Meteorological drivers and mortality associated with O3 and PM2.5 air pollution episodes in the UK in 2006

Meteorological drivers and mortality associated with O_3 and $PM_{2.5}$ air pollution episodes in the UK in 2006

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Abstract.

In this study we examine the meteorological drivers resulting in concurrent high levels of ozone (O_3) and particulate matter smaller than 2.5 µm in diameter ($PM_{2.5}$) during two five-day air pollution episodes in 2006 (1st - 5th July and 18th – 22nd July) using an air quality model (AQUM) at 12 km horizontal resolution to simulate air pollutant concentrations. The resultant UK health burden associated with short-term exposure to simulated maximum daily 8-hour O_3 (MDA8 O_3) and daily mean $PM_{2.5}$ is estimated at the national and regional level.

Both episodes were found to be driven by anticyclonic conditions with light easterly and south easterly winds and high temperatures that aided pollution build up in the UK. The estimated total mortality burden associated with short-term exposure to MDA8 O_3 is similar during the chosen episodes with about 70 daily deaths brought forward (summed across the UK) during the first and second episode, respectively. The estimated health burden associated with short-term exposure to daily mean $PM_{2.5}$ concentrations differs between the first and second episode resulting in about 43 and 36 daily deaths brought forward, respectively. The corresponding percentage of all-cause mortality due to short-term exposure to MDA8 O_3 and daily mean $PM_{2.5}$ during these two episodes and across the UK regions, ranges from 3.4% to 5.2% and from 1.6% to 3.9%, respectively. The attributable percentage of all-cause mortality differs between the regions depending on the pollution levels in each episode, but the overall estimated health burdens are highest in regions with higher population totals. We estimate that during these episodes the short-term exposure to MDA8 O_3 and daily mean $PM_{2.5}$ is between 36-38% and 39-56% higher, respectively, than if the pollution levels represented typical

seasonal-mean concentrations. This highlights the potential of air pollution episodes to have substantial short-term impacts on public health.

Key Words.

$PM_{2.5}$; O_3; Air pollution episode; Health impact assessment

1. Introduction

Air pollution has been identified as one of the top global mortality risk factors by the 2015 Global Burden of Disease (GBD, 2016). Short-term exposure to ozone (O_3) and particulate matter with an aerodynamic diameter less than 2.5 μm ($PM_{2.5}$) has been linked to negative effects on lung function (Chen et al., 2015), increased hospital admissions and mortality (Atkinson et al., 2014; Bell et al., 2006, 2005; COMEAP, 2015; Di et al., 2017; Ito et al., 2005; Levy et al., 2005).

Meteorology is a key factor in determining concentrations of O_3 and $PM_{2.5}$, through its impact on chemical reaction rates via temperature, deposition of pollutants, boundary layer depth, stagnation of air and long-range transport. In the northern mid-latitudes and over the UK, the highest O_3 concentrations typically occur in spring and summer (e.g. Anderson et al., 1996; Derwent et al., 1998; Monks, 2000), while the highest $PM_{2.5}$ levels occur most often in spring, autumn and winter (e.g. AQEG, 2012; Harrison et al., 2012a; Harrison et al., 2012b). However, in anticyclonic weather conditions with low wind speeds, high levels of these two pollutants may occur concurrently (Fischer et al., 2004; Stedman, 2004) and in summer may also be associated with heatwaves (Schnell and Prather, 2017; Solberg et al., 2008; Tong et al., 2010). Analysing the mechanisms responsible for the O_3 build-up over South East England during the August 2003 heatwave using the Community Multiscale Air Quality (CMAQ) model, Francis et al. (2011) found that convergence of westerly and easterly flows over the UK led to trapping of transported O_3 from mainland Europe, thus leading to increased O_3 levels. Using surface and satellite observations, Pope et al. (2016) also found that in the summer period (April-September 2006), anticyclonic conditions with low wind speeds and easterly flows significantly enhanced O_3 concentrations over the UK relative to summer-time average values and further show that the UK Met Office regional air quality model (AQUM) successfully reproduces UK increased O_3 concentrations under such synoptic conditions for this period.

Several studies have examined the impact of short-term exposure to O_3 and particulate matter (PM) on human health during air pollution episodes. Rooney et al. (1998) estimated that 619 extra deaths occurred during the heatwave between July and August 1995 in England and Wales of which 62% of the excess mortality was attributable to concurrent increases in air pollution. During the first two weeks of August 2003, a major heatwave occurred across much of Europe with temperatures in the UK reaching a record of 38.5 °C associated with a persistent high pressure system over Europe (Johnson et al., 2005; Lee et al., 2006; Solberg et al., 2008; Vieno et al., 2010). For the first two weeks in August 2003 in England and Wales, an estimated 83 and 29 deaths per day were associated with short-term exposure to daily maximum 8hr running mean (MDA8) O_3 and 24 hour mean PM_{10} (particulate matter with an aerodynamic diameter less than 10 μm), respectively (Stedman, 2004). This represented an increase of 38 (O_3) and 13 (PM_{10}) deaths per day compared with the previous year. In the Netherlands, Fischer et al. (2004) estimated that between June and August 2003, 15 and 16 daily deaths brought forward were associated with short-term exposure to O_3 and PM_{10}, respectively. Compared to the O_3 and PM_{10}-realted deaths brought forward between June and August 2000, approximately 4 and 2 additional daily deaths were brought forward in 2003.

In a recent study over the UK, a spring time air pollution episode in 2014 (totalling 10 days) was associated with ~ 60 daily deaths brought forward from short-term exposure to $PM_{2.5}$ when assuming a 1.04% increase in mortality for a 10 μg m^{-3} increase in 24-hour mean $PM_{2.5}$ (Macintyre et al., 2016). Using observed $PM_{2.5}$ levels from other years, it was estimated that the mortality burden was 2.0 to 2.7 times that associated with typical urban background levels of $PM_{2.5}$ at this time of year (Macintyre et al., 2016). Differences in health estimates amongst these previous studies are mainly due to differences in the concentration response coefficients used, as well as the magnitude of pollutant concentrations and the baseline mortality estimates (which vary for each country and from year to year). The method for calculating the excess deaths is also not consistent between the studies and thus a direct comparison between the different studies is not possible.

Under climate change, the risk of heatwaves in the future will likely increase (IPCC, 2014). In the absence of air quality abatement measures, this could give rise to increases in the occurrence of air pollution episodes and UK health burdens associated with short-term exposure to O_3 and $PM_{2.5}$. However, the frequency of air pollution episodes will likely also be affected by climate change through changes in the frequency of large-scale blocking episodes

which have been shown to decrease in winter and summer over Europe in the 21st century (Masato et al., 2013).

In this study, we focus on air pollution episodes during a heatwave in summer 2006 during which stagnant weather conditions resulted in high O_3 and $PM_{2.5}$ concentrations occurring concurrently across the UK. Emissions have a major influence on air pollutant concentrations, however in this study we focus on how air pollution levels are linked to anticyclonic weather conditions. To identify these episodes, we used the Daily Air Quality Index (DAQI) which gives information on the air pollution levels in the UK and provides recommended actions and health advice (Defra, 2013). The index ranges from 'low' (1) to 'very high' (10) and is divided into four bands ('low', 'moderate', 'high' and 'very high'). The DAQI is determined by the highest concentrations of any of the following five pollutants: nitrogen dioxide (NO_2), sulphur dioxide (SO_2), O_3, PM_{10} and $PM_{2.5}$ (uk-air.defra.gov.uk). For this study, episodes were defined based on when the DAQI reached 'moderate' to 'high' values in the majority of the England regions, Scotland and Wales.

Between the 16th and 28th of July 2006, there was a 4% increase in baseline all-cause mortality (~ 680 excess deaths, Office for National Statistics, 2006). The then UK Health Protection Agency (HPA) used measurements of pollution from fixed site monitors to estimate that between June and July 2006, 11 and 7 additional daily deaths brought forward in England and Wales were associated with increased O_3 and PM_{10} concentrations compared to 2004 (assuming a 0.3% and 0.75% increase in deaths brought forward for a 10 $\mu g\ m^{-3}$ increase in O_3 and PM_{10}, respectively) (HPA, 2006). Monitoring of $PM_{2.5}$ concentrations only became routine in the UK since 2008/9 following the 2008 ambient air quality directive (EU, 2008) and thus due to lack of measurement data, the $PM_{2.5}$ health burden has not previously been quantified for this period. This study presents new estimates of health burdens associated with short-term exposure to MDA8 O_3 and daily mean $PM_{2.5}$ occurring concurrently during two air pollution episodes in July 2006 using detailed spatio-temporal air pollution modelling for the UK and links to the underlying meteorology for the air pollution episodes in this period. Here, the short term health effects associated with O_3 and $PM_{2.5}$ air pollution episodes are examined rather than long term health impacts that would occur under future emission and climate change. Pollutant concentrations are simulated using the UK Met Office's air quality model (AQUM) at 12 km horizontal grid resolution. The impact on all-cause mortality was calculated both at a national level, also for the nine Government Office Regions (GOR) in England, and for Scotland and Wales.

The paper is organised as follows: Section 2 describes the modelling framework used to simulate the O_3 and $PM_{2.5}$ pollutant concentrations and the methods used to calculate the health burdens associated with short-term exposure to MDA8 O_3 and daily mean $PM_{2.5}$ for each region. Section 3 presents the observed long-term daily time series of MDA8 O_3 and daily mean $PM_{2.5}$ concentrations for multiple years which are used to identify the air pollution episodes. For both July 2006 episodes we first analyse the temporal variability of simulated pollutant concentrations and meteorological drivers of the air pollution episodes in Section 4.1, then we analyse the spatial variability of MDA8 O_3 and daily mean $PM_{2.5}$ concentrations across the UK in Section 4.2. The health impact assessments for the July 2006 air pollution episodes are then presented in Section 5 followed by conclusions in Section 6.

2. Methods

2.1. Air Quality in the Unified Model - AQUM

The model used in this study is the air quality model AQUM (Air Quality in the Unified Model) which is a limited–area model configuration based on the UK Met Office Unified Model (MetUM, 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 Western Europe. The model has 38 vertical levels from the ground surface up to 39 km (with the lowest model level centred 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 (FFOC), 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. Gas-phase chemistry is simulated within AQUM by 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 reanalyses fields

(Flemming et al., 2009) whilst meteorology is obtained from the UK Met Office Unified Model (MetUM). Further details on AQUM, including evaluation, can be found in Savage et al. (2013).

Model simulations are performed for the year 2006 (allowing for model spin up) from which hourly pollutant concentrations during the chosen episodes are then extracted, and from which the MDA8 O_3 and daily mean $PM_{2.5}$ are calculated. A statistical post-processing bias correction technique (SPPO) (Neal et al., 2014) is applied to correct O_3 and $PM_{2.5}$ simulated concentrations. As outlined above, all simulated O_3 and $PM_{2.5}$ concentrations shown in this study are taken from the lowest model vertical level having a midpoint at 20 m. Bi-linear interpolation is used to extract simulated O_3 and $PM_{2.5}$ concentrations at measurement sites for observation-model comparison. The SPPO technique is applied to the pollutant concentrations quoted in Section 4.1 and in the health section of this study (Section 5) while pollutant concentrations shown in Section 4.2 are not bias corrected to enable comparison with the simulated meteorological fields.

2.2. Measurement data

Modelled MDA8 O_3 and daily mean $PM_{2.5}$ concentrations for 2006 are evaluated against measurements from the Automatic Urban and Rural Network (AURN). As monitoring of $PM_{2.5}$ concentrations only became routine in the UK since 2008/9, measurements for $PM_{2.5}$ concentrations for this period are only available at three sites: London Bloomsbury (urban background), Rochester Stoke (south east UK, rural background) and Harwell (south east UK, rural background). However, an evaluation of AQUM against measurements from the AURN network presented in the model description paper by Savage et al (2013) demonstrates good model performance for later periods compared to a larger set of available measuring sites. Results between 1 May 2010 and 30 April 2011 suggest a positive bias in simulated O_3 concentrations (~ +8 μg m^{-3}), whilst simulated $PM_{2.5}$ concentrations exhibit a negative bias (~-3 μg m^{-3}) (Savage et al., 2013). In addition, Savage et al. (2013) evaluate the model representation of O_3 concentrations for the period July 2006. Results show that for July 2006, the AQUM exhibits a small positive bias for simulated O_3 concentrations (~ +1.99 μg m^{-3}).

Wind measurements for 2006 were only recorded at Rochester Stoke, therefore we use this site to show temporal variability in modelled and observed pollutant concentrations and meteorological variables in Sections 3 and 4.1. Surface temperatures are not recorded for any of the three AURN sites, therefore we use observed temperatures at the three closest sites to

these air pollution measurement sites from the Met Office Integrated Data Archive System (MIDAS) network (located at St James Park, London, East Malling and Upper Lambourn). Site locations from the AURN and MIDAS networks are illustrated in Fig. 4 and Fig. 6, respectively.

2.3. Heath impact assessment

Estimated health burdens attributable to short-term exposure to MDA8 O_3 and daily mean $PM_{2.5}$ are calculated as follows for each of the nine GOR for England, and for Scotland and Wales (shown in Fig. 1):

$$M_r = \sum_{i=1}^{N} BM_{ir} \times AF_{ir} \quad (1)$$

where

$$AF_{ir} = \frac{RR_{ir}-1}{RR_{ir}} \quad (2)$$

and

$$RR_{ir} = \exp(CRF \times x_{ir}) \quad (3)$$

and

$$x_{ir} = \frac{\sum_{j \in region} (x_{ij} \times p_j)}{\sum_{j \in region} p_j} \quad (4)$$

In equation 1, M_r is the all-cause mortality associated with short-term exposure to MDA8 O_3 or daily mean $PM_{2.5}$ for each region, r (Figure 1) summed over each day of the air pollution episode, i; N is the total number of days in the air pollution episode, BM_{ir} is the total regional daily mortality (henceforth referred to as baseline mortality) and AF_{ir} is the daily attributable fraction associated with short-term exposure to MDA8 O_3 or daily mean $PM_{2.5}$ that is calculated for each region using equation 2. Daily all-cause BM_{ir} for each region was obtained from the Office of the National Statistics for England and Wales (ons.gov.uk) and from the National Records of Scotland (nrscotland.gov.uk). In equations 2 and 3, RR_{ir} is the daily regional relative risk associated with short-term exposure to MDA8 O_3 or daily mean $PM_{2.5}$. In equation 3, *CRF* is the concentration-response function coefficient and x_{ir} is the regional daily population-weighted pollutant concentration. The CRFs used in this study for short-term exposure to MDA8 O_3 and daily mean $PM_{2.5}$ are taken from COMEAP (2015) and from a meta-

analysis of time series epidemiological studies (Atkinson et al., 2014), respectively. For short-term exposure to O_3-related health impacts, we use a CRF of 0.34 % (95% confidence interval (CI): 0.12%, 0.56%) per 10 μg m^{-3} increase in MDA8 O_3, and for short-term exposure to $PM_{2.5}$-related health impacts we use a CRF of 1.04 % (CI: 0.52%, 1.56%) per 10 μg m^{-3} increase in 24-hr mean $PM_{2.5}$. As limited evidence is available for a threshold below which no adverse effects for short-term exposure to MDA8 O_3 and daily mean $PM_{2.5}$ exist, no threshold was applied to pollutant concentrations (Atkinson et al., 2014; COMEAP 2015).

The daily regional population-weighted pollutant concentrations x_{ir} are calculated by first counting the total residential gridded population data (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 daily pollutant concentration within each grid cell x_{ij}, summed over every grid cell within the region r, and divided by the total population of the region.

To quantify the deaths brought forward associated with air pollution during the episodes we first calculate the 'typical' air pollution related daily deaths brought forward that would have occurred in the absence of an air pollution episode. This is done by replacing the modelled pollutant concentration which varies for each day of the air pollution episode (x_{ij}) with the mean pollutant concentration for June, July and August (summer mean) following the method in Macintyre et al. 2016), which we assume to be representative of air pollution levels when an episode does not occur. All other variables are left unchanged, therefore when calculating the 'typical' daily deaths, the AF_{ir} for each day is the same, with baseline mortality (BM) still varying daily. The excess deaths are then estimated by subtracting the 'typical' estimated deaths brought forward from the episode estimated deaths brought forward.

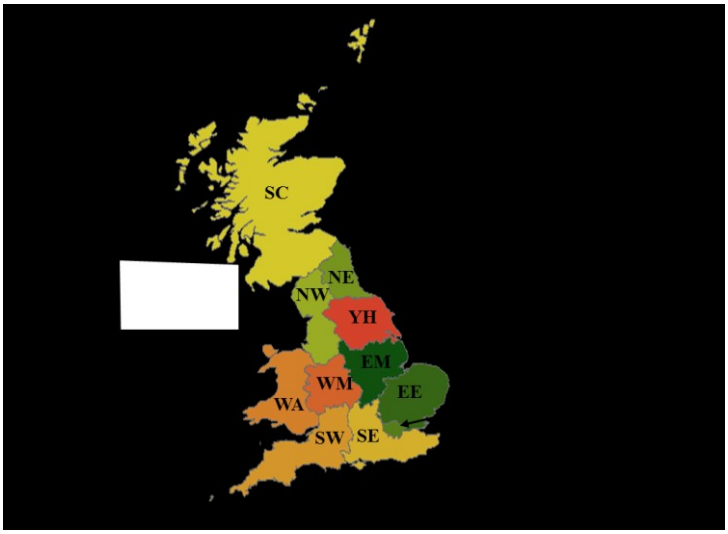

Figure 1: Government Office Regions (GOR) for England, Scotland and Wales used in this study

3. Identification of air pollution episodes from observations

Measured MDA8 O_3 and daily mean $PM_{2.5}$ concentrations from 2005 to 2007 (inclusive) at the Rochester Stoke site (rural background) are shown in Fig. 2. The seasonal profile of MDA8 O_3 concentrations is similar for all three years, with the highest concentrations occurring in the summer months and the lowest in winter (Fig. 2a). Superimposed on this seasonal cycle are daily variations in MDA8 O_3. A seasonal cycle is less evident for daily $PM_{2.5}$ concentrations between 2005 and 2007 (Fig. 2b), although a background level of 5-10 µg m^{-3} is evident with substantial day-to-day variability. We focus our analysis on periods during 2006 using two criteria to define air pollution episodes: a) high MDA8 O_3 and daily mean $PM_{2.5}$ concentrations occurring concurrently and b) a DAQI reaching a 'moderate' or 'high' level over the majority of the regions across the UK. This resulted in the selection of two 5-day periods from the 1st -5th July 2006 and from the 18th -22nd July 2006, which are described below. We note that these selection criteria will not yield the peak $PM_{2.5}$ concentrations in 2006 as these occurred in autumn 2006 (Figure 2).

In July 2006, measured MDA8 O_3 concentrations reached 150 µg m^{-3} on 2nd July 2006 at Rochester Stoke (Fig. 2a) which are higher compared to adjacent years. Daily mean $PM_{2.5}$ concentrations at the same station also show peaks occurring during July 2006 (up to 26 µg m^{-3} on the 4th July, Fig. 2b). Similarly peaks for MDA8 O_3 and daily mean $PM_{2.5}$ concentrations during this same period were noted at the London Bloomsbury and Harwell stations (refer to Fig. S1 in the Supplement to this manuscript).

Between the 1st and 5th of July 2006 the DAQI reached a 'moderate' or 'high' level in 96% of the regions in England, Scotland and Wales and in 88% of the regions between the 18th and the 22nd of July 2006 (uk-air.defra.gov.uk). This suggests that overall, more extensive high pollutant concentrations occurred in the first episode compared to the second episode. In particular, in the south east region, the DAQI reached a 'high' level during most days of the first episode and a 'moderate' level during most of the second episode.

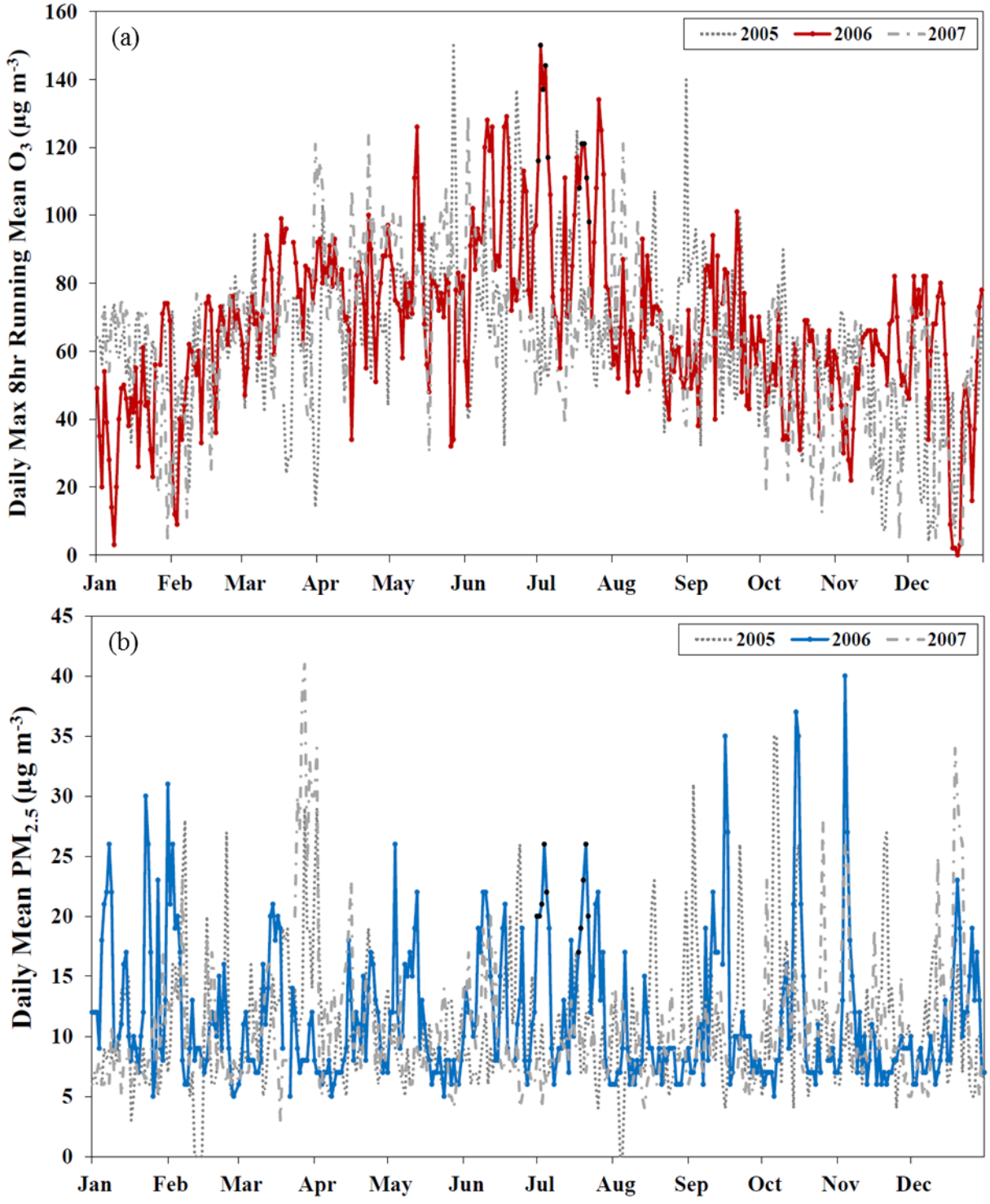

Figure 2: (a) Daily maximum 8-hr running mean (MDA8) O_3 and (b) daily mean $PM_{2.5}$ at the Rochester Stoke AURN rural background station from 2005 (dotted), 2006 (red for O_3 and blue for $PM_{2.5}$), and 2007 (dot-dashed). Black points mark the two 5-day air pollution episode days for July 2006 (1st-5th July and 18th-22nd July 2006).

4. Meteorological factors contributing to the air pollution episodes

In this section, we discuss the temporal and spatial variability of simulated pollutant concentrations and meteorological variables; analysing the main meteorological factors contributing to high modelled concentrations of MDA8 O_3 and daily mean $PM_{2.5}$.

4.1. Temporal variability of pollutants and meteorology during the pollution episodes

Simulated (original and bias corrected - as described in Section 2.1) and observed MDA8 O_3 and daily mean $PM_{2.5}$ concentrations for July 2006 are shown in Fig. 3a for the Rochester Stoke site (rural background). Using the raw model output for both episodes, MDA8 O_3 concentrations are underestimated (mean bias = -10.72 μg m^{-3} and r = 0.67; Table 1) while daily mean $PM_{2.5}$ concentrations are overestimated (mean bias of 26.29 μg m^{-3} and r = 0.67; Table 1). When applying the SPPO bias correction technique (Section 2.1), the temporal variability of MDA8 O_3 and daily mean $PM_{2.5}$ concentrations is largely unaltered with peaks captured well during both episodes (Fig. 3a) and with mean bias errors reducing to -2.76 μg m^{-3} (r = 0.89) and 12.03 μg m^{-3} (r = 0.81), respectively (Table 1).

Table 1: Statistics comparing daily mean model and observed pollutant concentrations and meteorological variables for both original (Raw) and bias corrected (SPPO – Statistical Post-Processing technique) model output averaged over the period 1st -5th July and 18th to 22nd July at Rochester Stoke.

		Bias	R
MDA8 O_3 (μg m^{-3})	Raw	-10.72	0.67
	SPPO	-2.76	0.89
24-hr mean $PM_{2.5}$ (μg m^{-3})	Raw	26.29	0.67
	SPPO	12.03	0.81
24-hr mean temperature (°C)	Raw	0.85	0.69
24-hr mean wind direction (°)	Raw	-7.59	0.95
24-hr mean wind speed (m s^{-1})	Raw	-0.44	0.85

Figure 3b shows simulated and observed daily mean meteorological variables (surface temperature, wind speed and wind direction) during July 2006 at the Rochester Stoke site. Daily variations of temperature, wind direction and wind speed are well captured by the model during both episodes, with r values equal to 0.69, 0.95 and 0.85 respectively (N.B. measured temperatures are taken from St East Malling and not Rochester Stoke). However, the magnitudes of daily mean wind speed is generally underestimated (mean bias = -0.44 m s^{-1}; Table 1), while temperature is generally overestimated (mean bias = 0.85 °C; Table 1). At Rochester Stoke, wind speeds during both air pollution episodes are generally low (between ~2 and ~ 4 m s^{-1}; Fig. 3b) compared to the rest of July, though wind speeds are fairly low throughout this month (not exceeding 7 m s^{-1}; Fig. 3b). Compared to the rest of July, daily mean temperatures are higher during the two episodes as well as at the end of the month (ranging from ~20 °C to ~27 °C; Fig. 3b). During the first episode, measured and simulated daily mean wind directions ranged from ~ 60° (north east; Fig. 3b) to ~ 200° (south west; Fig. 3b). In the later July episode wind directions ranged from north east to south west (Fig. 3b). However the prevailing wind during both episodes is from a north easterly and easterly direction.

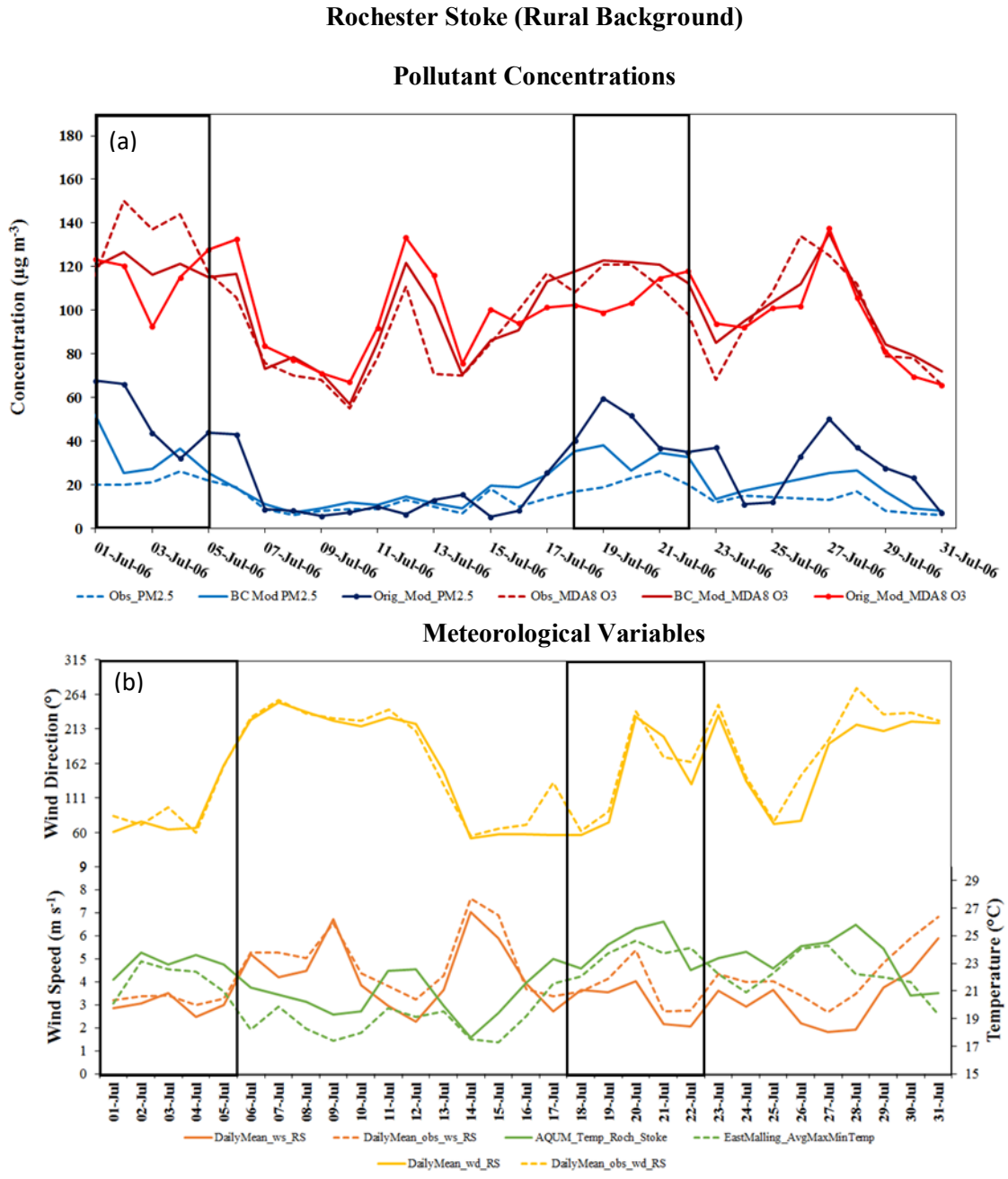

Figure 3: Daily time series during July 2006 of modelled (solid lines) and observed (dashed lines) a) $PM_{2.5}$ (blue) and MDA8 O_3 (red) b) wind direction (degrees are taken clockwise starting from the north - yellow), wind speed (orange) and temperature (green) at the rural background AURN station in Rochester Stoke (observed temperatures were obtained from the nearest MIDAS station to Rochester Stoke which is at East Malling.) Black boxes represent the two 5-day air pollution episodes in July 2006 (1^{st}-5^{th} July and 18^{th}-22^{nd} July)). Solid lines with dots indicated the original simulated concentrations while the solid lines with no dots show the bias-corrected concentrations using the SPPO technique.

4.2. Spatial variability of O_3 and $PM_{2.5}$ concentrations and meteorology during the pollution episodes across the UK

The spatial distributions of simulated surface MDA8 O_3 and daily mean $PM_{2.5}$ averaged for all of July, and for the two 5-day air pollution episodes are shown in Fig. 4 (concentrations shown here are not bias corrected to enable comparison with meteorology; however spatial patterns for bias-corrected pollutant concentration are similar (refer to Supplement Fig. S2)). Simulated MDA8 O_3 concentrations during all three averaging periods generally compare well with observations (to within 15 $\mu g\ m^{-3}$; Fig 4a-c). However, simulated MDA8 O_3 concentrations at London Bloomsbury are higher than observed concentrations during the first episode (Fig. 4b). July mean MDA8 O_3 concentrations range between ~ 80 $\mu g\ m^{-3}$ in Scotland to ~ 120 $\mu g\ m^{-3}$ in south eastern and eastern England (Fig. 4a). For the first episode, simulated MDA8 O_3 concentrations are highest in the west of England with concentrations reaching ~ 180 $\mu g\ m^{-3}$ in Wales and up to ~ 210 $\mu g\ m^{-3}$ in South West England (Fig. 4b). Simulated MDA8 O_3 concentrations during the second episode are lower reaching ~ 150 $\mu g\ m^{-3}$ in the south of England and also less variable across the UK than for the first episode as noted for the DAQI in Section 4.1 (Fig. 4c).

Simulated daily mean $PM_{2.5}$ concentrations during July and the second episode are generally in agreement with observations at Harwell, but are overestimated over the London Bloomsbury and Rochester Stoke locations during the two episodes (Fig 4 d-f). Neal et al. (2017) also find an all year round small positive bias in simulated $PM_{2.5}$ concentrations for a 5-year period at two background observational sites. For all three periods shown, higher daily mean $PM_{2.5}$ concentrations are simulated over England and Wales compared to Scotland with a stronger North-South spatial gradient occurring during the air pollution episodes compared to July (Fig. 4d-f). Simulated daily mean $PM_{2.5}$ concentrations are highest during the first episode reaching ~ 45 $\mu g\ m^{-3}$ in the west of England (Fig. 4e). The spatial pattern of simulated $PM_{2.5}$ concentrations in the second episode differs from that found in the first episode. Simulated daily mean $PM_{2.5}$ concentrations are higher in south east England compared to the west of England for this later episode (~ 45 $\mu g\ m^{-3}$ compared to ~ 20 $\mu g\ m^{-3}$, Fig. 4f).

Simulated MDA8 O_3 and $PM_{2.5}$ concentrations during the two episodes are also compared to summer (JJA) mean concentrations. The spatial distribution of summer mean pollutant concentrations remains unaltered with low concentrations in Scotland and higher concentrations in the southern and eastern regions. Summer mean pollutant concentrations are

however lower than the July mean concentrations shown in Fig. 4a and 4d. MDA8 O_3 concentrations in summer range between ~75 µg m^{-3} in Scotland and ~97 µg m^{-3} in East England while $PM_{2.5}$ concentrations range between ~7 µg m^{-3} in Scotland and ~15 µg m^{-3} in East Midlands and East of England (not shown).

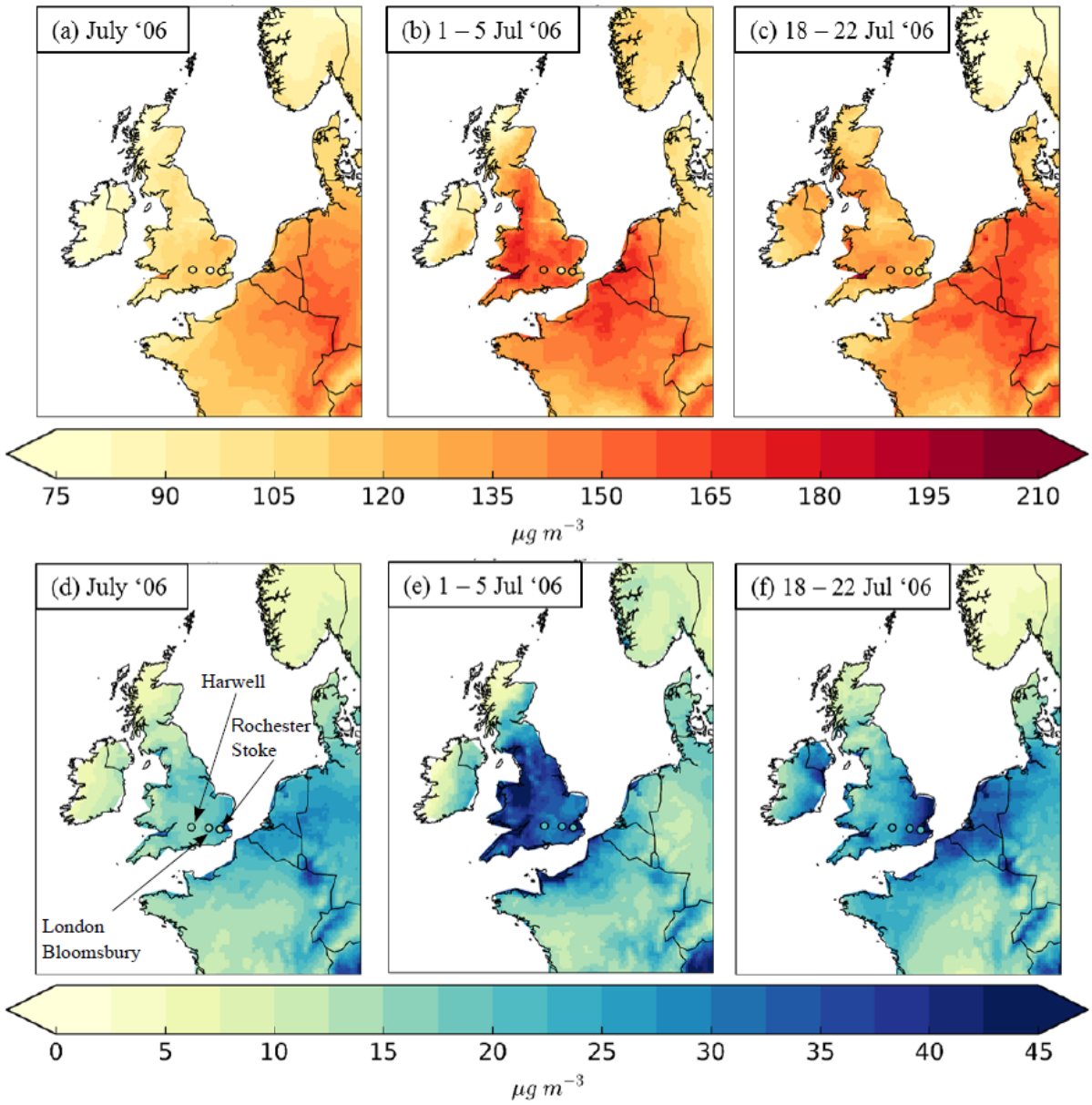

Figure 4: Simulated daily maximum 8-hr running mean O_3 concentrations for (a) July mean, (b) 1-5 July mean and (c) 18-22 July 2006 mean and simulated daily mean $PM_{2.5}$ concentrations for the same time periods (d, e and f) (NB concentrations shown here are not bias corrected to enable comparison with meteorology; however spatial patterns are similar (refer to Supplement)). The circles in each plot are coloured with measured data of the respective pollutant at 3 AURN sites - Rochester Stoke, Harwell and London Bloomsbury.

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Many factors can contribute to high MDA8 O_3 and daily mean $PM_{2.5}$ concentrations during these two episode periods. We first focus on spatial variations in meteorology (surface wind direction, wind speed, pressure and temperature), following on from section 4.1 . Wind direction and wind speeds across the UK are illustrated in Fig 5. For July 2006, the simulated
405 prevailing hourly wind direction is from the south west with a mean wind speed of 1.6 m s^{-1} (Fig. 5a). Simulated mean wind speeds during the two episodes are of similar magnitude to the whole July period. However, the dominant wind direction varies between the two episodes. Over most of the UK easterly and south easterly winds occur during the first episode (Fig 5b), whilst for the later episode south easterly and southerly winds are more prevalent (Fig 5c).
410 Thus light winds bringing air from continental Europe are characteristic of both the episodes in July 2006. Note that the general wind direction across the UK differs slightly from that at the Rochester Stoke location where the prevailing wind direction is north easterly and easterly during both episodes (Section 4.1). The different wind direction for Rochester Stoke (Section 4.1) might be linked to the coastal location of this site.

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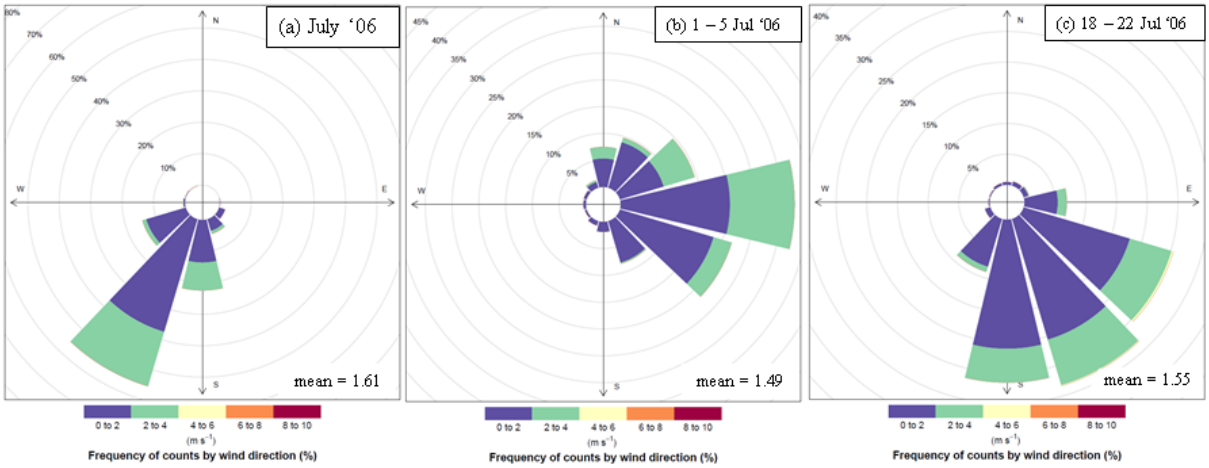
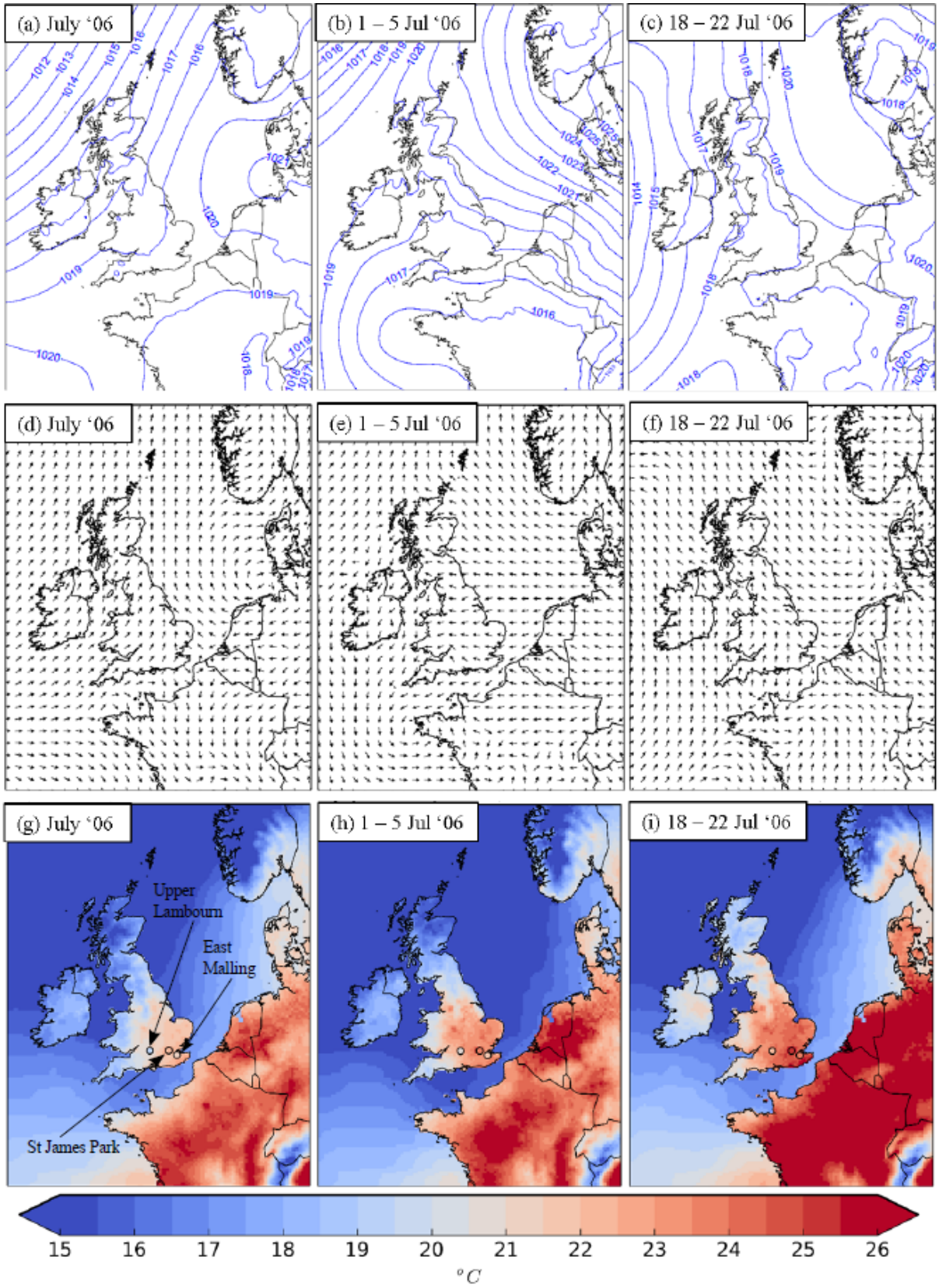


Figure 5: Wind roses showing the frequency of hourly wind directions for each land grid box in the UK for (a) July 2006, (b) 1st – 5th July, (c) 18th- 22nd July 2006. Colours indicate the wind speed.

Simulated spatial variations in daily mean surface pressure, wind direction and temperature are shown in Fig. 6. In July light winds of a south to south-westerly direction over southern and central England are indicated by the widely spaced simulated pressure contours and wind vectors in Fig. 6a and 6d, respectively. During this same period, anticyclonic conditions are simulated over northern Europe (> 1020 hPa, Fig. 6a). As discussed above, during the first episode, a change in wind direction compared to the July mean can be noted with winds blowing from an easterly and south easterly direction over most of the UK (Fig. 6e, Fig 5b). Also, higher pressures over central and northern UK (>1019 hPa) occur compared to the July mean with the high pressure centred over Norway and Sweden (Fig. 6b). A high pressure system is also characteristic of the second air pollution episode, but in this case is centred over the North Sea (Fig. 6c, f.). The high pressure simulated during both episodes leads to light winds and hence favourable stagnant weather conditions for high pollutant concentrations as well as slow transport of pollution from the European continent. Other possible reasons for high pollutant concentrations may include lack of cloud and precipitation, enhancing photolysis as well as reducing wet deposition. Using the same model Pope et al. (2016) also found O_3 concentrations to be high under anticyclonic and south-easterly conditions for the summer (Apr-Sept) 2006. High temperatures are also associated with anticyclonic conditions as discussed in Section 4.1. An increase in simulated daily mean surface temperatures during the two episodes compared to the July mean can be clearly seen (Figs. 6g-i). Measured daily mean surface temperatures from three MIDAS stations are also shown as coloured circles (Upper Lambourn, St James Park London and East Malling). Good agreement can be seen between modelled and observed July mean temperatures (to within 0.5

°C) at the St James Park station in Greater London. However, over the other two locations, July-mean temperatures are overestimated by the model by about 1°C for the Upper Lambourn and East Malling sites. Simulated July mean temperatures range from 15°C to 23°C and are highest in south east England (Fig. 6g). While the spatial distribution of simulated temperatures during the two episodes is similar to those in July (low temperatures in the north and high temperatures in the south), daily-mean values are generally between 16°C and 24°C for the first episode and range between 19°C and 26°C during the later episode (Fig. 6h and 6i). These higher temperatures in the central and south eastern England may partly explain the higher simulated MDA8 O_3 concentrations towards the south east of England for the July mean and two episodes as temperature and sunlight promote photochemical formation of O_3 (Fig. 4). $PM_{2.5}$ concentrations may also be influenced by temperature (c.f. Fig. 4d-f; Fig 6g-i). Higher temperatures may lead to increased sulphate aerosol formation due to increased reaction rates; however levels of other secondary components of $PM_{2.5}$ (e.g. nitrate aerosol) may be reduced due to increased partitioning from the aerosol phase into the gas phase (Doherty et al., 2017; Fiore et al., 2012).



460 **Figure 6: Daily mean air pressure at mean sea level (top panel), wind vectors (middle panel) and surface temperature (bottom panel) averaged for all of July (left column), 1st to 5th July (middle column) and 18th to 22nd July (right column). Measured levels of daily mean temperature averaged for the same time periods from three MIDAS stations (St James Park, East Malling and Upper Lambourn) are shown in coloured circles (bottom panel)**

5. Mortality from short term exposure to O_3 and $PM_{2.5}$

In this section we present our estimates for the short-term exposure to MDA8 O_3 and daily mean $PM_{2.5}$ concentrations using bias-corrected population-weighted pollutant concentrations for the two episodes in July 2006 (refer to Fig. S1). For each region, the total population and the population-weighted MDA8 O_3 concentrations for the summer (June-July-August, JJA) and for the two episode periods together with the percentage of all-cause mortality and the estimated number of deaths brought forward are shown in Table 2. In summer, population-weighted simulated MDA8 O_3 concentrations range from ~75 $\mu g\ m^{-3}$ in Scotland to ~ 96 $\mu g\ m^{-3}$ in east England (Table 2). In contrast, MDA8 O_3 concentrations range from ~ 98 $\mu g\ m^{-3}$ to ~ 155 $\mu g\ m^{-3}$ in the first episode (for Scotland and South West England, respectively; Table 2) and from ~ 112 $\mu g\ m^{-3}$ to ~ 147 $\mu g\ m^{-3}$ in the second episode (for Scotland and East England, respectively; Table 2). These regions having the highest simulated population-weighted bias corrected MDA8 O_3 concentrations are the same regions that have the highest uncorrected pollutant concentrations as discussed in Section 4.2. The south west and the east of England regions are also the regions with the highest percentage of all-cause mortality attributed to air pollution (AF) during the first and second episode, respectively (5.16% and 4.87%; refer to Table S1 for confidence intervals for all the regions) as this value only depends on the population-weighted pollutant concentration which is highest in these regions (refer to Eq. 2-4 in Section 2.3). The AF due to short-term exposure to MDA8 O_3 is higher for the first compared to second episode which again reflects the higher concentrations discussed in Section 4.2.

The total health burden associated with short-term exposure to MDA8 O_3 summed over all the regions for the first and second episode is similar: 69 (CI: 25,111; based on the 95% confidence interval of the concentrations response coefficient) and 70 (CI: 25,113) daily deaths brought forward, respectively (Table 2; refer to Table S1 for confidence intervals for all the regions). However, as discussed in Section 4.2 higher MDA8 O_3 concentrations are simulated during the first compared to the second episode. In this paper we only account for the uncertainty associated with the CRFs used (e.g. Heal et al. 2013, Macintyre et al. 2016). Furthermore Kushta et al. (2018) have shown that uncertainties in ambient $PM_{2.5}$-related mortality estimates are dominated by the estimated CRFs derived from epidemiological studies which result in statistical uncertainties in the mortality estimates within about ±30% compared to uncertainties associated with horizontal and vertical model resolutions in their study which give rise to mortality rates that differ by 2.4% and 0.6%, respectively. Apart from uncertainties associated with model resolution Kushta et al. (2018) suggest that estimates based on $PM_{2.5}$

concentrations derived from satellite data are within 10% of the model results. On the other hand, in a multi-model study, Silva et al. (2016) suggest the contribution to the overall uncertainty from modeled air pollution concentrations exceeds that from the CRFs.

The calculation to determine the health burden does not depend solely on pollutant concentrations but also depends on the population of the region as well as the baseline mortality. For each region, the all-cause baseline mortality in the second episode is higher compared to that in the earlier episode (by up to 17 daily deaths in the West Midlands) which may be linked to higher temperatures over that period. Therefore differences in baseline mortality between the two episodes appear to balance differences in simulated MDA8 O_3 concentrations when calculating the mortality attributable to short-term exposure to MDA8 O_3 (as described in Eq. 1 Section 2.3). The regions with the highest health burden during both air pollution episodes are the North West and South East England (Table 2) which is due to a combination of high pollutant concentrations and a larger total population in these two regions (Table 2).

Table 2: GOR regions for England, Scotland and Wales and their populations together with the regional bias corrected population-weighted daily maximum 8-hour O_3 concentrations for summer (JJA) and averaged between the 1st-5th July and the 18th-22nd July 2006. For each of the episodes the percentage of all-cause mortality and the daily deaths brought forward are also included.

		JJA 2006	1-5 July 2006			18-22 July 2006		
				Daily deaths brought forward			Daily deaths brought forward	
Region	Pop. (1000s)	MDA8 O_3 ($\mu g\ m^{-3}$)	MDA8 O_3 ($\mu g\ m^{-3}$)	Percent of all-cause	Number	MDA8 O_3 ($\mu g\ m^{-3}$)	Percent of all-cause	Number
East Midlands	4356	86.2	137.0	4.57%	5	135.6	4.51%	5
East England	5591	96.3	148.3	4.94%	7	147.1	4.87%	7
London	7237	89.7	136.8	4.58%	7	139.6	4.65%	7
North East	2433	77.4	123.0	4.05%	3	119.6	3.95%	3
North West	6637	77.8	146.6	4.80%	9	125.6	4.26%	9
Scotland	4802	74.7	98.0	3.35%	4	112.1	3.74%	5
South East	8084	94.1	146.5	4.86%	10	144.5	4.81%	10
South West	4930	90.3	155.4	5.16%	7	134.0	4.56%	7
Wales	2811	86.2	152.4	5.09%	4	138.1	4.63%	4
West Midlands	5323	84.0	148.0	4.91%	7	137.2	4.59%	7
Yorkshire and The Humber	4967	78.7	126.5	4.20%	5	119.9	4.03%	5
Total					69			70

The population-weighted daily mean $PM_{2.5}$ concentrations during summer range from 9.4 $\mu g\ m^{-3}$ in Scotland to 14.7 $\mu g\ m^{-3}$ in east midlands and east England. Higher concentrations occur during the first and second episode with concentrations range from 17 $\mu g\ m^{-3}$ to 38 $\mu g\ m^{-3}$ (in Scotland and North West; Table 3) and from 15.4 $\mu g\ m^{-3}$ to 28.5 $\mu g\ m^{-3}$ (in Scotland and East England; Table 3), for the first and second episodes, respectively. The percentage of attributable all-cause mortality is highest in the North West and East England regions for the first (3.9%) and second episode (2.9%), respectively, again reflecting high $PM_{2.5}$ concentrations in these regions (Table 3; refer to Table S2 for confidence intervals in mortality estimates for all the regions). Due to higher population-weighted $PM_{2.5}$ concentrations during the first episode compared to the later episode, the percentage of attributable all-cause mortality is overall also higher during the first episode. The total number of deaths brought forward attributable to short-term exposure to $PM_{2.5}$ during the first and second episodes are 43 (CI: 22, 64) and 36 (CI: 18, 54) daily deaths brought forward, respectively (Table 3). In this case

differences in $PM_{2.5}$ concentrations between the two episodes outweigh differences in baseline mortality when calculating the mortality attributable to short-term exposure to $PM_{2.5}$ thus resulting in a higher number of estimated deaths brought forward in the first compared to the second episode (as described in Eq. 1 Section 2.3). The regions having the highest number of deaths brought forward during the first and second episode are again the North West and South East regions, respectively as high $PM_{2.5}$ concentrations and a large population density coincide.

Table 3: GOR regions for England, Scotland and Wales and their populations together with the regional bias corrected population-weighted daily mean $PM_{2.5}$ concentrations for summer (JJA) and averaged between the 1st-5th July and the 18th-22nd July 2006. For each of the episodes the percentage of all-cause mortality and the daily deaths brought forward are also included.

		JJA 2006	1-5 July 2006			18-22 July 2006		
				Daily deaths brought forward			Daily deaths brought forward	
Region	Pop. (1000s)	Mean Daily $PM_{2.5}$ ($\mu g\ m^{-3}$)	Mean Daily $PM_{2.5}$ ($\mu g\ m^{-3}$)	Percent of all-cause	Number	Mean Daily $PM_{2.5}$ ($\mu g\ m^{-3}$)	Percent of all-cause	Number
East Midlands	4356	14.7	27.5	2.88%	3	21.9	2.27%	3
East England	5591	14.7	23.2	2.39%	3	28.5	2.92%	4
London	7237	14.6	22.5	2.25%	3	26.8	2.72%	4
North East	2433	13.3	28.2	2.81%	2	19.1	2.02%	1
North West	6637	14.2	38.0	3.91%	7	20.5	2.13%	4
Scotland	4802	9.4	17.0	1.72%	2	15.4	1.63%	2
South East	8084	14.3	24.8	2.50%	5	24.9	2.54%	5
South West	4930	14.3	33.8	3.43%	5	24.9	2.62%	4
Wales	2811	13.3	36.1	3.72%	3	23.2	2.42%	2
West Midlands	5323	14.1	32.4	3.28%	4	20.0	2.04%	3
Yorkshire and The Humber	4967	14.6	30.0	3.11%	4	21.3	2.16%	3
Total					43			36

Using the summer mean (JJA) pollutant concentrations discussed above as 'typical' concentrations that would occur in the absence of an air pollution episode, we estimate that the mortality burden associated with short-term exposure to MDA8 O_3 in the first and second episode is 38 % and 36 % higher than the 'typical' summer mean estimate, respectively (Table 4; refer to Section 2.3). Similarly, we find that the mortality burden associated with short-term exposure to daily mean $PM_{2.5}$ during the first and second episode is 56 % and 39% higher than if the concentrations were more similar to those occurring in the absence of an air pollution episode (Table 4; regional estimates for 'typical' concentrations can be found in Tables S1 and S2).

Table 4: Estimated deaths brought forward from short-term exposure to 'typical' summer time population-weighted MDA8 O_3 and daily mean $PM_{2.5}$ concentrations compared to the population-weighted pollutant concentrations simulated between the 1st-5th July and the 18th-22nd July 2006. Excess deaths brought forward from short-term exposure to MDA8 O_3 and daily mean $PM_{2.5}$ are calculated as a percentage using (((Episode Estimate – Typical Estimate)/Episode Estimate) * 100)

				Daily Deaths Brought Forward		
		Pollutant Concentrations ($\mu g\ m^{-3}$)		Number	Percentage of all-cause	Excess (%)
1-5 July 2006	MDA8 O_3	Episode	138.0	69 (25,111)	4.59 (1.65,7.45)	
		Typical *	85.0	43 (15,69)	2.85 (1.02,4.65)	38
	$PM_{2.5}$	Episode	28.5	43 (22,64)	2.91 (1.47,4.33)	
		Typical *	13.8	19 (15,25)	1.28 (1.00,1.70)	56
18-22 July 2006	MDA8 O_3	Episode	132.1	70 (25,113)	4.42 (1.58,7.17)	
		Typical *	85.0	45 (16,73)	2.85 (1.02,4.65)	36
	$PM_{2.5}$	Episode	22.4	36 (18,54)	2.32 (1.17,3.45)	
		Typical *	13.8	22 (11,33)	1.42 (0.71,2.13)	39

*** Simulated population-weighted summer mean (JJA) concentrations for 2006 for each region are used as 'typical' concentrations. This is kept constant for each day of the episode.**

Estimated health burdens attributable to exposure to MDA8 O_3 during both episodes are lower than estimates for the first two weeks of August 2003 derived by Stedman (2004) (~70 compared with 88 daily deaths). This difference is due primarily to the use of a concentration response coefficient by Stedman (2004) that is about double that used in this study (0.6% compared with 0.34% for a 10 $\mu g\ m^{-3}$ increase in MDA8 O_3) as well as different baseline mortality rates as discussed in Section 1. Our $PM_{2.5}$-related estimates differ from estimates of Macintyre et al. (2016) of approximately 59 daily deaths brought forward from short-term exposure to $PM_{2.5}$ during a total of 10 days across the UK in spring 2014 (compared to 43 and 36 daily deaths brought forward during the first and second episode in this study). The $PM_{2.5}$-related concentration response coefficient used in our study and that of Macintyre et al. (2016) is the same, however population-weighted daily mean $PM_{2.5}$ concentrations during the air pollution episode studied in Macintyre et al. (2016) reach a mean concentration of 50

µg m^{-3}, which is approximately 12 µg m^{-3} higher than the highest mean daily population-weighted $PM_{2.5}$ concentrations we observe during the first episode in North West England in this study (38 µg m^{-3}, Table 3). $PM_{2.5}$ concentrations for both July episodes in this study are unusually high compared to other years (Figure 2b for 2005 and 2007) and although slightly lower, are of similar magnitude to those reported in spring 2014 by Macintyre et al. (2016). Hence the health burden we estimate for the July 2006 episodes is somewhat comparable to the $PM_{2.5}$-related health burden found by Macintyre et al. (2016) in spring 2014.

In this paper the CRFs used are derived from single-pollutant studies that do not take into account any potential overlap or interaction in relationships between O_3 and $PM_{2.5}$ concentrations in terms of joint or interactive health effects. This can lead to an underestimation of the true impact of the pollution mixture, especially if other pollutants also affect the same health outcome (COMEAP, 2015). On the other hand, the effects of O_3 and PM have been shown to be relatively independent (WHO, 2006) and for this reason we have not added the burdens estimated for each pollutant to avoid any double counting. The uncertainties presented in this paper are related to the CRFs used however, other uncertainties are associated with health impact assessments. These include uncertainties associated with the simulated air pollutant concentrations, baseline mortality and population data. In addition, uncertainties may arise from the different air quality models used which are typically addressed in multi-model studies and cannot be accounted for in this paper. Also, in our calculation to estimate the excess deaths during the chosen episodes compared to 'typical' conditions we only take into account 'typical' pollutant concentrations but not a 'typical' baseline mortality which changes both daily and annually. Therefore our estimates of the impact of the air pollution episode are conservative.

6. Conclusions

Air pollution episodes are typically driven by stagnant weather conditions. Between the 1st - 5th July and the 18th - 22nd July 2006, the driving meteorology which included persistent anticyclonic conditions and prevailing winds from the East and South East led to high MDA8 O_3 and daily mean $PM_{2.5}$ levels occurring simultaneously, likely emanating from both local emissions trapping and processing, and slow transport of pollution from continental Europe.

Over the two episodes in July 2006, the estimated total mortality burden associated with short-term exposure to MDA8 O_3 is around 70 daily deaths brought forward summed across

the UK. By using summer 2006 (JJA) population-weighted simulated MDA8 O_3 concentrations as 'typical' concentrations during this time of year, we estimate the health burden to be 38% and 36% higher during the first and second episode, respectively compared to the summer average. The estimated health burden associated with short-term exposure to $PM_{2.5}$ varies between the two episodes resulting in 43 and 36 daily deaths brought forward during the first and second episode, respectively. Using $PM_{2.5}$ concentrations representative of the summer average, we estimate the health burden to be 56% and 39% higher than if the pollution levels represented typical season-mean concentrations.

The regions with the highest percentage of all-cause mortality (AF) associated with short-term exposure to MDA8 O_3 and daily mean $PM_{2.5}$ varied between the episodes, as this depends on population-weighted pollutant concentrations. During the first episode in July 2006 the regions with the highest simulated population-weighted MDA8 O_3 and daily mean $PM_{2.5}$ concentrations and thus the highest AF due to exposure to MDA8 O_3 and daily mean $PM_{2.5}$ were the South West and North West regions. During the second air pollution episode, MDA8 O_3 and daily mean $PM_{2.5}$ concentrations were highest in the East of England resulting in the highest AF in this region. In contrast, the estimate mortality also depends on the baseline mortality and thus for all episodes, regions with the greatest total population which coincided with relatively high pollutant concentrations had the highest mortality estimates (e.g. North West and South East regions).

Our results show that episodes of high MDA8 O_3 and daily mean $PM_{2.5}$ such as those presented in this study can lead to an increase in deaths brought forward up to double that expected from typical concentrations over the same period. These conservative results are of importance for future policy making as they provide an estimate of the scale of the health impacts associated with such air pollution episodes in different UK regions, which can help raise public and political awareness of the issue and strengthen public health preparedness.

This study provides insights into the health effects of short-term exposure to MDA8 O_3 and daily mean $PM_{2.5}$ which occur together during air pollution episodes in July 2006 in the UK as well as the meteorological drivers. Using modelled pollutant concentrations at a 12 km resolution, this study also provides an indication of the regional variability of such impacts which is difficult to achieve with the paucity of observational data.

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