., Vieno, M., Stevenson, D.S., Vardoulakis, S., 2013. Health burdens of surface ozone in the UK for a range of future scenarios. Environ. Int. 61, 36–44.

Hedegaard, G.B., Christensen, J.H., Brandt, J., 2013. The relative importance of impacts from climate change vs. emissions change on air pollution levels in the 21st century. Atmos. Chem. Phys. 13, 3569–3585.

Hoek, G., Krishnan, R.M., Beelen, R., Peters, A., Ostro, B., Brunekreef, B., Kaufman, J.D., 2013. Long-term air pollution exposure and cardio- respiratory mortality: a review. Environ. Heal. 12, 43.

Im, U., Christensen, J.H., Geels, C., Hansen, K.M., Brandt, J., Solazzo, E., Alyuz, U., Balzarini, A., Baro, R., Bellasio, R., Bianconi, R., Bieser, J., Colette, A., Curci, G., Farrow, A., Flemming, J., Fraser, A., Jimenez-Guerrero, P., Kitwiroon, N., Liu, P., Nopmongcol, U., Palacios-Peña, L., Pirovano, G., Pozzoli, L., Prank, M., Rose, R., Sokhi, R., Tuccella, P., Unal, A., Vivanco, M.G., Yarwood, G., Hogrefe, C., Galmarini, S., 2018. Influence of anthropogenic emissions and boundary conditions on multi-model simulations of major air pollutants over Europe and North America in the framework of AQMEII3. Atmos. Chem. Phys. Discuss. 1–43.

Jerrett, M., Burnett, R.T., Pope, C.A., Ito, K., Thurston, G., Krewski, D., Shi, Y., Calle, E., Thun, M., 2009. Long-Term Ozone Exposure and Mortality. N. Engl. J. Med. 360, 1085–1095.

Jones, A., Roberts, D.L., Woodage, M.J., Johnson, C.E., 2001. Indirect sulphate aerosol forcing in a climate model with an interactive sulphur cycle. J. Geophys. Res. 106, 20293–20310.

Jones, B., O'Neill, B.C., 2017. Global Population Projection Grids Based on Shared Socioeconomic Pathways (SSPs), 2010-2100. [WWW Document]. Socioecon. Data Appl. Cent. URL https://doi.org/10.7927/H4RF5S0P (accessed 7.3.18).

Krewski, D., 2009. Extended Follow-Up and Spatial Analysis of the American Cancer Society Study Linking Particulate Air Pollution and Mortality, Health Effects Institute.

Lamarque, J.F., Bond, T.C., Eyring, V., Granier, C., Heil, a., Klimont, Z., Lee, D., Liousse, C., Mieville, a., Owen, B., Schultz, M.G., Shindell, D., Smith, S.J., Stehfest, E., Van Aardenne, J., Cooper, O.R., Kainuma, M., Mahowald, N., McConnell, J.R., Naik, V., Riahi, K., Van Vuuren, D.P., 2010. Historical (1850-2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: Methodology and application. Atmos. Chem. Phys. 10, 7017–7039.

Lamarque, J.F., Kyle, P.P., Meinshausen, M., Riahi, K., Smith, S.J., van Vuuren, D.P., Conley, A.J., Vitt, F., 2011. Global and regional evolution of short-lived radiatively-active gases and aerosols in the Representative Concentration Pathways. Clim. Change 109, 191–212.

Masui, T., Matsumoto, K., Hijioka, Y., Kinoshita, T., Nozawa, T., Ishiwatari, S., Kato, E., Shukla, P.R., Yamagata, Y., Kainuma, M., 2011. An emission pathway for stabilization at 6 Wm-2radiative forcing. Clim. Change 109, 59–76.

Morgenstern, O., Braesicke, P., O'Connor, F.M., Bushell, A.C., Johnson, C.E., Osprey, S.M., Pyle, J.A., 2009. Evaluation of the new UKCA climate-composition model – Part 1: The stratosphere. Geosci. Model Dev. 2, 43–57.

Nakicenovic, N., Swart, R., Alcamo, J., Davis, G., Vries, B., Fenhann, J., Gaffin, S., Gregory, K., Gruebler, A., 2000. IPCC Special Report of Working Group III on Emission Scenarios, Working Group III of the Intergovenmental Panel on Climate Change (IPCC). Cambridge.

Neal, L.S., Dalvi, M., Folberth, G., McInnes, R.N., Agnew, P., O'Connor, F.M., Savage, N.H., Tilbee, M.,

2017. A description and evaluation of an air quality model nested within global and regional composition-climate models using MetUM. Geosci. Model Dev. 10, 3941–3962.

Neu, J.L., Prather, M.J., Penner, J.E., 2007. Global atmospheric chemistry: Integrating over fractional cloud cover. J. Geophys. Res. Atmos. 112, 1–12.

O'Connor, F.M., Johnson, C.E., Morgenstern, O., Abraham, N.L., Braesicke, P., Dalvi, M., Folberth, G. a., Sanderson, M.G., Telford, P.J., Voulgarakis, A., Young, P.J., Zeng, G., Collins, W.J., Pyle, J.A., 2014. Evaluation of the new UKCA climate-composition model-Part 2: The troposphere. Geosci. Model Dev. 7, 41–91.

ONS, 2017. National Population Projections: 2016-based statistical bulletin 1–10.

Pacifico, F., Folberth, G.A., Jones, C.D., Harrison, S.P., Collins, W.J., 2012. Sensitivity of biogenic isoprene emissions to past, present, and future environmental conditions and implications for atmospheric chemistry. J. Geophys. Res. Atmos. 117, 1–13.

Pacifico, F., Harrison, S.P., Jones, C.D., Arneth, A., Sitch, S., Weedon, G.P., Barkley, M.P., Palmer, P.I., Serça, D., Potosnak, M., Fu, T.M., Goldstein, A., Bai, J., Schurgers, G., 2011. Evaluation of a photosynthesis-based biogenic isoprene emission scheme in JULES and simulation of isoprene emissions under present-day climate conditions. Atmos. Chem. Phys. 11, 4371–4389.

Pannullo, F., Lee, D., Neal, L., Dalvi, M., Agnew, P., O'Connor, F.M., Mukhopadhyay, S., Sahu, S., Sarran, C., 2017. Quantifying the impact of current and future concentrations of air pollutants on respiratory disease risk in England. Environ. Heal. 16, 29.

Pozzer, A., Tsimpidi, A.P., Karydis, V.A., de Meij, A., Lelieveld, J., 2017. Impact of agricultural emission reductions on fine particulate matter and public health. Atmos. Chem. Phys. Discuss. 1–19.

Pye, H.O.T., Liao, H., Wu, S., Mickley, L.J., Jacob, D.J., Henze, D.J., Seinfeld, J.H., 2009. Effect of changes in climate and emissions on future sulfate-nitrate-ammonium aerosol levels in the United States. J. Geophys. Res. Atmos. 114.

Riahi, K., Rao, S., Krey, V., Cho, C., Chirkov, V., Fischer, G., Kindermann, G., Nakicenovic, N., Rafaj, P., 2011. RCP 8.5-A scenario of comparatively high greenhouse gas emissions. Clim. Change 109, 33–57.

Riahi, K., van Vuuren, D.P., Kriegler, E., Edmonds, J., O'Neill, B.C., Fujimori, S., Bauer, N., Calvin, K., Dellink, R., Fricko, O., Lutz, W., Popp, A., Cuaresma, J.C., KC, S., Leimbach, M., Jiang, L., Kram, T., Rao, S., Emmerling, J., Ebi, K., Hasegawa, T., Havlik, P., Humpenöder, F., Da Silva, L.A., Smith, S., Stehfest, E., Bosetti, V., Eom, J., Gernaat, D., Masui, T., Rogelj, J., Strefler, J., Drouet, L., Krey, V., Luderer, G., Harmsen, M., Takahashi, K., Baumstark, L., Doelman, J.C., Kainuma, M., Klimont, Z., Marangoni, G., Lotze-Campen, H., Obersteiner, M., Tabeau, A., Tavoni, M., 2017. The Shared Socioeconomic Pathways and their energy, land use, and greenhouse gas emissions implications: An overview. Glob. Environ. Chang. 42, 153–168.

Savage, N.H., Agnew, P., Davis, L.S., Ordonez, C., 2013. Air quality modelling using the Met Office Unified Model (AQUM OS24-26): model description and initial evaluation. Geosci. Model Dev. 6, 353–372.

Silva, R.A., West, J.J., Lamarque, J.F., Shindell, D.T., Collins, W.J., Dalsoren, S., Faluvegi, G., Folberth, G., Horowitz, L.W., Nagashima, T., Naik, V., Rumbold, S.T., Sudo, K., Takemura, T., Bergmann, D., Cameron-Smith, P., Cionni, I., Doherty, R.M., Eyring, V., Josse, B., MacKenzie, I.A., Plummer, D., Righi, M., Stevenson, D.S., Strode, S., Szopa, S., Zengast, G., 2016. The effect of future ambient air pollution on human premature mortality to 2100 using output from the ACCMIP model ensemble. Atmos. Chem. Phys. 16, 9847–9862.

Squire, O.J., Archibald, A.T., Griffiths, P.T., Jenkin, M.E., Smith, D., Pyle, J.A., 2015. Influence of isoprene chemical mechanism on modelled changes in tropospheric ozone due to climate and land use over the 21st century. Atmos. Chem. Phys. 15, 5123–5143.

Thomson, A.M., Calvin, K. V., Smith, S.J., Kyle, G.P., Volke, A., Patel, P., Delgado-Arias, S., Bond-Lamberty, B., Wise, M.A., Clarke, L.E., Edmonds, J.A., 2011. RCP4.5: A pathway for stabilization of radiative forcing by 2100. Clim. Change 109, 77–94.

Turner, M.C., Jerrett, M., Pope, C.A., Krewski, D., Gapstur, S.M., Diver, W.R., Beckerman, B.S., Marshall, J.D., Su, J., Crouse, D.L., Burnett, R.T., 2015. Long-Term Ozone Exposure and Mortality in a Large Prospective Study. Am. J. Respir. Crit. Care Med. 193, 1134–1142.

van Vuuren, D.P., Edmonds, J., Kainuma, M., Riahi, K., Thomson, A., Hibbard, K., Hurtt, G.C., Kram, T., Krey, V., Lamarque, J.F., Masui, T., Meinshausen, M., Nakicenovic, N., Smith, S.J., Rose, S.K., 2011a. The representative concentration pathways: An overview. Clim. Change 109, 5–31.

van Vuuren, D.P., Stehfest, E., den Elzen, M.G.J., Kram, T., van Vliet, J., Deetman, S., Isaac, M., Goldewijk, K.K., Hof, A., Beltran, A.M., Oostenrijk, R., van Ruijven, B., 2011b. RCP2.6: Exploring the possibility to keep global mean temperature increase below 2°C. Clim. Change 109, 95–116.

Vieno, M., Heal, M.R., Williams, M.L., Carnell, E.J., Nemitz, E., Stedman, J.R., Reis, S., 2016. The sensitivities of emissions reductions for the mitigation of UK PM2.5. Atmos. Chem. Phys. 16, 265–276.

West, J.J., Smith, S.J., Silva, R. a, Naik, V., Zhang, Y., Adelman, Z., Fry, M.M., Anenberg, S., Horowitz, L.W., Lamarque, J.-F., 2013. Co-benefits of Global Greenhouse Gas Mitigation for Future Air Quality and Human Health. Nat. Clim. Chang. 3, 885–889.

WHO, 2013. Health Risks of Air Pollution in Europe – HRAPIE project: Recommendations for concentration-response functions for cost-benefit analysis of particulate matter, ozone and nitrogen dioxide.

Wild, O., Fiore, A.M., Shindell, D.T., Doherty, R.M., Collins, W.J., Dentener, F.J., Schultz, M.G., Gong, S., Mackenzie, I.A., Zeng, G., Hess, P., Duncan, B.N., Bergmann, D.J., Szopa, S., Jonson, J.E., Keating, T.J., Zuber, A., 2012. Modelling future changes in surface ozone: A parameterized approach. Atmos. Chem. Phys. 12, 2037–2054.

Williams, M.L., Lott, M.C., Kitwiroon, N., Dajnak, D., Walton, H., Holland, M., Pye, S., Fecht, D., Toledano, M.B., Beevers, S.D., 2018. The Lancet Countdown on health benefits from the UK Climate Change Act: a modelling study for Great Britain. Lancet Planet. Heal. 2, e202–e213.

Supplement

S1. The emissions driven impact on ammonium sulphate and nitrate concentrations in the UK

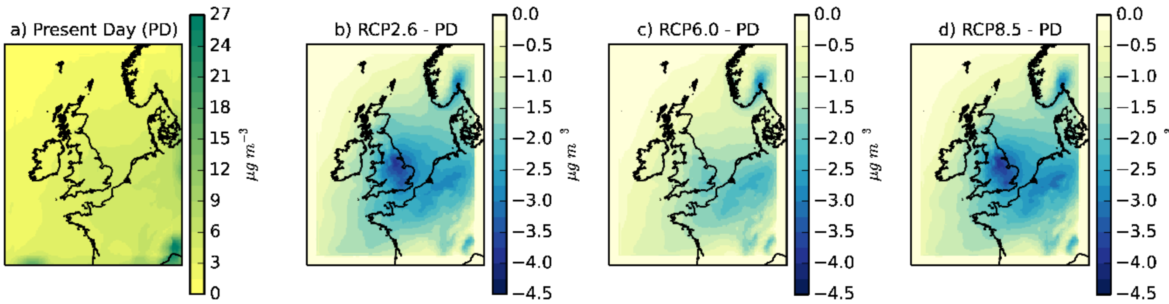

Figure S1: Simulated annual mean ammonium sulphate concentrations for (a) present-day (PD), (b-d) differences in simulated annual mean annual mean ammonium sulphate concentrations between present-day (2000s) and future (2050s) for each future pathway

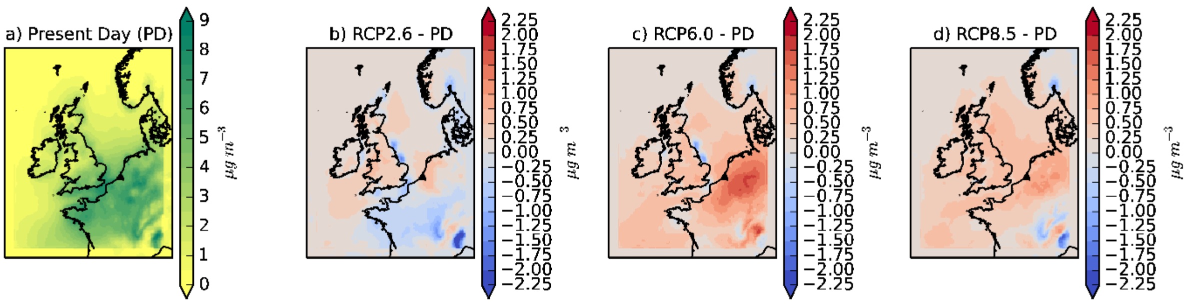

Figure S2: Simulated annual mean ammonium nitrate concentrations for (a) present-day (PD), (b-d) differences in simulated annual mean annual mean ammonium nitrate concentrations between present-day (2000s) and future (2050s) for each future pathway

S2. Health impacts associated with long-term exposure to annual mean pollutant concentrations

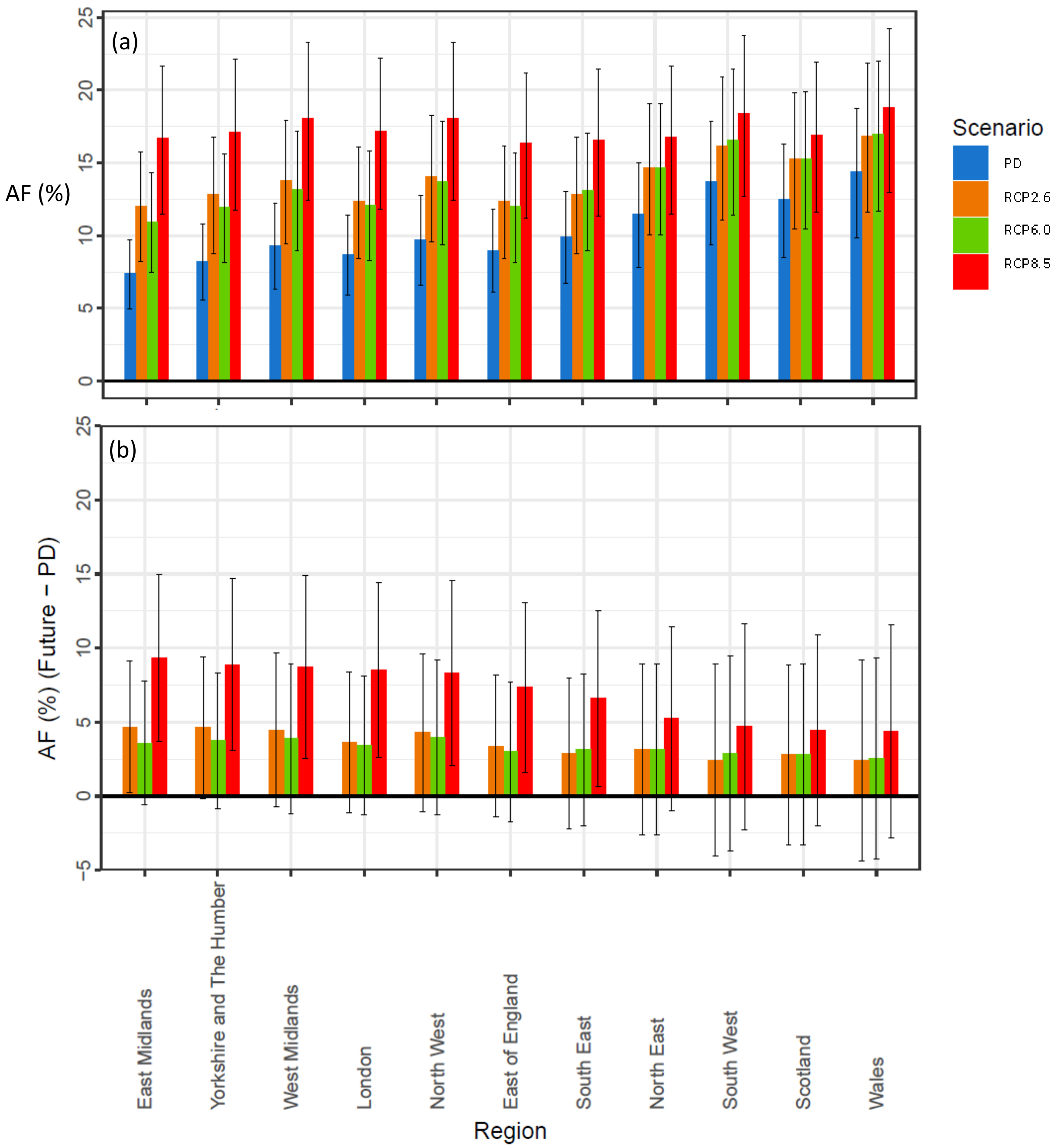

Figure S3: (a) Attributable fraction (AF) of respiratory mortality associated with long-term exposure to annual mean MDA8 O_3 for present-day (PD – 2000s) and each RCP for 2050s expressed as a percentage of total annual respiratory mortality. (b) Difference in AF between present-day and future expressed as a percentage for each regions in England, Scotland and Wales ($AF_{future} – AF_{present\text{-}day}$). Error bars show the 95% CI which represents uncertainties associated only with the concentrations-response coefficient used.

695

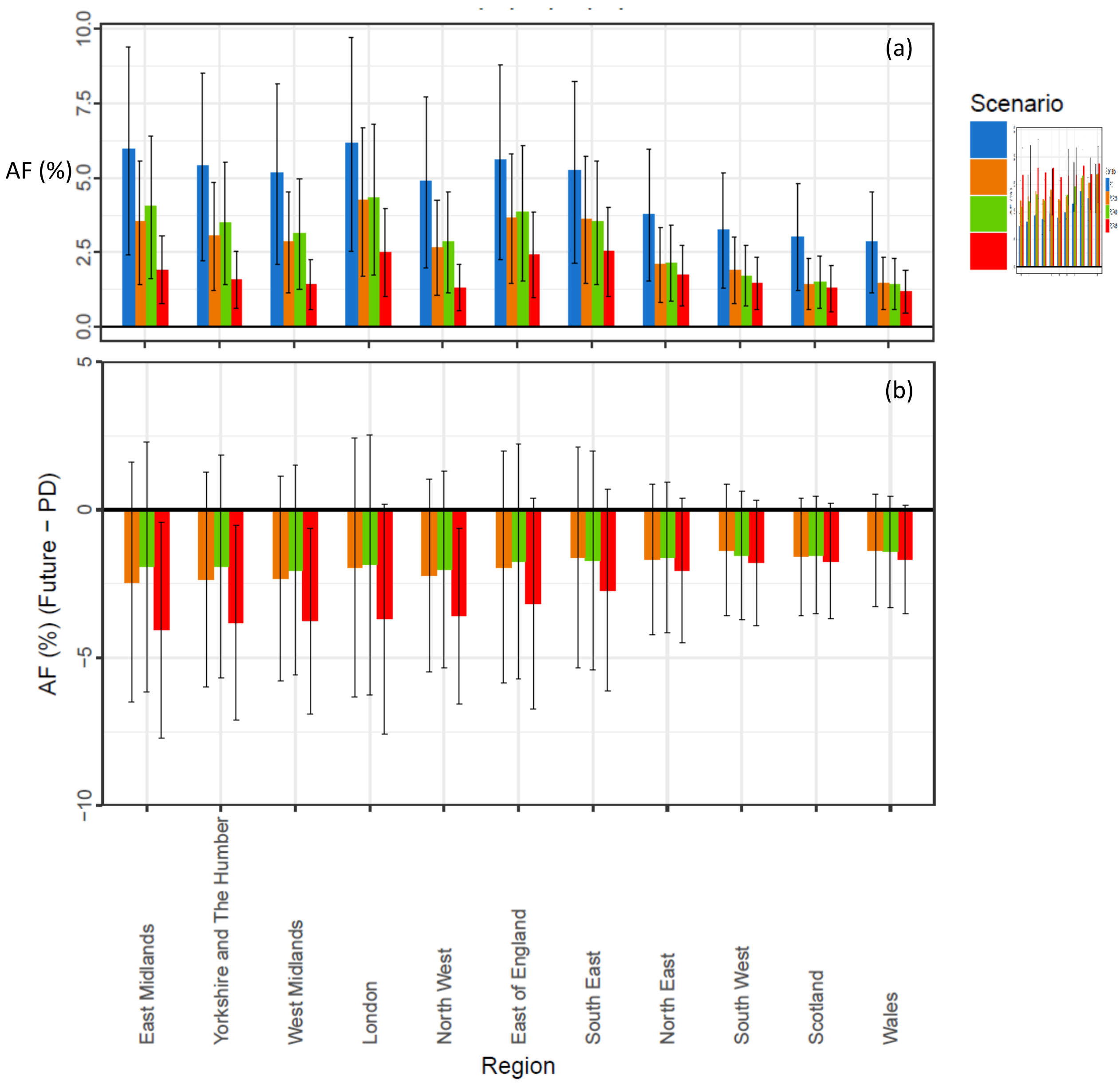

Figure S4: (a) Attributable fraction (AF) of all-cause (excluding external) mortality associated with long-term exposure to annual mean NO_2 for present-day (PD) in 2000s and each RCP for 2050s expressed as a percentage of total annual respiratory mortality. (b) Difference in AF between present-day and future expressed as a percentage for each regions in England, Scotland and Wales ($AF_{future} – AF_{present\text{-}day}$). Error bars show the 95% CI which represents uncertainties associated only with the concentrations-response coefficient used.

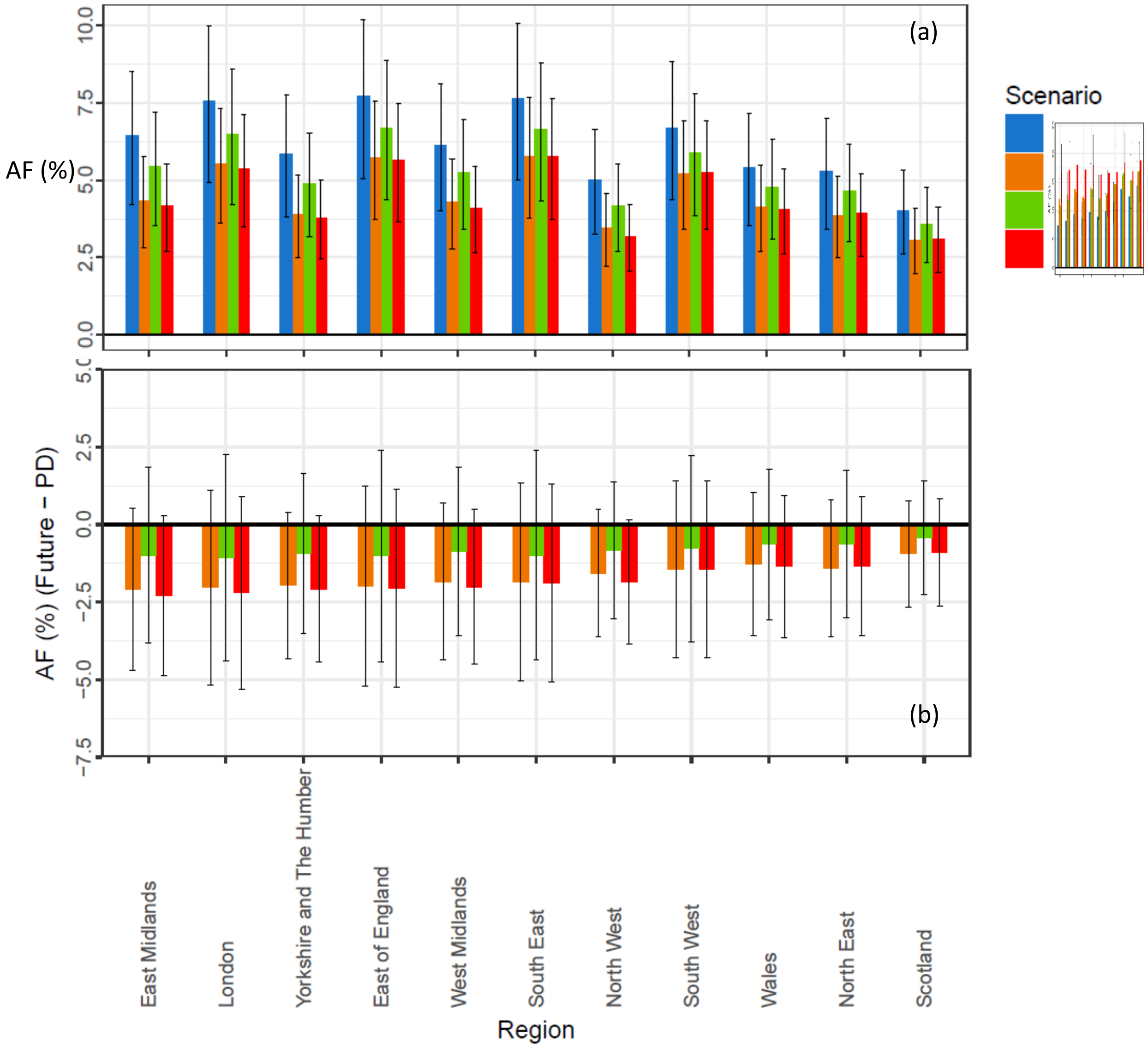

Figure S5: (a) Attributable fraction (AF) associated with long-term exposure to annual mean $PM_{2.5}$ for present-day (PD – 2000s) and each future RCP for 2050s expressed as a percentage of total annual respiratory mortality. (b) Difference in AF between present-day and future expressed as a percentage for each regions in England, Scotland and Wales (AF_{future} – $AF_{present\text{-}day}$). Error bars show the 95% CI which represents uncertainties associated only with the concentrations-response coefficient used.

700

S3. Sensitivity of estimated regional mortality burdens to future population projections

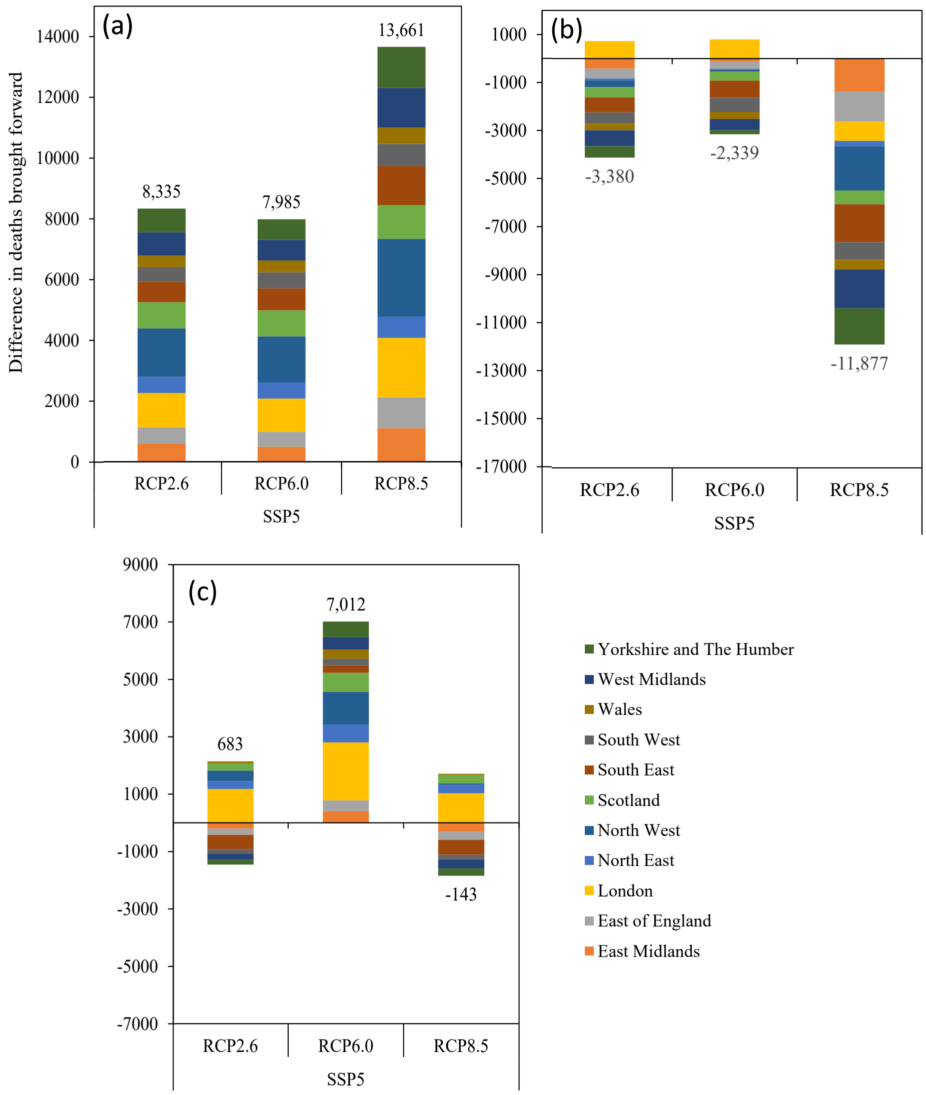

Figure S6: Differences in UK annual attributable deaths associated with long-term exposure to annual mean (a) MDA8 O_3 (b) NO_2 and (c) $PM_{2.5}$ between present-day (PD) for the 2000s pathway and the future under RCP2.6, RCP6.0 and RCP8.5 including future population projections following SSP5. Colours indicate differences in annual attributable deaths for each region in England and Scotland and Wales.

Differences in the estimated UK mortality burdens between present-day (2000s) (using present day population totals) and 2050s (including the SSP1 and SSP5 population projections) are presented in Fig. 11 and Fig. S6a, respectively. Using population projections that follow SSP1, the estimated differences in the UK mortality burden due to O_3 between present-day and future ranges from 4,949 in RCP6.0 to 8,647 in RCP8.5 (Fig. 11a), while differences range from 7,985 to 13,661 additional attributable deaths for SSP5 (Fig. S6a). For all RCPs under both population projections, the differences in estimated MDA8 O_3- related mortality burdens between present-day and future are amplified compared to the differences in mortality burdens with static populations levels (c.f. Fig 9b and Fig S6a). Future O_3-related mortality burdens are further amplified under SSP5 with results up to a factor of ~3 greater in 2050s than if population totals are kept at present-day levels. Under both SSPs, all regions exhibit increases in O_3-related mortality burden following the different RCPs in 2050s. There are large increases in London especially under RCP8.5 following the SSP5 population pathway (Yellow box Fig. S6a)

Differences in UK regional mortality burdens associated with long-term exposure to annual mean NO_2 between present-day and future estimates including future population pathways range from 5,179 (RCP6.0) to 13,547 (RCP8.5) avoided attributable deaths for SSP1 (Fig. 11b) and from 2,339 (RCP6.0) to 11,877 (RCP8.5) for SSP5 (Fig. S6b). When including these future SSP population projections estimated future mortality burdens in 2050s are higher compared to when using present-day population levels. Therefore, reductions in the UK mortality burden between present-day and all three future pathways are diminished (Fig. S6b). Under SSP1, differences in NO_2-related mortality burdens in 2050s vary from those using present day population totals by up to a factor of 0.9, and under SSP5 by up to a factor of 0.8. For most of the regions, the mortality burdens in 2050s are lower compared to present-day when accounting for future population projections (Fig S6b). The exception is London for SSP5, where the future NO_2-realted health burden is higher compared to present-day for RCP2.6 and RCP6.0 (yellow box; Fig S6b). This increase in the NO_2-related mortality burden occurs despite estimated reductions in NO_2 concentrations in London under all three RCPs, highlighting the impact of the large increase in the future population for this region (from 4.5 million people for present-day to 8.3 million people for 2050s in London under SSP5).

The reduction in $PM_{2.5}$-related mortality is 0.4 times that estimated using present-day population totals, indicating that total UK-wide future mortality burdens are still lower compared to present-day under the RCPs that incorporate population increases in 2050s. As a result of a further increase in total population for SSP5, the estimated future mortality burdens are higher overall compared to present-day for RCP2.6 and RCP6.0 with 683 and 7,012 additional attributable deaths (Fig. S6c). In contrast, for most of the regions under RCP8.5 with SSP5 population levels for 2050s, the $PM_{2.5}$-related UK mortality burden is lower overall compared to present-day with a total of 143 avoided attributable deaths (Fig. S6c).