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Impact of weather types on UK ambient particulate matter concentrations

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ABSTRACT

Each year more than 29,000 premature deaths in the UK are linked to long term-exposure to ambient particulate matter (PM) with a diameter less than 2.5 μm ($PM_{2.5}$). Many studies have focused on the long-term impacts of exposure to PM, but short-term increases in pollution can also exacerbate health effects, leading to deaths brought forward within exposed populations. This study investigates the impact of different atmospheric circulation patterns on UK $PM_{2.5}$ concentrations and the relative contribution of local and transboundary pollutants to variations in $PM_{2.5}$ concentrations. Daily mean $PM_{2.5}$ observations from 42 UK background sites indicate that easterly, south-easterly and southerly wind directions and anticyclonic circulation patterns enhance background concentrations of $PM_{2.5}$ at all UK sites by up to 12 μg m^{-3}. Results from back trajectory analysis and the European Monitoring and Evaluation Programme for UK model (EMEP4UK) show this is due to the transboundary transport of pollutants from continental Europe. While back trajectories indicate under easterly, south-easterly and southerly flow 25–50% of the total accumulated primary $PM_{2.5}$ emissions originate outside of the UK, with a very polluted footprint (0.25–0.35 μg m^{-2}). Anticyclonic conditions, which occur frequently (21%), also lead to increases in $PM_{2.5}$ concentrations (UK multi-annual mean 14.7 μg m^{-3}). EMEP4UK results indicate this is likely due the build-up of local emissions due to slack winds. Under westerly and north-westerly flow 15–30% of the total accumulated primary $PM_{2.5}$ emissions originate outside of the UK, and are much less polluted (0.1 μg m^{-2}) with model results indicating transport of clean maritime air masses from the Atlantic. Results indicate that both wind-direction and stability under anticyclonic conditions are important in controlling ambient $PM_{2.5}$ concentrations across the UK. There is also a strong dependence of high $PM_{2.5}$ Daily Air Quality Index (DAQI) values on easterly, south-easterly and southerly wind-directions, with >70% of occurrences of observed 48–71+ μg m^{-3} concentrations occurring under these wind directions. While north-westerly and cyclonic conditions reduce $PM_{2.5}$ concentrations at all sites by up to 8 μg m^{-3}. $PM_{2.5}$ DAQI values are also lowest under these conditions, with >80% of 0–11 μg m^{-3} concentrations and >50% of 12–23 μg m^{-3} concentrations observed during westerly, north-westerly and northerly wind directions. Indicating that these conditions are likely to be associated with a reduction in the potential health effects from exposure to ambient levels of $PM_{2.5}$.

1. Introduction

Air pollution is the fourth highest-ranking risk factor for mortality globally, with 85% of the global population living in areas where the WHO recommended air quality guidelines (10 μg m^{-3} for particulate matter with a diameter less than 2.5 μm ($PM_{2.5}$)) are exceeded (GBD, 2018). Exposure to air pollutants, including $PM_{2.5}$, on both short and long-time scales has been shown to be strongly associated with mortality and morbidity (GBD, 2018). Exposure to $PM_{2.5}$ is associated with increases in diseases such as cardiovascular disease, ischemic heart disease, stroke, lower respiratory tract infections and chronic obstructive pulmonary disorder (Atkinson et al., 2014; Cohen et al., 2017). In the UK, it is estimated that more than 29,000 premature deaths each year are linked to long term-exposure to ambient $PM_{2.5}$ (COMEAP, 2010). Short-lived high pollution episodes can lead to acute health impacts from exposure to $PM_{2.5}$ over shorter time periods, leading to deaths being brought forward among an exposed population (Stedman, 2004). $PM_{2.5}$ is composed of both solid and liquid droplets suspended in

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the atmosphere, which are small enough to be inhaled deep into the lungs (Raaschou-Nielsen et al., 2013). Emissions of primary $PM_{2.5}$ and secondary $PM_{2.5}$ precursors come from a wide range of sources including combustion for power generation, heating and from vehicles as well as dust and sea spray. There is little evidence to suggest which chemical constituents of the PM present an increased health risk and whether there is a safe limit of exposure for health effects (COMEAP, 2009).

Previous research on UK air pollution has focussed on the health impacts or mechanisms of short-term high pollution event case-studies (Macintyre et al., 2016; Vieno et al., 2016; Stedman, 2004). Macintyre et al. (2016) found exposure to high $PM_{2.5}$ concentrations (maximum hourly concentration - 83 μgm^{-3} at urban background sites) during a 10-day spring pollution episode in 2014 brought forward 600 deaths, 840 emergency respiratory and 730 emergency cardiovascular hospital admissions. This equated to a doubling of hospital admissions compared with those under typical springtime conditions. Stedman (2004) quantified the impact of high ozone and particulate matter with a diameter less than 10 μm (PM_{10}) concentrations during the summer 2003 heatwave using a dose-response function. They found that 471 deaths were brought forward, attributable to exposure to PM_{10} during the two-week pollution event, representing an increase of 207 deaths compared to the same period in 2002. This agrees with previous work that found a large proportion of the deaths brought forward resulted from elevation of pollutant concentrations rather than the direct impact of high temperatures (Rooney et al., 1998).

Since UK concentrations of $PM_{2.5}$ were not routinely monitored until 2008, when the New Air Quality Directive was introduced by the European Union, previous studies focussing on the drivers of high pollution episodes have analysed PM_{10} observations. These studies used back trajectories to link observations of high PM_{10} concentrations with possible source regions (King and Dorling, 1996; Stedman, 1996). King and Dorling (1996) found that on days where PM_{10} concentrations exceeded 50 $\mu g\ m^{-3}$ in 12 UK cities and at two rural sites, local emissions represented a small fraction of the total concentration and each episode was dominated by easterly flow. They suggested that, since the back trajectories emanated from mainland Europe, long-range transport has a large contribution in the overall PM_{10} concentrations observed. They concluded that more work was required to confirm this, over a longer period with observations at rural sites.

Harrison et al. (2012) used 37 urban-background observational sites from the UK Automated Urban and Rural Network (AURN) to examine $PM_{2.5}$ concentrations for the year 2009 in order to better understand processes affecting concentrations across the UK. The study used meteorological data from 8 sites to determine the wind direction and wind speed at the AURN sites. They found that $PM_{2.5}$ concentrations were below the annual mean when winds were from westerly flows, while for south-easterly, easterly and north-easterly flows they were above the annual mean. This was attributed to emissions from continental Europe under easterly and south-easterly flow. The work of Harrison et al. (2012) suggests that the long-range transport of pollutants to the UK is associated with specific meteorological conditions. However, due to the short observational record at the time, the research used only one year of $PM_{2.5}$ observations, the sample size for individual wind directions was small. This meant relationships between wind direction and $PM_{2.5}$ observations could not be established over a longer period of time to be statistically robust.

This study builds upon the work of Harrison et al. (2012) with observations that have increased coverage both spatially (42 sites compared to 37) and temporally (2010–2016). Additionally, we use Lamb Weather Types (LWTs) rather than local meteorological observations to investigate the relationship between synoptic meteorology and the transport of pollutants. LWTs reflect the synoptic-scale conditions, rather than local meteorology, and so are more closely related to the transport of pollutants.

In recent years Lamb weather types (LWT) (Lamb, 1972) and circulation weather types (CWT) have become an increasingly popular method of investigating the impact of regional atmospheric circulation patterns on pollutant concentrations (Russo et al., 2014; Grundstrom et al., 2015, 2017; Tang et al., 2009, 2011; Pope et al., 2014; 2016; Demuzere et al., 2009). These can be used to classify synoptic scale atmospheric circulation patterns over regions such as the UK using wind direction, speed and circulation strength. The application of LWTs alongside observations of pollutant concentrations (PM_{10}, NO_2, O_3 and birch pollen) allows the association of different wind directions with the long-range transport of pollutants and the build-up or dispersion of pollutants for large areas. This allows relationships to be derived between specific weather types and higher pollutant concentrations over longer time periods. Previous work by Demuzere et al. (2009) in the Netherlands found that PM_{10} concentrations increased when air was transported from the east and south during summer when there were dry conditions and high temperatures. Liu et al. (2017) also found that $PM_{2.5}$ concentrations in the United States were closely controlled by temperature, finding tropical weather types were associated with significantly higher $PM_{2.5}$ concentrations and polar weather types with low $PM_{2.5}$ concentrations.

The UK, given its close proximity to Europe, is often subject to pollution episodes propagating from the continent. Pope et al. (2014, 2016) used LWTs to investigate the influence of meteorology on NO_2 and O_3 concentrations in the UK. The research found that both pollutants are strongly influenced by wind and circulation patterns. The highest O_3 concentrations occurred under summer anticyclonic conditions due to large scale subsidence limiting vertical mixing. The study also identified that south-easterly and north-easterly flow increased mean UK ozone concentrations by between 10 and 15 $\mu g\ m^{-3}$ (Pope et al., 2016). NO_2 concentrations were found to significantly increase under winter-time anticyclonic conditions through pollutant accumulation and were enhanced under south-easterly flow due to long-range transport of pollutants from continental Europe (Pope et al., 2014). They attributed the winter increase to be a result of the combined effect of increased emissions, more stable conditions and decreased photolysis allowing accumulation over emission sources.

Here, we present the first study to use long-term (2010–2016) observations of $PM_{2.5}$, sub-sampled under LWTs, back trajectories and an atmospheric chemistry transport model, to investigate how climatological weather regimes influence UK surface particulate matter air quality.

2. Data and methods

An overview of the methods used in this study can be found in Fig. 1 for reference.

2.1. Observations

2.1.1. Lamb Weather Types

Lamb Weather Types (LWTs) are a synoptic classification of daily weather patterns across the UK (Lamb, 1972). LWTs are a useful tool for UK air pollution studies. They indicate the large-scale atmospheric flow and air mass origins, linking each air mass to specific dispersion conditions and mesoscale meteorology that control the regional transport of air pollution (Dayan and Levy, 2004). In this work we use LWTs calculated automatically (using the algorithm from Jenkinson and Collinson (1977)) from NCEP reanalysis between 1948 and present (Jones et al., 2013). NCEP reanalysis are available at 2.5° at 00, 06, 12 and 18Z each day (Kalnay et al., 1996). The 12Z reanalysis is used to calculate the LWT each day. We have confidence in the reliability of LWT classification from NCEP reanalysis since Jones et al. (2013) found LWT calculated from NCEP reanalysis correlated well (0.65–0.79) with the subjective LWTs of Lamb (1972). Each LWT is calculated using the daily mean of three variables from NCEP reanalysis, which characterise the circulation at the surface over the UK at 1200Z. Variables used are (i) the mean flow direction, (ii) the strength of mean flow and (iii) the mean

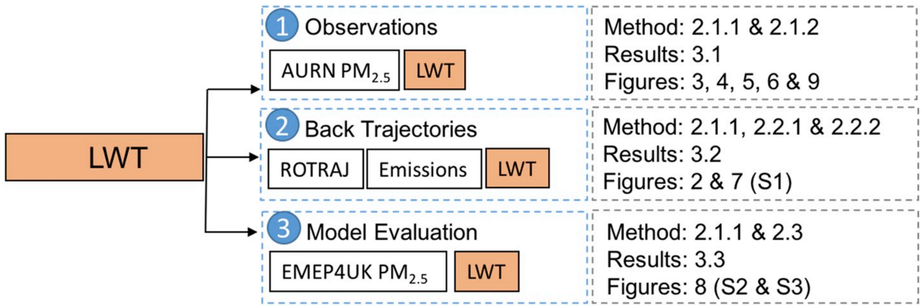

Fig. 1. An overview of the datasets and method used in this study. Lamb weather types (LWT) are combined with observations of $PM_{2.5}$ concentrations from the Automated Urban and Rural Network (AURN), back trajectories from the Reading Offline Trajectory Model (ROTRAJ) and a gridded emission dataset. We also compare our results to a chemistry transport model (EMEP4UK, 2018) (Fig. 2).

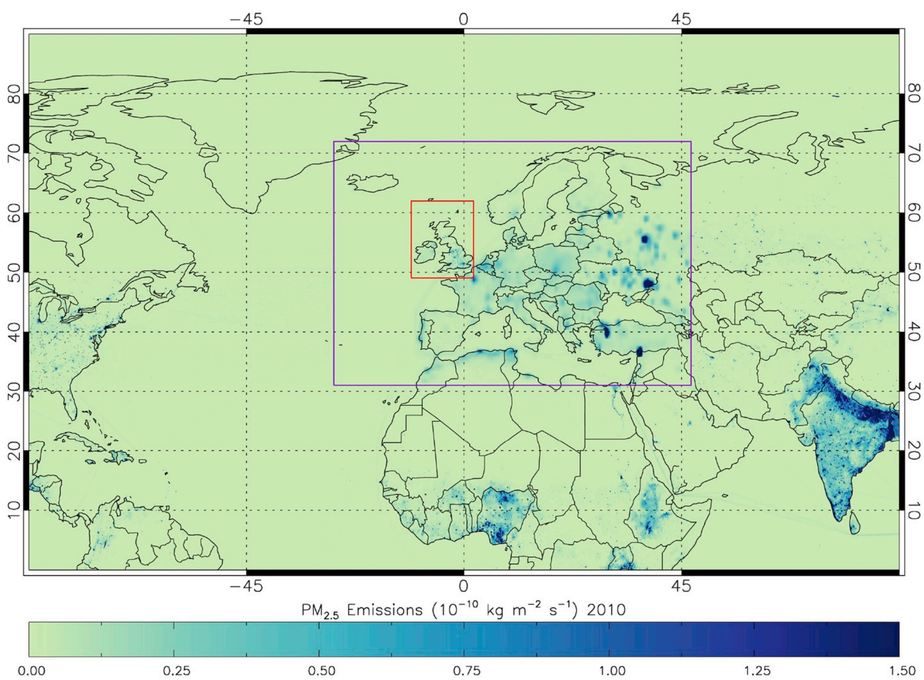

Fig. 2. Gridded emissions of primary $PM_{2.5}$ for 2010 are shown as an example (annual varying emissions were used between 2010 and 2014). For the outer domain (Purple Box), gridded annual EMEP emissions at 0.5° resolution from the Centre for Emission Inventories and Projections (CEIP, 2018, www.ceip.at) are used. While for the inner domain (Red Box) gridded annual National Atmospheric Emissions Inventories (NAEI, 2015) emissions at 0.01° resolution are aggregated to 0.05° resolution. Emissions outside of Europe are provided by the Emission Database for Global Atmospheric Research with Task Force on Hemispheric Transport of Air Pollution (EDGAR-HTAP) version 2.2 emissions for 2010 at 0.1° resolution (Janssens-Maenhout et al., 2015). More information in Supplementary Material: Sect. 1.2.

strength of the circulation pattern (vorticity) (Jones et al., 2013). Based on this analysis, conditions on a given day are classified as one of 28 LWTs. The 28 different LWTs comprise of three circulation types: Anticyclonic (0), Cyclonic (20) and Unclassified (−1), and eight wind types: N, NE, E, SE, S, SW, W, NW. We use a similar grouping method to O'Hare and Wilby (1995) and Pope et al., 2014, grouping the LWTs into eight wind types (Table 1). We however, use 0, 20 and −1 to classify the synoptic types, like Otero et al. (2018). This allows the independent examination of circulation and wind direction on pollutant concentrations.

2.1.2. Automated Urban and Rural Network

We use observed $PM_{2.5}$ concentrations taken from the Automated Urban and Rural Network (AURN). AURN is the largest automated air quality monitoring network in the UK with 145 sites measuring species including $PM_{2.5}$, NO_2, SO_2 and O_3. AURN sites are classified as urban traffic/kerbside, urban or suburban background, and rural background. For this study background sites are used (urban background, suburban background and rural background). Background sites are chosen as they are considered to be more representative of the surrounding region than kerbside sites. This is because their locations are chosen so as to be influenced by the integrated contribution of all sources upwind rather than by a single souce or street (DEFRA, 2018). Data from 42 sites is used; 39 of which are urban background (UB), 2 rural background (RB) and 1 background suburban (BS) (Table S1). We use daily mean $PM_{2.5}$ concentrations (calculated from hourly measurements) from the 42

Table 1
27 LWT classifications (Jenkinson and Collinson, 1977), the 11 LWTs used in this study are NE, E, SE, S, SW, W, NW, N, Anticyclonic, Unclassified & Cyclonic. There are 8 wind types NE, E, SE, S, SW, W, NW & N, shown in the left columns, and 2 circulation types (anticyclonic, cyclonic), in the top row, and unclassified days, where wind speed and shear were too low to allow classification. Following our grouping of LWT into 11 types, LWT index 3 (ASE) would fall under the south-easterly classification (see outer column and row of Table 1). There is also one other LWT (−9: non-existent day) not used in this study.

This Study	Anticyclonic	Neutral	Cyclonic
		−1 UC	
–	0 A	–	20 C
NE	1 ANE	11 NE	21 CNE
E	2 AE	12 E	22 CE
SE	3 ASE	13 SE	23 CSE
S	4 AS	14 S	24 CS
SW	5 ASW	15 SW	25 CSW
W	6 AW	16 W	26 CW
NW	7 ANW	17 NW	27 CNW
N	8 AN	18 N	28 CN

background sites for the period of 1st January 2010–31st December 2016. Quality assurance checks are performed by DEFRA before data release (DEFRA, 2009). Thus we only perform basic data quality control on the daily data on two different time scales: annual and monthly. For annual statistics, sites are only used if fewer than 10% of days (per year) are missing. Monthly data for seasonal statistics is only used if fewer than 10% of days (per month) have missing data.

2.2. Back trajectories and integrated emissions

Reddington et al. (2014) showed that the use of back trajectories and emissions can be a powerful tool in understanding the influence of emissions on local air quality due to long-range transport in air masses arriving in Singapore. Following a similar method, we quantify the importance of the relationship between LWT and AURN $PM_{2.5}$ concentrations.

2.2.1. Emissions

Emissions from the National Atmospheric Emission Inventory (NAEI), European Monitoring and Evaluation Programme (EMEP) and Emission Database for Global Atmospheric Research with Task Force on Hemispheric Transport of Air Pollution (EDGAR-HTAP) are combined to create a gridded emission dataset (Fig. 2). More details in Supplementary Material: Sect. 1.2.

2.2.2. Reading Offline Trajectory Model (ROTRAJ)

We combine back trajectories and bottom-up emission estimates in order to investigate the influence of long range transport of pollutants on ambient pollutant concentrations under different LWTs in the UK. We use primary $PM_{2.5}$ emissions integrated over air mass back trajectories to determine the relative influence of direct $PM_{2.5}$ emissions on air masses (i.e. ROTRAJ back trajectories) arriving at different times and locations over the UK. Back trajectories are calculated using the ROTRAJ offline Lagrangian transport model (Methven et al., 2003). The model uses ERA-Interim reanalysis from the European Centre for Medium Range Weather Forecasting (ECMWF) to provide velocity fields for the simulations at 1.0125° horizontal resolution. After a trajectory parcel is released the location of each trajectory parcel is calculated every 6 h by vertical cubic Lagrange interpolation and horizontal bilinear interpolation. This method accounts for large scale advection since the winds are resolved but does not resolve small scale sub-grid turbulent transport.

In this study, ROTRAJ back trajectories were initialised from just above the surface (0.99 sigma level; a terrain following coordinate system where 1 is the surface) at 12:00 UTC to match the LWT dataset between 2010 and 2014 at all background AURNsites (42 sites), extending back 4 days in 6-hourly time steps. $PM_{2.5}$ emissions were accumulated along each trajectory over 4 days at 15-min time intervals (interpolated linearly from 6-hourly position output). $PM_{2.5}$ emissions were only accumulated when the trajectory path was at pressures greater than 850 hPa (as an approximation of being within the boundary layer). At each location, we accumulate the entire emission within an emission grid box over which the trajectory passes. The surface area of each grid box that the trajectory points passed over is also accumulated over time. To approximate for dilution and chemical loss of $PM_{2.5}$ along the trajectory path, e-folding lifetimes were applied to the total $PM_{2.5}$ accumulated emission in the air parcel. A range of lifetimes of 1, 3, 7 and 14 days were applied to investigate the sensitivity of the final $PM_{2.5}$ accumulation, on arrival to the respective AURN sites, to loss processes (Supplementary Material: Sect. 5.3). The along-trajectory emission accumulation can be represented by Eq. (1):

$$E_i = [E_{i-1} + \varphi_i.\Delta t.\alpha_i]e^{-\Delta t/\tau} \quad i = 1,\ N\ (=384) \text{ and } E_0 = 0.0 \tag{1}$$

where E_N is total accumulated $PM_{2.5}$ mass (kg), N is the number of time steps within the trajectory (384), E_i is accumulated $PM_{2.5}$ (kg) at any given point i along the trajectory, φ_i is the emissions flux of $PM_{2.5}$ (kg m^{-2} s^{-1}) at point i, Δt is the 15-min time step, α_i is the surface area of the grid box (m^2) at point i and τ is the assumed e-folding $PM_{2.5}$ lifetime(s).

To remove the dependence on emission grid resolution (since we assumed the air mass has the same width as the emission grid box), the total accumulated $PM_{2.5}$ mass (E) was divided by accumulated surface area (S) and then scaled by 10^9. This results in E having units of μg m^{-2}. S is given by Eq. (2):

$$S = \sum_{i=1}^{N} \alpha_i \tag{2}$$

E_{UK} is also determined using the same approach, but only implemented when the trajectories enter the UK region defined by a longitudinal-latitudinal box (8°W-2°E, 50–60°N). To derive E_{UK} in units of μg m^{-2} the accumulated $PM_{2.5}$ mass from the UK is divided by the accumulated surface area (S) over the full trajectory path. The ratio between E_{UK}/E represents the fractional contribution of UK sources towards the total accumulated $PM_{2.5}$ emissions.

Finally, the daily (12:00 UTC) total accumulated emission and E_{UK}/E ratios from all sites are binned by the LWTs. This methodology provides a powerful tool to identify which flow directions, as classified by the LWTs, are the most polluted and the proportion of pollutant emissions from long range transport (e.g. continental Europe) versus local sources.

2.3. European Modelling and Evaluation Programme for the UK (EMEP4UK) $PM_{2.5}$ data

Since the UK observational network is very sparse and so only gives limited spatial coverage we sample the European Modelling and Evaluation Programme for the UK (EMEP4UK) (v4.17) model (EMEP4UK, 2018) under different LWTs to look further into the spatial distribution of $PM_{2.5}$ concentrations. The model covers the UK at 0.05° resolution using a nested approach from the coarser European wide EMEP model (Simpson et al., 2012). Further details of the model set-up can be found in the Supplementary Material: Sect. 1.4.

We tested the model's skill in reproducing variability in UK $PM_{2.5}$ concentrations both temporally and spatially. The model captures the variability in $PM_{2.5}$ concentrations and their relationship with LWT well and shows strong correlation with observations and anomalies at each site (r = 0.887 and 0.905 respectively) with only a small negative bias (1 μg m^{-3}) (Supplementary Material: Sect. 1.4). Therefore, we can have good confidence in the model's ability to represent ambient $PM_{2.5}$ concentrations.

3. Results

3.1. AURN $PM_{2.5}$ observations 2010–2016

We find a strong dependency of observed $PM_{2.5}$ abundance on wind flow and circulation pattern, as characterised by the LWTs, with enhanced $PM_{2.5}$ concentrations under easterly, south-easterly and southerly flow and anticyclonic and unclassified weather types. Fig. 3 a shows the daily mean AURN concentrations of $PM_{2.5}$ binned into the 11 different LWT regimes for the years 2010–2016. The multi-annual mean for all sites and all LWTs is 11 μg m^{-3}. We find that the average $PM_{2.5}$ concentrations binned according to LWT regimes follow a coherent pattern; mean concentrations of $PM_{2.5}$ in easterly, southerly and south-easterly flow directions are elevated above the annual mean (15–20 μg m^{-3}). Easterly, southerly and south-easterly flow regimes also have 90th percentile concentrations of 28–35 μg m^{-3} (10–20 μg m^{-3} higher than other flow directions). 10th percentile concentrations under these regimes are also elevated (2.5–4.5 μg m^{-3} higher than other regimes). These flow types occur on 3, 5 and 8% of days . Northerly, north-easterly, south-westerly, westerly and north-westerly flows all give mean $PM_{2.5}$ concentrations below the multi-annual mean. The lowest concentrations in the 75th and 90th percentiles also occur under

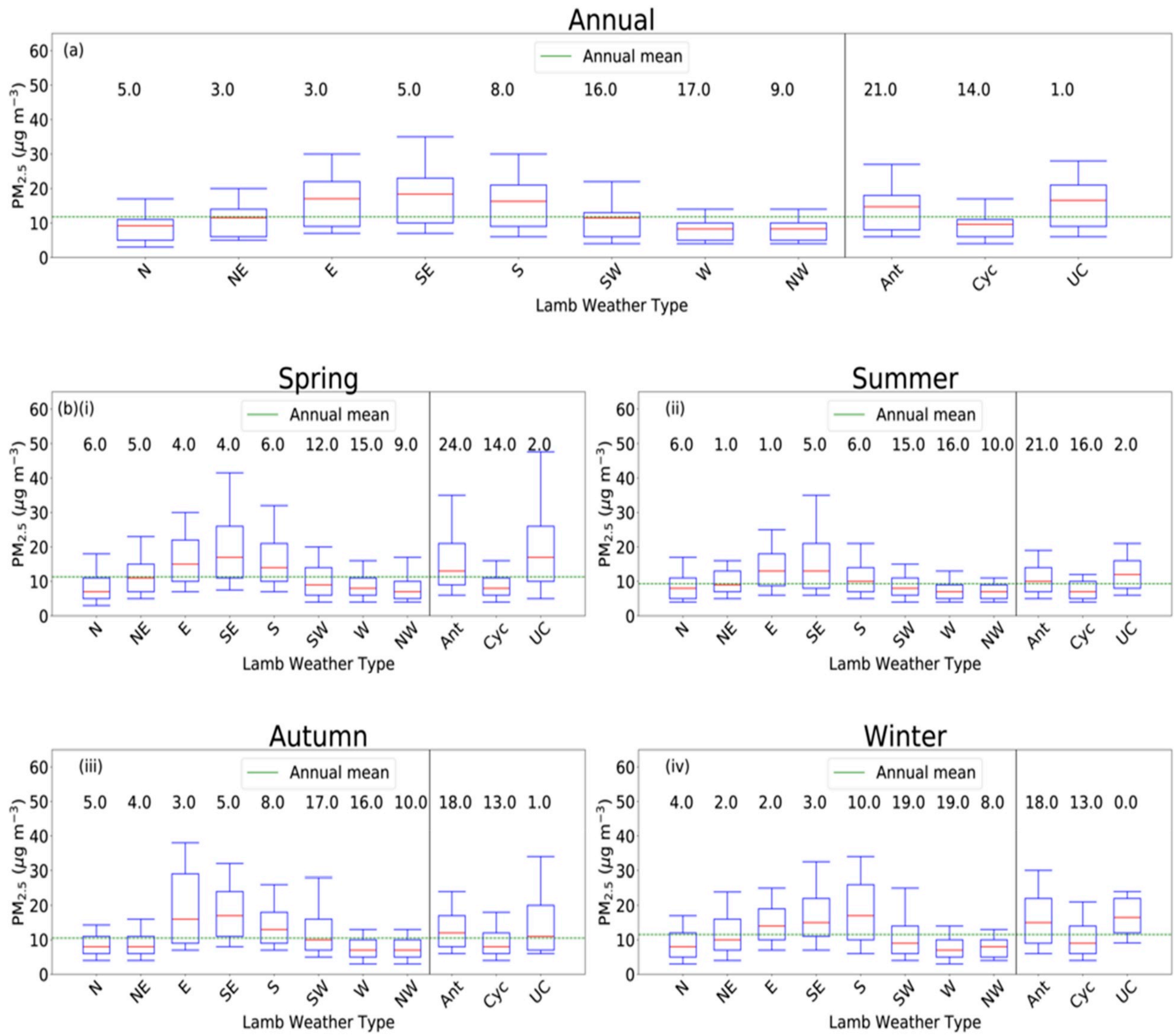

Fig. 3. (a) Annual observations and (b) seasonal: (i) spring (ii) summer (iii) autumn (iv) winter observations of $PM_{2.5}$ concentrations from 42 UK AURN sites between 2010 and 2016 under different Lamb Weather Types (LWTs) ($\mu g\ m^{-3}$). Mean concentrations are shown in red, with the 10th, 25th, 75th and 90th percentiles in blue. The mean of all LWTs, is shown by the green dashed line. The frequency of each LWT (in %) for the 2010–2016 period, annually and seasonally is also indicated.

westerly, north-westerly and northerly flow types (<10.0, <11.0 & <20.0 $\mu g\ m^{-3}$ respectively). Westerly, north-westerly and south-westerly weather types occur on a larger proportion of days each year (17, 9 and 16% of days). Mean $PM_{2.5}$ concentrations are also affected by the circulation type; elevated concentrations are found under anticyclonic and unclassified conditions (mean concentrations of 14.7 and 16.6 $\mu g\ m^{-3}$), exceeding the annual mean concentration. Although anticyclonic conditions are associated with lower mean $PM_{2.5}$ concentrations than easterly, south-easterly and southerly flow, they occur much more frequently (21% of days). Therefore, they have a more important contribution to the annual mean concentration and thus, the population's long-term exposure to $PM_{2.5}$. In contrast, $PM_{2.5}$ below the annual mean concentrations are found under cyclonic flows (9.6 $\mu g\ m^{-3}$), occurring on 14% of days.

The distribution of observed concentrations with LWT and proportion of occurrences of LWT for spring (MAM), summer (JJA) and winter (DJF) follows a similar pattern as that seen annually (Fig. 3 b (i, ii, iv)), although there is some seasonal variability. In autumn (SON) (Fig. 3 b (iii)) the highest $PM_{2.5}$ concentrations are found under easterly, south-easterly and unclassified flows, occurring 2–8% of the time. Whereas in winter the highest concentrations are found in southerly, south-easterly and anticyclonic flows, with a small increase in the number of occurrences of southerly types (10%). 90th percentile concentrations are highest in spring under the unclassified type and are the highest observed of any season (47.6 $\mu g\ m^{-3}$), although they only occur 2% of the time.

Fig. 4 shows the geographical distribution of annual mean $PM_{2.5}$ concentrations under all LWTs. Concentrations are highest in the south of England (12–16 $\mu g\ m^{-3}$) and decrease northward, with the lowest concentrations observed in Scotland and Northern Ireland (0–4 $\mu g\ m^{-3}$). 33 of the 42 sites in England have multi-annual mean concentrations above 10 $\mu g\ m^{-3}$, the WHO recommended limit, and 20 are above the multi-annual mean of all sites (11 $\mu g\ m^{-3}$).

To examine the geographic distribution of the effect of LWT on $PM_{2.5}$ concentrations (Fig. 5 a-f), we calculate the $PM_{2.5}$ anomaly under each LWT for individual sites with respect to the multi-annual mean concentration at that site (Fig. 4). We also test for statistical significance at each AURN site under each LWT using a one million sample Monte Carlo simulation, in which we randomly sample $PM_{2.5}$ concentrations for all LWT between 2010 and 2016 to build up a distribution of concentrations containing one million random samples. We then take the mean $PM_{2.5}$ concentration for a given LWT and site (e.g. SE site 1), if this lies above

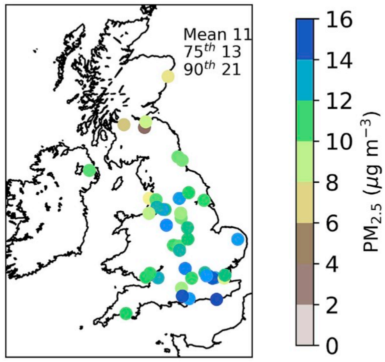

Fig. 4. Multi-annual mean $PM_{2.5}$ concentrations from 42 UK AURN monitoring sites (2010–2016 average in μg m^{-3}) averaged over all LWT regimes. The mean, 75th and 90th percentile $PM_{2.5}$ concentration calculated from all sites is shown on the top right of each panel.

the 95th or below the 5th percentile of the one million-sample distribution we can conclude that the concentration observed did not occur by chance and is significantly different statistically. The process is repeated for each LWT, creating a new distribution each time. Statistically significant anomalies ($p < 0.05$) are subsequently circled in black (Fig. 5 a-f). In line with the previous analysis, $PM_{2.5}$ concentrations are enhanced by between 28 and 35% under easterly, south-easterly and southerly flow (Fig. 5 a, b & c). Some sites experience LWT flow direction anomalies of up to 12 μg m^{-3} (Wigan Centre), and 40 of the 42 sites exhibit a positive anomaly under south-easterly and southerly flow with a mean anomaly of 6.1 μg m^{-3} and 4.4 μg m^{-3} respectively under these flows. $PM_{2.5}$ concentrations are affected by LWT across the whole of the UK with the northernmost extent reaching to Scotland and Northern Ireland. Northerly, westerly and north-westerly are the three flow directions associated with the largest $PM_{2.5}$ reductions (−5 μg m^{-3}, equivalent to a 30–44% reduction) (Fig. 5 e, f & g). The negative anomalies under these flow regimes are present at the same number of sites (40 of 42) but the anomaly is smaller in magnitude than the positive anomaly from the easterly, south-easterly and southerly flows with mean negative anomalies of −2 to −3 μg m^{-3}.

The effect of circulation pattern on $PM_{2.5}$ concentrations is generally weaker than that of flow direction. This suggests that long-range transport of $PM_{2.5}$ rather than the build-up of local pollutant emissions is more important in controlling $PM_{2.5}$ concentrations. Despite this, the presence of anticyclonic and cyclonic conditions has an influence on $PM_{2.5}$ across the UK, with a maximum multi-annual anomaly of 4.6 and −4.4 μg m^{-3} respectively (Fig. 6 a & b). This represents a 20% increase and 24% decrease, respectively. Both of these flow types also occur more frequently (21% and 14%), meaning they are more important in contributing to the annual mean concentration and thus, the population's long-term exposure to $PM_{2.5}$.

3.2. Back trajectories and integrated emissions

Variability in the back-trajectory integrated emissions sampled at the UK AURN sites further supports the relationships between in-situ observed $PM_{2.5}$ and wind direction discussed above. Fig. 7 shows the median accumulated primary $PM_{2.5}$ emissions along ROTRAJ back trajectories arriving between 2010 and 2014, binned by the LWT flow directions. Here a representative 7-day e-folding lifetime (Seinfeld and Pandis, 2016) is used to approximate for physical/chemical loss processes from the air parcel. In the supplementary material (Supplementary Material: Sect. 1.3) we explore the sensitivity of the accumulated $PM_{2.5}$ emissions to different e-folding lifetimes. This showed that for shorter e-folding lifetimes (1 and 3 days) integrated emissions are dominated by UK emissions and there is little change between the total summed emission with different LWT. While, at larger e-folding lifetimes (7 and 14 days) the total integrated emission UK contribution and

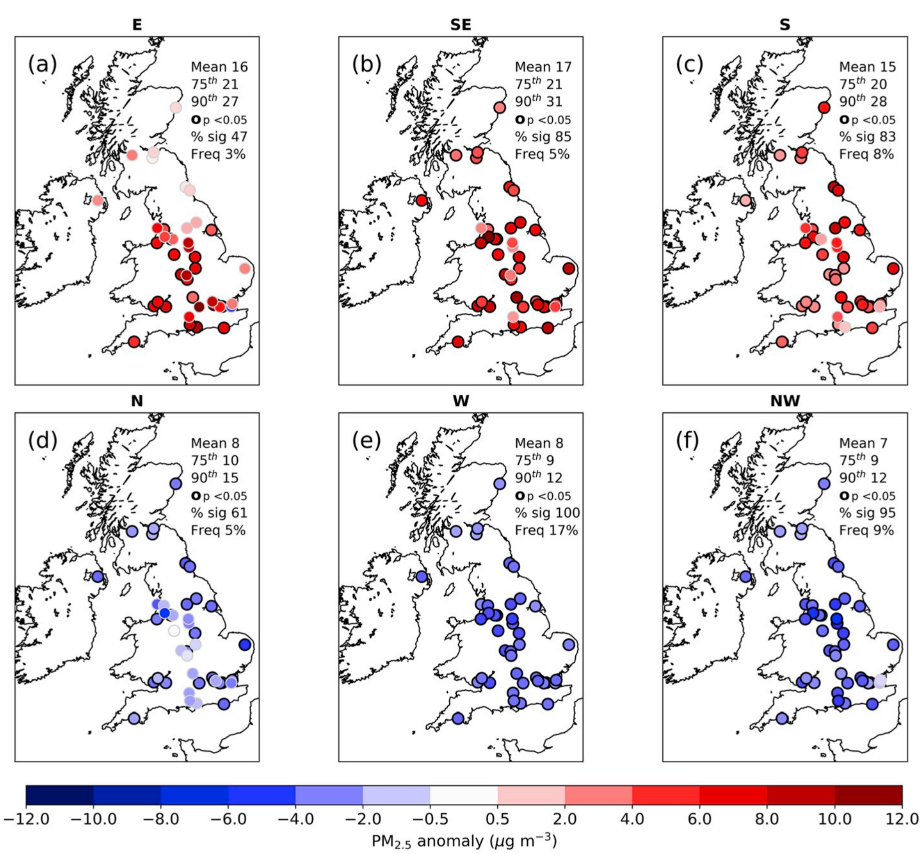

Fig. 5. The multi-annual mean $PM_{2.5}$ anomaly relative to annual mean concentration averaged over all LWT regimes (relative to multi-annual mean concentration averaged over all LWT regimes (2010–2016) (in μg m^{-3}), shown in Fig. 4) under different flows directions is shown in panels (a) to (f). For clarity, we show the three flow directions with the largest positive anomaly ((a) easterly, (b) south-easterly and (c) southerly) and the three flow directions with the largest negative anomaly ((d) northerly, (e) westerly and (f) north-westerly). The mean, 75th and 90th percentile $PM_{2.5}$ concentrations calculated from all sites are shown on the top right of each panel. Sites where the anomaly is statistically significant ($p < 0.05$) are indicated by black contouring and the percentage of sites where anomalies are statistically significant is also indicated in the top right panel (% sig). The frequency of each LWT (in %) for the 2010–2016 period is also indicated.

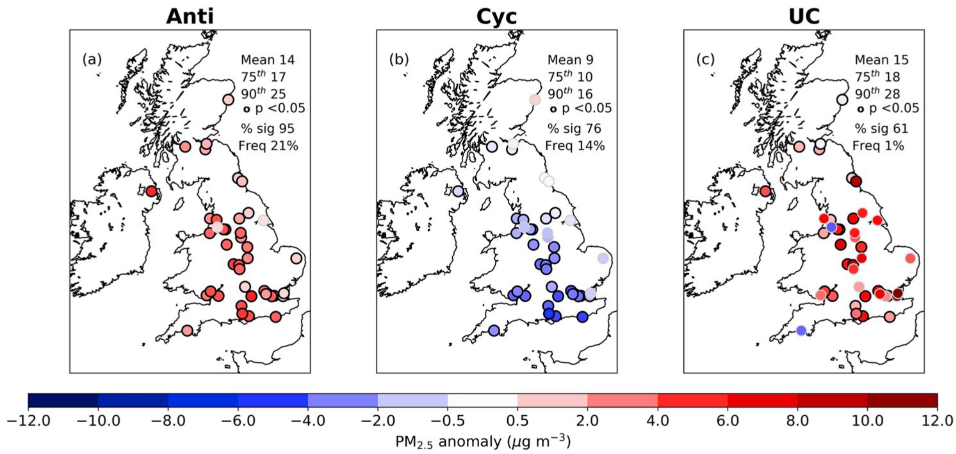

Fig. 6. AURN annual mean $PM_{2.5}$ anomalies (relative to multi-annual mean concentration averaged over all LWT regimes (2010–2016 (in μg m^{-3}), shown in Fig. 4). Concentrations and anomalies sampled under (a) anticyclonic (b) cyclonic and c) unclassified weather types are shown. The mean, 75th and 90th percentile $PM_{2.5}$ concentration calculated from all sites is shown on the top right of each panel. Sites where the anomaly is statistically significant ($p < 0.05$) are indicated by black contouring and the percentage of sites where anomalies are statistically significant is also indicated in the top right panel (% sig). The frequency of each LWT (in %) for the 2010–2016 period is also indicated.

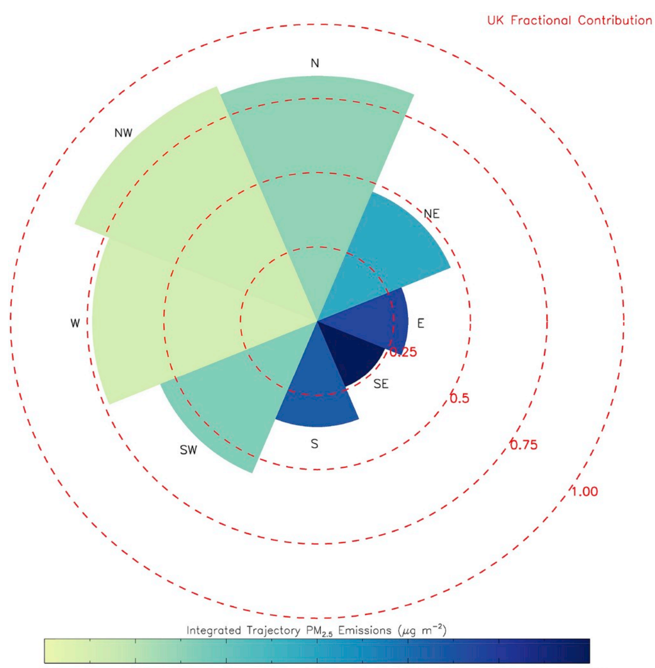

Fig. 7. Median UK (background AURN sites) integrated $PM_{2.5}$ emissions (μg m^{-2}) accumulated over the daily (12 UTC, 2010–2014) ROTRAJ back trajectories (4 days–15-min time steps), with a 7-day e-folding lifetime, binned by LWT flow directions. Red circles represent the UK fractional contribution to trajectory accumulated $PM_{2.5}$ emissions.

the total summed emission varies more between LWT. Since this method cannot account for secondary $PM_{2.5,}$ the total accumulated $PM_{2.5}$ emissions should be interpreted as a proxy of how polluted each air mass is, and the fractional contribution of emissions inside and outside of the UK to the total loading (akin to using CO as a tracer), rather than an estimate of $PM_{2.5}$ in the atmosphere.

Overall, the results support the LWT-AURN $PM_{2.5}$ relationships with peak median accumulated emissions (E) from the south-easterly, southerly and easterly directions (0.25–0.3 μg m^{-2} accumulated primary $PM_{2.5}$ emission). This supports the idea that continental European primary emissions are contributing to poor UK air quality when UK-bound air masses pass over polluted source regions (e.g. the Benelux region and west Germany). The fractional contribution from UK emissions under these flow directions is between 25 and 50%, indicating that under these flows more than 50% of emissions contributing to UK primary particulate pollution originate in continental Europe. The north-westerly and westerly flow directions correspond to the cleanest air masses (<0.1 μg m^{-2} accumulated primary $PM_{2.5}$ emission), again in agreement with the LWT-AURN $PM_{2.5}$, as the back trajectories primarily originate from over the North Atlantic. Here, the UK fractional contribution is much larger (~70–85%) as the majority of the accumulated $PM_{2.5}$ emission (E) is from within the UK domain (i.e. E_{UK} is relatively large). Exterior emissions sources will include Ireland and potentially sources where back trajectories tails originate in Europe, over source regions, but loop around to the UK West coast.

3.3. EMEP4UK modelled $PM_{2.5}$ and LWT

Since AURN observations give sparse coverage of the UK, we use EMEP4UK surface $PM_{2.5}$ fields to further investigate the processes affecting ambient $PM_{2.5}$ concentrations under different LWT classifications. In the supplementary material, we show that the model has skill in reproducing $PM_{2.5}$ concentrations ($r = 0.887$) and anomalies ($r = 0.905$) under different LWTs when compared with AURN observations. Therefore, we have confidence in EMEP4UK's representation of ambient $PM_{2.5}$ concentrations when sub-sampled under the LWTs.

EMEP4UK reproduces the back trajectory and AURN-LWT analysis with the largest positive $PM_{2.5}$ anomalies observed under easterly, south-easterly and southerly weather types due to the long-range transport of $PM_{2.5}$ from continental Europe (positive anomalies of 6–12 μg m^{-3}). The addition of the model 10-m winds, also sub-sampled under the LWTs, adds valuable information of the flow characteristics. Here, the flow clearly originates from the continent (typically around 5 m s^{-1}) and is closely aligned with the spatial anomaly features (Fig. 8).

The largest negative anomalies (−10 to −4 μg m^{-3}) are associated with the transport of clean air masses from the Atlantic, as indicated by the back-trajectory analysis. Northerly, north-westerly and westerly flow all have wind speeds between 5 and 10 m s^{-1} transporting $PM_{2.5}$ offshore away from source regions. However, under north-easterly and south-westerly flow directions, a strong $PM_{2.5}$ anomaly gradient can be observed across the UK. Negative anomalies (−6 to −2 μg m^{-3}) over the northern (south-western) UK represent the more gradual replacement of polluted air masses under north-easterly (south-westerly) flow. Transport of the polluted air mass yields positive anomalies (2–6 μg m^{-3}) over the southern (north-eastern) UK region. Here, the model adds important spatial details which are less reliably captured in the observations.

Circulation influences are also further investigated using the model, where anticyclonic conditions show reduced onshore transport of pollutants due to relatively weak easterly winds (under 5 m s^{-1}) from continental Europe. This leads to conditions favourable for the build-up of $PM_{2.5}$ (2–6 μg m^{-3}) predominantly from local emissions/formation. While under cyclonic conditions, $PM_{2.5}$ is transported offshore, into the North Sea, by strong westerly winds (10 m s^{-1}) from the Atlantic leading

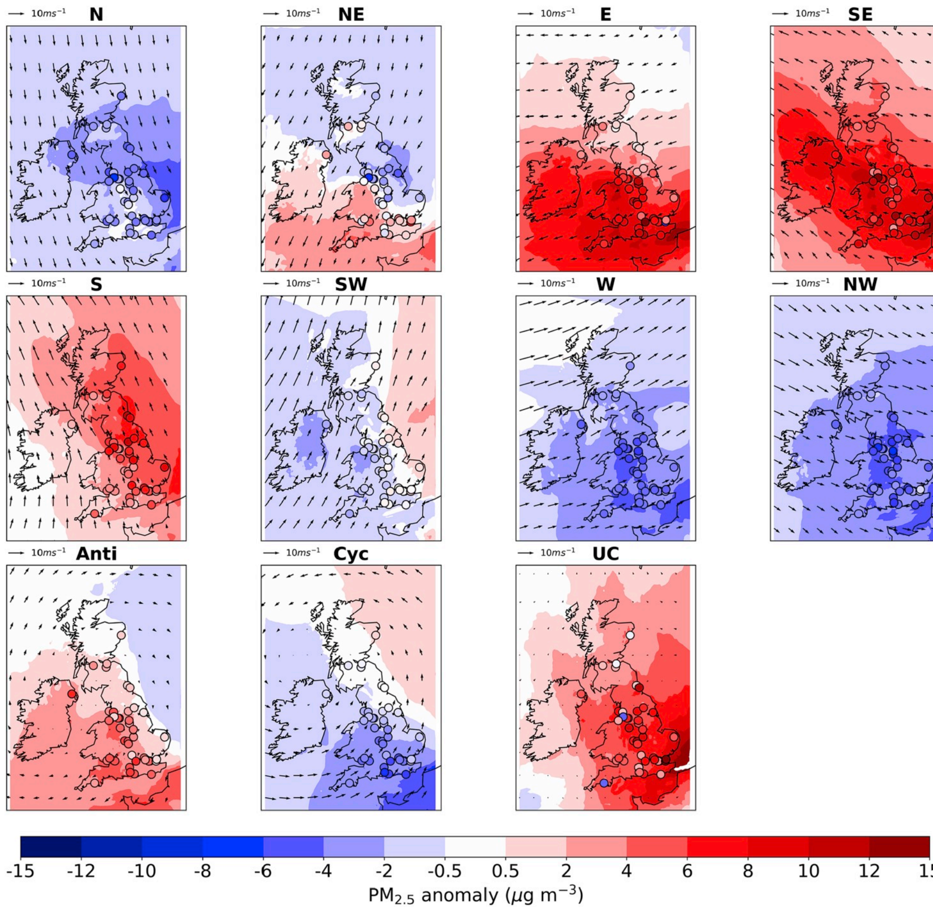

Fig. 8. The multi-annual mean $PM_{2.5}$ anomaly relative to annual mean concentration averaged over all LWT regimes (relative to multi-annual mean concentration averaged over all LWT regimes (2010–2016) (in μg m^{-3})) under different flow directions from the EMEP4UK model. 10m winds, also from the EMEP4UK model (nudged to 3-hourly GFS analysis), are over plotted. All LWT wind directions and synoptic types are shown.

to decreased concentrations over the UK mainland (−6 to −2 μg m^{-3}). Unclassified weather types are characterised by slack winds (0–2 m s^{-1}) over the UK leading to the build-up of local emissions due to stagnant air masses (4–10 μg m^{-3}). This all further supports the importance of how flow characteristics (i.e. long-range transport and stagnation) influence UK $PM_{2.5}$ concentrations.

3.4. Contribution of LWTs to the Daily Air Quality Index (DAQI)

To put the results of Sect. 3.1 in a public health context, we bin the daily mean $PM_{2.5}$ concentrations under the different LWTs according to the 10 UK Daily Air Quality Index (DAQI, 2018) $PM_{2.5}$ concentration bands. The UK Daily Air Quality Index (DAQI) is a public health air quality warning system used by the UK Department for Environment, Food and Rural Affairs (DEFRA) to communicate current and future pollutant levels in the UK to the general public (COMEAP, 2011). Five key pollutants have been identified to monitor by the Committee on the Medical Effects of Air Pollutants (PM_{10}, O_3, NO_x, SO_2 and $PM_{2.5}$). In order to gain an overall DAQI each of these pollutants are given an individual score between 1 (low) and 10 (very polluted). The overall DAQI is then assigned based on the highest individual DAQI value for each of the 5 pollutants at a given time. For example, if O_3 scored a DAQI value of 9, and all other species scored a value of 2, the overall DAQI would be 9. As we only investigate $PM_{2.5}$, we can only comment on the effect of LWT on $PM_{2.5}$ within the DAQI (PM DAQI). However, since the DAQI score is assigned the highest individual species score from each of the 5 pollutants, air masses with high or very high $PM_{2.5}$ scores are likely to have the same overall DAQI score.

We find that 71% of days classed as “very polluted” (PM DAQI of 10) occur with south easterly, southerly and south westerly flows, whereas only 12% of days with the cleanest air (PM DAQI of 1) occur with these air masses (Fig. 9). North westerly, northerly and westerly air flows dominate the cleanest air days (59% of days with PM DAQI of 1 occur with these flows) and there are no occurrences of the highest PM DAQI values (9 and 10) on days with north westerly, northerly or westerly air flows (Fig. 9). For PM DAQI values of 4 and above, at-risk individuals (e.g. those with asthma or heart conditions) are advised to reduce strenuous activity if they experience symptoms. These results suggest a strong dependence of periods of increased risk for such individuals on meteorological conditions.

4. Discussion and conclusions

This study investigated the role that synoptic weather plays in controlling variability of ambient $PM_{2.5}$ concentrations in the UK.

Observations of $PM_{2.5}$ concentrations under different LWTs indicate that both annually and seasonally, anticyclonic circulation and easterly, south-easterly and southerly flow increase the mean $PM_{2.5}$ concentrations observed. Results from the EMEP4UK model suggest transboundary transport is likely responsible for the increases in $PM_{2.5}$ observed under the wind types (easterly, south-easterly and southerly flow) and the build-up of local emissions under stagnant air masses under anticyclonic and unclassified types. Results also indicate that although $PM_{2.5}$ concentrations are higher under easterly, south-easterly and southerly flow than under anticyclonic conditions, anticyclonic conditions occur on a much larger fraction of days and so have a larger

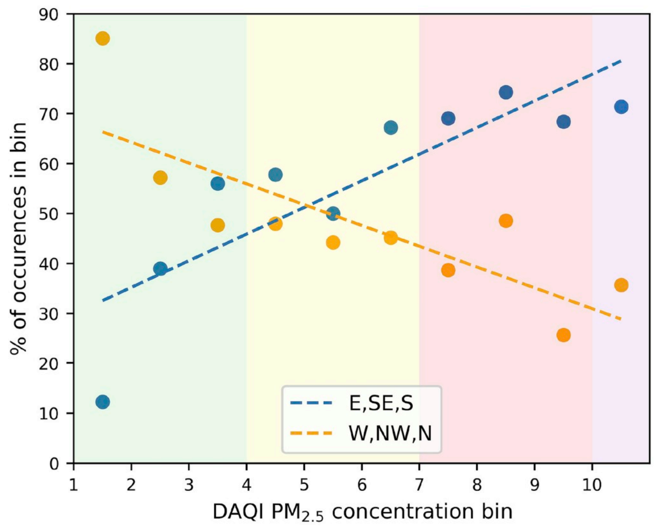

Fig. 9. Percentage occurrence (defined as the percentage of occurrences of easterly, south-easterly and southerly (and westerly north-westerly and northerly) LWTs in each bin) of easterly, south-easterly and southerly weather types and westerly, north-westerly and northerly in each DAQI $PM_{2.5}$ concentration bin. Bins 1–10 indicate $PM_{2.5}$ concentrations of 0–11, 12–23, 24–35, 36–41,42-47,48–53,54-58,59–64,65-70, >71 (all in μg m^{-3}).

impact on the annual $PM_{2.5}$ concentration and the population's exposure to increased $PM_{2.5}$ concentrations. These findings are in agreement with previous work which has examined different species and short-lived pollution episodes as case studies (Pope et al., 2014; 2016; Vieno et al., 2010, 2014, 2016). Pope et al., 2014 also found that under anti-cyclonic conditions NO_2 concentrations were significantly increased through pollutant accumulation and that south-easterly flow enhanced NO_2 concentrations. They attributed this to long-range transport from continental Europe. A similar relationship was found for ozone in summer months, with enhanced concentrations under north-easterly and south-easterly flow and anticyclonic conditions leading to increased ozone concentrations due to large scale subsidence and little vertical mixing (Pope et al., 2016). Demuzere et al. (2009) found PM_{10} concentrations were highest in the Netherlands under easterly, south-easterly and southerly wind directions, attributing the increase in PM_{10} concentrations to air masses passing over large source regions.

The results of the back-trajectory analysis indicate that the transport of pollutants from large source regions outside of the UK is an important contributor to the total accumulated emission under easterly, south-easterly and southerly flow. Since secondary $PM_{2.5}$ typically represents 3–8 μg m^{-3} (20–50%) of the total $PM_{2.5}$ concentration at background sites in Europe (Querol et al., 2004) a method accounting for secondary $PM_{2.5}$ (nitrate, sulphate and ammonium) would need to be applied to quantify the contribution of non-UK emissions to total $PM_{2.5}$ in the UK. Nevertheless, our results suggest a substantial non-UK burden on UK pollution under continental air masses.

This study further reinforces that synoptic weather in the UK plays an important role in controlling $PM_{2.5}$. It is important, therefore, that air quality models are able to accurately simulate synoptic meteorology in order to reliably forecast $PM_{2.5}$ concentrations in forecasts. Given the large impact on health that short-term exposure to $PM_{2.5}$ has been shown to have in previous studies (e.g. Macintyre et al., 2016), the ability of air quality forecast models to accurately predict $PM_{2.5}$ concentrations is key in preparing for and mitigating the associated health impacts of exposure.

The results of the back-trajectory analysis indicate that quantifying the contribution of UK and non-UK pollution sources is extremely important in evaluating the impact of local emission controls on UK pollutant concentrations. This is particularly relevant given that we have shown variations in background $PM_{2.5}$ concentrations are highly variable under different weather patterns.

Author contributions section

Ailish Graham – conceptualization, methodology, software, formal analysis, data curation and writing (original and review), visualisaton and investigation, with help and guidance from Stephen Arnold, Richard Pope, Kirsty Pringle, James McQuaid.

Stephen Arnold, Richard Pope, Kirsty Pringle, James McQuaid – Supervision.

James McQuaid – funding acquisition.

Ellen Stirling, Richard Pope and Ailish Graham– Development of back-trajectory analaysis after Ailish Graham ran the back trajectories.

Massimo Vieno - data curation (EMEP4UK data).

Ed Butt and Luke Conibear – advice on method.

Ellen Stirling, Richard Pope and Ailish Graham– Development of back-trajectory analysis and visualisation after Ailish Graham ran the back trajectories.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.aeaoa.2019.100061.

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Further reading

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