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Detecting and quantifying the contribution made by aircraft emissions to ambient concentrations of nitrogen oxides in the vicinity of a large international airport

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Abstract

Plans to build a third runway at London Heathrow Airport (LHR) have been held back because of concerns that the development would lead to annual mean concentrations of nitrogen dioxide (NO₂) in excess of EU Directives, which must be met by 2010. The dominant effect of other sources of NOₓ close to the airport, primarily from road traffic, makes it difficult to detect and quantify the contribution made by the airport to local NOₓ and NO₂ concentrations. This work presents approaches that aim to detect and quantify the airport contribution to NOₓ at a network of seven measurement sites close to the airport. Two principal approaches are used. First, a graphical technique using bivariate polar plots that develops the idea of a pollution rose is used to help discriminate between different source types. The sampling uncertainties associated with the technique have been calculated through a randomised re-sampling approach. Second, the unique pattern of aircraft activity at LHR enables data filtering techniques to be used to statistically verify the presence of aircraft sources. It is shown that aircraft NOₓ sources can be detected at least 2.7 km from the airport, despite that the airport contribution is very small at that distance. Using these approaches, estimates have been made of the airport contribution to long-term mean concentrations of NOₓ and NO₂. At the airport boundary we estimate that approximately 28 % (34 μg m⁻³) of the annual mean NOₓ is due to airport operations. At background locations 2-3 km downwind of the airport we estimate that the upper limit of the airport contribution to be less than 15 % (< 10 μg m⁻³). This work also provides approaches that would help validate and refine dispersion modelling studies used for airport assessments.
Key words: London, urban air quality, Heathrow Airport, aircraft emissions, source apportionment.

1. Introduction

1.1 Background

The aviation sector in the UK has grown five-fold in the past 30 years and is expected to increase by another 2-3 times by 2030 compared with current day estimates (DfT, 2003). In December 2003, the UK Department for Transport (DfT) published the UK Government’s Aviation White Paper, setting out a strategic framework for the development of UK aviation (DfT, 2003). Currently, the future development of London Heathrow (LHR) is supported by the UK Government, including the building of a third runway, but only if it can be shown that the development does not exceed EU Limit Values for ambient air pollution. The White Paper also called for an urgent programme of work to tackle the air quality problems at Heathrow. The principal concern is whether the annual mean nitrogen dioxide (NO$_2$) limit of 40 µg m$^{-3}$, which must be met by 2010 as part of the EU Daughter Directive (1999/30/EC), can be achieved. A third runway is seen as essential for the economy also. The airport supports 100,000 jobs and a short third runway would yield net economic benefits of some £6 billion (DfT, 2003). In the context of air pollution, the contribution made by the airport and its operation to concentrations of NO$_2$ and NO$_x$ is therefore of key importance. However, to determine this impact, a detailed knowledge of the sources of NO$_x$ close to LHR is essential, together with their contribution to measured concentrations, if current and future assessments of annual mean NO$_2$ concentrations are to be reliable.

Comparatively little source apportionment work has been undertaken in the vicinity of airports to determine the extent to which aircraft emissions affect local air quality. Yu et al. (2004) used a nonparametric approach based on kernel smoothing to identify aircraft sources at Los Angeles International Airport (LAX) and Hong Kong International Airport. Sulphur dioxide (SO$_2$) was found to be a useful tracer of aircraft emissions at both airports. This work usefully extended that of Henry et al. (2002), which only considered concentrations by wind direction. The additional insight provided by incorporating wind speed into the nonparametric approach greatly enhanced the method by providing some discrimination between ground-
level and elevated plumes. The analysis by Yu et al. (2004) additionally showed that CO and NO\(_X\) concentrations at LAX were dominated by road traffic sources. At Heathrow, the primary interest is NO\(_X\) and NO\(_2\), which presents a difficulty in quantifying due to the overwhelming influence of ground-level traffic sources of NO\(_X\). Furthermore, no measurements of SO\(_2\) were available at the monitoring sites around Heathrow.

This paper aims to develop methods to discriminate between road traffic and aircraft sources of NO\(_X\) in air pollution data sets. Of particular interest is whether aircraft emissions can be detected and the contribution quantified in hourly data sets of NO\(_X\) from routine monitoring sites in the vicinity of the airport. Because the airport is situated within Greater London, it is embedded in a region of high emissions of NO\(_X\), which makes the analysis of its impact on local NO\(_X\) and NO\(_2\) concentrations problematic because of the confounding influence of other sources of NO\(_X\).

2. Method

2.1 Description of site and data used

Heathrow Airport is situated within the Greater London Authority boundary in west London approximately 25 km from central London. In 2002 the airport was responsible for an estimated 4222 t yr\(^{-1}\) NO\(_X\), 4.9 % of total NO\(_X\) emissions in London (accounting for aircraft emissions up to 1 km). Heathrow Airport has two runways: a northern and a southern runway separated by approximately 1.4 km. Heathrow operates a ‘runway alternation’ system of operation for noise mitigation reasons. During daytime westerly operations (taking off and landing into the prevailing westerly wind), landing aircraft use one runway from 07:00 until 15:00 and switch to the parallel runway from 15:00 until 23:00. Runway operation also operates on weekly basis so that communities in west London situated under the final approach tracks may benefit from predictably quieter periods at certain times of the day. Heathrow also operates a 'westerly preference'. The preference provides for westerly operations to continue when there is a light easterly following wind up to 5-knots (2.5 m s\(^{-1}\)), if the runways are dry and any cross-wind does not exceed 12-knots. These features of runway use at LHR provide an important and unique activity profile, which is very different to other major NO\(_X\) sources such as road transport. Runway alternation characteristics are exploited in section 3.3 when
aerial contributions to measured NO\textsubscript{X} concentrations are detected. Data from National Air Traffic Services (NATS), made available as part of the project, were used to provide information on runway alternation. These data provided hourly information on the number of aircraft movements during each hour including: runway used, whether the aircraft was arriving or departing and the direction of take off or landing. Later in section 3.3, aircraft are classified as departing or arriving on the northern or southern runways for westerly operation. For the vast majority of hours this classification is straightforward. For some hours (notably around 3 pm) when runway use switches it is less straightforward. For these hours, a runway is assumed to be used for taking off if there are more departures on it compared with the other runway.

There are several routine monitoring sites close to LHR that belong either to national, London or British Airways Authority networks. More information and most data for these sites can be obtained from www.heathrowairwatch.org.uk. A range of pollutants are measured at these sites, but the focus here is on measurements of NO\textsubscript{X} and NO\textsubscript{2}, which are measured using the chemiluminescent technique. Data from these sites undergoes quality assurance and control procedures consistent with that of the national network in the UK (AQEG, 2004). In total there are eight monitoring sites within 2 km of the airport boundary. Table 1 summarises the data available from these sites together with their distances from the northern or southern runway. Of principal importance is the LHR2 site situated 180 m north of the northern runway. This site is a few metres within the airport boundary, close to the eastern end of the northern runway. The LHR2 site is therefore ideally placed for considering airport sources when the wind is from a southerly or south-westerly direction i.e. the prevailing wind direction. The airport boundary road is approximately 15-20 m north of LHR2. With the exception of the Hillingdon site, most of the other sites are located either close to minor roads or in background locations, as borne out by the measured NO\textsubscript{X} and NO\textsubscript{2} concentrations, which are typical of background concentration in London (Fuller, 2005). Note, that only one year of Harlington data were available. Hourly meteorological data were obtained from the Met Office Heathrow site. These data underwent several adjustments to account for equipment changes at the site (see ref for more details).
Table 1. NO\textsubscript{X} and NO\textsubscript{2} monitoring sites used in the analysis.

<table>
<thead>
<tr>
<th>Site</th>
<th>Data period</th>
<th>Distance to northern runway (m)</th>
<th>Distance to southern runway (m)</th>
<th>Mean NO\textsubscript{X} (μg m\textsuperscript{-3})</th>
<th>Mean NO\textsubscript{2} (μg m\textsuperscript{-3})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Main Road</td>
<td>Jul. 2001-Dec. 2004</td>
<td>1060</td>
<td>550</td>
<td>81</td>
<td>39</td>
</tr>
<tr>
<td>Hillingdon</td>
<td>Jul. 2001-Dec. 2004</td>
<td>2060</td>
<td>3460</td>
<td>120</td>
<td>48</td>
</tr>
</tbody>
</table>

2.2 Graphical analysis for source apportionment

Yu et al. (2004) showed how accounting for wind speed in addition to wind direction yielded information on the types of source in the vicinity of a monitoring site. Here, a similar approach is used, with several modifications. First, data were averaged into different wind speed (0-1, 1-2 m s\textsuperscript{-1}...) and wind direction (0-10, 10-20°...) categories (cells) and the mean concentration of NO\textsubscript{X} calculated. The choice of the cell size affects the bivariate surface generated. An inappropriate cell size can lead to unnecessary imprecision: too small a cell causes the plot to become excessively noisy due to a small sample size, and too large a cell leads to an excessively coarse partition and a loss of information. Henry et al. (2002) and Yu et al. (2004) used a nonparametric kernel smoothing technique to determine the optimum balance between a plot that is too noisy or too coarse. The choice used here was in part determined by the meteorological data, which were provided rounded to the nearest 10°. Section 2.3 considers the affect of cell sample size on the estimated error of the mean concentration in a cell, which provides additional information on the most appropriate choice of cell size. This process yielded a surface in Cartesian coordinates, which was then converted into polar coordinates. Henry et al. (2002) did not favour converting to polar coordinates because data are compressed close to the centre. However, it will be shown later that when data from several monitoring sites are available, the polar coordinate system is a very effective one. The data were then interpolated using a Kriging technique to produce a bivariate polar plot.

Fig. 1a shows an example of a NO\textsubscript{X} bivariate polar plot for a monitoring site located approximately 40 m north of a motorway. There are several points that should be noted. First, the highest concentrations are recorded when the wind blows from the south. This is entirely expected because of the dominant motorway
source to the south of the site. Second, as the wind speed increases from any direction, the concentration of 
NO\textsubscript{X} decreases. This pattern of decrease is what would be expected from a ground level source where the 
concentration takes the form of a function that is inversely proportional to the wind speed. Fig. 1b shows 
the bivariate polar plot for SO\textsubscript{2} at a monitoring site in east London, which is affected by industrial sources. 
For SO\textsubscript{2} there are three clear regions where a source has an influence (approximately 60, 120 and 160\textdegree). 
Unlike Fig. 1a, the concentration of SO\textsubscript{2} increases with increasing wind speed. In fact, a consideration of 
potential sources highlights an oil refinery source at 12 km, a power station source at 6 km and other 
industrial sources at 4 km.

Increases in concentration with wind speed are indicative of a buoyant plume from a source such as a 
chimney stack, where the plume is brought down to ground-level when the wind speed increases. Note that 
it is not the case that a high-level emission from a stack gives rise to the pattern shown in Fig. 1b, but the 
presence of plume buoyancy. These features can be shown by considering the basic Gaussian plume 
dispersion equation (see Seinfeld and Pandis, 1998). In the absence of plume rise, the ground-level 
centreline concentration, \( c(x, 0, 0) \) is proportional to \( \bar{u}^{-1} \), where \( \bar{u} \) is the mean wind speed. In the presence 
of plume buoyancy, \( c(x, 0, 0) \) is a function of \( \bar{u}^a \), where \( a \) is a constant > 0, such that the lower the wind 
speed, the greater the plume rise. As the wind speed increases for a buoyant plume the ground-level 
centreline concentration increases to a maximum and then decreases. These plots potentially provide an effective 
graphical method of source discrimination between buoyant plumes such as those from power stations and 
plumes with little or no buoyancy such as road traffic sources.

The bivariate polar plot approach has been applied to the LHR2 site. The availability of monitoring sites 
around LHR (see Table 1 and Fig. 3) allows the subtraction of a background NO\textsubscript{X} concentration for certain 
wind sectors. Fig. 1c shows the effect of subtracting the Oaks Road NO\textsubscript{X} from LHR2 on an hourly basis 
with the purpose of highlighting the effect of airport sources of NO\textsubscript{X} from the south. The Figure shows the 
presence of a large source of NO\textsubscript{X} south, which does not decrease in concentration as the wind speed 
increases. Fig. 1c also shows that there is a sharp decrease in NO\textsubscript{X} concentration at about 150\textdegree. The angle 
between LHR2 and the end of the runway is approximately 110\textdegree, which suggests that aircraft plumes should
be detected from 110-150°. The reason for the sharp change at 150° is that aircraft take off in an easterly direction (on the southern runway) for easterly wind conditions. Fig. 1c on its own does not provide conclusive proof of the presence of an aircraft source. However, it does highlight a very different pattern compared with a road source (Fig. 1a).

![Fig. 1a) Bivariate polar plot for NO\textsubscript{X} (\mu g m^{-3}) at the Hillingdon monitoring site located approximately 40 m north of the M4 motorway, b) bivariate polar plot for SO\textsubscript{2} (ppb) at the Thurrock background monitoring site located close to areas of industrial activity c) bivariate polar plot for NO\textsubscript{X} (\mu g m^{-3}) at LHR2 with background NO\textsubscript{X} concentrations subtracted from Oaks Road for 06:00-23:00 on an hourly basis, d) estimated error surface (at 2\sigma) for (c) based on the re-sampling approach described in section 2.3. In each plot the wind speed increases radially outwards towards the circumference to 10 m s\textsuperscript{-1}.](image)

### 2.3 Sources of uncertainty and estimation of errors

The pattern of concentration shown in Fig. 1c is influenced by many factors related to meteorology and emissions. Furthermore, as shown by Yu et al. (2004), the choice of wind speed and direction interval size is also a factor. However, the principal influence on the uncertainty associated with Fig. 1c is the number of data points that exist in any wind speed – wind direction cell. Even with several years of data, some cells
only have a few data points; most notably those at high wind speeds (≥ 9 m s⁻¹) with an easterly component. As the sample size decreases in a cell, the representativeness of the mean concentration in that cell will become more uncertain. Estimates of the sampling errors were made using a re-sampling technique. For concentrations affected most by airport sources, a wind angle from 180-220° was considered. By considering several wind direction sectors affected by the same dominant source together, enough data points were available to test the effect of sample size on the mean concentration. Within each wind speed category (e.g. from 1-2, 2-3 m s⁻¹...) different sample sizes from 2 to 300 were randomly selected without replacement 500 times. For each ensemble, the standard deviation of the mean was calculated. Fig. 2 shows the effect of sample size on the error at LHR2. It shows that for cells with relatively few measurements the standard deviation of the mean NOₓ concentration is high. For example, for a cell with only 10 measurements, the standard deviation is approximately 10 μg m⁻³. The importance of the error does, however, depend on the magnitude of the concentration due to a particular source. In the case of LHR2 in the direction of the aircraft, the concentration is typically around 70-100 μg m⁻³, which is much larger than the sample error and suggests that sample sizes smaller than 10 would be adequate to highlight the source. Nevertheless, the plot shows that the error declines sharply as the number of measurement points in a cell increases.

The errors shown in Fig. 1d also depend on other factors. In the direction of the aircraft the concentration of NOₓ varies little by wind speed. Furthermore, there was found to be very little variation in the sampling error with wind speed for wind directions from the airport, as shown in Fig. 2. However, for a road source (not shown), an increase in wind speed resulted in a decrease in the mean NOₓ concentration and the error estimate. For wind directions from the north at LHR2 (i.e. from the nearby road), it was found that the error term decreased typically by a factor of 2-3 as the wind speed increased from 1-2 to 8-9 m s⁻¹. It was also found that there was an approximately linear dependence between the error estimate and the mean concentration of NOₓ. Because the focus of this work is aircraft sources, the error estimates derived in Fig. 1c have been used to derive an error surface as shown in Fig. 1d. In Fig. 1d, the largest errors were calculated for wind speeds > 8 m s⁻¹ from all wind directions except the south-west sector. The lower estimated error at high wind speeds from the south-west is due to the high proportion of wind angles from that direction, which is the prevailing wind direction. Overall, the pattern of concentration shown in Fig. 1c
does not change much if estimated sampling errors are accounted for. In addition to the sample population errors, errors are associated with the interpolation routine used. However, these errors are small in comparison with the sample population errors because over 95% of the wind speed/direction cells are populated with one or more measurements. Increasing the resolution of the grid spacing led to a smoother plot rather than a plot with a different distribution of concentration. Furthermore, the concentration distribution was not found to be highly sensitive to the interpolation technique used. These calculations do, however, highlight the sample size necessary for the bivariate polar plots to represent a particular source. Nevertheless, care would need to be exercised when applying this approach to sparse data sets.

![Graph showing the dependence of sample size on the standard deviation of the mean NO\textsubscript{X} concentration for a wind direction 180-220° and different wind speed intervals at LHR2.]

Fig. 2 Dependence of sample size on the standard deviation of the mean NO\textsubscript{X} concentration for a wind direction 180-220° and different wind speed intervals at LHR2.

3. Results and Discussion

3.1 Spatial analysis

Bivariate polar plots for NO\textsubscript{X} have been derived for seven monitoring sites shown in Fig. 3. Each of these plots considers hours from 06:00-22:00, to maximise the NO\textsubscript{X} signal from aircraft sources. Appropriate background NO\textsubscript{X} concentrations were subtracted, consistent with the assumptions shown in Table 2, with the aim of highlighting potential airport sources. Most plots highlight elevated NO\textsubscript{X} concentrations even at wind speeds up to 10 m s\textsuperscript{-1} when the wind direction is from the airport. At Harlington and Hounslow NO\textsubscript{X} concentrations of between 30-50 μg m\textsuperscript{-3} are observed for wind speeds around 5-6 m s\textsuperscript{-1}. For the sites to the
east of LHR (Slough, Green Gates and Main Road), and for wind speeds typically > 3 m s\(^{-1}\), NO\(_X\) concentrations will be detected for ‘easterly operations’ i.e. take off east on the southern runway and landing on the northern runway. This potentially explains the relatively high concentrations of NO\(_X\) recorded at Main Road (550 m from the southern runway) due to aircraft taking off on that runway. Similarly, at Green Gates, lower NO\(_X\) concentrations might be expected because aircraft land on the northern runway during easterly operations. On this basis, the NO\(_X\) concentrations shown in the Slough plot look anomalously high, which might suggest the influence of other sources. Finally, the site at Oaks Road, also highlights a potential aircraft NO\(_X\) source, which is most apparent in the direction from 340-80°. Taken together, these plots appear to show the presence of a NO\(_X\) source in the direction of LHR that does not have the characteristics of typical ground-level sources. However, the analysis is qualitative and does not provide statistically robust proof of the presence of aircraft sources. Section 3.3 seeks to quantify the presence of aircraft by applying a statistical test.

Fig. 3. Bivariate NO\(_X\) polar plots for monitoring sites close to Heathrow Airport with background concentrations subtracted (\(\mu g \text{ m}^{-3}\)). These plots are for hours from 06:00–22:00. The grey lines highlight major roads and the two rectangles show the location of the northern and southern runways.
The patterns of concentration observed in Fig. 3 depend on the choice of background site used for subtraction. Ideally, background sites would be located at the airport perimeter, directly upwind of the site of interest. In most cases the choice of site was limited by the availability of background sites. However, for some sites there were several possible background sites available for subtraction. The choice of background site is important for two principal reasons. First, it is important to confirm whether the pattern of concentration generated in the bivariate polar plots is strongly affected by the choice of background site. Second, in section 3.4, background subtraction is used to estimate the upper limit of the airport contribution to NO\textsubscript{X} and NO\textsubscript{2} concentrations and the choice of background site is likely to affect the estimated airport contribution. Fig. 4 shows the NO\textsubscript{X} bivariate polar plot for the Oaks Road site to the south of LHR with three different background sites used for subtraction (Hounslow, Slough and Green Gates). The pattern of concentration is similar for all cases with strong evidence of high concentrations of NO\textsubscript{X} (> 50 \(\mu\text{g}\text{ m}^{-3}\)) at wind speeds > 8 m s\textsuperscript{-1} and for wind angles from 350-70°. However, Fig. 4 highlights that using Hounslow as a site for background subtraction leads to lower concentrations of NO\textsubscript{X} than either Slough or Green Gates. This result is expected because the Hounslow site is more directly upwind of Oaks Road (see Fig. 3) and is also in the same direction of central and inner London. Air trajectories from that direction will therefore tend to have a higher concentration of NO\textsubscript{X} than from other directions, thus leading to lower NO\textsubscript{X} concentrations in the bivariate polar plot. Considering all these factors, it was assumed that the Hounslow site was the most appropriate in this case. For other possible combinations of site pairs it was found that the pattern of concentrations was not strongly affected by the choice of site used for background subtraction.

Fig. 4 Effect of choice of background site used for subtraction for NO\textsubscript{X} concentrations measured at Oaks Road (\(\mu\text{g}\text{ m}^{-3}\)) for data from 06:00-22:00.
3.2 Wind speed and runway dependence

Section 2.2 highlighted that the wind speed dependence of NO\textsubscript{X} concentrations at airport-influenced sites is markedly different compared with road sources. This section considers that dependence in more detail and also aims to highlight how the pattern of runway alternation operating at LHR can be highlighted by data filtering. To illustrate the effect on NO\textsubscript{X} concentrations of runway operation, hours were extracted for westerly operation, which were further separated into northerly runway take off and southerly runway take off. Data were also filtered by wind direction (see Table 2) and for hours between 06:00-22:00. Fig. 5 shows the results at LHR2 and Harlington and highlights the very clear difference between the two runway operation modes. At LHR2, for example, the difference between northern and southern runway operation is clear: concentrations of NO\textsubscript{X} are over a factor of two less when aircraft take off from the southern runway. The difference is also clear at Harlington (1230 m and 2670 m from the northern and southern runways, respectively). Considering northern runway take off, these results indicate that NO\textsubscript{X} is diluted by a factor of approximately 5 between LHR2 and Harlington; a distance of 1 km.

Also highlighted in Fig. 5 is the wind speed dependence of NO\textsubscript{X} for LHR2, Harlington and the Hillingdon site that is dominated by a local road source. The Hillingdon results highlight that increasing wind speeds decrease the concentration of NO\textsubscript{X}. By contrast, the LHR2 and Harlington results show that concentrations of NO\textsubscript{X} vary little in the wind speed range 2-12 m s\textsuperscript{-1}. These results highlight the very different behaviour or aircraft plume dilution compared with non-buoyant road traffic sources. Taken together, these characteristics strongly suggest the presence of an aircraft source that can be detected to at least 2600 m from the airport.
Fig. 5. Wind speed dependence of NO\textsubscript{X} concentrations for LHR2, Harlington and Hillingdon. All data sets have been filtered by wind direction and a background concentration removed (see text). The LHR2 and Harlington relationships show the effect on concentrations of different runway operation modes.

3.3 Detection of an aircraft NO\textsubscript{X} signal

There exist certain conditions where the detection of aircraft emissions at monitoring sites close to LHR would be most likely and these can be used to apply a statistical test to determine whether they can be detected in the ambient data sets. As discussed previously, these conditions include: filtering data by wind direction such that emissions from LHR are upwind of the monitoring sites, selecting higher wind speed conditions where the effect of ground-level sources such as roads are diminished, selecting hours of the day (06:00-22:00) when aircraft activity is high, and removing a local background concentration to highlight airport sources. Filtering for these conditions does not, however, result in the unambiguous identification of airport or aircraft emissions because of the remaining dominant effect of road traffic NO\textsubscript{X} sources.

A quantitative approach for detecting aircraft sources at LHR is to exploit the unique activity of aircraft movements and in particular the pattern of runway alternation described in section 2.1 together with the data filtering described above. A statistical test can be constructed that compares measurements of NO\textsubscript{X} when aircraft take-off on the northern runway (and land on the southern runway) with take-off on the southern runway (and land on the northern runway) for westerly operation. These two modes of operation yield two
independent hourly data sets that can be analysed for a statistical difference between them because aircraft take-off emissions are many times higher than taxing or landing emissions (see Herndon et al. (2004) and the International Civil Aviation Organization (ICAO) emissions databank at http://www.caa.co.uk/). This unique activity profile of aircraft movements at Heathrow therefore results in emissions that vary spatially and temporally in a way that is different from other sources such as road transport and thus distinguishes them from other emission sources. Here, use is made of the nonparametric Mann-Whitney U test for two independent samples. This approach was chosen rather than the t-test because the latter assumes normality, which rarely exists in air pollution data sets. Table 2 summarises the results of the test applied to the monitoring sites and highlights the value of $p$, the probability that there is no difference in the means, and the test statistic $Z$.

Table 2. Results of the Mann-Whitney test applied to filtered data at monitoring sites.

<table>
<thead>
<tr>
<th>Site</th>
<th>Wind direction</th>
<th>Background site</th>
<th>P</th>
<th>Z</th>
</tr>
</thead>
<tbody>
<tr>
<td>LHR2</td>
<td>150-260</td>
<td>Oaks Road</td>
<td>0.000</td>
<td>64.8</td>
</tr>
<tr>
<td>Harlington</td>
<td>160-260</td>
<td>Oaks Road</td>
<td>0.000</td>
<td>10.2</td>
</tr>
<tr>
<td>Hounslow</td>
<td>200-260</td>
<td>Oaks Road</td>
<td>0.000</td>
<td>6.2</td>
</tr>
<tr>
<td>Oaks Road</td>
<td>340-80</td>
<td>Hounslow</td>
<td>0.000</td>
<td>5.3</td>
</tr>
<tr>
<td>Main Road</td>
<td>80-170</td>
<td>Oaks Road</td>
<td>0.002</td>
<td>3.1</td>
</tr>
<tr>
<td>Green Gates</td>
<td>100-170</td>
<td>Oaks Road</td>
<td>0.000</td>
<td>3.5</td>
</tr>
<tr>
<td>Slough</td>
<td>100-170</td>
<td>Oaks Road</td>
<td>0.803</td>
<td>0.3</td>
</tr>
<tr>
<td>Hillingdon</td>
<td>130-230</td>
<td>Oaks Road</td>
<td>0.268</td>
<td>1.1</td>
</tr>
</tbody>
</table>

1Data in bold show results that are statistically significant at the $p=0.01$ level.

2Data have also been filtered by hour of day (06:00-22:00); wind speeds $> 3$ m s$^{-1}$.

The strongest signal of aircraft emissions is at LHR2, which is not surprising given the proximity of this site to the airport. At this site there was also a large difference in NO$_X$ concentration between aircraft taking off or landing on the northern runway (136.2 vs. 44.8 μg m$^{-3}$). At other sites the difference in measured NO$_X$ due to aircraft operation is much less significant. At the Hounslow site, for example, the difference was 29.8 vs. 29.2 μg m$^{-3}$ and yet there is a highly statistically significant difference between the two modes of aircraft operation. Only two of the monitoring sites did not show an indication of an aircraft source: Hillingdon and Slough. The Hillingdon site is about 2 km from the northern runway and is dominated by a nearby motorway source of NO$_X$. Because runway alternation is not used for easterly operation, it is difficult to use this approach to detect aircraft sources to the east of the airport. It is not possible to say with confidence...
therefore, whether the concentration pattern shown for Slough in Fig. 3 is due to the airport or other sources of NOX. These results show that the unique pattern of aircraft operation at LHR2 is a very effective characteristic that can be used to detect the influence of the airport even at locations where the contribution to NOX is small and dominated by other sources of NOX.

It is possible that some of the results in Table 2 could have arisen by chance because of variations in meteorology and emissions that are not associated with the airport. Therefore, as an additional check, the Mann-Whitney test was also applied to a sample of 10 other NOX monitoring sites across London more than 5 km away from LHR (5 background and 5 roadside) using the same data filtering techniques to determine whether there was a statistically significant difference due to aircraft operation. At all of these sites p >0.1 suggesting that there was no statistically significant difference in NOX concentration due to different runway operation modes. These results provide strong evidence that aircraft emissions can only be detected at monitoring sites within a few km of LHR.

3.4 Quantification of airport contribution to concentrations of NOX

Estimates of the upper limit of the airport contribution to NOX can be made by considering the wind sector where the airport is likely to have an effect and by removing a background contribution. The choice of background site and wind sector is shown in Table 2. The estimate is an upper limit because of the presence of other sources of NOX between the background site and the site analysed. In the case of LHR2, the upper estimate should also be a best estimate because there are very few other sources between LHR2 and Oaks Road. For all the other sites, there are other sources (principally roads) that would also contribute. A better estimate of the airport contribution is likely to be made by filtering for wind speeds > 3 m s⁻¹. The filtering has the effect of reducing the influence of road sources while maximising aircraft sources. This approach is, however, an approximation and dispersion modelling would be needed to reduce the uncertainty.

An upper limit to the airport contribution to NO₂ concentrations has also been estimated, as shown in Table 3. At LHR2 it is estimated that the airport contributes 15.0 μg m⁻³ (27.3 %) of the total measured NO₂, which is similar to the NOX contribution. This estimate yields a NO₂/NOX ratio of 0.44. Estimates at
Harringhton and Hounslow were similar, and contributed 17.4 and 18.0 % of the total NO₂ measured as an upper limit at these sites, respectively. Using the 3 m s⁻¹ wind speed filtering suggests that a contribution of about 10 % is probably closer to the actual contribution at these two sites. These results also show that NO₂ accounts for a greater proportion of the total NO₂ than for NOₓ at Harrington and Hounslow (ca. 10 vs. 18 %). This is probably because the airport plume is “aged” and that there is enough time for the plume to be well-mixed and for NO to react with ozone to form NO₂.

Table 3. Estimated upper limit of airport NOₓ and NO₂ contribution to measured NOₓ and NO₂ concentrations.

<table>
<thead>
<tr>
<th>Location</th>
<th>Upper limit for airport NOₓ contribution (µg m⁻³)</th>
<th>Upper limit for airport NOₓ contribution (%)</th>
<th>NOₓ range (µg m⁻³)*</th>
<th>Upper limit for airport NO₂ contribution (µg m⁻³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LHR2</td>
<td>33.9</td>
<td>26.7</td>
<td>21.5-33.9</td>
<td>15.0</td>
</tr>
<tr>
<td>Harlington</td>
<td>9.9</td>
<td>14.0</td>
<td>5.7-9.9</td>
<td>6.6</td>
</tr>
<tr>
<td>Hounslow</td>
<td>9.5</td>
<td>12.0</td>
<td>5.7-9.5</td>
<td>6.5</td>
</tr>
<tr>
<td>Green Gates</td>
<td>3.0</td>
<td>4.0</td>
<td>1.1-3.0</td>
<td>1.5</td>
</tr>
<tr>
<td>Main Road</td>
<td>7.1</td>
<td>8.8</td>
<td>3.3-7.1</td>
<td>4.2</td>
</tr>
<tr>
<td>Slough</td>
<td>1.8</td>
<td>2.6</td>
<td>1.7-1.8</td>
<td>1.5</td>
</tr>
<tr>
<td>Oaks Road</td>
<td>5.9</td>
<td>8.9</td>
<td>2.2-5.9</td>
<td>2.0</td>
</tr>
</tbody>
</table>

*numbers in bold are considered to be closest to the actual contribution.

Although it is not the focus of this paper, hourly measurements of particulate matter below 10 µm diameter (PM₁₀) made by the Tapered Element Oscillating Microbalance (TEOM) technique were available at LHR2 and Oaks Road, which enabled similar analyses to be undertaken for PM₁₀ as for NOₓ. The overall contribution made by aircraft to PM₁₀ concentrations is small, but can, however, be detected in a statistically significant way by considering the variation in PM₁₀ concentrations by runway alternation. The same methodology used to calculate the contribution that airport NOₓ emissions was also applied to PM₁₀. The analysis was based on the use of TEOM instruments located at LHR2 and Oaks Road. It is estimated that at LHR2 a contribution of 0.9 µg m⁻³ due to the airport out of a total of 21.6 µg m⁻³ i.e. 4.2 % of the total. It is useful to compare the PM₁₀/NOₓ ratio calculated from these results because the ratio can be compared with that of road transport sources. A mean PM₁₀/NOₓ ratio of 0.015 (on a mass basis) was estimated, which is lower than that for road traffic exhaust emissions of 0.041, based on average vehicle emissions across the
LAEI for 2002. This is consistent with the interpretation that aircraft are a more important source of NOX than PM10 compared with road traffic.

4. Conclusions

A major factor that will determine whether a third runway is built at London Heathrow is the compliance with the EU annual mean Directive for NO2. The complexity of NOX sources close to Heathrow makes it difficult to undertake source apportionment analysis. Even though Heathrow is an important emission source of NOX, concentrations of NOX close to the airport are dominated by road traffic sources. Detecting and quantifying the contribution made by the airport to local concentrations of NOX is therefore difficult.

A graphical technique has been developed, extending the work of Yu et al. (2004), which can help discriminate between sources of NOX emitted at ground level with little or no buoyancy (e.g. road traffic) and sources of NOX with significant amounts of buoyancy (e.g. aircraft and large point sources). Bivariate polar plots have been derived that extend the idea of pollution roses by also accounting for wind speed. It is shown that for a small network of monitoring sites, where enough sites exist to subtract a background concentration, that bivariate polar plots are effective at highlighting the presence of aircraft NOX emissions. By removing a local background contribution a much clearer indication of airport source characteristics can be gained, such as the wind speed dependence of aircraft jet plumes. Although bivariate polar plots have been used in this work to distinguish between sources where plume buoyancy is important or not, they are useful in other situations where there is a complex relationship between the concentration of a species, wind speed and direction. Examples include complex flows in street canyon locations, where the presence of building can affect the flows in a complex manner and particle sources where wind-blown re-suspension is important and where particle concentrations can increase with wind speed.

The unique activity profile of aircraft movements at Heathrow has been exploited to discriminate between airport and other sources of NOX. In particular, the two runways at Heathrow alternate in their use daily and weekly and this contrasts with the pattern of activity for road transport, which is the major contributor to NOX emissions and concentrations close to the airport. The aircraft activity patterns can be used together
with other data filtering techniques that maximise the possibility of observing aircraft sources, to quantify whether aircraft emissions can be detected in hourly measurements of NO\textsubscript{X} and NO\textsubscript{2}. It is found that aircraft NO\textsubscript{X} emissions can be unambiguously detected at monitoring sites at least 2.6 km from the airport. Close to the airport boundary downwind of the prevailing wind direction, we find that the airport accounts for 27\% of the measured NO\textsubscript{X}. However, at about 1.0-1.5 km it is estimated that the airport contribution is diluted by a factor of about 5 and the airport accounts for 12-14\% of the measured NO\textsubscript{X} at these locations. However, because of the presence of other sources (primarily road traffic), estimates made beyond the airport boundary are considered to be upper limits and the actual contribution will be less than these estimates.

This work has also highlighted the contrasting wind speed-dependence of road traffic and aircraft plumes. In the case of aircraft it is found that approximately 180 m from the runway, concentrations of NO\textsubscript{X} vary little across a wind speed range from 2-12 m s\(^{-1}\). These results indicate that the buoyant nature of aircraft plumes is an important characteristic that should be accounted for in dispersion modelling studies. These results should also provide an effective additional means of validating dispersion models used for the prediction of concentrations in the vicinity of airports.

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