Quantification of air quality impacts of London Heathrow Airport (UK) from 2005 to 2012

Masiol, Mauro; Harrison, Roy M.

DOI:

[10.1016/j.atmosenv.2015.06.048](https://doi.org/10.1016/j.atmosenv.2015.06.048)

License:

Creative Commons: Attribution-NonCommercial-NoDerivs (CC BY-NC-ND)

Document Version

Peer reviewed version

Citation for published version (Harvard):

Masiol, M & Harrison, RM 2015, 'Quantification of air quality impacts of London Heathrow Airport (UK) from 2005 to 2012', *Atmospheric Environment*, vol. 116, pp. 308-319. <https://doi.org/10.1016/j.atmosenv.2015.06.048>

[Link to publication on Research at Birmingham portal](#)

Publisher Rights Statement:

After an embargo period this document is subject to the terms of a Creative Commons Attribution Non-Commercial No Derivatives license

Checked October 2015

General rights

Unless a licence is specified above, all rights (including copyright and moral rights) in this document are retained by the authors and/or the copyright holders. The express permission of the copyright holder must be obtained for any use of this material other than for purposes permitted by law.

- Users may freely distribute the URL that is used to identify this publication.
- Users may download and/or print one copy of the publication from the University of Birmingham research portal for the purpose of private study or non-commercial research.
- User may use extracts from the document in line with the concept of 'fair dealing' under the Copyright, Designs and Patents Act 1988 (?)
- Users may not further distribute the material nor use it for the purposes of commercial gain.

Where a licence is displayed above, please note the terms and conditions of the licence govern your use of this document.

When citing, please reference the published version.

Take down policy

While the University of Birmingham exercises care and attention in making items available there are rare occasions when an item has been uploaded in error or has been deemed to be commercially or otherwise sensitive.

If you believe that this is the case for this document, please contact UBIRA@lists.bham.ac.uk providing details and we will remove access to the work immediately and investigate.

1
2
3
4
5
6 **QUANTIFICATION OF SOME AIR QUALITY**
7 **IMPACTS OF LONDON HEATHROW**
8 **AIRPORT (UK) FROM 2005 TO 2012**
9

10
11 **Mauro Masiol and Roy M. Harrison^{*†}**
12

13
14 **Division of Environmental Health and Risk Management**
15 **School of Geography, Earth and Environmental Sciences**
16 **University of Birmingham**
17 **Edgbaston, Birmingham B15 2TT**
18 **United Kingdom**
19
20
21

^{*} To whom correspondence should be addressed.

Tele: +44 121 414 3494; Fax: +44 121 414 3708; Email: r.m.harrison@bham.ac.uk

[†]Also at: Department of Environmental Sciences / Center of Excellence in Environmental Studies, King Abdulaziz University, PO Box 80203, Jeddah, 21589, Saudi Arabia

22 **HIGHLIGHTS**

- 23 ➤ Eight years of hourly air pollution data from 8 sites around Heathrow are analysed
- 24 ➤ Temporal analysis reveals diurnal, weekly and seasonal patterns and annual trends
- 25 ➤ Statistical tools are applied to depict the inter-site relationships
- 26 ➤ The relationships with weather parameters and atmospheric circulation are studied
- 27 ➤ The contributions of airport and motorway traffic are quantified

28 ABSTRACT

29 Among other emission sources in the Greater London area, the international airport of Heathrow is
30 recognised to be a major source of air pollution and is one of the UK locations where European air
31 quality Limit Values are currently breached. However it is very difficult to differentiate between
32 pollutants arising from airport operations and those from the large volumes of road traffic generated
33 by the airport, as well as the nearby M4 and M25 motorways, A4 and A30 major roads, the
34 conurbation of London and other external sources. In this study, eight years (January 2005–
35 December 2012) of measurements of various air pollutants (NO, NO₂, NO_x, O₃, CO, PM₁₀ and
36 PM_{2.5}) were investigated from 10 sites: eight sites are located within a distance of 2.5 km from the
37 airport, while two sites representative of the regional background and of background air quality in
38 London (Harwell (60 km WNW) and North Kensington (17 km ENE), respectively) were included.
39 A series of statistical tools was thus applied to: (1) investigate the time series by analysing hourly
40 data as diurnal, weekly, seasonal and annual patterns; (2) reveal the effects of the atmospheric
41 circulation upon air pollution by analysing background-corrected polar plots and (3) quantify the
42 impact of the airport upon air quality in the local area using the inter-site differences of measured
43 concentrations. The results show different diurnal patterns in emissions of NO_x from the airport and
44 from the motorways. The concentration increment arising from passage of air across the airport
45 during airport activity (6am-10pm) and with wind speed > 3 m s⁻¹ is ca. 1-9 µg m⁻³ of NO₂ and 2-20
46 µg m⁻³ of NO_x at background stations. Such results are slightly lower than in a previous study
47 analysing the 2001-2004 period. Air quality impacts of the M25 and M4 motorways are substantial
48 only at the Hillingdon site (30 m from M4). Concentration increments of particulate matter can take
49 either small positive or negative values.

50

51 **Keywords:** Airport; aircraft; road traffic; emissions; nitrogen oxides; particulate matter

52

53 1. INTRODUCTION

54 During the last decades, an increasing number of epidemiological studies have established a direct
55 association between the exposure to some ambient air pollutants and adverse effects on human
56 health due to respiratory and cardiovascular diseases (e.g., Dockery, 2009; Katsouyanni et al., 2009;
57 Raaschou-Nielsen et al., 2013). Recently, outdoor air pollution has been classified as known
58 carcinogenic to humans (Group 1) by the IARC. However, in the last decades, most European
59 countries have experienced a general drop of ambient levels for many air pollutants. Generally, this
60 air quality improvement has followed the implementation of legislation, technological advances, the
61 application of successful abatement technologies and other mitigation measures. However, air
62 pollution in Europe remains an actual and serious concern. Under this scenario, the identification,
63 characterisation and quantification of the most relevant sources is amongst the main objectives
64 addressed in research by policy-makers and stakeholders.

65
66 In Europe, air quality is monitored by local and national authorities through an extended monitoring
67 network and data are managed to meet EC Directive requirements. In case of the exceeding of Limit
68 Values or even lower assessment thresholds, such data can be used to inform the population about
69 air quality and potential impacts upon health. Moreover, such data represent a valuable resource to
70 develop and implement possible mitigation measures.

71
72 Among the EU-27 countries, UK has fewer critical issues in relation to air pollution than some
73 other regions, such as Benelux, Northern Italy and some Eastern European countries (EEA, 2014).
74 However, high levels of air pollutants exceeding the European air quality Limit Values are still
75 recorded in the Greater London urban area (GL), where an extensive and densely populated
76 conurbation hosts more than 9 million inhabitants, with the related high traffic and energy demand
77 for domestic heating. In particular, those pollutants which currently do not fulfil the EU and UK air

78 quality standards and objectives (DEFRA, 2013a) are nitrogen dioxide ($>$ Limit Value) and ozone
79 ($>$ target value).

80

81 Among other emission sources in the Greater London area, the airport of Heathrow (LHR) is
82 recognised to be a major source of nitrogen oxides (e.g., Carslaw et al., 2006; 2008; Stettler et al.,
83 2011; Yim et al., 2013) and NO_2 concentrations have breached the EU and UK annual mean Limit
84 Value ($40 \mu\text{g m}^{-3}$) at some locations around the terminals in the last decade (UK Department of
85 Transport, 2006; HAL, 2011). The Airports Council International (ACI, 2014) reported that LHR is
86 amongst the busiest airports for arriving and departing passengers (~ 72 million passengers y^{-1} in
87 2013), and consequently has congested flight traffic with near capacity utilisation during many
88 hours of the day (e.g., Gelhausen et al., 2011; Bernhart et al., 2012). In the past decade some studies
89 have attempted to estimate the contribution of LHR to local air quality, especially for nitrogen
90 oxides ($\text{NO} + \text{NO}_2 = \text{NO}_x$). For example, Carslaw et al. (2006) estimated that airport operations
91 accounted for $\sim 27\%$ of the annual mean NO_x and NO_2 at the airfield boundary and less than 15%
92 ($< 10 \mu\text{g m}^{-3}$) at background locations 2–3 km downwind of the airport. Carslaw et al. (2008)
93 investigated the nitrogen oxides levels in individual plumes from aircraft departing on the LHR
94 northern runway and found that aircraft operational factors such as take-off weight and aircraft
95 thrust setting have effects on concentrations of NO_x . Results of a model evaluation for the 2008/9
96 period by AEA (2010) indicated that the source attribution from airport operations at surrounding
97 monitoring sites was similar to that calculated by Carslaw et al. (2006). Stettler et al. (2011)
98 estimated that emissions due to the landing and take-off (LTO) cycles accounted for $\sim 8.19 \times 10^6$ kg
99 NO_x in 2005, of which more than 80% are in form of NO. HAL (2011) reported that 46% of the
100 total ground level NO_x from aircraft in 2010 was emitted during take-off roll, 21% in taxi-in and
101 taxi-out phases, 19% by the auxiliary power units (APUs), while the remaining 14% is attributed to
102 hold, landing roll and engine testing. Carslaw et al. (2012) quantified the impact of the flight-ban
103 due to the eruption of the Icelandic volcano Eyjafjallajökull on concentrations of NO_x in April 2010

104 and stated that airport closure resulted in an unambiguous effect on NO_x and NO₂ concentrations.
105 Yim et al. (2013) applied a multi-scale air quality modelling approach to assess the air quality
106 impacts of UK airports and calculated that 24% of UK-wide aviation-attributable early deaths could
107 be avoided in 2030 if Heathrow were replaced by a new airport the in Thames Estuary, because the
108 location is generally downwind of London, and at greater distance.

109

110 This study analyses an eight year hourly time series (January 2005– December 2012) of air
111 pollutants measured at 10 monitoring sites. Eight sites are located in the surroundings of LHR,
112 while two stations were selected to be representative of regional background and GL pollution,
113 respectively. The main aims are to investigate the time series for patterns and trends, and study the
114 potential location and strength of the main sources and their impact upon air quality.

115

116

117 **2. MATERIALS AND METHODS**

118 Data were measured at 10 sites managed by the UK Department for Environment, Food and Rural
119 Affairs (DEFRA; <http://uk-air.defra.gov.uk/>) and London Heathrow authorities
120 (<http://www.heathrowairwatch.org.uk/>). A map of the sites is shown in Figure 1, with greater detail
121 of the sites local to Heathrow in Figure SI1, while the site names, acronyms, some characteristics,
122 the monitored pollutant and the periods of available data are summarized in Table 1. One site
123 (LHR2) is situated 180 m north to the northern runway centreline and a few metres inside the
124 airport boundary. Four sites (GRG, OAK, HAT, HOA) are positioned close (<330 m) to the outer
125 perimeter of the airport, while three sites (HRL, HIL, SLC) are located farther from the airport (> 1
126 km). The maximum distance between any pair of sites is 6 km (SLC-HOA). A very similar set of
127 monitoring stations was used in a previous study (Carslaw et al., 2006) which investigated data up
128 to 2004. Because of their relative proximity, the eight sites are affected to differing degrees by the
129 same set of sources, which include airport activities (aircraft, ground support equipment, auxiliary

130 power units), road traffic (mainly due to the M4 and M25 motorways, A4, A30 and minor local
131 roads) and urban emissions (domestic heating). However, due to the high density of potential
132 emission sources in the study area, sites are categorized differently (Table 1). Two supplementary
133 sites were selected to provide comparative data for regional (HAR) and urban London (LNK)
134 background pollution. Despite being classified as “urban background” the Hillingdon site is only 30
135 metres from the busy M4 motorway, and hence heavily influenced by it.

136

137 Analysed pollutants were measured hourly using automatic instruments according to European
138 protocols. Quality assurance and quality control procedures follow the standards for the Automatic
139 Urban and Rural Network (AURN) and the London Air Quality Network (LAQN): all instruments
140 are routinely calibrated, and every six months are fully serviced and undergo an intercalibration
141 audit. Weather data measured at Met Office Heathrow (station ID no. 708) including wind direction
142 and speed, atmospheric pressure, air temperature and relative humidity (RH) were provided by the
143 Met Office (<http://www.metoffice.gov.uk>) and BADC (<http://badc.nerc.ac.uk/data/>).

144

145 Data were analysed using R version 3.0.1 (R Core Team, 2013) and a series of supplementary
146 packages, including ‘Openair’ (Carslaw and Ropkins, 2012; Carslaw, 2013). Preliminary data
147 handling and clean-up were carried out to check the datasets for outliers and anomalous records.
148 Particulate matter (PM₁₀ and PM_{2.5}) was measured automatically using TEOM or TEOM-FDMS
149 (Table 1). However, the main concern with the use of the TEOM technique is the loss of the more
150 volatile component (principally some semi-volatile hydrocarbons and nitrates) because the inlet is
151 held at a temperature of about 50 °C. A simple adjustment applied to the UK data is to apply a
152 factor of 1.3 to TEOM-measured concentrations to give approximate comparability with the
153 European gravimetric reference method. Recently, the use of more sophisticated techniques
154 (TEOM-FDMS and the Volatile Correction Method (VCM)) has allowed robust estimations of PM
155 mass. To harmonise the datasets and obtain comparable data, PM₁₀ were reported as gravimetric

equivalent (TEOM x 1.3), VCM corrected and TEOM-FDMS, depending on the technique used. Unfortunately, no suitable correction method yet exists for PM_{2.5} and the sampling stations are equipped with differing instruments, which were sometimes changed during the study period (Table 1). The best compromise is thus to use TEOM for LHR2, GRG, OAK (full period, if available) and TEOM-FDMS for HRL, HAR, LNK (starting about in 2009). Due to this, a cross-comparison between the two groups is not possible.

Data for traffic on the M4 and M25 motorways is provided by the UK Department for Transport, which commissions manual counts of traffic for a number of count points every year. The counts take place between 7 am and 7 pm; each road link is counted a maximum of one day in a year. Data for LHR air traffic is provided by Heathrow authorities.

3. RESULTS AND DISCUSSION

Data frequency distributions for each pollutant during the whole study period are given for all sites as boxplots in Figure SI2, while the time series of monthly averaged concentrations calculated from hourly data are shown in Figure 2. In this study NO_x mass concentrations are expressed as NO₂. The average concentrations of NO over the 8 years at eight sites at Heathrow varied from 18 µg m⁻³ (OAK) and 41 µg m⁻³ (LHR2), while NO₂ ranged from 31 µg m⁻³ (SLC) to 51 µg m⁻³ (at both LHR2 and HIL) and NO_x from 59 µg m⁻³ (SLC) to 114 µg m⁻³ (LHR2). Low levels of nitrogen oxides were recorded at the rural background site (HAR: 2, 11 and 14 µg m⁻³ for NO, NO₂ and NO_x, respectively). All Heathrow sites have NO levels significantly higher than LNK (14 µg m⁻³), while NO₂ concentrations are comparable (37 µg m⁻³ at LNK). Since vehicular traffic is the major source of nitrogen oxides at LNK, this result gives a first indication that in the surroundings of Heathrow Airport there is an anomaly in NO levels. In recent years there has been growing attention towards NO_x emissions and the NO-NO₂ partitioning in Europe because of the evident discrepancy between achieving NO_x emission reductions and NO₂ ambient concentrations, which

do not meet the targets in many locations (e.g., Grice et al., 2009; Cyrus et al., 2012). In the UK, electricity generation is recognized to be the main anthropogenic source of emissions (29.8%), followed by road traffic (~27.5%, of which 14.5% is from passenger cars and 13% from heavy duty vehicles), other stationary combustion sources (25%) and off-road transport (16.6%) (DEFRA, 2013b). However, it is evident that road traffic is the main contributor to ambient ground-level concentrations of nitrogen oxides in urban environments, and the recent increase in NO₂ levels in Europe has been related to the growing proportion of diesel-powered vehicles, which are known to have higher primary (direct) emissions of NO₂ (Carslaw et al., 2007). Aircraft engines also emit NO_x, and emissions increase monotonically with engine thrust, i.e. are higher during take-off and lower in taxi and idle phases. The NO-NO₂ partitioning in the emissions of modern high by-pass turbofan engines is also thrust-dependent: NO₂ is principally emitted at idle, while NO is dominant at higher thrust regimes (Wormhoudt et al., 2007). Other in-airport sources of nitrogen oxides may be attributed to: (i) the use of auxiliary power units (APUs), which are small on-board gas-turbine engines; (ii) the ground power units (GPUs) directly provided by airports and (iii) the airport ground service equipment (GSE), which refers to most of the equipment that an airport offers as a service for flights and passengers and includes a large number of vehicles. In this study, the NO₂/NO_x ratio was calculated and results show lowest ratios at LHR2, HIL and HOA.

Comparing results averaged over 8 years with the annual EC Limit Value for NO₂ (40 µg m⁻³ averaged over 1 year), it is evident that the limit is exceeded at LHR2, HIL and HOA. However, the HIL and HOA sites are strongly influenced by the M4 motorway (HIL) or A4 highway (HOA), and LHR2 is within the airport boundary where the limit values do not apply. Moreover, it should be remembered that NO₂ levels are much lower than those normally recorded in many hotspots in Europe, such as Northern Italy and some areas of Benelux and Germany.

207 Data for ozone are available only for HAR, LNK, HRL and HIL (8 years), while at LHR2 and OAK
 208 measurements finished in 2007: highest concentrations were recorded at the rural site, followed by
 209 OAK ($39 \mu\text{g m}^{-3}$) and LNK ($38 \mu\text{g m}^{-3}$), whereas lower levels were recorded at HIL ($27 \mu\text{g m}^{-3}$).
 210 The information and alert thresholds were exceeded only on a limited number of days. Carbon
 211 monoxide and sulphur dioxide are emitted from both vehicular traffic (very little in recent years)
 212 and aircraft engines. However, data for CO and SO₂ are available only at 4 and 3 sites, respectively,
 213 and at no sites around Heathrow do such data cover the entire study period (generally measurements
 214 finished in 2007). The concentrations of CO and SO₂ are well below the limits set by EU Directives
 215 or recommended by the WHO (WHO, 2000). Because of the complex photochemistry of the NO-
 216 NO₂-O₃ system, the level of total oxidants (OX=O₃+NO₂ expressed in ppbv) is frequently reported
 217 in the literature (e.g. Anttila et al., 2011; Mavroidis and Chaloulakou, 2011; Notario et al., 2012) to
 218 give insights into the oxidative potential in the atmosphere (Kley et al., 1999). The highest OX
 219 levels are recorded at LHR2, however such data refer to measurements before April 2007, while the
 220 values were lower at HAR and HRL.

221
 222 The concentrations of PM₁₀ calculated over 8 years never exceeded the European annual Limit
 223 Value of $40 \mu\text{g m}^{-3}$ and varied from $28 \mu\text{g m}^{-3}$ (HIL) to $18 \mu\text{g m}^{-3}$ (HAR). PM_{2.5} levels were
 224 recorded only at HAR and OAK (full period), GRG (missing data for about 20 months), LHR2
 225 (from 2007), HRL and LNK (from 2009). Despite the sparse coverage of data for some sites, it is
 226 evident that the average concentrations are similar at all sites, varying from $15 \mu\text{g m}^{-3}$ (LNK) to 11
 227 $\mu\text{g m}^{-3}$ (HAR, LHR2, GRG, OAK), and the European target value of $25 \mu\text{g m}^{-3}$ averaged over a
 228 calendar year is far from being breached at any of the sites.

229

230 **3.1 Seasonal and Weekly Variations**

231 Figure SI3 and Figure 3 show the monthly time series and weekly cycles for all the monitored
 232 pollutants, calculated over eight years. For all the measured pollutants, similar seasonal trends and

233 weekly patterns are recorded at all the sites, except HAR. Generally, the cycles derive from the
234 interaction of emissions, dispersion and atmospheric chemical processes. NO, NO₂ and NO_x show
235 typical seasonality at all the road traffic-influenced sites, with maxima in the coldest seasons (Nov-
236 Feb) and minima in the warmest months (May-Aug) and two daily peaks corresponding to the hours
237 with higher traffic, i.e. morning 7-9 am and evening, as previously observed at London, North
238 Kensington (Bigi and Harrison, 2010). Figure SI1 reports the average daily road traffic and aircraft
239 movement profiles. Such patterns are the mirror image of the levels of ozone, which exhibit
240 increased levels in the April-July period and two daily maxima at 2-4 am and 1-4 pm.

241

242 Particulate matter (PM₁₀ and PM_{2.5}) exhibits two monthly peaks in spring and autumn, while
243 minima are in August. This behaviour is evident at all the sites, except PM₁₀ at HIL, which presents
244 an additional increase of monthly-averaged concentrations in Jun-Jul, although data for this site
245 only refer to two years of observations. The weekly cycles are similar to nitrogen oxides at all the
246 sites: two peaks of concentration were generally recorded daily corresponding to the peaks of
247 traffic. However, as for gaseous pollutants, particulate matter is also affected by the dispersion
248 driven by the daily cycles of the mixing layer. Figure 3 also shows the weekday/weekend
249 differences: nitrogen oxides, CO and PM₁₀ clearly show lower concentrations during weekends,
250 while PM_{2.5} shows a much smaller effect. On the other hand, O₃ increases during the weekends,
251 further underlining its interplay with nitrogen oxides.

252

253 **3.3 Long-Term Trends**

254 The long-term trends of the pollutants have been analysed by calculating the smooth trends of the
255 monthly averages. This procedure is essentially determined using generalized additive modelling:
256 further details of the adopted methods are provided in Carslaw (2013). Data were firstly
257 deseasonalized using the seasonal-trend decomposition procedure of time series based

on ‘loess’ (STL). Results are provided in Figure 4 and Figure SI4: along with the fit smooth lines, which represent the long-term trends, the figure also shows the 95% confidence intervals of the fits as grey bands. Such intervals are calculated by bootstrapping the data (n=2000).

Generally, concentrations of nitrogen oxides show constant or slightly decreasing tendencies at all the sites, except in HIL, where a notable increase of NO₂ was recorded, i.e. annual means increased from 45 µg m⁻³ in 2005 to 57 µg m⁻³ in 2012. Decreases in nitrogen oxide emissions have been reported over all Western Europe in the last decades and were essentially attributed to the EU mitigation measures adopted since 1990 (Vestreng et al., 2009). However, the NO₂ levels have not decreased at the same rate as those of NO_x (e.g., Carslaw et al., 2007; Zamboni et al., 2009; Anttila et al., 2011). It is likely that the increase of NO_x levels at HIL is the result of an increased vehicular traffic on the adjacent M4 motorway. Despite trends for ozone having been computed for only 4 sites, it is evident that a slight increase of concentrations occurred in the rural background, while at remaining sites levels were almost constant. The increasing levels of ozone at HAR are not surprising as the same behaviour was predicted over recent decades for many rural regions in Europe, including the southern UK (e.g. Colette et al., 2011; Paoletti et al., 2014). Decreasing trends of PM₁₀ were instead observed at all the sites, particularly for LHR2, while trends of PM_{2.5} were almost constant at HAR, LNK, LHR, HRL and slightly decreasing at GRG and OAK. In summary, all the pollutants at almost all the sites underwent a decline of concentrations in the past eight years. In addition, the quantification and the assessment of the significance of the trends were evaluated by applying the Theil-Sen nonparametric estimator of slope (Sen, 1968; Theil, 1992) on the de-seasonalized monthly means (Carslaw, 2013). Since missing data can significantly affect this method, only months having at least 75% of available data were included in the computations and missing months were linearly interpolated. The trends are listed in Table SI1 along with the upper and lower 95th confidence intervals in the trends and the *p*-values, which indicate the statistical significance of the slope estimation.

284 **3.4 Polar Plot Analysis**

285 A preliminary investigation on potential sources of atmospheric pollutants at each site was assessed
286 by mean of polar plot analysis. Polar plots essentially map the pollutant concentrations by wind
287 speed and direction as a continuous surface (Carslaw and Ropkins, 2012). Simple polar plots
288 computed for each site over the whole dataset are provided as Figures SI5 and SI6. Most polar plots
289 show increasing average concentrations of nitrogen oxides and PM_{2.5}, and decreasing levels of
290 ozone when the wind comes from both the airport and motorway sectors, while PM₁₀ appears to
291 have major sources toward main roads and urban settlements. This is an environment with
292 relatively high concentrations of NO_x and of VOCs. It is behaving as NO_x-saturated, whereby a
293 reduction in NO_x will be accompanied by an increase in ozone, and vice versa. However, even if
294 the sites are strategically located around the main sources, the concurrent effects of multiple
295 emission sources makes it difficult to assess the contribution made by any specific sources.

296
297 According to Carslaw et al. (2006), the subtraction of “background” concentrations for certain wind
298 sectors was further adopted in order to better investigate the effects of single sources. Pairs of sites
299 were therefore selected on the basis of their locations with respect to the main sources and
300 prevailing wind regimes: a reference site downwind of the investigated emission source and a
301 background site located upwind, and hence not directly influenced. In this analysis, each
302 background site is selected as representative of the general levels of air pollutants in the study area
303 before the air masses pass over the investigated sources, i.e., the airfield and motorways. Since the
304 study by Carslaw et al. (2006) only focused on the airport emissions, a larger number of site pairs
305 were selected in this study to include a view on the motorway emissions. Table 2 lists the selected
306 pairs. Resulting polar plots corrected for upwind sites are reported in Figure 5 and are computed
307 over a wind sector spanning ca. 180° toward the background site to account all the potential
308 sources. Generally, pairs of sites selected as indicative of airport emissions clearly indicate a rise of
309 concentrations after passage of air over the airport sector. For example, the maximum average

310 increases of NO_x in the polar plots cells for some selected site pairs shown in Figure 5 were: LHR2-
 311 OAK ($\sim 30 \mu\text{g m}^{-3}$ for NO, $\sim 60 \mu\text{g m}^{-3}$ for NO_2 , $\sim 90 \mu\text{g m}^{-3}$ for NO_x), HRL-OAK ($\sim 20 \mu\text{g m}^{-3}$ for
 312 NO, up to $20 \mu\text{g m}^{-3}$ for NO_2 , $\sim 35 \mu\text{g m}^{-3}$ for NO_x). In a similar way, pairs of sites affected by a
 313 motorway highlighted significant increases: HIL-HRL ($\sim 70 \mu\text{g m}^{-3}$ for NO, $\sim 50 \mu\text{g m}^{-3}$ for NO_2 , up
 314 to $150 \mu\text{g m}^{-3}$ for NO_x); SLC-GRG ($\sim 10 \mu\text{g m}^{-3}$ for NO, $\sim 12 \mu\text{g m}^{-3}$ for NO_2 , $\sim 25 \mu\text{g m}^{-3}$ for NO_x).
 315 Despite few sites measuring ozone, an opposite behaviour was generally observed, with decreasing
 316 concentrations when air comes over the main sources, as a consequence of the NO- NO_2 - O_3 reaction
 317 system. For example, a drop of up to $30 \mu\text{g m}^{-3}$ for ozone is observed for the HIL-HRL pair toward
 318 the M4 motorway, while a decrease of about $25 \mu\text{g m}^{-3}$ is seen for the LHR2-OAK pair toward the
 319 airfield. Despite the drop in O_3 , OX ($= \text{NO}_2 + \text{O}_3$) is still increasing in such pairs toward the main
 320 sources. Results for PM_{10} reveal elevated concentrations when air masses moved over motorways,
 321 while a slight $\text{PM}_{2.5}$ increase seems to be mostly linked to airport emissions for the LHR2-OAK
 322 pair.

323
 324 The polar plot analysis with background subtraction is a proven useful method to check the location
 325 of the main sources in the study area. However, as already reported by Carslaw et al. (2006), it
 326 gives only qualitative results and cannot be used to quantify the source emissions. A reliable
 327 quantification should include all wind sectors and not only those when the source contributions are
 328 highest.

330 **3.5 Quantification of Airport Contributions**

331 A further strategy aiming to quantify the source contributions was thus applied to site pairs which
 332 were proven as unambiguously representative of airport, M4 or M25 emissions by the polar plot
 333 analysis. Since the sites are located around the airport perimeter, the approach is based on the
 334 assumption that the difference in the levels of pollutants between pairs of sites located respectively
 335 upwind and downwind of a source may reflect the contribution of that source.

336 The first step of the approach was to follow the method employed by Carslaw et al. (2006) for
337 estimating the upper limit of airport contributions. Briefly, it is performed by subtracting upwind
338 background contributions from each site to give deltas, (ΔX) for appropriate wind direction sectors
339 in the hours most affected by airport activities, i.e. between 06:00 and 22:00. These can be
340 estimated separately for different wind speed classes. Since most of the sites can be affected by
341 multiple sources (most sites are located near roads), wind speeds $> 3 \text{ m s}^{-1}$ were selected to remove
342 periods with strong local contributions of pollutants. For example, this effect is evident in the polar
343 plot for HRL-OAK (Figure 5) located close a secondary road whose effect cannot be disregarded, or
344 in polar plots for LHR2, HOA and GRG, which are potentially affected by both airport and road
345 traffic emissions (Figures SI5 and SI6).

346
347 Additional pairs were also selected to account the contributions of M4 and M25 motorways. Table 2
348 reports the selected site pairs and wind sectors. The final upper limit estimates of airport
349 contribution were thus obtained by computing the average concentrations and frequencies of
350 measurements in each wind speed/direction cell in the range as a proportion of the total number of
351 hourly measurements (Carslaw et al., 2006).

352
353 Since the deltas may be affected by the strength of the sources and the subsequent dispersion of
354 pollutants, the location of sampling sites and their closeness to the sources may play an important
355 role that cannot be disregarded in the emission assessment. For example, the dilution effect is
356 clearly evident from the polar plot of LHR2-OAK and HRL-OAK, which are computed over similar
357 wind sectors, but return very differing results. In order to isolate the signal of the source under
358 consideration, and thus reduce any interference due to other emission sources in the study area, a
359 further strategy was adopted: deltas were calculated over both directions, i.e. using the two sites
360 reciprocally as background or reference (both ΔX_{ji} and ΔX_{ij} are thus computed). This latter action
361 may also give important indications about the differences amongst sites: it is plausible to expect that

362 pairs of sites having comparable deltas in both directions are similarly affected by sources, while
363 pairs having very different delta values over the two directions indicate that one site is affected by
364 the sources much more than the other.

365

366 Results are also listed in Table 2. Generally, most of the pairs selected for assessing the airport
367 emission show significant increases in levels of nitrogen oxides and particulate matter and
368 decreases of ozone over both directions. In general, the upper limit contributions of NO₂ and NO_x
369 from the airport are slightly lower than those calculated by Carslaw et al. (2006) for the period
370 2001-2004, which is consistent with the drop of pollutants recorded from 2005 to 2012 over the
371 study area (Figures 4 and SI4).

372

373 For LHR2-OAK, which was originally chosen by Carslaw et al. (2006) as the best estimate for
374 airport emissions, results of this study apportion ~27-29% of nitrogen oxides to airport operations,
375 i.e. 12 µg m⁻³ (29%) of NO, 13.3 µg m⁻³ (25.9 %) of NO₂, 31.5 µg m⁻³ (27.6%) of NO_x, but a
376 relatively low contribution of particulate matter, i.e. 1.5 µg m⁻³ (5.5%) of PM₁₀ and 0.5 µg m⁻³
377 (4.7%) of PM_{2.5}. Beside those results, it can be noted that the airport operations are responsible for a
378 reduction of 6.1 µg m⁻³ (-18.6%) of ozone, but the total amount of oxidants is slightly increased
379 (OX +3.5 ppbv; 7.9%). However, the LHR2-OAK pair is the only pair having an opposite trend
380 over the two directions, clearly indicating that the influence of the airport emissions on LHR2 is
381 extremely high and it is not possible to view it as a background site. Because of this, upper limit
382 estimates having LHR2 as reference site are strongly affected by the location of the site, which is
383 very close both to the runway and to the North Perimeter Road and therefore may give interesting
384 information about the direct airport emissions, but cannot be used as indicative for the assessment
385 of airport emissions over the entire study area.

386

387 Airport emissions in remaining pairs account for an average of $1-9 \mu\text{g m}^{-3}$ of NO_2 , $2-20 \mu\text{g m}^{-3}$ of
388 NO_x , an average decrease of -2 to $-5 \mu\text{g m}^{-3}$ of O_3 (computed only for one pair), while particulate
389 matter changes are quite low and variable. Generally, results also show that the levels of all the
390 monitored pollutants decline rapidly with distance from the airport. On the other hand, upper limit
391 estimates for non-LHR2 pairs selected to be representative of the airport emissions resulted in more
392 comparable average levels over both directions. The increment in NO_x differs for a 180° change in
393 wind sector: there will be a number of reasons for this. Specifically, the wind speed and stability
394 may differ leading to differing dispersion characteristics on the two wind directions. Secondly, the
395 distribution of emissions within the airport is not homogeneous and the proximity of emission
396 sources to the airport boundaries closest to the sampling sites will have a major influence upon
397 measured concentrations.

398

399 The effect of selecting wind speeds $> 3 \text{ m s}^{-1}$ for deltas was also investigated by separately
400 computing ΔX for the pairs LHR-OAK (OAK-LHR) and GRG-OAK (OAK-GRG) over wind
401 speeds in the range of 0.5 to 3 m s^{-1} and including a 1 h lag (time difference) between the two sites
402 in a pair (Table SI2). Results indicate significantly lower airport contributions. The difference in
403 results can be explained by: (i) the effect of strong local sources, i.e. LHR2, GRG and OAK are all
404 located near busy roads and are strongly affected by non-airport sources of pollutants when wind
405 speed are low; (ii) the fluctuations in wind direction at low wind speeds causing a disconnection
406 between the sites. The results clearly indicate that the choice of selecting wind speeds $> 3 \text{ m s}^{-1}$
407 must be interpreted as the upper limit of airport contributions.

408

409 The assessment of the M4 motorway emissions resulted in very high values for most pollutants
410 when HIL was taken as reference (downwind) site. As with LHR2 for airport emissions, the results
411 are strongly affected by the location of the site, which is very close to the motorway and cannot be
412 used as indicative for the assessment of traffic emissions over the entire study area. However, upper

413 limit estimates are positive (except for ozone) in both directions, indicating that the traffic signal is
 414 high. Deltas for the SLC/GRG site pair indicative of the M25 motorway resulted in comparable
 415 distributions in both directions with the motorway emissions accounting for an average increase of
 416 $0.6\text{--}0.8\ \mu\text{g m}^{-3}$ of NO, $0\text{--}2.6\ \mu\text{g m}^{-3}$ of NO₂, $1\text{--}4\ \mu\text{g m}^{-3}$ of NO_x and $0.2\text{--}0.4\ \mu\text{g m}^{-3}$ of PM₁₀.
 417
 418 Despite the substantial variability of the data, the results expressed as ppbv indicate that upper limit
 419 delta values indicative of airport emissions for NO and NO₂ are quite similar, while estimates for
 420 vehicular traffic show higher values for NO than NO₂. Some of NO₂ is a product of the NO + O₃
 421 reaction. Such results can give some insights into the NO_x partitioning of the two sources. Several
 422 studies have reported that the majority of the NO_x emitted from modern turbofan engines at idle is
 423 in the form of NO₂, while NO is dominant in high power regimes (Song and Shon, 2012; Masiol
 424 and Harrison, 2014 and references therein). In addition, HAL (2011) estimated that the emissions
 425 from take-offs at Heathrow account for 46% of total emissions, while other sources are APU (19%),
 426 taxi-out (13%), hold (10%), taxi-in (8%), landing roll (3%) and engine testing (1%). While data on
 427 APU emissions are sparse, most of the non-takeoff flight phases and aircraft operations involve
 428 engines at low thrusts and therefore NO_x partitioning can be expected toward NO₂ for those sources.
 429 The small differences between the deltas of NO and NO₂ suggest that the airport-related emissions
 430 of NO_x are the result of different processes: it can be speculated that the takeoff provides most of
 431 the NO, while the other operational phases emit mainly NO₂. However, external or unaccounted
 432 sources may also have a role in the NO_x partitioning, as well as NO atmospheric oxidation. More
 433 information on this point can be derived from the data for OX, available only for the HRL/OAK site
 434 pair (excluding the heavily source-influenced LHR2 and HIL sites). An increase in OX on the
 435 distance scale of the airport is indicative of primary nitrogen dioxide emissions, as emission of NO_x
 436 purely as NO would give an OX increment of zero. The substantial increment in OX for OAK-HRL
 437 is consistent with appreciable emissions of primary NO₂. Hence, although take-offs are the main

438 source of NO_x, an appreciable contribution from other aircraft operational phases and other sources
439 seems likely.

440

441 Ozone concentrations in the study area appear to be determined by the upwind background and
442 local NO emissions, which cause a suppression of ozone. Although the area of the airport is an
443 appreciable source of NO_x and VOC emissions, any contribution to ozone formation is likely to
444 occur only at large downwind distances.

445

446 Data for PM₁₀ indicate that the motorways are a significant source of particulate matter (mainly for
447 HIL-GRG). Road dust resuspension may play a role in enhancing the levels of particulate matter
448 arising from the motorway source, as indicated by a large number of studies (e.g., Thorpe and
449 Harrison, 2008). In a similar manner, the resuspension of particles due to the turbulence created by
450 the aircraft movements may also be a significant source of particulate matter close to the airport, as
451 for example is demonstrated by the Gatwick Airport emission inventory (British Airports Authority,
452 2006). In summary, even if subject to large variability, the results obtained applying this method
453 demonstrate that both the LHR airport and the two motorways have a clear effect upon air quality
454 but neither appears strongly dominant over the other. The data do however suggest that the
455 influence of the airport is experienced over a greater geographic area.

456

457 **3.6 Hourly Contributions of Motorway Traffic and Airport Emissions**

458 Since all the air pollutants present characteristic diurnal and weekly patterns (Figure 3) which are
459 strongly influenced by local sources, a further investigation was conducted to determine whether the
460 contributions of traffic and airport emissions have different or covariant daily behaviours. The
461 diurnally averaged cycles of the differences between pairs of sites were thus re-computed. Results
462 are then investigated with airport and motorway traffic data (Figure SI1). As for the upper limit
463 estimation, only hours between 06:00 and 22:00 were taken in account because: (i) the contributions

464 of both airport and motorways at other hours was minor; (ii) no data on airport and motorway traffic
465 are available during nighttime, and there is no significant flight activity. Results are reported in
466 Figure 6 and show that on average the contributions of motorway traffic and airport operations have
467 different patterns. Generally, NO, NO₂ and NO_x estimated from site pairs indicative of airport
468 emissions show an often dominant evening peak on both wind directions, while paired sites for
469 vehicular traffic have higher morning peaks.

470

471 As similar mixing layer dynamics are expected over the entire study area due to the closeness of the
472 sites, and aircraft traffic schedules are normally constant from 6am to 8pm (Figure SI1), this result
473 indicates that the increased concentration of nitrogen oxides due to airport emissions are mainly
474 driven by the variation in atmospheric turbulence/stability and wind speed. On the other hand,
475 traffic mainly contributes to NO_x in the morning.

476

477 Ozone has the opposite behaviour relative to nitrogen oxides, further demonstrating the key role of
478 nitrogen oxides in ozone behaviour. PM₁₀ values generally show quite variable behaviour and some
479 pairs have different patterns over the two directions (e.g., LHR2-OAK, HRL-OAK, SLC GRG).

480 This result indicates that PM pollution is more sensitive to the local site characteristics than for the
481 gaseous pollutants and no further information can be extracted.

482

483 **CONCLUSIONS**

484 This study gives some indication of the impact of Heathrow Airport activities upon air quality.

485 However, the greatest difficulty in determining the contribution of the airport to local air pollution
486 is the presence of other major sources in the study area, i.e. the two motorways and other main
487 roads and the urban emissions of London. A series of tools has been therefore applied to analyse the
488 levels of pollutants with respect to the spatial distribution of sites around the airport and the wind
489 regimes. The main results for each monitored pollutant can be summarised as follows:

- 490 • nitrogen oxides deserve particular attention, mainly due to exceedence of the annual mean
491 Limit Value for NO₂ at some sites around Heathrow. However, the only local monitoring sites
492 that exceed the limit values for NO₂ are strongly influenced by busy roads (HOA from the A4
493 and HIL from the M4), or are on-airport (LHR2), where the limit values do not apply.
494 Nitrogen oxides present their highest concentrations in colder periods, and two different daily
495 peaks at all of the sites. Generally, LHR2 and HIL show the highest levels of nitrogen oxides
496 during the whole study period, but while the levels at LHR2 are decreasing slowly, the
497 concentrations of NO_x are increasing at HIL;
- 498 • Measurement of concentration differences (deltas) between a carefully selected downwind
499 and upwind site is an effective means of expressing the impact of the airport upon ambient air
500 quality;
- 501 • The results of the upper limit assessment study show that both road traffic and airport
502 emissions are responsible for marked increments upon nitrogen oxide levels: in particular the
503 peaks of concentration in the morning are the result of traffic, while the peaks in the late
504 evening are mainly due to the airport emissions;
- 505 • The increments upon nitrogen oxide levels recorded for the period 2005-2012 are similar or
506 slightly lower than those calculated for the period 2001-2004. The changes may reflect the
507 reduction in emissions which some pollutants underwent from 2005 to 2012;
- 508 • There is evidence for emissions of primary nitrogen dioxide within the airport, consistent with
509 jet engines operating at low thrust settings;
- 510 • ozone generally follows an opposite behaviour with respect to nitrogen oxides. This finding
511 reflects the key role of the photostationary state, and the rapid consumption of ozone by the
512 reaction with NO to form NO₂. Ozone levels are slowly increasing at most monitoring sites;
- 513 • particulate matter concentrations are always below the limit imposed by the EC, and the long-
514 term analysis reveals that their concentrations are declining further. However, a moderate
515 impact of road and flight traffic on PM₁₀ concentrations can be seen, deriving from exhaust

and non-exhaust emissions including the resuspension of road dust from both motorways and airport runways. PM_{2.5} seems not to be significantly affected by local sources.

ACKNOWLEDGEMENTS

We gratefully acknowledge: (i) the European Union for funding the Marie Curie Intra-European Fellowship for career development to M. Masiol through the project entitled ‘Chemical and Physical Properties and Source Apportionment of Airport Emissions in the context of European Air Quality Directives (Project CHEERS, call: FP7-PEOPLE-2012-IEF, proposal no. 328542); (ii) the UK Department for Transport, Road Traffic and Road Freight Statistics, for providing traffic data; (iii) Heathrow and Ricardo-AEA for supplying aircraft movement data and for the valuable exchange of information and discussion, in particular David Wovles, Katherine Rolfe, Elizabeth Hegarty (Heathrow) and Brian Stacey (Ricardo-AEA); (iv) DEFRA Automatic Urban and Rural Network, London Air Quality Network, Heathrow airport and Airwatch website for providing pollutant data; (v) Met Office and BADC for weather data.

530 REFERENCES

- 531 ACI, 2014. ACI Releases its 2013 World Airport Traffic Report. Media Release, Airports Council
532 International, Montreal. Available at: <http://www.aci.aero/News/Releases/Most-Recent/2014/09/16>.
533
- 534 AEA, 2010. Heathrow Airport Air Quality Modelling for 2008/9: Results and Model Evaluation.
535 Report by AEA Energy & Environment on behalf of BAA, July 2010. AEAT/ENV/R/2948/Issue 1.
536
- 537 Anttila, P., Tuovinen J.P., Niemi J.V., 2011. Primary NO₂ emissions and their role in the
538 development of NO₂ concentrations in a traffic environment. *Atmospheric Environment* 45, 986-
539 992.
540
- 541 Barnhart, C., Fearing, D., Odoni, A., Vaze, V., 2012. Demand and capacity management in air
542 transportation. *EURO Journal on Transportation and Logistics* 1, 135-155.
- 543 Bigi, A., Harrison, R.M., 2010. Analysis of the air pollution climate at a central urban background
544 site. *Atmospheric Environment*, 44, 2004-2012.
545
- 546 British Airports Authority, 2006. Gatwick 2010 Baseline Emission Inventory. Available at:
547 [http://83.98.24.64/Documents/business_and_community/Publications/2006/2010_baseline_emission](http://83.98.24.64/Documents/business_and_community/Publications/2006/2010_baseline_emissions_inventory.pdf)
548 [s_inventory.pdf](http://83.98.24.64/Documents/business_and_community/Publications/2006/2010_baseline_emissions_inventory.pdf) (last accessed September, 2013).
549
- 550 Carslaw, D.C., Beevers, S.D., Ropkins, K., Bell, M.C., 2006. Detecting and quantifying aircraft and
551 other on-airport contributions to ambient nitrogen oxides in the vicinity of a large international
552 airport. *Atmos. Environ.* 40, 5424–5434.
553
- 554 Carslaw, D.C., Beevers, S.D., Bell, M.C., 2007. Risks of exceeding the hourly EU limit value for
555 nitrogen dioxide resulting from increased road transport emissions of primary nitrogen dioxide.
556 *Atmospheric Environment* 41, 2073-2082.
557
- 558 Carslaw, D.C., Ropkins, K., Laxen, D., Moorcroft, S., Marner, B., Williams, M. L., 2008. Near-
559 field commercial aircraft contribution to nitrogen oxides by engine, aircraft type, and airline by
560 individual plume sampling. *Environmental Science & Technology* 42, 1871-1876.
561
- 562 Carslaw, D.C., Williams, M.L, Barratt, B., 2012. A short-term intervention study – Impact of
563 airport closure due to the eruption of Eyjafjallajökull on near-field air quality. *Atmospheric*
564 *Environment* 54, 328-336.
565
- 566 Carslaw, D.C., Ropkins, K., 2012. openair - an R package for air quality data analysis.
567 *Environmental Modelling and Software* 27-28, 52-61.
- 568 Carslaw, D.C., 2013. The openair manual — open-source tools for analysing air pollution data.
569 Manual for version 0.8-0, King's College London.
570
- 571 Colette, A., Granier, C., Hodnebrog, Ø., Jakobs, H., Maurizi, A., Nyiri, A., Bessagnet, B.,
572 D'Angiola, A., D'Isidoro, M., Gauss, M., Meleux, F., Memmesheimer, M., Mieville, A., Rouil, L.,
573 Russo, F., Solberg, S., Stordal, F., Tampieri, F., 2011. Air quality trends in Europe over the past
574 decade: a first multi-model assessment. *Atmospheric Chemistry & Physics* 11, 11657-11678.
575
- 576 Cyrus, J., Eeftens, M., Heinrich, J., Ampe, C., Armengaud, A., Beelen, R., Bellander, T.,
577 Beregszaszi, T., Birk, M., Cesaroni, G., Cirach, M., de Hoogh, K., De Nazelle, A., de Vocht, F.,
578 Declercq C., Dedele, A., Dimakopoulou, K., Eriksen, K., Galassi, C., Grauleviciene, R., Grivas, G.,
579 Gruzieva, O., Hagenbjörk Gustafsson, A., Hoffmann, B., Iakovides, M., Ineichen, A., Krämer, U.,

580 Lanki, T., Lozano, P., Madsen, C., Meliefste, K., Modig, L., Mölterm, A., Mosler, G.,
581 Nieuwenhuijsen, M., Nonnemacher, M., Oldenwening, M., Peters, A., Pontet, S., Probst-Hensch,
582 N., Quass, U., Raaschou-Nielsen, O., Ranzi, A., Sugiri, D., Stephanou, E.G., Taimisto, P., Tsai, M.-
583 Y., Vaskövi, E., Villani, S., Wang, M., Brunekreef, B., Hoek, G., 2012. Variation of NO₂ and NO_x
584 concentrations between and within 36 European study areas: Results from the ESCAPE study.
585 *Atmospheric Environment* 62, 374–390.
586

587 DEFRA, 2013a. Air Pollution in the UK 2012. UK Department for Environment, Food and Rural
588 Affairs. Issue of September 2013. Available at: [http://uk-](http://uk-air.defra.gov.uk/library/annualreport/air_pollution_uk_2012_issue_1.pdf)
589 [air.defra.gov.uk/library/annualreport/air_pollution_uk_2012_issue_1.pdf](http://uk-air.defra.gov.uk/library/annualreport/air_pollution_uk_2012_issue_1.pdf) (last accessed: November
590 2013).
591

592 DEFRA, 2013b. Emissions of Air Quality Pollutants 1970 – 2011. UK Department for
593 Environment, Food and Rural Affairs. AQPI Summary Report. Available at: [http://uk-](http://uk-air.defra.gov.uk/reports/cat07/1305031312_EoAQP1970-2011_pq.pdf)
594 [air.defra.gov.uk/reports/cat07/1305031312_EoAQP1970-2011_pq.pdf](http://uk-air.defra.gov.uk/reports/cat07/1305031312_EoAQP1970-2011_pq.pdf) (last accessed: January
595 2014).
596

597 Dockery, D.W. (2009). Health effects of particulate air pollution. *Annals of Epidemiology* 19, 257-
598 263.
599

600 EEA, 2014. AirBase—The European Air Quality Database. European Environment Agency.
601 Available from: <http://www.eea.europa.eu/themes/air/airbaseS> (last access: July 28, 2014).
602

603 Gelhausen, M.C., Berster, P., Wilken D., 2011. Do airport capacity constraints have a serious
604 impact on the future development of air traffic? *Journal of Air Transport Management* 28, 3-13.
605

606 Grice, S., Stedman, J., Kent, A., Hobson, M., Norris, J., Abbott, J., Cooke S., 2009. Recent trends
607 and projections of primary NO₂ emissions in Europe. *Atmospheric Environment* 43, 2154-2167.
608

609 HAL, 2011. Heathrow Air Quality Strategy 2011–2020. Heathrow Airport Ltd. Available at:
610 http://www.heathrowairport.com/static/Heathrow/Downloads/PDF/air-quality-strategy_LHR.pdf
611 (last accessed: August 2013).
612

613 Katsouyanni, K., Samet, J.M., Anderson, H.R., Atkinson, R., Le Tertre, A., Medina, S., Samoli, E.,
614 Touloumi, G., Burnett, R.T., Krewski, D., Ramsay, T., Dominici, F., Peng, R.D., Schwartz, J.,
615 Zanobetti A., 2009. Air pollution and health: a European and North American approach
616 (APHENA). Research Report, Health Effects Institute 142, 5-90.
617

618 Kley, D., Kleinmann, M., Sanderman, H., Krupa, S., 1999. Photochemical oxidants: state of the
619 science. *Environmental Pollution* 100, 19-42.

620 Masiol, M., Harrison, R.M., 2014. Aircraft engine exhaust emissions and other airport-related
621 contributions to ambient air pollution: A review. *Atmos. Environ.* 95, 409-455.
622

623 Mavroidis, I., Chaloulakou, A., 2011. Long-term trends of primary and secondary NO₂ production
624 in the Athens area. Variation of the NO₂/NO_x ratio. *Atmospheric Environment* 45, 6872-6879.
625

626 Notario, A., Bravo, I., Adame, J. A., Díaz-de-Mera, Y., Aranda, A., Rodríguez, A., Rodríguez, D.,
627 2012. Analysis of NO, NO₂, NO_x, O₃ and oxidant (OX= O₃+ NO₂) levels measured in a
628 metropolitan area in the southwest of Iberian Peninsula. *Atmospheric Research* 104, 217-226.
629
630

631 Paoletti, E., De Marco, A., Beddows, D.C.S., Harrison, R.M. and Manning, W.J., 2014.
 632 Ozone levels in European and USA cities are increasing more than at rural sites, while peak values
 633 are decreasing. *Environmental Pollution* 192, 295-299.
 634
 635 R Core Team, 2013. R: A language and environment for statistical computing. R Foundation for
 636 Statistical Computing, Vienna, Austria. URL <http://www.R-project.org/>.
 637
 638 Raaschou-Nielsen, O., Andersen, Z.J., Beelen, R., Samoli, E., Stafoggia, M., Weinmayr, G.,
 639 Hoffmann, B., Fischer, P., Nieuwenhuijsen, M.J., Brunekreef, B., Xun, W.W., Katsouyanni, K.,
 640 Dimakopoulou, K., Sommar, J., Forsberg, B., Modig, L., Oudin, A., Oftedal, B., Schwarze, P.E.,
 641 Nafstad, P., De Faire, U., Pedersen, N.L., Östenson, C.-G., Fratiglioni, L., Penell, J., Korek, M.,
 642 Pershagen, G., Eriksen, K.T., Sørensen, M., Tjønneland, A., Ellermann, T., Eeftens, M., Peeters,
 643 P.H., Meliefste, K., Wang, M., Bueno-de-Mesquita, B., Key, T.J., de Hoogh, K., Concin, H., Nagel,
 644 G., Vilier, A., Grioni, S., Krogh, V., Tsai, M.-Y., Ricceri, F., Sacerdote, C., Galassi, C., Migliore,
 645 E., Ranzi, A., Cesaroni, G., Badaloni, C., Forastiere, F., Tamayo, I., Amiano, P., Dorronsoro, M.,
 646 Trichopoulou, A., Bamia, C., Vineis, P., G. Hoek, 2013. Air pollution and lung cancer incidence in
 647 17 European cohorts: prospective analyses from the European Study of Cohorts for Air Pollution
 648 Effects (ESCAPE). *The Lancet Oncology*, doi:10.1016/S1470-2045(13)70279-1.
 649
 649 Sen, P.K., 1968. Estimates of the regression coefficient based on Kendall's tau. *The Journal of the*
 650 *American Statistical Association* 63, 1379-1389.
 651
 652 Song, S.-K., Shon, Z.-H., 2012. Emissions of greenhouse gases and air pollutants from commercial
 653 aircraft at international airports in Korea. *Atmospheric Environment* 61, 148-158.
 654
 655 Stettler, M.E.J., Eastham, S., Barrett, S.R.H., 2011. Air quality and public health impacts of UK
 656 airports. Part I: emissions. *Atmospheric Environment* 45, 5415–5424.
 657
 658 Theil, H., 1992. A rank-invariant method of linear and polynomial regression analysis. In Henri
 659 Theil's Contributions to Economics and Econometrics. Springer, Netherlands, 345-381.
 660
 661 Thorpe, A., Harrison, R.M., 2008. Sources and properties of non-exhaust particulate matter from
 662 road traffic: A review. *Science of the Total Environment* 400, 270-282.
 663
 664 UK Department for Transport, 2006. Project for the Sustainable Development of Heathrow: Report
 665 of the Airport Air Quality Technical Panels.
 666
 667 Vestreng, V., Ntziachristos, L., Semb, A., Reis, S., Isaksen, I.S.A., Tarrasón, L., 2009. Evolution of
 668 NO_x emissions in Europe with focus on road transport control measures. *Atmospheric Chemistry &*
 669 *Physics* 9, 1503-1520.
 670
 671 WHO, 2000. Air Quality Guidelines for Europe. European Series No 91, World Health
 672 Organization. WHO Regional Publications, Geneva.
 673
 674 Wormhoudt, J., Herndon, S.C., Yelvington, P.E., Lye-Miake, R.C., Wey, C., 2007. Nitrogen oxide
 675 (NO/NO₂/HONO) emissions measurements in aircraft exhausts. *Journal of Propulsion & Power* 23,
 676 906-911.
 677
 678 Yim, S.H.L., Stettler, M.E.J., Barrett, S.R.H., 2013. Air quality and public health impacts of UK
 679 airports. Part II: Impacts and policy assessment. *Atmospheric Environment* 67, 184-192.
 680

681 Zamboni, G., Capobianco, M., Daminelli, E., 2009. Estimation of road vehicle exhaust emissions
682 from 1992 to 2010 and comparison with air quality measurements in Genoa, Italy. Atmospheric
683 Environment 43, 1086-1092.
684

685 **TABLE LEGENDS**

686

687 **Table 1.** Site characteristics: site name and acronym, geographic coordinates (decimal
688 degrees, WGS 84 system), site categorization (if available) and analyzed gaseous
689 pollutants. Periods of data availability are given in brackets.
690

691 **Table 2.** Site pairs used in bivariate polar plot analysis with background subtraction and
692 quantification of upper limit for source contributions following the method proposed
693 by Carslaw et al. (2006). Data were filtered for hour of day (6:00-22:00) and for
694 wind speeds $> 3 \text{ m s}^{-1}$.
695

696

697 **FIGURE LEGENDS**

698

699 **Figure 1.** Map of the study area showing the sampling sites.
700

701 **Figure 2.** Time series of monthly average concentrations of measured air pollutants. Only
702 months with more than 75% of available data are included. Note that $\text{PM}_{2.5}$ data are
703 measured with TEOM (LHR2, GRG, OAK) and TEOM-FDMS (HRL, HAR, LNK).

704 **Figure 3.** Weekly and hourly-resolved averages calculated over 8 years. Data are corrected for
705 DST. Note that $\text{PM}_{2.5}$ data are measured with TEOM (LHR2, GRG, OAK) and
706 TEOM-FDMS (HRL, HAR, LNK).
707

708 **Figure 4.** Long-term trends of analysed pollutants computed from the monthly averages.

709 **Figure 5.** Bivariate polar plots for selected sites with background concentrations subtracted.
710 All values are expressed as $\mu\text{g m}^{-3}$, except OX (ppbv).
711

712 **Figure 6.** Daily patterns computed from the differences between pairs of sites (reference site -
713 background site) shown in Table 2. Data were filtered for hour of day (6:00-22:00)
714 and for wind speeds $> 3 \text{ m s}^{-1}$.

715 **Table 1.** Site characteristics: site name and acronym, geographic coordinates (decimal degrees, WGS 84 system), site categorization (if available) and
716 analyzed gaseous pollutants. Periods of data availability are given in brackets.
717

Site	Lat.; Long.	Categorization	Analyzed compounds (periods)
Harwell (HAR)	51.571078, -1.325283	Rural background	NO, NO ₂ , NO _x , O ₃ , SO ₂ , PM ₁₀ , PM _{2.5} (2005-2013)
London N. Kensington (LNK)	51.521050, -0.213492	Urban background	NO, NO ₂ , NO _x , CO, O ₃ , SO ₂ , PM ₁₀ (2005-2013); PM _{2.5} (from Dec 2008)
Heathrow LHR2 (LHR2)	51.479268, -0.440556	Airport	NO, NO ₂ , NO _x , PM ₁₀ , (2005-2013); PM _{2.5} (from Feb 2010); CO, O ₃ (until Apr 2007)
London Harlington (HRL)	51.488790, -0.441614	Urban Industrial	NO, NO ₂ , NO _x , O ₃ , PM ₁₀ (2005-2013); PM _{2.5} ; (from Apr 2008); CO (until Mar 2008)
London Hillingdon (HIL)	51.496330, -0.460861	Urban background	NO, NO ₂ , NO _x , O ₃ (2005-2013); PM ₁₀ , SO ₂ , CO, (until Sep 2007)
Heathrow Green Gates (GRG)	51.481478, -0.486675	—	NO, NO ₂ , NO _x , PM ₁₀ (2005-2013); PM _{2.5} (2005-Mar 2006 and Nov 2007-2013)
Slough Colnbrook (SLC)	51.480372, -0.508729	Urban background	NO, NO ₂ , NO _x , PM ₁₀ (2005-2013)
Heathrow Oaks Road (OAK)	51.459577, -0.479445	Urban background	NO, NO ₂ , NO _x , PM ₁₀ , PM _{2.5} (2005-2013); O ₃ (until Jul 2007)
Hounslow Hatton Cross (HAT)	51.463319, -0.427225	Roadside (10 m)	NO, NO ₂ , NO _x , PM ₁₀ (2005-2013)
London Hillingdon Oxford Avenue (HOA)	51.481130, -0.423760	Urban centre	NO, NO ₂ , NO _x , PM ₁₀ (2005-2013)

Table 2. Site pairs used in bivariate polar plot analysis with background subtraction and quantification of upper limit for source contributions following the method proposed by Carslaw et al. (2006). Data were filtered for hour of day (6:00-22:00) and for wind speeds > 3 m s⁻¹.

Source	Site pairs	Wind sector ^a	NO ₂		NO _x		NO	O ₃ ^b	OX	PM ₁₀ ^c	PM _{2.5} ^d
			2001-2004 ^e	2005-2012	2001-2004 ^e	2005-2012	2005-2012	2005-2012	2005-2012	2005-2012	2005-2012
		degree	µg m ⁻³ (%)	µg m ⁻³ [ppbv] (%)	µg m ⁻³ (%)	µg m ⁻³ [ppbv] (%)	µg m ⁻³ [ppbv] (%)	µg m ⁻³ [ppbv] (%)	ppbv (%)	µg m ⁻³ (%)	µg m ⁻³ (%)
Airport	LHR-OAK	150-260	15 (27.3%)	13.3 [7] (25.9%)	33.9 (26.7%)	31.5 [16.5] (27.6%)	12 [9.6] (29%)	-6.1 [-3.1] (-18.6%)	3.5 (7.9%)	1.5 (5.5%)	0.5 (4.7%)
	OAK-LHR	340-80	—	-0.3 [-0.2] (-0.8%)	—	-5.8 [-3] (-9.5%)	-3.6 [-2.9] (-20.5%)	0.1 [0.1] (0.2%)	-0.1 (-0.2%)	-0.7 (-3.2%)	-0.1 (-0.7%)
	HRL-OAK ^f	160-260	6.6 (17.4%)	5.3 [2.8] (14.9%)	9.9 (14%)	8.2 [4.3] (12.6%)	1.9 [1.5] (9.8%)	-4.7 [-2.4] (-13.7%)	0.4 (1%)	-1.2 (-5.7%)	— ^h
	OAK-HRL	340-80	—	3.6 [1.9] (10.4%)	—	6.8 [3.6] (11.1%)	2.1 [1.7] (11.9%)	-1.9 [-1] (-4.8%)	0.8 (2.1%)	0.5 (2.1%)	— ^h
	HOA-OAK ^g	200-260	6.5 (18.1%)	9.2 [4.8] (21.5%)	9.5 (12%)	19.7 [10.3] (23.4%)	6.9 [5.5] (25%)	—	—	0.7 (3.3%)	—
	OAK-HOA ^g	340-80	2 (5.9%)	3.8 [2] (11.1%)	5.9 (8.9%)	7.4 [3.9] (12%)	2.3 [1.8] (12.9%)	—	—	0.6 (2.6%)	—
	GRG-OAK	100-170	1.5 (3.9%)	1.2 [0.6] (3.3%)	3 (4%)	1.9 [1] (2.8%)	0.5 [0.4] (2.3%)	—	—	-0.1 (-0.4%)	0 (0%)
	OAK-GRG	340-80	—	3.2 [1.7] (9.2%)	—	6.4 [3.3] (10.4%)	2.1 [1.7] (12%)	—	—	0.7 (3%)	0.2 (2%)
	SLC-OAK ⁱ	100-170	1.5 (4.2%)	1.2 [0.6] (3.9%)	1.8 (2.6%)	2.6 [1.4] (4.4%)	0.9 [0.7] (5%)	—	—	0.1 (0.4%)	—
	OAK-SLC	350-80	—	2.9 [1.5] (8.3%)	—	5.7 [3] (9.3%)	1.9 [1.5] (10.6%)	—	—	0.5 (2.1%)	—
	GRG-HAT	100-200	—	2.8 [1.5] (7.4%)	—	4.5 [2.4] (6.6%)	0.8 [0.6] (4%)	—	—	0.5 (2.4%)	—
	HAT-GRG	260-30	—	3.5 [1.8] (9.4%)	—	9 [4.7] (13.6%)	4.1 [3.3] (18.4%)	—	—	0.5 (2.3%)	—
M4	HIL-HRL	100-260	—	16.4 [8.6] (32%)	—	47 [24.6] (42%)	20.1 [16.1] (50.4%)	-8.3 [-4.2] (-30.4%)	4.4 (11%)	4.1 (14.7%)	—
	HRL-HIL	280-80	—	1.2 [0.6] (3.4%)	—	2.6 [1.4] (4%)	0.9 [0.7] (4.6%)	-1.1 [-0.6] (-3.2%)	0.1 (0.4%)	1.2 (5.6%)	—
	HIL-GRG	100-260	—	16.9 [8.8] (32.9%)	—	46.5 [24.3] (41.5%)	19.3 [15.5] (48.7%)	—	—	3.3 (12%)	—
	GRG-HIL	340-70	—	1.1 [0.6] (3%)	—	1.7 [0.9] (2.5%)	0.4 [0.3] (2%)	—	—	0.4 (1.9%)	—
M25	SLC-GRG	30-180	—	0 [0] (0%)	—	1 [0.5] (1.7%)	0.6 [0.5] (3.5%)	—	—	0.4 (1.9%)	—
	GRG-SLC	240-340	—	2.6 [1.4] (7.1%)	—	3.9 [2] (5.7%)	0.8 [0.6] (3.9%)	—	—	0.2 (0.9%)	—

a) Selected wind sectors were kept identical to those used in Carslaw et al. (2006), whereas wind sectors for new pairs of sites were selected on the basis of polar plot analysis. b) O₃ was measured until ca. mid-2007 in LHR2 and OAK; c) PM₁₀ was measured until mid-2007 in HIL. d) PM_{2.5} measurements in LHR started in 2010. e) Data from Carslaw et al. (2006); f) HRL-OAK in Carslaw et al. referred to 2001 only. g) Hounslow was used in Carslaw et al. whereas Hillingdon Oxford Avenue (HOA) was used in this study. h) Stations are equipped with differing instruments and a cross-comparison is not possible. i) SLC-OAK was used in Carslaw et al. for quantifying the airport emission, but is also potentially affected by M25 motorway emissions.

Note: The percentage values in parentheses express the source contribution as a percentage of the average concentration at the reference (upwind) site.

Figure 1. Map of the study area showing the sampling sites.

Figure 2. Time series of monthly average concentrations of measured air pollutants. Only months with more than 75% of available data are included. Note that $\text{PM}_{2.5}$ data are measured with TEOM (LHR2, GRG, OAK) and TEOM-FDMS (HRL, HAR, LNK).

Figure 3. Weekly and hourly-resolved averages calculated over 8 years. Data are corrected for DST. Note that PM_{2.5} data are measured with TEOM (LHR2, GRG, OAK) and TEOM-FDMS (HRL, HAR, LNK).

Figure 4. Long-term trends of analysed pollutants computed from the monthly averages.

Figure 5. Bivariate polar plots for selected sites with background concentrations subtracted. All values are expressed as $\mu\text{g m}^{-3}$, except OX (ppbv). The location of the airfield is highlighted with dashed arcs, while the location of motorways with dotted arcs.

Figure 6. Daily patterns computed from the differences between pairs of sites (reference site - background site) shown in Table 2. Data were filtered for hour of day (6:00-22:00) and for wind speeds $> 3 \text{ m s}^{-1}$.