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RESEARCH ARTICLE

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Quantifying the transboundary contribution of nitrogen oxides to UK air quality

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Abstract

Nitrogen dioxide (NO_2) pollution is an important contributor to poor air quality (AQ) and a significant cause of premature deaths in the UK. Although transboundary (i.e., international) transport of pollution to the UK is believed to have an impact on UK pollutant concentrations, large uncertainties remain in these estimates. Therefore, the extent to which emission reductions in neighbouring countries would benefit UK AQ relative to local emission reductions also remains unknown. We have used a back-trajectory model in conjunction with synoptic scale classifications of UK circulation patterns (Lamb Weather Types [LWT]), to quantify the accumulation of nitrogen oxide ($NO_x = NO_2 + NO$) emissions in air masses en-route to the UK. This novel method presents a computationally inexpensive and useful method of quantifying the accumulation of pollutants under different circulation patterns. We find the highest accumulated NO_x totals occur under south-easterly and southerly flows (>15 $\mu g \cdot m^{-2}$), with a substantial contribution from outwith the UK (>25%). In contrast, the total accumulated NO_x under northerly and westerly flows is lower (~10 $\mu g \cdot m^{-2}$), and dominated by UK emissions (>95%). This indicates that European emissions can contribute substantially to UK local-scale pollution in urban areas under south-easterly and southerly flows. The sensitivity of integrated NO_x emission totals under different air masses is investigated by modelling future European emission contributions based on emission reduction targets. Under targets set by the European Union, there would be a decrease in accumulated NO_x emissions in London under most wind directions except for north-westerly, westerly and northerly flow. The largest benefits to UK AQ from transboundary contributions occur with emission reductions in the Benelux region, due to its close proximity and high NO_x emission rates, emphasising the importance of international cooperation in improving local AQ.

KEYWORDS

back trajectories, lamb weather type, NO_x, transboundary pollution

1 | INTRODUCTION

Poor air quality (AQ) has a significant impact on human health, inducing heath ailments such as asthma, cancer, diabetes and heart disease (Royal College of Physicians, 2016). Nitrogen dioxide (NO_2) is an air pollutant emitted through high temperature combustion in motor vehicles and power production (US EPA, 2017). It is estimated that NO_2 pollution was responsible for 9,600 premature deaths in the UK alone in 2015 (EEA, 2018).

Under EU directive 2008/50/EC Ambient AQ regulation, the UK must meet pollution concentration targets. However, many UK cities currently exceed the limit for NO_2 (DEFRA, 2018a). In 2017, the 200 $\mu g \cdot m^{-3}$ 1-hr limit for NO_2 was exceeded within the UK in two zones—The Greater London Urban Area and South Wales. A further 37 zones failed to meet the annual mean limit value of 40 $\mu g \cdot m^{-3}$ for NO_2 (DEFRA, 2018b).

Pollutant contributions from both local and transboundary emission sources can lead to poor air quality. Multiple studies (Tang *et al*., 2009; Pope *et al*., 2014; Grundstrom *et al*., 2015; Pope *et al*., 2016) have used classifications of atmospheric circulations (e.g., the Lamb Weather Type [LWT]) to analyse long-term pollutant variability and relationships with atmospheric transport. Grundstrom *et al*. (2015) and Tang *et al*. (2009) used LWT classifications to investigate responses to AQ in southern Sweden. Grundstrom *et al*. (2015) concluded during winter NO_2 regulation exceedances occurred most frequently under north and northwest flow regimes, while Tang *et al*. (2009) identified that summer southeast and southwest airflows yielded increased O_3 concentrations. Pope *et al*. (2014, 2016) found that UK NO_2 and summer O_3 concentrations were enhanced under anticyclonic conditions and south-easterly flow, attributing the latter to the transport of pollutants from continental Europe.

Few studies have quantified the contribution of transboundary pollution sources in enhancing local pollutant concentrations. Schaub *et al*. (2005) found that during an elevated pollution episode in February 2001, 50% of NO_2 pollution measured at a Swiss site originated from transboundary sources. Kindap (2008) found that under westerly airflow in Istanbul, pollutants transported from European cities substantially contributed to poor AQ events. Vieno *et al*. (2014) concluded that transboundary sources of $PM_{2.5}$ contributed 63% and 41% of South–West England and Central Scotland total concentrations, respectively. Using back-trajectories, Reddington et al. (2014) concluded that AQ in Singapore was significantly impacted by fire emissions in Kalimantan and Central and Southern Sumatra.

Quantifying the contribution of transboundary pollution using complex models is computationally expensive. Here, we present a computationally inexpensive method of quantifying the accumulation of pollutants along 4-day trajectories arriving at several UK cities between 2010 and 2013. In this case, we address nitrogen oxide ($NO_x = NO_2 + NO$) pollution under different atmospheric circulation patterns over the UK. Using a back-trajectory model, in conjunction with LWT classifications, we quantify the contribution of pollution emitted outside of the UK to the summed emission within back-trajectories. We also estimate the potential benefits to be gained from neighbouring countries achieving future emission targets.

2 | METHODS AND DATA

2.1 | Surface measurements

The Automated Urban and Rural Network (AURN) is maintained and funded by the Department for Environment, Food and Rural Affairs (DEFRA). It has been routinely monitoring AQ across the UK since 1973 at over 100 sites (DEFRA, 2018c). Daily average surface NO_2 observations between 2010 and 2017 were obtained from both Urban Background and Urban Traffic sites in Leeds (Leeds Centre Background and Headingley Kerbside) and London (North Kensington Background and Marylebone Kerbside). These two cities were selected as example urban regions that were in exceedance of the EU annual limit for NO_2 (40 $\mu g \cdot m^{-3}$) in 2017. Urban background sites are more representative of a surrounding urban area, while Urban Traffic sites are subject to larger concentrations and diurnal variations from traffic activity (DEFRA, 2018d).

2.2 | Lamb weather types

The LWTs were originally presented by Lamb (1972) to classify daily circulation patterns across Europe in accordance with wind direction and circulation type. Jones *et al*. (2013) used the automated scheme created by Jenkinson and Collison (1977) with the National Centers for Environment Prediction (NCEP) reanalysis data (Kalnay *et al*., 1996) to generate an objective LWT time-series based on grid-point mean sea level pressure at midday (12:00 UTC). LWTs are calculated using daily mean flow strength, flow direction and circulation strength, with conditions falling into 28 possible categories (Table 1). For this study, we combine all circulation types under each wind direction to give flow classification by only wind direction—N, NE, E, SE, S, SW, W and NW. For example, Anticyclonic North Easterly (1 ANE),

TABLE 1 The 28 LWT classifications with their number coding, grouped into circulation type and wind direction

	Anticyclonic	Neutral vorticity	Cyclonic
	0 A		20 C
North-easterly	1 ANE	11 NE	21 CNE
Easterly	2 AE	12 E	22 CE
South-easterly	3 ASE	13 SE	23 CSE
Southerly	4 AS	14 S	24 CS
South-westerly	5 ASW	15 SW	25 CSW
Westerly	6 AW	16 W	26 CW
North-westerly	7 ANW	17 NW	27 CNW
Northerly	8 AN	18 N	28 CN

Note: LWT also include −1 (unclassified) and − 9 (non-existent day) which were not used in this study. Adapted from Jones *et al.* (2013).

Neutral North Easterly (11 NE) and Cyclonic North Easterly (21 CNE) all fall under the NE classification. The use of LWT classification allows for long term patterns to be more robustly identified rather than just focusing on one pollution event.

2.3 | Emissions

We use NO_x emissions from the Emissions Database for Global Atmospheric Research (EDGAR), obtained from the European Commission's Science Hub (http://edgar.jrc.ec.europa.eu), and the National Atmospheric Emissions Inventory (NAEI) from http://naei.beis.gov.uk. EDGAR is a global inventory (resolution 0.1° × 0.1°), while the NAEI inventory covers the UK (0.025° × 0.025° used here, covering 8°W–2°E and 50°N–60°N). Our study focuses on the 2010–2013 period, but EDGAR represents emissions for 2010–2012, while the NAEI data is for 2015 (previous years were not publicly available). Here, the EDGAR emissions were mapped onto the higher spatial resolution and the NAEI emissions were nested within the domain, replacing the EDGAR equivalent (i.e., EDGAR-NAEI emissions). As the NAEI emissions were for 2015, they were scaled for years 2010–2013 based on the respective annual UK total NO_x emissions reported by the NAEI. The 2012 EDGAR emissions were repeated for 2013. Figure S1 gives an example of the merged emissions for 2010. Both EDGAR and the NAEI express NO_x emissions (i.e., emissions of both NO and NO_2) as NO_2.

2.4 | Back-trajectory model

We use the Reading Offline Trajectory Model (ROTRAJ), a Lagrangian atmospheric transport model, which uses analysed meteorology from the ERA-Interim product of the European Centre for Medium-Range Weather Forecasts (ECMWF), to generate air mass back-trajectories (Methven *et al.*, 2003). The velocity fields at the Lagrangian particle positions are obtained from the reanalysis data and determined analyses (1.0125° horizontal resolution) and determined by cubic Lagrange interpolation in the vertical followed by bilinear interpolation in the horizontal and linear interpolation in time. These were used in conjunction with the EDGAR-NAEI emissions datasets, to determine the quantity of NO_x accumulated by air masses reaching the UK and the fraction of which originated from non-UK sources. The back trajectories were binned by the wind flow classifications on the day they were released in order to identify which wind directions are associated with the highest integrated emission totals.

Kinematic back-trajectories (4 days with 6-hr output) were calculated, initialised daily at 12 UTC (to match the timing of the LWT classifications) from four background AURN sites over the period 2010–2013—Leeds Centre, Edinburgh St. Leonards, London Bexley (South-East London) and London North Kensington (North-West London) (see Figure 1). These trajectories account for large-scale advection by the resolved model winds, and neglect convective and turbulent transport.

Each trajectory air mass path was linearly interpolated in 15-min intervals, with NO_x emissions at each 15-min location accumulating over time. The boundary layer height was assumed to be 850 hPa and any trajectory points below this pressure were removed from the analysis (i.e., not exposed to emissions). Given the relatively short lifetime of NO_x (Nunnermacker *et al.*, 2000; Alvarado *et al.*, 2010; Romer *et al.*, 2016; Schaub *et al.*, 2007), we imposed representative e-folding lifetimes of 3, 6, 9 and 12 hr to account for loss processes within the air mass and to test the sensitivity of the final integrated emissions along the trajectory to the assumed timescale for loss processes. Here, an e-folding lifetime is defined as the time required for a quantity to reduce by a factor of *1/e*. Following a similar methodology using the ROTRAJ model in Arnold *et al.* (2010), we calculate integrated NO_x emission totals according to:

$$E_i = [E_{i-1} + \phi_i.\Delta t.\alpha_i]e^{-\Delta t/\tau},\ i = 1, N \text{ and } E_0 = 0 \quad (1)$$

where E_N is total accumulated NO_x mass (kg), N is the number of time steps within the trajectory (384), E_i is accumulated NO_x (kg) at any given point i along the trajectory, ϕ_i is the emissions flux of NO_x ($kg{\cdot}m^{-2}{\cdot}s^{-1}$) at point i, Δt is the 15-min time step, α_i is the surface area

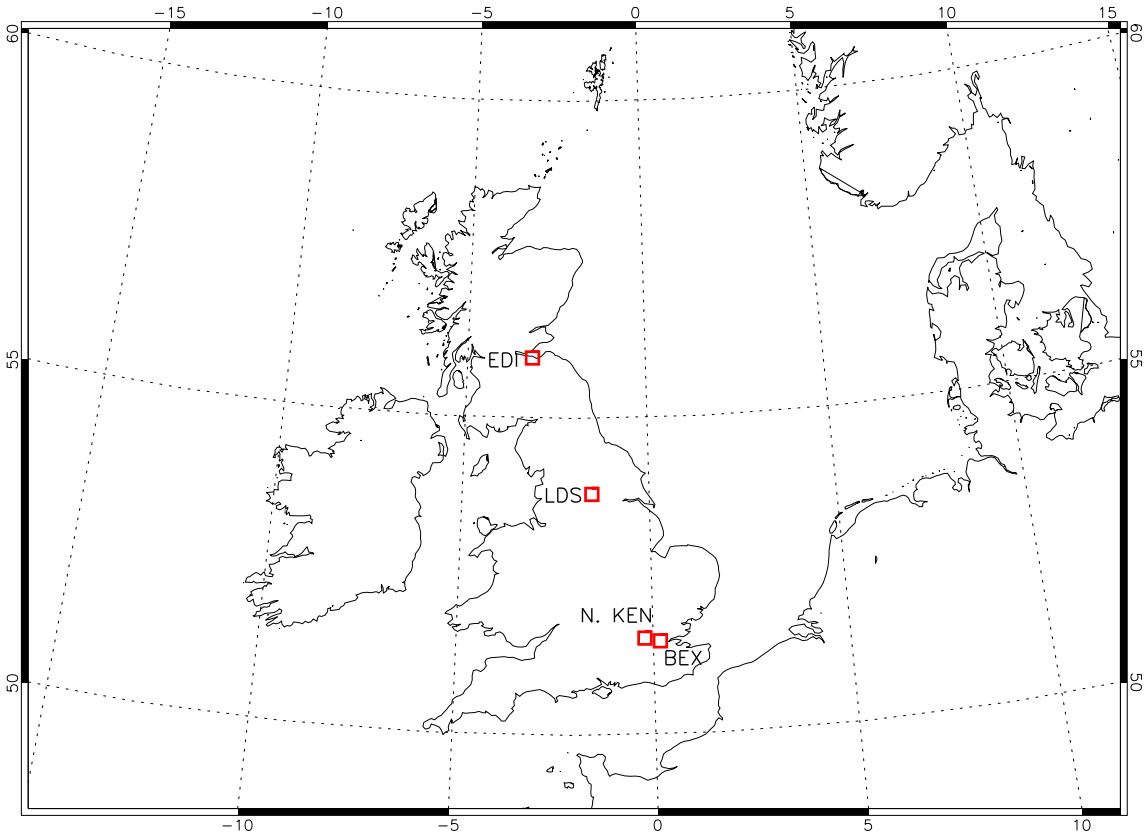

FIGURE 1 Location of background AURN sites used for the back-trajectory analysis. BEX, Bexley; EDI, Edinburgh; LDS, Leeds; N. KEN, North Kensington

of the grid box (m^2) at point i and τ is the assumed e-folding NO_x lifetime.

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

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

To obtain the UK fractional contribution of E, the same approach was applied only when trajectories entered the UK domain (8°W–2°E and 50°–60°N) to obtain E_{UK} (kg). E_{UK} was also divided by S to get units of μm^{-2}. E_{UK}/E, then represents the fractional contribution of UK emissions to the integrated NO_x emissions total.

Figure 2 demonstrates this methodology where integrated NO_x totals (using Equations 1 and 2) over the 4-year period (2010–2013) for London Bexley (background AURN site), but with no e-folding lifetime (i.e., no decay). Trajectories coming from the north and west originate in the Atlantic, where there are fewer and more dispersed (i.e., shipping) pollution sources, whereas trajectories from the south and east will originate from mainland Europe. These trajectories are likely to have passed over a number of NO_x sources in neighbouring countries including France, Germany and the Benelux region before reaching the UK, accumulating NO_x en route.

To evaluate this methodological approach, we have correlated the trajectory-integrated NO_x emission totals with the surface AURN NO_2 concentrations, sampled at 12:00 UTC to match the LWT classification and de-seasonalised as the emissions used represent annual rates. Comparisons show robust positive correlation at the 99% confidence level between the two quantities, despite their representation of two different measures of pollutant burden. Correlations of 0.45, 0.42, 0.45 and 0.25 are obtained at North Kensington, Bexley, Leeds and Edinburgh, respectively. This indicates likely non-local transport-driven variability in measured NO_2 at the London sites and Leeds providing some confidence in this approach outlined in this study, while the AURN site in Edinburgh appears to be less affected by non-local sources, which is to be expected given its geographical

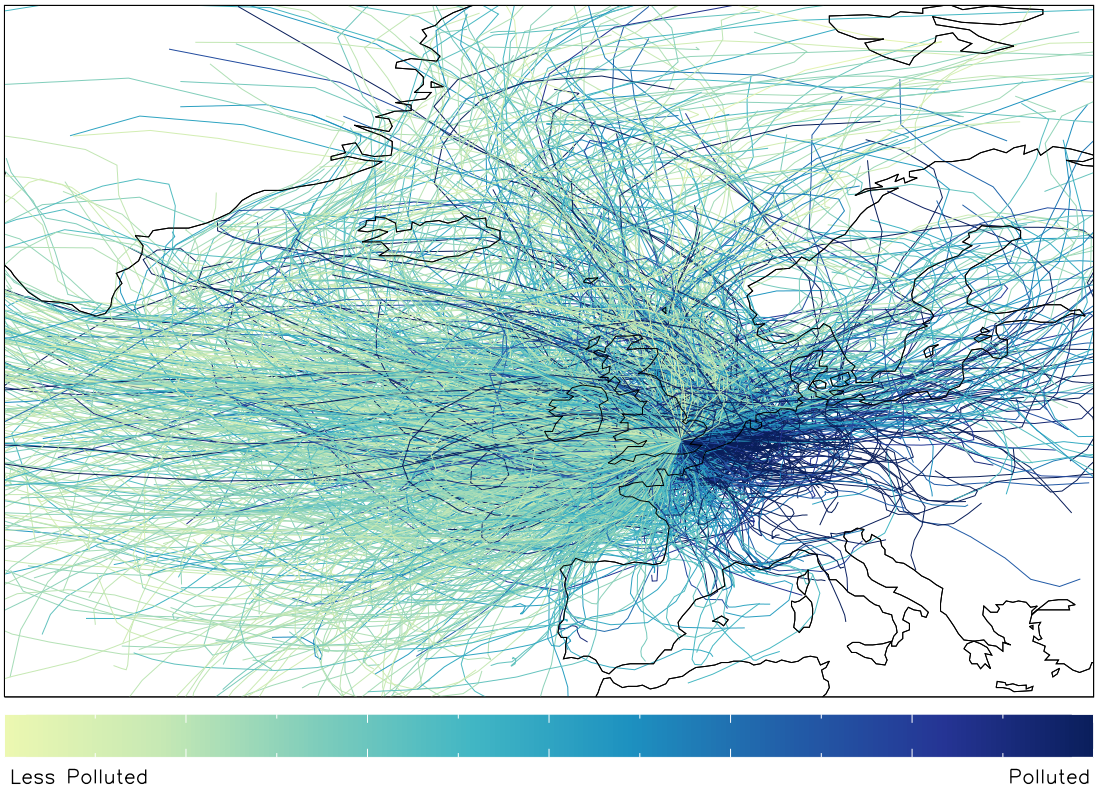

FIGURE 2 Four-day back trajectories with 6-hr time steps arriving at the Bexley AURN background site between 2010 and 2013. Trajectories accumulated NO_x over time, with darker trajectories indicating higher levels of accumulated NO_x with no e-folding lifetime (i.e., no decay)

position which means reduced susceptibility to emissions from Southern England and continental Europe.

3 | RESULTS

3.1 | Surface AURN concentrations

Between 2010 and 2017, annual mean NO_2 concentrations at Marylebone, London (kerbside site) (Figure 3c) remained above the EU annual mean target of 40 $\mu g \cdot m^{-3}$, with no significant change over time (concentrations remained between 85–100 $\mu g \cdot m^{-3}$). At North Kensington, London (background site) annual mean concentrations remained below 40 $\mu g \cdot m^{-3}$ over the study period (Figure 3a). However, the upper range (*SD*) indicates NO_2 concentrations at this site frequently exceeded this limit.

When NO_2 concentrations are sub-sampled under the flow classifications, a clear relationship between NO_2 and wind direction exists. At the North Kensington site, concentrations exceed 40 $\mu g \cdot m^{-3}$ under E and SE flows. At Marylebone, concentrations are largest under S, SW and W flows and in excess of 100 $\mu g \cdot m^{-3}$. The lowest mean concentration of 65 $\mu g \cdot m^{-3}$ is observed under NE flow, which is 25 $\mu g \cdot m^{-3}$ above the 40 $\mu g \cdot m^{-3}$ target. These results potentially highlight the contribution of continental European sources to local concentrations from long-range transport. A similar pattern exists at background and kerbside sites in Leeds (see Figure S2), which is likely typical of many other UK cities.

3.2 | Integrated NO_x emission totals

Here, we present our results primarily based on an e-folding NO_2 lifetime of 6 hr. Multiple studies have shown that European NO_2 lifetimes range from a few hours in summer to over a day in winter (Nunnermacker *et al.*, 2000; Ryerson *et al.*, 2003; Alvarado *et al.*, 2010; Romer *et al.*, 2016; Schaub *et al.*, 2007). Therefore, as we use annual emission inventories, we assume a median lifetime of 6 hr. However, we present the sensitivity of our results to different lifetimes (3, 9 and 12 hr) in the SM (Figures S3-5) to provide context to our results. Without the application of an e-folding lifetime to the London Bexley back trajectories, there is a clear pattern of elevated integrated NO_x emission totals in trajectories originating from continental Europe, while lower totals originate from the North Atlantic.

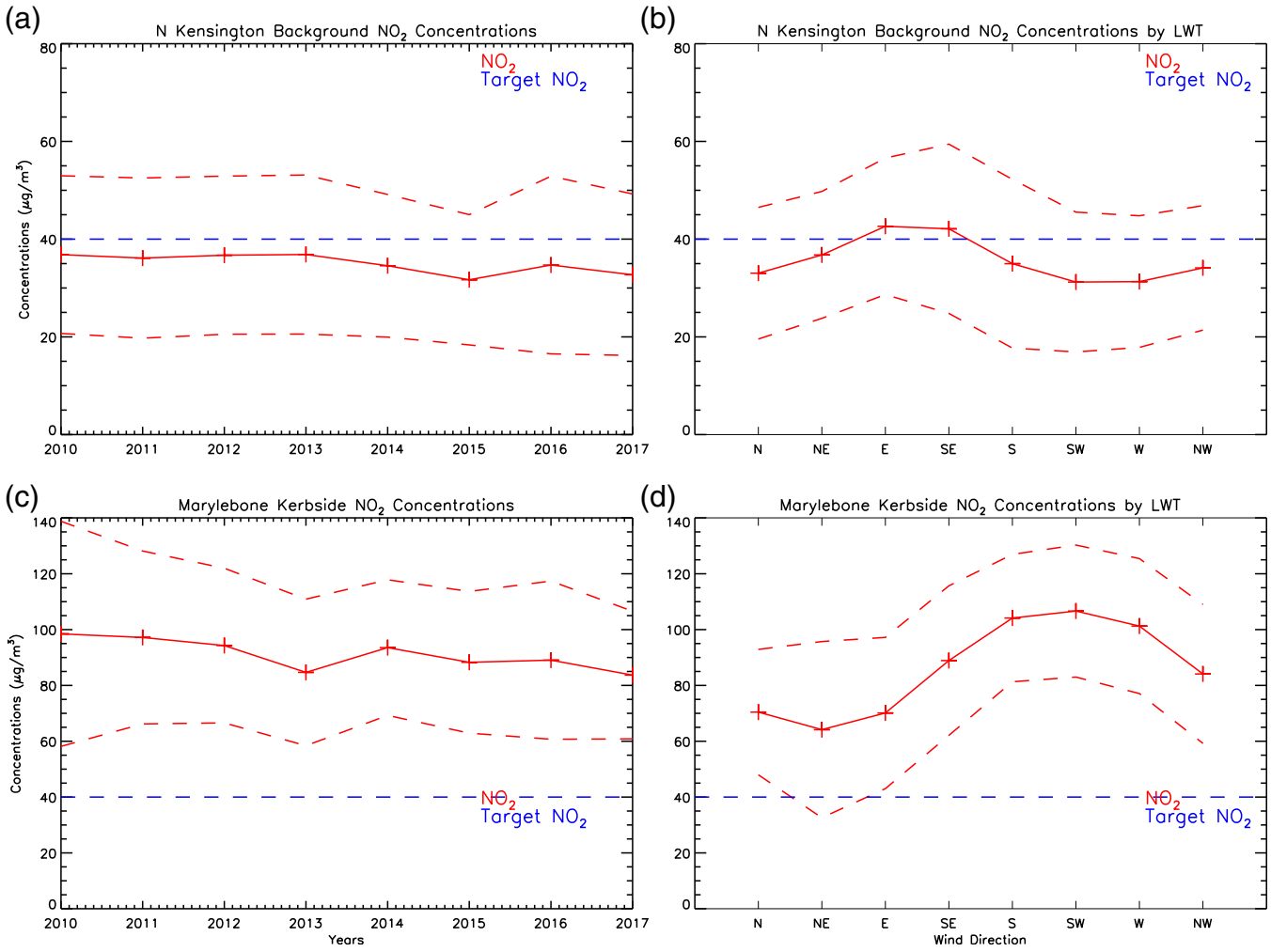

FIGURE 3 Annual NO_2 concentrations ($\mu g \cdot m^{-3}$), between 2010 and 2017, for London AURN sites. (a) North Kensington background and (c) Marylebone Kerbside. The NO_2 concentrations sub-sampled under the flow regimes are shown for (b) North Kensington background and (d) Marylebone Kerbside. Solid red lines show the annual mean concentration and dashed red lines are ±1 *SD* sampled from daily data each year. The blue dashed lines represent the EU annual threshold limit for NO_2 (40 $\mu g \cdot m^3$)

The largest integrated NO_x emissions totals, assuming a 6-hr-folding lifetime, are found at the two London sites (North Kensington and Bexley), exceeding 16 $\mu g \cdot m^{-2}$ under SE flow and 13 $\mu g \cdot m^{-2}$ under S and SW flows (Figure 4). The cleanest flows are N, NE, E and NW where integrated emission totals range between 9 and 13 $\mu g \cdot m^{-2}$. Under SE flow, the UK contribution to London integrated emissions totals is approximately 75%, suggesting a substantial contribution (~25%) from transboundary sources (e.g., continental Europe and shipping emissions in the English Channel). The largest UK contributions (95–100%) occur under W, NW and N flows, as most UK sources are situated to the west and north of London. For longer e-folding lifetimes, the integrated NO_x emissions totals increase to over 25 $\mu g \cdot m^{-2}$ and the UK contribution decreases to 50–60% under SE flow (Figure S4 and S5). Air masses originating from the N, NW and W show little change as the trajectories primarily originate in the North Atlantic, with few NO_x sources. With a 3-hr e-folding lifetime, the integrated NO_x emission totals peak at approximately 12 $\mu g \cdot m^{-2}$ under S, SW and SE flow. The totals are lower and the UK contribution is higher (>90%), due to the shorter assumed lifetime limiting the contribution of European emissions. The N, NW and W integrated NO_x emission totals decrease to 6–10 $\mu g \cdot m^{-2}$ and represent nearly 100% UK sources.

At the Leeds site, peak integrated NO_x emission totals range from 8–12 $\mu g \cdot m^{-2}$ for the SE, S, SW and W flows using a 6-hr e-folding lifetime (Figure 4). The NW, N, NE and E flows are ~50% lower (3–5 $\mu g \cdot m^{-2}$), typically representing cleaner air masses. North of Leeds, there are fewer NO_x sources (i.e., predominantly national parks), while south of Leeds there are a number of urban regions including Manchester, Sheffield and Wakefield. The E and SE flows, while not the most polluted air masses,

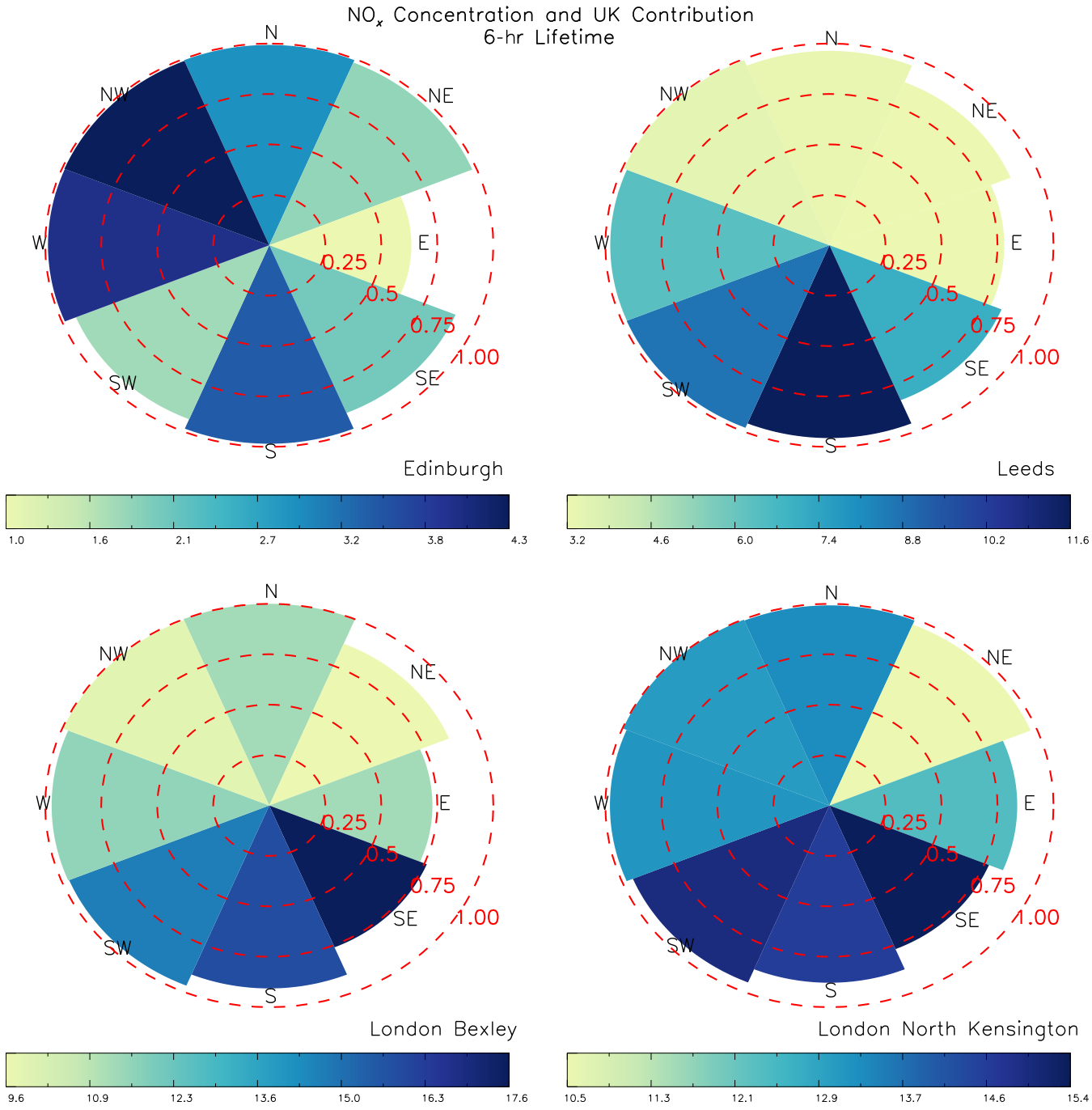

FIGURE 4 Integrated NO_x emission totals ($\mu g \cdot m^{-2}$) for multiple UK sites (Edinburgh, Leeds, London Bexley and London North Kensington) displayed by wind direction (as determined by the LWTs) using ROTRAJ back trajectories between 2010 and 2013. The integrated emission totals assume an e-folding lifetime of 6 hr. Red dashed circles mark the UK fractional contribution to the emissions total defining UK sources within the box of 8°W–2°E, 50°–60°N. Outside of this region, emission sources are defined as transboundary (e.g., continental European emissions)

have the lowest UK contribution (~75–80%). This suggests that Leeds experiences relatively moderate pollution contributions from European, shipping and off-shore sources. At longer e-folding lifetimes, the integrated NO_x emission totals increase and the UK contribution decreases to 50–60% whereas at a 3-hr lifetime, UK contributions increase to nearly 100% (Figures S3–S5). However, in all cases, the UK contributions are smallest under the E and SE flows, highlighting the potential impact of non-UK sources on Leeds pollution levels.

The lowest integrated NO_x emission totals of all locations are at the Edinburgh site (Figure 4), peaking

at ~4–5 μg·m^{-2} under NW and W flows (nearly 100% UK contribution) using a 6-hr e-folding lifetime. There are several regional pollution sources which can explain this, including the M9 and Longannet Power Station to the northwest and the M80 and Glasgow to the west. The smallest total and UK contribution is under E flow (1 μg·m^{-2}; ~60–65%), likely due to the location of the city on the east coast. As the e-folding

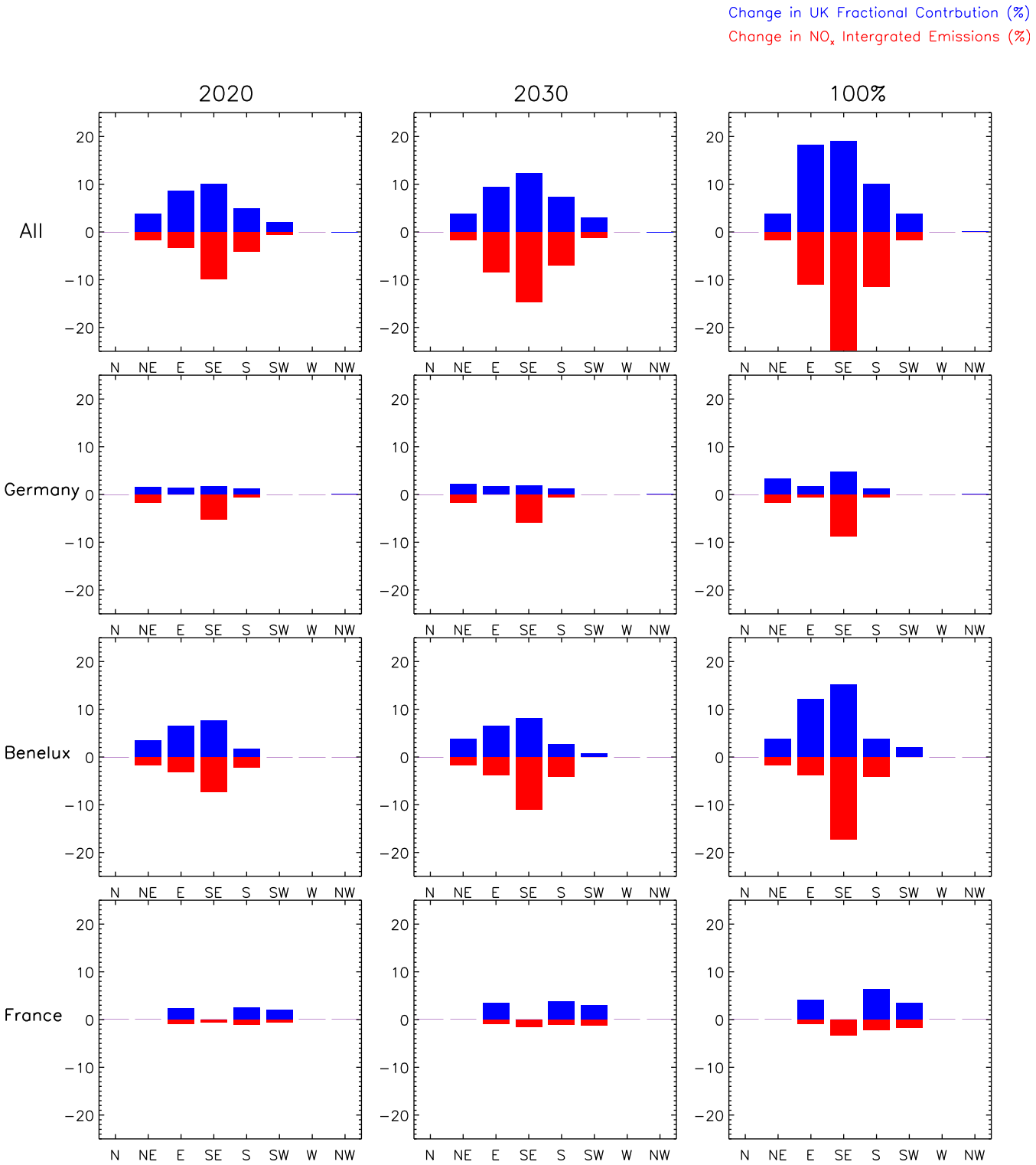

FIGURE 5 Change to integrated NO_x emissions totals (%) and UK contribution (%), using 6 hr e-folding lifetime, due to reduction and removal of country emissions, binned by flow regime. Positive values (blue) show the increase in UK contribution, negative values (red) show the decrease in integrated NO_x emission totals

lifetime increases from 3- to 12-hr, the dominant source changes from westerly to southerly direction (see Figures S3–S5) and the UK contribution from the S flow decreases (∼100% to 85%).

The largest integrated NO_x emissions totals are derived at the London sites, with larger absolute contributions from transboundary sources. Typically, southerly (S and SE) flows are the most polluted, as substantial quantities of emitted NO_x are transported into the UK from continental Europe. Here, non-UK sources from transboundary pollution (continental Europe) contribute up to approximately 25% of the total contribution. This relationship intensifies (peaking at ∼50% from transboundary sources) at longer e-folding lifetimes (9 and 12 hr). However, at a 3-hr e-folding lifetime, transboundary pollutants are less important yielding long-range contributions of only 10–15% under SE flow. At Leeds and Edinburgh, the integrated NO_x emission totals are reduced with a lower non-UK fractional contribution than in London, as the chemical lifetime of NO_x limits the distance over which it can be transported. The main exception being E flow, with substantial (30–40%) contribution from non-UK sources, however with much lower emissions totals. Here, pollution is transported from Scandinavia, off-shore and shipping sources, but with the majority lost en route over the North Sea. As there are fewer sources within the UK to the east of Leeds and Edinburgh, the transboundary contribution is larger than seen in other flow directions.

3.3 | Emission control

Finally, we analyse the potential benefits to UK AQ which may be gained from other European countries meeting their 2020 and 2030 National Emission Ceiling Directive (NECD) targets (see Table S1). Although these targets are based on 2005 emission levels, we apply them to the 2010–2013 emissions used in this study. Therefore, our results are potentially a conservative estimate given the decrease in UK NO_x emissions already achieved between 2005 and 2010. The potential benefits gained from the transition to zero emissions in neighbouring countries are represented in the "100%" column in Figure 5. By running the ROTRAJ back trajectory model from London North Kensington with the reduction or removal of emissions from selected European countries, assuming a 6-hr e-folding lifetime, we have been able to estimate the benefits of individual countries' emission controls to UK NO_2 AQ. The grouping defined as "All" indicates combined emissions reductions occurring in all three regions of interest: France, Germany and the Benelux region (Belgium, The Netherlands and Luxembourg).

Generally, a decrease in emissions in the Benelux region leads to the largest decrease in accumulated NO_x and an increase in UK contribution. As it is the closest of the three regions to the UK, NO_x is less likely to be lost before reaching London. The largest decrease in NO_x is found under SE flow (∼16%), with a corresponding increase in UK contribution of ∼15% under the same directions. There is a negligible change in both accumulated NO_x and UK contribution under N, W or NW flow. In contrast, the smallest change to NO_x totals and UK contribution occurs with reductions in French emissions, where the largest reduction in NO_x occurring under SE, S and SW flow (2–3%). Similar to Benelux, emission reductions in Germany lead to the largest decrease in NO_x totals under SE flow of ∼8% under 100% removal.

If all regions of interest were to achieve their 2020 emissions targets, there would be reductions in accumulated NO_x totals reaching North Kensington under the majority of wind directions apart from NW, N and W. The greatest decrease occurs under SE flow (∼10%). Trajectories from the north and west are unlikely to pass over these countries before reaching the UK, therefore their NO_x levels remain unaffected by changes in emissions. The decrease in NO_x totals and increase in UK contribution only grows larger as emissions in "All" are cut to 2030 NECD targets and 100% emissions removal. The largest change in trajectory totals continues to be under SE flow, with a 25% decrease under the zero-emission scenario, followed by E and S flow where NO_x totals would decrease ∼10–12%.

4 | DISCUSSION AND CONCLUSIONS

We have shown that transboundary pollution can be an important contributor to NO_x accumulated by air masses arriving at UK urban locations. Airflow from the south and east leads to the highest accumulated NO_x. When comparing UK cities, AQ in London is more strongly influenced by transboundary pollution than cities further north, with transboundary pollution contributing to up to 25% of accumulated NO_x under SE flow. Trajectories under E, SE and S flows are likely to have passed over emissions sources in continental Europe, accumulating emissions before entering the UK domain. In contrast, trajectories under W, NW and N flows originate over the clean maritime environment of the Atlantic and are therefore less polluted with a higher UK contribution (∼99–100%).

For the first time, this study has estimated the contribution of individual European regions to accumulated NO_x in the UK. If Germany, France and the Benelux region were to achieve their 2030 NECD targets (applied

to 2010–2013 emissions used here and not 2005 baseline), the largest decrease in London's accumulated NO_x would occur under SE flow (>20%) and E and S flows (>10%). Due to the small continental European contribution to trajectories from the north and the west, there is a negligible change in NO_x totals under N, NW and W flows.

Overall, the LWT classifications and back trajectories, in conjunctions with emissions inventories, are useful tools in quantifying the contribution of European emissions to UK AQ. The conclusions of this research are applicable to the creation of future AQ policy and reinforce the need for international cooperation to improve regional AQ. This research has also allowed for a greater understanding of the role of local and long-range transport of emissions to the UK, without the need for a complex AQ model.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of this article.

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