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# Evaluation of ultrafine particle concentrations and size distributions at London Heathrow Airport

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6	<b>Evaluation of Ultrafine Particle Concentrations</b>
7	and Size Distributions at London Heathrow
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8	Airport
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## 20 ABSTRACT

A study to monitor UFP at Heathrow Airport was undertaken in the autumn of 2016. The objective 21 was to assess the context of measurements at the airport compared to measurements at "typical" 22 23 traffic, background and rural locations in the south east of England. Measurements were made at two airport locations (called LHR2 and Oaks Road) at opposite ends of the airfield, to further 24 understand the contribution of the airport to local air quality. Average concentrations showed that 25 total particle number concentrations at the airport are typically lower than a traffic location and 26 higher than an urban background location in London, matching the trends seen for NO<sub>x</sub>, PM<sub>10</sub>, 27 PM<sub>2.5</sub> and BC pollutants. However, the size distribution of the submicrometre particles at the airport 28 29 is completely different to the London monitoring stations, with the airport PSD dominated by particles with a mode of 20nm. In contrast, measurements of PN in London have a significantly 30 larger mode of 30nm. This study demonstrated that measurements of particle number from within 31 the airport perimeter are dominated by the smallest particles and are closely associated with aircraft. 32 Analysis of the operating modes at the airport showed that aircraft departing from the airport emit 33 34 particles in much higher numbers than those arriving. Nucleation mode particles are commonly associated with emissions from combustion processes. However, measurement of these particles at 35 the airport are not strongly correlated with Black Carbon. There does appear to be some 36 37 correlation of nucleation mode particles with UV active BC particles (brown carbon, typically associated with biomass combustion or wood smoke) at the Heathrow airside monitoring station, 38 LHR2. There is also modest association between nucleation mode particles and NO<sub>2</sub>. The study 39 showed that the classical air pollutants measured at Heathrow are very similar in concentration to 40 typical urban environments in London and south east England, but particle numbers in the sub 41 42 30nm size range are markedly different to those measured in London.

43 44

## 46 1. INTRODUCTION

47 Heathrow Airport is the busiest two-runway airport in the world. In 2016, the airport handled over
48 75.7 million passengers and approximately 470,000 aircraft movements

49 (https://www.heathrow.com/file\_source/Company/Static/PDF/Investorcentre/Heathrow-(SP)-

- 50 FY2016-results-release-(FINAL).pdf).
- 51

Heathrow Airport has undertaken automatic monitoring of air quality continuously since 1992. In
2016, there were 4 locations around the airport. These focus on classical air pollutants; NO, NO<sub>2</sub>,
PM<sub>10</sub> and PM<sub>2.5</sub>. Black Carbon is measured at 2 of the 4 locations, while O<sub>3</sub> and meteorological
measurements are each measured at one station. This data and background information is available
to view and download at http://heathrowairwatch.org.uk.

57

The literature review by Stacey (2019) collects the most relevant literature into a single document and informs the direction of research and analysis throughout this paper. Evidence, for example from Fanning et al. (2007), Fleuti et al. (2017), Hudda and Fruin (2016), Masiol et al. (2017) and Peters et al. (2016), identifies that concentrations of ultrafine particles (UFP) close to airports are substantially different to UFP concentrations in more conventional urban environments.

63

As of the end of 2015, the literature was incomplete in the identification, assessment and nature of the ambient concentrations of UFP emitted from airports and aircraft. Research conducted by, for example, Donaldson et al. (2001) and Health Effects Institute (2013) has identified links between exposure to UFP and detrimental health impact and specifically argue that the smallest particles are likely to carry the highest risk to adverse health outcome. These health impact studies focussed primarily on particles emitted from road transport and energy use into the ambient environment – few studies had been conducted on the health impact of exposure to UFP around airports.

71	Ellermann et al. (2011) undertook research at Copenhagen Airport to assess exposure of airport
72	workers, but no associations with health impacts were presented in that report.

74	Studies undertaken by, for example Durdina et al. (2014), Lobo et al. (2015), Abegglen et al.
75	(2016), Turgut et al. (2015), and Vander Wal et al. (2016), measured emissions directly from the
76	exhaust of aircraft. These largely focussed on non-volatile particles and showed that, generally,
77	these particles are mostly carbon based and not significantly different in composition to other
78	combustion sources.
79	
80	As the exhaust plume emerges from the engine and interacts with the atmosphere, combustion
81	products cool and can condense and/or interact with other components to form secondary aerosols.
82	A study by Beyersdorf et al. (2014), looked at volatile and non-volatile UFP exhaust emissions with
83	increasing distance from the source and found that as the exhaust plume cools and evolves, large
84	quantities of very fine particles are detected.
85	
86	Studies of ambient concentrations by, for example Ellermann et al. (2012), Fanning et al. (2007),
86 87	Studies of ambient concentrations by, for example Ellermann et al. (2012), Fanning et al. (2007), Hudda and Fruin (2016), Westerdahl et al. (2008), Peters et al. (2016), Keuken et al. (2015),
87	Hudda and Fruin (2016), Westerdahl et al. (2008), Peters et al. (2016), Keuken et al. (2015),
87 88	Hudda and Fruin (2016), Westerdahl et al. (2008), Peters et al. (2016), Keuken et al. (2015), Bezemer et al. (2015), Riley et al. (2016), Fleuti et al. (2017), and Shirmohammadi et al. (2017),
87 88 89	Hudda and Fruin (2016), Westerdahl et al. (2008), Peters et al. (2016), Keuken et al. (2015), Bezemer et al. (2015), Riley et al. (2016), Fleuti et al. (2017), and Shirmohammadi et al. (2017),
87 88 89 90	Hudda and Fruin (2016), Westerdahl et al. (2008), Peters et al. (2016), Keuken et al. (2015), Bezemer et al. (2015), Riley et al. (2016), Fleuti et al. (2017), and Shirmohammadi et al. (2017), have all shown that high PN concentrations can be seen close to airports.
87 88 89 90 91	<ul><li>Hudda and Fruin (2016), Westerdahl et al. (2008), Peters et al. (2016), Keuken et al. (2015),</li><li>Bezemer et al. (2015), Riley et al. (2016), Fleuti et al. (2017), and Shirmohammadi et al. (2017),</li><li>have all shown that high PN concentrations can be seen close to airports.</li><li>Masiol et al. (2017) undertook a pair of studies at Harlington, 1 km north of the airport in 2014 and</li></ul>
87 88 89 90 91 92	<ul> <li>Hudda and Fruin (2016), Westerdahl et al. (2008), Peters et al. (2016), Keuken et al. (2015),</li> <li>Bezemer et al. (2015), Riley et al. (2016), Fleuti et al. (2017), and Shirmohammadi et al. (2017),</li> <li>have all shown that high PN concentrations can be seen close to airports.</li> <li>Masiol et al. (2017) undertook a pair of studies at Harlington, 1 km north of the airport in 2014 and</li> <li>2015, which further confirmed that emissions of UFP from airports are different in size distribution</li> </ul>
87 88 90 91 92 93	<ul> <li>Hudda and Fruin (2016), Westerdahl et al. (2008), Peters et al. (2016), Keuken et al. (2015),</li> <li>Bezemer et al. (2015), Riley et al. (2016), Fleuti et al. (2017), and Shirmohammadi et al. (2017),</li> <li>have all shown that high PN concentrations can be seen close to airports.</li> <li>Masiol et al. (2017) undertook a pair of studies at Harlington, 1 km north of the airport in 2014 and 2015, which further confirmed that emissions of UFP from airports are different in size distribution to typical urban and road traffic environments. The studies led by Hudda, Peters, Keuken and</li> </ul>

96 detect airport related UFP emissions 18 km from Los Angeles International Airport, LAX , while

97 the Keuken et al. (2015) research detected airport related UFP over 40 km from Amsterdam Airport98 Schiphol .

99

106	2. METHODS
105	
104	airport.
103	September and November 2016, to determine UFP concentrations and size distributions near the
102	warranted. This paper presents the results from the measurement campaign, undertaken between
101	UFP measurements, and in particular the particle size distribution of aircraft emissions was
100	Assessment of the research by Masiol and others made it clear that further robust investigation of

## 107 **2.1 Monitoring Locations**

This measurement campaign was designed to compare measurements of UFP at Heathrow against
measurements made at other measurement stations in the South East of the UK, and then to further
explore the local nature of UFP at the airport.

111 The network of air quality monitoring stations at Heathrow Airport is presented in Figure 1:

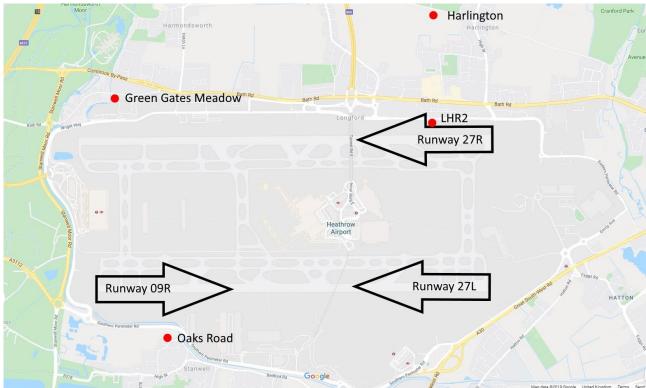




Figure 1. Locations of Heathrow monitoring stations. Runway 27R, Runway 27L and Runway 114 09R denote the three operating modes of the airport, indicating here the runway assigned for 115 departing aircraft. Note that aircraft never depart in an easterly direction on the northern runway. 116 117 118 Because of the dominant south-westerly nature of the winds in the UK, two of the four locations are ideally suited to explore the contribution of the airport to local air quality; Oaks Road and LHR2. 119 120

121 **Oaks Road** – Located in a residential area to the south west of the airport, approximately 600m

from the southern runway. It has been in continuous operation since 2001, measuring PM<sub>10</sub>, PM<sub>2.5</sub>, 122

BC and NOx. 123

124

- LHR2 located airside in the north eastern corner of the airfield, 170m from the northern runway 125
- and less than 20m from the northern perimeter road. It has operated continuously since 1993, 126

127 measuring PM<sub>10</sub>, PM<sub>2.5</sub>, BC, NOx and meteorology.

128

129

#### **2.2 UFP Measurement Campaign** 130

Measurement of UFP at the LHR2 and Oaks Road monitoring stations was undertaken between 30<sup>th</sup>
September and 25<sup>th</sup> November 2016.

133

134 The following equipment was used:

135	•	Butanol based TSI Model 3775 CPCs (TSI inc., MN, USA) to count particle numbers.
136	•	At Oaks Road, TSI Model 3080 with long DMA (Model 3081) classifier and soft X-ray
137		neutraliser. Automatic on-board software correction was enabled for diffusive losses and
138		multiple charge. Analyser operation and data storage was managed on a laptop running AIM
139		v9.0.0.0, which was used to control the operation of the TSI Model 3080/ Model 3775 setup.
140	•	At LHR2, TSI Model 3082 with long DMA (Model 3081) classifier and soft X-ray
141		neutraliser. Automatic on-board software correction was enabled for diffusive losses and
142		multiple charge. Analyser operation and data storage controlled by the Model 3082 running
143		AIM v10.1.0.6. Data was downloaded weekly from the 3082 to a USB stick for subsequent
144		analysis.
145		The operating methodology of the TSI Scanning Mobility Particle Sizer (SMPS) and
146		Condensation Particle Counter (CPC) has been extensively described in literature, for example
147		by Wiedensohler et al. (2012) and Wiedensohler et al. (2018). The only difference from the
148		recommendations of Wiedensohler et al. (2012) was the absence of a dryer.
149		

Both SMPS instruments were configured to sample in the range 14.6nm to 661.2nm. Sampling was
programmed to run for 3 minutes, sweeping up in size for 2 minutes 15 seconds, and returning
down for the remaining 45 seconds.

153

Both instruments were set up to be operated continuously for the entire measurement campaign; unattended automated operation 24 hours per day. Because of the proprietary nature of the TSI software and only a short window of opportunity to deploy the analysers, it was not possible to

establish remote communication to the analysers. The monitoring stations were visited weekly toensure correct operation and take remedial action if required.

159

The Heathrow CPCs and SMPSs were calibrated before and after the monitoring campaign at the
ISO/IEC17025 accredited Ricardo Energy and Environment (REE) calibration laboratory in
Harwell, UK. The classifiers and CPC were calibrated using a Jing miniCAST model 6003 (Jing
Ltd, Zollikofen, Switzerland) soot generator, which creates particles using a controlled burn
propane flame. The results of these calibrations showed both CPCs were accurate to within 1% of
the reference device and the SMPSs were able to size particles within 1 size bin in the range 14.6nm
to 680nm.

167

## 168 **2.3 Differences between Heathrow and National Monitoring UFP analyser setup**

169 The configuration of the Heathrow analysers matched, as far as possible, the configurations used in

170 the UK Particle Number monitoring network (<u>https://uk-air.defra.gov.uk/interactive-map</u>). This

171 network is managed by Kings' College London, while operation and QA/QC is provided by the

172 National Physical Laboratory. The national network stations use the following equipment:

- Butanol based TSI 3775 Condensation Particle Counters (CPC)
- TSI 3080 Scanning Mobility Particle Sizer (SMPS) with long DMA classifier and Kr-85
   neutraliser source

176 • Nafion dryer

177 • Laptop running AIM v9.0.0.0

The SMPS / CPCs in the national network are also configured to sample in the range 14.6nm to
661.2nm. Sampling is also programmed to run for 3 minutes, sweeping up in size for 2 minutes 15
seconds, and returning down for the remaining 45 seconds. This will allow measurements between
the airport and national network analysers to be directly and robustly compared with each other.

182 The authors believe that this is the first time such a robust concurrent comparison has been made

183 between UFP measured at airports and background locations.

184

185 In 2016, the 3 national network stations measuring UFP were located at:

London Marylebone Road – a heavily trafficked roadside location, near Baker Street in the
 centre of London.

- London North Kensington located in a school in a residential area of West London, less
   than 4km west of the Marylebone Road station.
- Chilbolton Observatory located in a rural environment, 25km north of the centre of
   Southampton, 78km WSW of Heathrow Airport and 95km from Marylebone Road.
- 192

193 There were two main differences between measurements made at Heathrow and those made in the194 national network:

A nafion dryer is used in the national network station analysers. As noted above, no drying
 was installed in line for the Heathrow study. It was considered unnecessary: studies (e.g.
 Stanier et al., 2004) have shown that relative humidity contributes little to increased particle
 size even for hygroscopic particles smaller than 50nm.

Because of transportation restrictions inside the airport, a radioactive source was impossible 199 • to deploy. Comparison studies within CEN TC264 WG32 and ISO/TC24/SC4/WG12 200 (standards in development), show that the measurement differences between particles 201 neutralised with soft X-rays and those neutralised with beta radiation are negligible, adding 202 further confidence that the X-ray and beta radiation neutralisers behave in a reasonably 203 similar manner. Additionally, the calibration of the Heathrow analysers at REE was 204 undertaken using a Kr-85 neutraliser for the reference device. The close agreement of the 205 Heathrow analysers, for both counting and sizing, reinforces the confidence that field 206 207 measurements are valid.

209	2.4 Data Analysis				
210	The plots and analysis undertaken in this paper make extensive use of the R and R Studio programs				
211	(R Foundation for Statistical Computing, Vienna, Austria, and R Studio Inc, MA, USA) and the				
212	OpenAir suite of analysis tools (Carslaw and Ropkins, (2012))				
213					
214	Unless specifically stated, particle number plots are aggregated into three size groups, defined here				
215	as:				
216	• Nucleation (particles smaller than 25nm),				
217	• Aitken (particles between 26 and 100nm)				
218	• Accumulation (particles larger than 100nm)				
219					
220	Measurements from the black carbon aethalometers are reported here from two of the seven				
221	components:				
222	• Black Carbon (BC) – the Particulate Matter concentration recorded from the attenuation of				
223	light by particles in the infra-red spectrum at 880nm				
224	• Ultra Violet Particulate Matter (UVPM) – defined here as the additional particulate matter				
225	concentration recorded from the attenuation in the UV region of the spectrum. It is calculated				
226	from the difference between the concentration recorded at 370nm and the concentration				
227	recorded at 880nm using a wavelength-adjusted absorption coefficient. Some other studies				
228	have referred to this variable as "Brown Carbon" or Delta-C and interpreted it as a measure of				
229	wood smoke concentrations (e.g. Wang et al. (2011)):				
230					
231	$UVPM = Conc_{ATT 370} - Conc_{ATT 880} $ (1)				
232					
233	2.5 Measurement Quality Assurance and Quality Control				

234	It is essential for the data collected in a measurement campaign to have clearly defined provenance.
235	Without descriptions of methodology, stated levels of accuracy, precision, harmonisation and
236	measurement uncertainty, it is extremely difficult to make meaningful comparisons between
237	different datasets and research. This was explored in Stacey (2019), where it was clear that,
238	historically, different UFP studies used a range of instrumentation, setups and calibration
239	methodologies, meaning only qualitative comparisons between them was realistically possible.
240	Wiedensohler et al. (2012) and Wiedensohler et al. (2018) emphasize the need for robust quality
241	control and standardised measurement methodologies; the Heathrow study reported here uses
242	quality assurance and quality control procedures that ensure consistency and comparability in UFP
243	data collection between the Heathrow and national network datasets.
244	For measurements of $NO_x$ , $PM_{10}$ , $PM_{2.5}$ , BC and meteorology, the measurements at Heathrow are
245	managed, collected and processed following guidance described in https://uk-
246	air.defra.gov.uk/assets/documents/reports/cat09/1902040953_All_Networks_QAQC_Document_20
247	<u>12</u> <u>Issue2.pdf</u> . Information about the analysers used at Heathrow is provided in the Supplemental
248	Information, Tables S1 and S2.
249	
250	3. RESULTS
251	3.1 Overall Summary
252	Timeseries data for the hourly measurements of particle number concentrations at LHR2 and Oaks
253	Road are presented in Supplemental Information, Figures S1, S2. Measurements of NO <sub>x</sub> , PM <sub>10</sub> ,
254	$PM_{2.5}$ and BC are also fully reported (Figures $S3 - S8$ ) and accessible through the
255	http://heathrowairwatch.org.uk webpages. Data from these analysers will be used to explore
256	associations and differences to typical ambient environments, but not considered in detail.
257	
258	

- 259

Pollutant	Mean	Median	Standard	Min-Max	Data
			deviation	(15 min data)	capture %
NO, ppb	46.9	30.0	56.7	0 – 540	100
NO₂, ppb	27.7	27.9	11.7	1-84	100
PM <sub>10</sub> , ug/m <sup>3</sup>	16.8	12.4	17.2	0.7 – 346.5	100
PM <sub>2.5</sub> , ug/m <sup>3</sup>	10.9	7.2	14.4	0.4 – 288.3	100
BC, ug/m <sup>3</sup>	3.11	2.30	2.79	0.08 - 28.08	100
UVPM, ug/m <sup>3</sup>	0.84	0.49	0.96	0.03 - 11.41	100
Nucleation,	7817	1871	15993	42 – 150000	87.6
dN/dlog Dp					
Aitken,	8638	5542	9704	93 – 107918	87.6
dN/dlog Dp					
Accumulation,	2088	1570	2110	70 – 30052	87.6
dN/dlog Dp					
Total PN,	8911	4756	12014	394 - 118726	87.6
#/cm³					

260 Measurement data for LHR2 and Oaks Road are summarised in Tables 1 and 2 respectively.

**Table 1.** Summary statistics for measurements at LHR2, 30 Sep to 25 Nov 2016

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Pollutant	Mean	Median	Standard	Min-Max	Data
			deviation	(15 min data)	capture %
NO, ppb	23.1	10.3	35.1	0 – 328	99.8
NO₂, ppb	19.5	19.1	10.3	0 - 68	99.8
PM10, ug/m <sup>3</sup>	14.3	11.0	12.5	0.8 – 186.6	100
PM <sub>2.5</sub> , ug/m <sup>3</sup>	10.2	6.9	11.1	0.4 – 172.4	100
BC, ug/m³	1.77	1.20	1.86	0.01 – 26.08	100
UVPM, ug/m³	0.55	0.37	0.71	0.01 - 6.83	100
Nucleation,	8476	2152	12064	0 – 86287	50.0
dN/dlog Dp					
Aitken,	7798	4723	8223	0 – 63372	50.0
dN/dlog Dp					
Accumulation,	1639	1370	1146	0 - 10280	50.0
dN/dlog Dp					
Total PN,	7408	3948	8180	0-62124	50.0
#/cm³					

**Table 2.** Summary statistics for measurements at Oaks Road, 30 Sep to 25 Nov 2016

266

267 Data for the first week of UFP measurements at LHR2 were rejected due to a software configuration error.

268 Data capture for the UFP analyser at Oaks Road was affected by a software fault with the controlling PC.

No data from this analyser was collected after 28 October 2016, data quality for the period 30 Sep to 28 Oct

270 was unaffected by the software fault.

As noted earlier, it was not possible to activate remote operation of the analysers by telemetry for this

survey. As a result, any instrumental faults arising during the campaign were assessed and corrected during

273 weekly calibration visits to the stations.

<sup>262</sup> 

274

275 Examination of historic NO<sub>x</sub>, PM and BC data at LHR2 and Oaks Road (available from

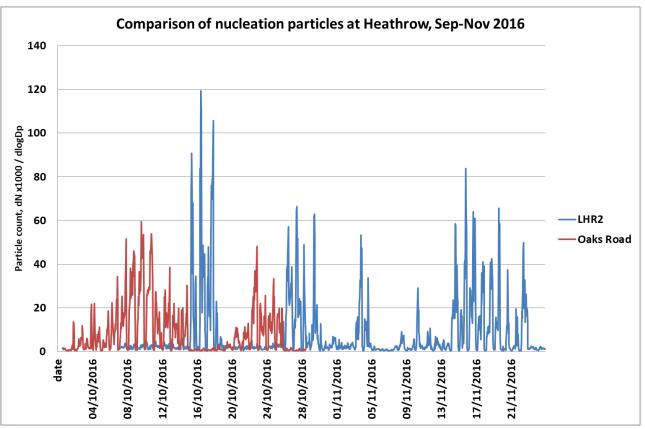
276 <u>http://www.heathrowairwatch.org.uk/reports</u>) has shown that measured concentrations and profiles for NOx,

277 PM mass and black carbon are not significantly different to those measured at nearby background and traffic

- 278 monitoring stations operated by Local Authorities or the national network stations in London.
- 279

It is clear from the particle number timeseries plots in Figures S1 and S2 that there are distinct periods where hourly average concentrations are significantly elevated from the baseline concentrations but equally periods where the PN hourly average concentrations are comparatively low. Figure 2 explores this for nucleation mode particles.





285

Figure 2. Time series of nucleation mode particles at the Heathrow Airport sampling sites, Octoberand November 2016

It is clear from this plot that high concentrations of nucleation mode particles are only observed at one location at a time, so wind direction is a critical factor in the presence and concentration of these particles.

293

Examination of LHR2 and Oaks Road particle number diurnal data averaged for the entire survey 294 (presented in Figure S9 and S10) shows enhanced PN concentrations between 06:00 and 23:00, 295 296 coinciding with typical increased activity around the airport. The diurnal plots also show close agreement with the two humped diurnal profiles of the NO<sub>x</sub> and BC pollutants, suggesting that 297 these pollutants mostly share common sources, including road traffic and commercial / domestic 298 299 energy use. The PM<sub>10</sub>, PM<sub>2.5</sub> and accumulation mode particle diurnal plots do not follow the exact same pattern as  $NO_x$ , so likely originate from different sources to  $NO_x$ . The accumulation mode PN 300 appears to follow a similar trend to the  $PM_{10}$  and  $PM_{2.5}$  diurnal profiles, but it is relatively flat and 301 significantly lower in number concentrations when compared to the Nucleation and Aitken mode 302 PN datasets. The diurnal plots for Nucleation and Aitken mode particles do not follow the trends 303 304 for the other pollutants, further confirming that they are not associated with the same sources.

305

## **306 3.2 Results in Context with Other Monitoring Data**

Measurements of UFP were coincident at LHR2 and Oaks Road for the period 7 – 28<sup>th</sup> October
2016. As noted earlier, there are three measurement stations within the UK national monitoring
network (https://uk-air.defra.gov.uk/interactive-map) that measure UFP – these stations were also
all in operation during this time. A summary of average concentrations measured at all 5 sites is
presented in Table 3.

Pollutant	Marylebone	North	Chilbolton	LHR2	Oaks Road
	Road	Kensington			
NO, ppb	80.4	9.1	1.6	43.9	21.8
NO <sub>2</sub> , ppb	39.2	18.9	8.4	27.5	20.7
PM <sub>10</sub> , ug/m <sup>3</sup>	21.6	17.1	13.4	15.9	13.8
PM <sub>2.5</sub> , ug/m <sup>3</sup>	12.8	11.1	7.3	9.5	9.4
BC, ug/m <sup>3</sup>	3.787	0.912	0.620	2.901	1.792

UVPM, ug/m <sup>3</sup>	0.305	0.198	0.277	0.615	0.537
Total PN,	10046	5384	2637	9053	7964
particles/cm <sup>3</sup>					

Table 3. Average pollutant concentrations at Heathrow and comparison stations, 7-28 Oct 2016
314
315

In this "averaged" scenario, concentrations for all pollutants near the airport can be seen to be largely in the range of the urban traffic and urban background environments of the two London locations, but substantially higher than the rural Chilbolton location.

319

320 Airport PM<sub>10</sub> and PM<sub>2.5</sub> concentrations are slightly lower than seen in London, but different measurement techniques are deployed, which may account for some of the differences. The 321 Heathrow sites use Fidas 200 analysers, while TEOM1400/FDMS8500 units were deployed at the 322 national network stations. There is ongoing work, in preparation for the UK Environment Agency 323 by Ricardo, Bureau Veritas and Kings' College London, discussed at a number of seminars, for 324 325 example http://www.scottishairquality.scot/assets/documents/reports/9 PM analyser replacement Brian St 326 327 <u>acey.pdf</u>, that suggests that there are differences in instrument signal performance that accounts for 328 most of the observed differences in concentrations. This work highlights that detailed knowledge of the operation and limitations of notionally similar measurement devices is essential before drawing 329 330 any conclusions about observed differences.

331

Higher concentrations of UVPM were measured at the airport stations, compared to the London
stations. They are likely to be real, although there are again differences in equipment used. The
airports use modern AE33-7 seven wavelength aethalometers, while the national network uses older
AE22-2 two wavelength instruments. It is possible that differences in attenuation correction
protocols (automatically corrected in the AE33, manually corrected post-collection for the AE22),
may account for a significant proportion of the differences in measurements. For example, studies
undertaken at University of Birmingham (yet to be published), comparing attenuation correction

protocols for the AE22 aethalometer published by Virkkula et al (2007) and Apte et al (2011) have found that significant differences in "corrected" concentrations are observed. It can therefore be justifiably argued that neither method for attenuation correction can be guaranteed to give data comparable to that produced by the AE33 aethalometer, where no correction for attenuation is required.

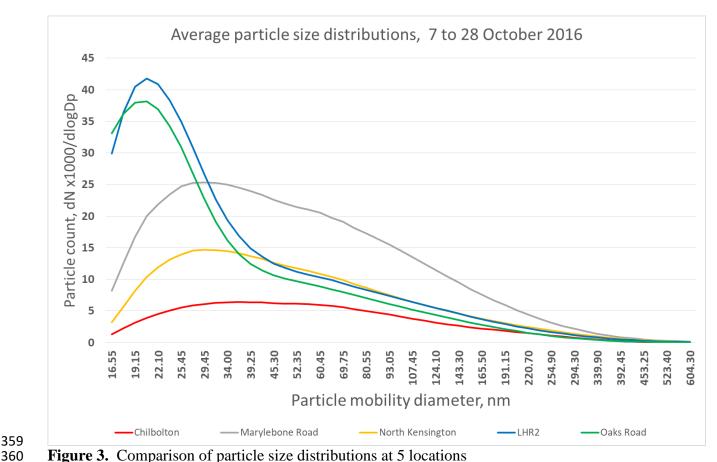
344

Averaged particle number concentrations, calculated by summing all of the particle counts from the
SMPS analysers from 14.6nm to 661.2nm, shows that a similar trend is observed to other
pollutants: concentrations at the airport locations fall between the traffic and background
measurements made at the London stations.

349

On initial investigation of the measurement datasets therefore, the ambient air environment at Heathrow appears reasonably similar to the rest of London. However, data from the SMPS analysers also provides valuable information about the particle size distribution at all 5 locations. It has already been demonstrated in earlier research that nucleation mode particles are strongly associated with airport activity. The plot in Figure 3 shows the average particle size distribution at each station for the period when all 5 SMPS were operational; the period between 7 and 28<sup>th</sup> October 2016.

357





363 There are many points that are striking about this data:

• The size distributions for Marylebone Road and North Kensington are remarkably similar,

differing only in magnitude. The mode value for both stations is ~30nm, suggesting that both
stations share commonality of source origins.

• Chilbolton concentrations are much lower, with a larger mode particle size value of ~37nm.

368 The distribution profile is otherwise reasonably similar to the London stations.

- For particle sizes larger than ~40nm, the LHR2 profile follows a very similar profile to the
   North Kensington station
- The Oaks Road particle distribution profile is very similar to LHR2 for particles up to about
- 150nm in size. For particles larger than 200nm, Oaks Road follows a profile similar to
- 373 Chilbolton, suggesting that these larger particles are more background in nature than the
- 374 LHR2 station.

The most obvious observation about the airport particle size distribution (PSD) is how the particle number concentrations smaller than 40nm differ significantly from the other three datasets. The mode value for LHR2 and Oaks Road is ~20nm, significantly smaller particle modes than at the other 3 stations. It is clear from this plot that the ambient environment close to the airport is significantly different for smallest particle numbers compared to typical urban environments.

The data are further analysed using a cumulative frequency plot, which sums the proportion of total particles within the increasing particle size dataset. The CFD plot in Figure 4 for all 5 locations supports the observation in Figure 3 that most particles at the airport are smaller in nature than in typical urban environments. At Marylebone Road and North Kensington, 50% of the particles are smaller than ~50nm, whereas at LHR2 and Oaks Road, 50% of all particles are smaller than ~25nm, suggesting a distinct and different source near the airport.

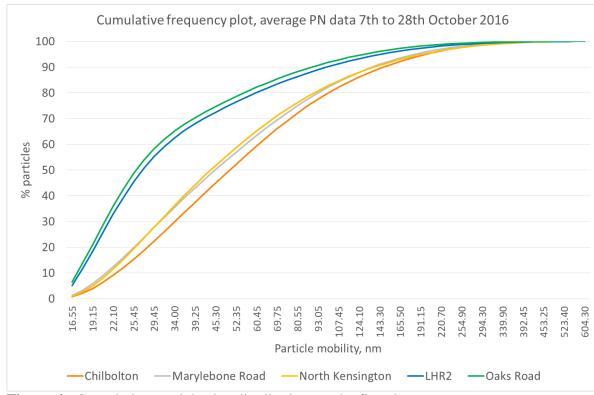


Figure 4. Cumulative particle size distributions at the five sites.

392

The directional nature of the UFP emissions can be explored further by looking at PSD at LHR2 and Oaks Road when winds are split into roughly northerly (the wind segment clockwise from 270 to 90 degrees) and southerly (clockwise from 90 to 270 degrees) segments. Figure 6 explores these data.

397

403

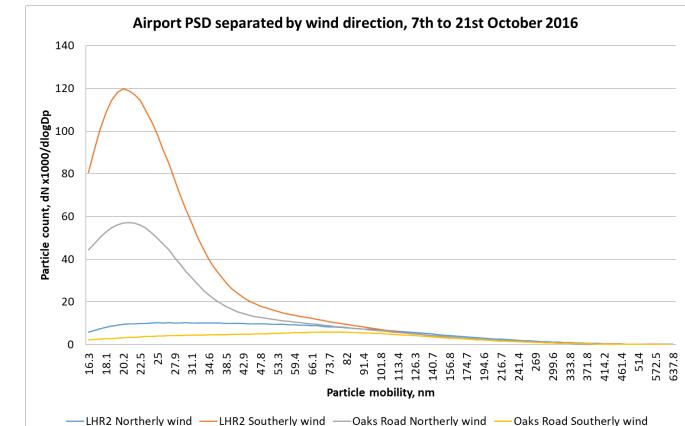


Figure 6. Airport PSD separated by wind direction for the Heathrow Airport sites. Note that the
particle mobility axis differs slightly from earlier figures due to the need to align Heathrow particle
size concentrations exactly with the data provided by the national network stations.

This plot clearly shows that when the wind does not originate from the airport (Northerly for LHR2 and Southerly for Oaks Road), the PSD profile of measurements is broadly similar to measurements made at the London urban locations. In contrast, when the stations are directly impacted by winds from the airport, the PSD profiles are dominated by very fine particles. LHR2, which is just 170m from the runway and unobstructed by buildings and other infrastructure, experiences much higher average particle counts than Oaks Road, over 600m from the runway and surrounded by residential buildings.

- 411
- 412
- 413
- 414

## 415 **3.3** Dependence of Airport Measurements on Meteorology

Meteorological measurements made at Heathrow allow for further analysis of the data using the
polarPlot function in OpenAir. The plots in Figure 7 examine the dependence of measurements on
wind speed and direction.



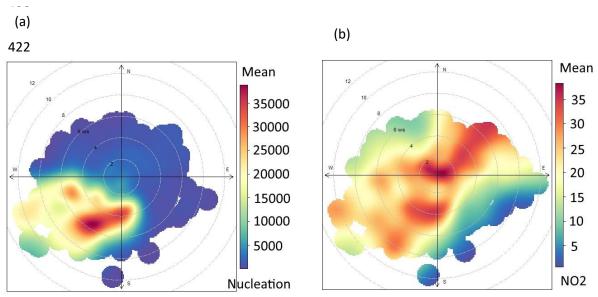


Figure 7. Polar plots for LHR2 data, (a) nucleation mode particles, (b) nitrogen dioxide
426

The plots for nucleation mode (Figure 7(a)) and Aitken mode (Figure S11) mode particles show a very strong influence from the airport, to the south and west of the measurement station. The nucleation mode particles plot shows very little influence from other directions, clearly pointing to airport activities as the dominating source of these particles at this location.

431

432 NO<sub>2</sub> at LHR2 (Figure 7(b)) is strongly associated with south west and north east wind directions,

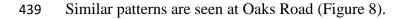
433 but also to a lesser extent from other directions. This reflects the multiple source nature of  $NO_2$  in

the environment; road traffic and domestic / commercial energy use from many sources around the

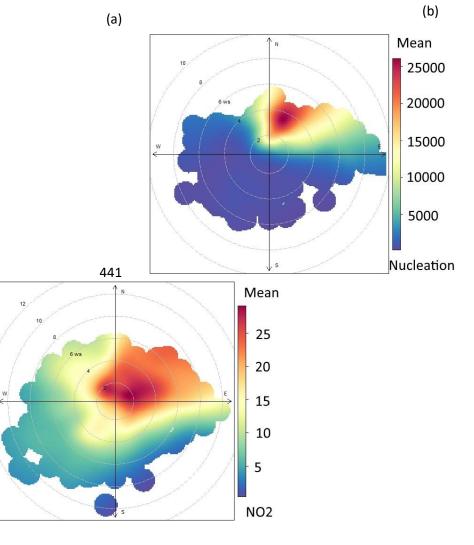
435 station are all seen to influence the polar plot.

436

- 437 NO, BC, UVPM and accumulation mode particles (presented in Figure S11) are associated with
- 438 most wind directions and also at low wind speeds.



440



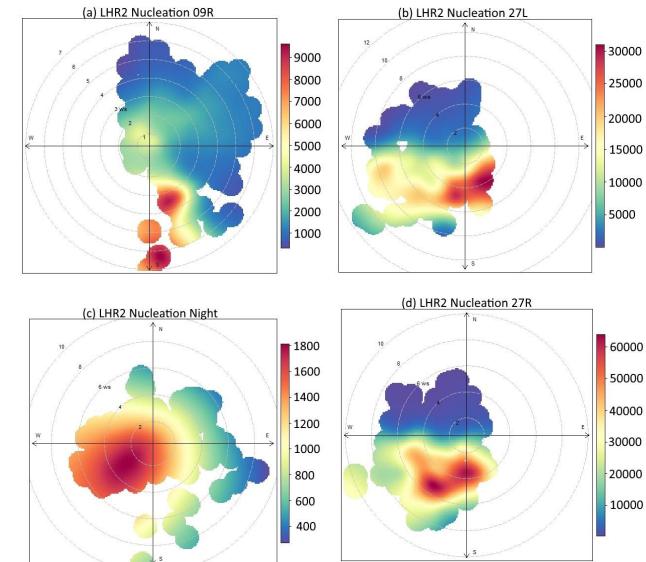
442 443

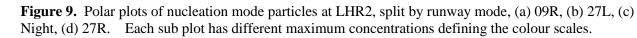
Figure 8. Polar plots for Oaks Road, (a) nucleation mode particles, (b) nitrogen dioxide
445
446

The plots show that high concentrations of nucleation mode particles, as well as NO<sub>2</sub>, are strongly
associated with winds from the airport. Aitken mode particles (in Figure S12) follow a similar

449	trend to nucleation mode particles, but are associated with a slightly wider range of wind directions
450	than the nucleation mode. It is clear from the two monitoring station datasets that nucleation mode
451	particles are predominantly associated with winds from the airport, suggesting that the airport is by
452	far the major source of emissions of these particles.
453	
454	In contrast, all other pollutants (Figure S12) are strongly influenced by low wind speeds, indicating
455	local sources, and the background environment as significant contributors. $PM_{10}$ and $PM_{2.5}$ appear
456	to originate largely from the same common sources, and PM mass sources appear to be mostly
457	independent from the other pollutants.
458	
459	<b>3.4</b> Dependence of Measurements on Airport Operation
460	The two runway configuration at Heathrow allows the airport to operate in a number of modes.
461	When winds are easterly, aircraft exclusively depart from Runway 09R, the southernmost runway,
462	and generally arrive on 09L, the northerly runway.
463	
464	When winds are westerly, the airport typically operates a shift-based departure system, departing on
465	one runway for half the day, and the other runway for the remainder. Landing occurs on the other
466	runway during these times.
467	
468	These operating modes are primarily chosen for practicality. For westerly departures, spreading the
469	distribution of landings and departures on Runways 27R and 27L equalises the wear and tear on the
470	landing zones on each runway, reducing the amount of maintenance required. For easterly
471	departures, the taxiways approaching the thresholds of Runway 09L are not suitable for modern
472	aircraft. This means that departures in this mode are exclusively from Runway 09R.
473	
474	The airport is typically closed to most air traffic during the hours 23:00 to 05:00 local time.

the measurement data to be examined in far greater detail. The three modes, departing from 09R,
27L and 27R, plus the overnight period, are presented as polar plots for nucleation mode particles in
Figure 9.





492 493

It is clear that nucleation mode particle number concentrations are highest when aircraft depart from 494 27R (closest to the monitoring station) and lowest when the airport is closed overnight. Nucleation 495 496 particle numbers are significantly higher when aircraft are departing on 27R compared to when they are landing on 27R (departing on 27L) Even when aircraft are departing from 09R, a small yet 497 clear peak in nucleation mode particles can still be seen from the airfield, presumably from 498 departing aircraft exhaust – arriving aircraft leave the runway before they are within 1km of the 499 500 LHR2 monitoring station and are thus not expected to significantly influence measurements during easterly winds. Overnight concentrations of nucleation particles are generally comparatively very 501 502 low, but still appear to be associated with winds from the airfield.

503

The plots for Aitken mode particles for 27R and 27L are very similar to those seen for nucleation mode particles (presented in Figure S13), suggesting that the largest influence for these particles still comes from the aircraft. In contrast, the polar plots for Aitken mode particles from 09R and overnight (presented in Figure S13) differ from the nucleation mode plots, being both significantly lower in concentration and showing more influence from lower wind speed meteorology. This suggests more diverse source origins than just the dominance of the airport in nucleation mode measurements.

511

The polar plots (presented in Figure S14) for black carbon, measured by the Aethalometer, illustrate that BC is neither strongly associated with airport activity or nucleation mode particles. This reinforces work conducted by Costabile et al. (2015), which found no strong links between aircraft emissions and elevated BC measurements.

516

517 The polar plots (presented in Figure S14) for UVPM, measured by the Aethalometer, suggest that518 elevated concentrations of UVPM at LHR2 might have an association with nucleation mode

particles when aircraft are departing from runway 27R. A similar link is not obvious when aircraft are landing on 27R, 09R or indeed any other aircraft operating modes at the airport, suggesting that high thrust exhaust emissions may be associated with production of black carbon particles that strongly attenuate UV light. In contrast (in Figure S15), UVPM at Oaks Road is dominated by association with low wind speeds. There is some indication of a contribution from the direction of the airport, but it is likely that a number of different sources contribute to measurements in this residential location.

526

For Oaks Road, a similar picture emerges (plots presented in Figure S15). Highest concentrations
of nucleation mode particles are associated with aircraft departing from 09R, closest to the
monitoring station, but high concentrations of nucleation mode particles in other polar plot modes
clearly also originate from the airfield.

531

Polar annuli for all pollutants at both sites are presented in Figures S16 and S17. These plots

533 further reinforce the directional and diurnal nature of emissions around the airport

534

## 535 **3.5** Examination of Fine Temporal Resolution Data

The monitoring station at LHR2 is 170m from the centre of the northern runway. Under favourable meteorology, plumes from aircraft departing and landing impact on the monitoring station, raising the possibility that these plumes can be further analysed and characterised by, for example, aircraft type, engine type, aircraft landing and aircraft departing.

540

541 On average (<u>https://www.heathrow.com/file\_source/Company/Static/PDF/Investorcentre/Heathrow-</u>

542 (SP)-FY2016-results-release-(FINAL).pdf), an aircraft departs from the airport every 90 seconds

between 06:00 and 23:00 every day. The SMPS/CPC configuration at LHR2 was set to provide a

full particle size sweep every three minutes, meaning that it was impossible (with this dataset) to

uniquely assign a single measurement to an individual aircraft. Nevertheless, some structure in the
PN measurements can be observed which bears excellent correlation to the runway operations. The
plot in Figure 10 presents a timeseries of PSD collected on 17 October.

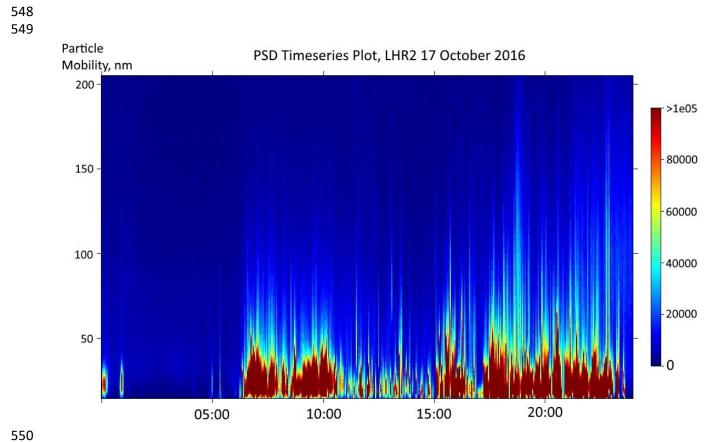


Figure 10. Particle Size Distribution plot, LHR2, 17 October

553

The plot shows that the smallest particles have the highest concentrations, and a clear temporal 554 correlation to airport activity (from around 6am to 11pm). Particle number concentrations are very 555 low between midnight and 06:00. Aircraft movement information obtained from the airport for 17 556 October confirmed that aircraft departed from 27R (closest to the station), between the hours of 557 06:00 and 10:00, and 15:00 to 23:00. Aircraft landed on 27R between 10:00 and 15:00. This 558 information supports the observations in Figure 6: PN concentrations are clearly lower between 559 10:00 and 15:00, suggesting that emissions of nucleation mode particles from landing aircraft are 560 significantly lower than those from departing aircraft. This observation was repeated throughout 561 the survey, though the meteorology made this most obvious on 17<sup>th</sup> October. 562

As was seen in Figure 9, the activities on the southern runway have an effect on measured concentrations at LHR2, so it is likely that the departing aircraft on the southerly runway 27L will also have an impact, albeit much less than the northern runway, on the measurements at LHR2. However, it is not possible to decouple these emissions from the landing aircraft on 27R to investigate this, and it assumed that, at least for aircraft departing on 27R, any contribution from emissions on 27L is overwhelmed by the proximity of the nearest emissions.

570

563

571 **3.6** Comparison with Receptor Modelling Outputs

Masiol et al. (2017) undertook a similar study at Heathrow Airport in 2014/15, investigating UFP
concentrations at a monitoring station 1km NE of LHR2 (London Harlington, part of the UK
national monitoring network). The data collected were analysed using *k*-mean clustering and
positive matrix factorisation (PMF), which revealed the contribution of the airfield to local particle
number concentrations (high concentrations, mode concentration ~20nm). Masiol et al. (2017)
calculated that at the London Harlington station, approximately one third of the total measured PN
concentrations originated from the airport.

579

580 Data collected from this 2016 LHR2 and Oaks Road study were analysed using the PMF5 positive

581 matrix factorisation source apportionment model (v5.0.14.21735, U.S. Environmental Protection

Agency, USA). Details of the model and usage methodologies are comprehensively described by

many authors including, for example Rizzo and Scheff (2007), Masiol et al. (2017), and in

584 USEPA's own guidance: <u>https://www.epa.gov/sites/production/files/2015-</u>

585 <u>02/documents/pmf\_5.0\_user\_guide.pdf</u> and

586 https://www3.epa.gov/ttnamti1/files/ambient/pm25/workshop/laymen.pdf

For LHR2 and Oaks Road, analysis was focussed on qualitative output. The factors identified by
the model were used to compare against the measurement data analysed using the tools in R and
OpenAir. No effort has been made at this stage to normalise the extremes of measured
concentrations in the model to allow for detailed quantitative assessments.

592

In order for the model to run more effectively, SMPS data from both locations was aggregated into hourly means and then further aggregated into a reduced number of size bins – 15, reduced from the 107 size fractions natively output by the SMPS in this configuration. The data from the 14.6nm and 680nm channels was rejected for this analysis, to remove any possible influence from spurious data at the start and end of the SMPS measurement cycles. Data from the other pollutants at the station: NO, NO<sub>2</sub>, NO<sub>x</sub>, BC, UVPM, PM<sub>10</sub>, and PM<sub>2.5</sub> were also included in the PMF runs.

599

600 Uncertainties and detection limits (DL) for all pollutants were derived from data provided in the 601 Supplementary Information, with the exception of PN, which was set to 100% uncertainty and DL 602 of 100 particles/cm<sup>3</sup>. Where measurements were lower than the stated detection limit, the DL value 603 was substituted into the uncertainties data. An additional 10% uncertainty was added to the model 604 before all runs.

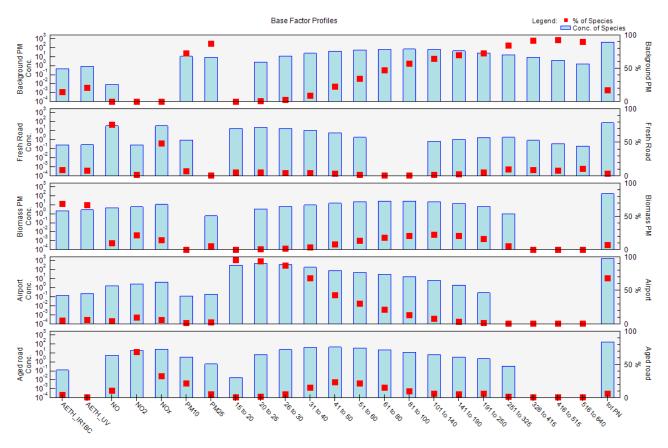
605

The model was run for 3 to 10 factor scenarios, with strong relationships set for all pollutants except PM<sub>10</sub> and PM<sub>2.5</sub>. The total variable was set to total PN (the sum of all PN data from 15-640nm) and assigned strong status. The base model was set to 100 runs, although there was little difference between this solution and a 20 run solution, confirming that both analyses are robust. Displacement analysis was run using default settings. Bootstrapping used default settings for 50 bootstraps. For BS-DISP, all Strong channels except Total PN were enabled for the analysis.

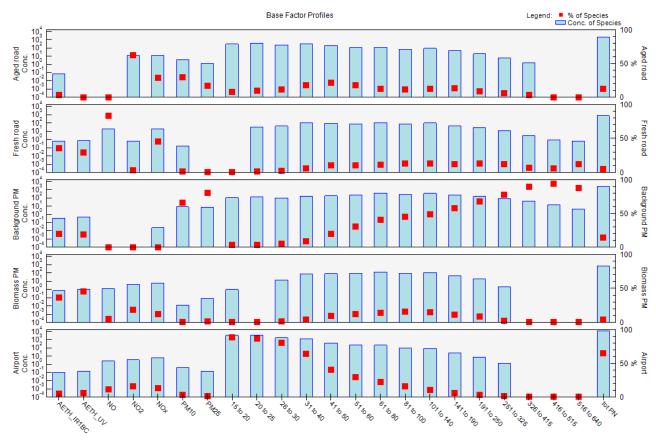
612

613	The model outputs were examined to check that all factors were unique and that factors had not
614	een subdivided unnecessarily. There was no adjustment required for rotational ambiguity.
615	
616	At both locations, 5 factors was identified as the optimal number, with factors identified as:
617	Airport. Factor dominated by PN <50nm, comparatively low contribution from all other
618	pollutants.
619	Fresh road traffic. Factor dominated by high concentrations of NO. Minor contribution also
620	from larger particles.
621	Aged road traffic. Factor dominated by NO <sub>2</sub> . PN from 30-80nm and PM <sub>10</sub> also observed
622	Biomass PM. Factor dominated by BC, UVPM and particles $80 - 250$ nm. NOx and PM <sub>2.5</sub>
623	also observed.
624	Background PM. Factor dominated by PM <sub>10</sub> , PM <sub>2.5</sub> and particles 80 – 640nm. BC and UVPM
625	also observed.
626	
627	The plots in Figures 11 and 12 show the base factor profiles for LHR2 and Oaks Road, with factors

628 labelled according to identified sources.



629630 Figure 11. PMF Base Factor Profiles for LHR2 site631



**Figure 12.** PMF Base Factor Profiles for the Oaks Road site

634

The model runs at both stations clearly identify the very fine particles associated with aircraft movements. The Aircraft factor from both LHR2 and Oaks Road models is overwhelmingly dominated by particles in the 15-50nm size range. 93 to 95% of the 15 to 25nm particles measured, and 68% of all particles smaller than 660nm measured at LHR2 originate from the Airport factor (At Oaks Road, these figures are 86 to 89% and 65% respectively). The factor explains very little of the variation in NO<sub>x</sub>, BC or PM however, suggesting that other sources dominate the contribution to local air quality. All iterations of the model runs from 3 to 10 factors were successful in separating this factor and its profile at both locations, further supporting the clear aircraft contributions at the stations. 

To add further confidence that the PMF model was extracting the airport factor consistently at both
LHR2 and Oaks Road, factor data for each factor run were input into regression analysis with
different factor scenarios. The results of these regressions showed extremely high correlation
between the different factor runs and are presented in Figures S20 to S23.

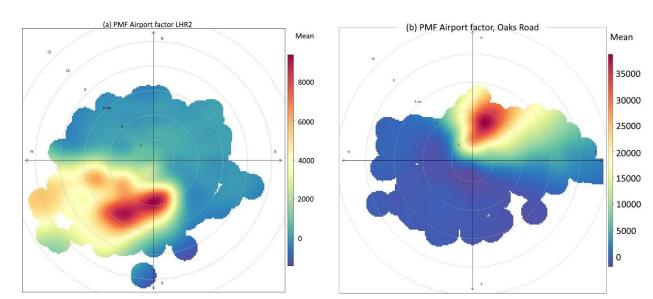


Figure 13. Polar plots of Aircraft PMF factors at (a) LHR2 and (b) Oaks Road
657
658

Analysis of the extracted aircraft factors from LHR2 and Oaks Road when combined with the meteorological data from LHR2 in polar plots shows the airport source very clearly in Figure 13 (to the south west at LHR2 and north east at Oaks Road) and compare exceptionally well to the polar plots for measured nucleation particles presented in Figures 7 and 8. This further confirms the robust analysis of the measurement data and the role of aircraft in the concentrations of the finest particle sizes measured near Heathrow Airport.

665

As a further data quality check, the model was run at LHR2 and Oaks Road with all SMPS channels retained in the model run unaggregated. The base model plots are presented in Figures S18 and S19 and confirm that the qualitative accuracy of splitting out the factors is unaffected by aggregating the PN size bins.

670

## 672 **4. CONCLUSIONS**

An extensive campaign to monitor UFP at Heathrow was undertaken in the autumn of 2016. The objective was to assess the context of measurements at the airport compared to measurements at "typical" traffic, background and rural locations in the south east of England.

676

Monitoring at the two locations at the airport was configured to ensure direct comparability withother measurements made in south east England.

679

Average concentrations at the airport, taking no account of particle size distributions, showed that total particle number concentrations the airport fits within the range of traffic and urban background locations in London, matching the trends seen for  $NO_x$ ,  $PM_{10}$ ,  $PM_{2.5}$  and BC. The distribution of particle sizes is however, completely different, with the airport PSD dominated by particles with a mode of 20nm. In contrast, measurements of PN in London have a significantly larger mode of 30nm. We believe that this is the first time this type of concurrent comparison of airport and urban UFP has been undertaken, providing valuable insight into the nature of the different environments.

687

Further investigation of the nucleation mode particles and meteorology reveals that measurements of particle number from within the airport perimeter are dominated by these smallest particles and are closely associated with aircraft. Analysis of the operating modes at the airport showed that aircraft departing from the airport emit particles in much higher numbers than those arriving.

692

Nucleation mode particles from the airport are not strongly associated with Black Carbon, though,
at LHR2, there does appear to be some correlation with BC particles that strongly absorb UV light.
There is a modest association between nucleation mode particles and NO<sub>2</sub>.

696

697	The Heathrow data were analysed using the USEPA PMF model to separate the contributions into 4
698	factors. A clear airport component was identified at both locations, where the largest proportion of
699	the factor was associated with nucleation mode particles. Examination of these factor datasets in
700	polar plots showed excellent agreement with the nucleation mode polar plots using data collected
701	from the analysers.
702	
703	
704	DATA AVAILABILITY
705	Data supporting this publication are openly available from the UBIRA eData repository at
706	https://doi.org/10.25500/edata.bham.00000349).
707	
708	ACKNOWLEDGEMENTS

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710 project.

711

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# 853 SUPPLEMENTARY INFORMATION:

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- 855 Evaluation of Ultrafine Particle Concentrations and Size Distributions at London Heathrow Airport.
- 856 Supplemental information.
- 857

Station	Equipment installed
LHR2	API T200 NOx analyser
	FIDAS 200 PM analyser
	Magee AE33-7 Black Carbon analyser
	Lufft WS-600 weather station (WS/WD/T/P/RH/Precipitation)
Oaks Road	API T200 NOx analyser
	FIDAS 200 PM analyser
	Magee AE33 Black Carbon analyser

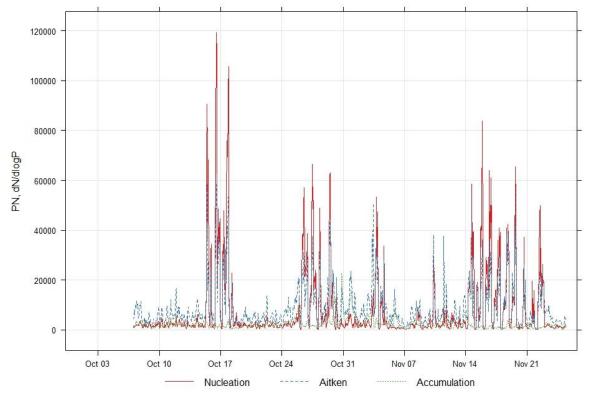
858 Table S1 – Conventional instrumentation at LHR2 and Oaks Road

859

Pollutant	Accuracy	Limit of detection
NO	±14.0%	±2ppb
NO <sub>2</sub>	±14.0%	±2ppb
PM <sub>10</sub>	±7.5%	±3μg/m³
PM <sub>2.5</sub>	±9.3%	±3μg/m³
BC	±15.4%	±0.1µg/m <sup>3</sup>
Particle Number	20%	20 particles /cm <sup>3</sup>

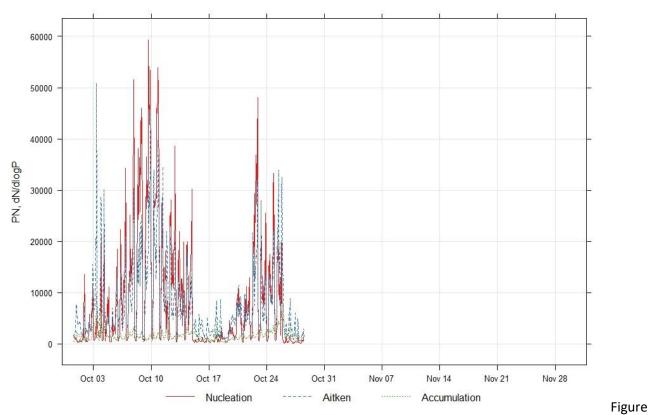
Table S2 – Accuracy and detection limits for instruments used for the survey.

LHR 2 Timeseries Plot





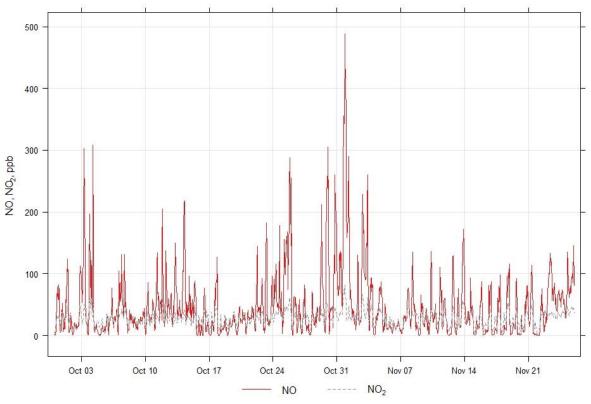
## Oaks Road Timeseries Plot







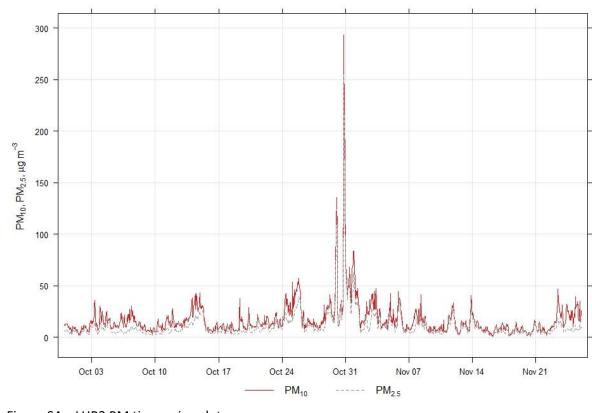




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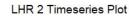
Figure S3 – LHR2 NOx timeseries plot

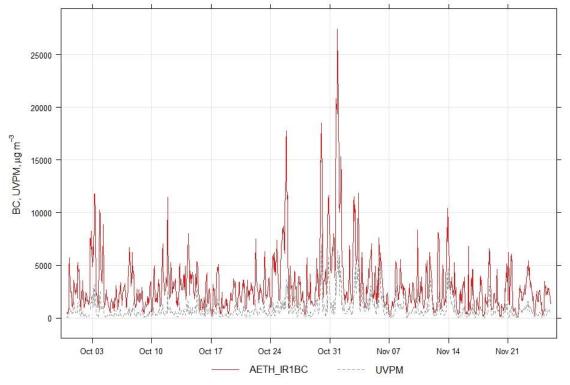
### LHR 2 Timeseries Plot



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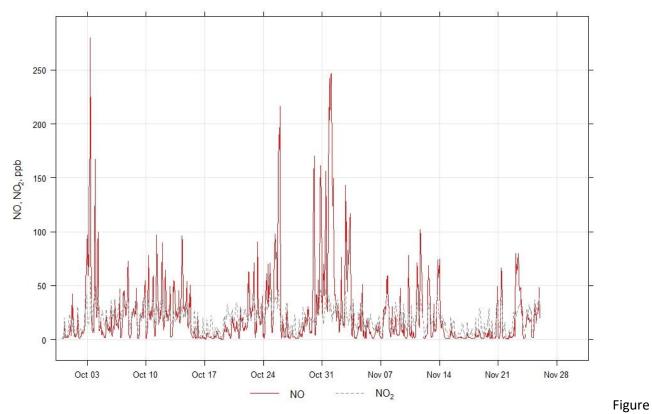
Figure S4 – LHR2 PM timeseries plot





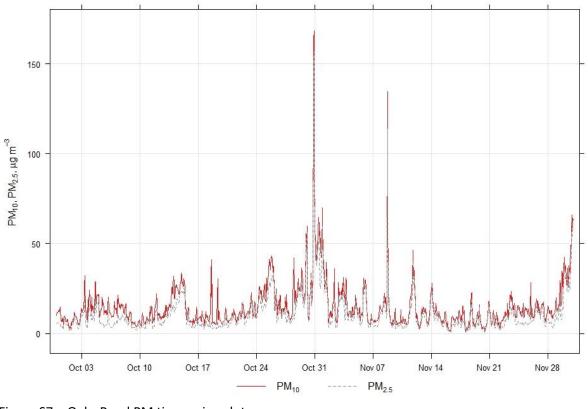
869 870 Figure S5 – LHR2 BC timeseries plot





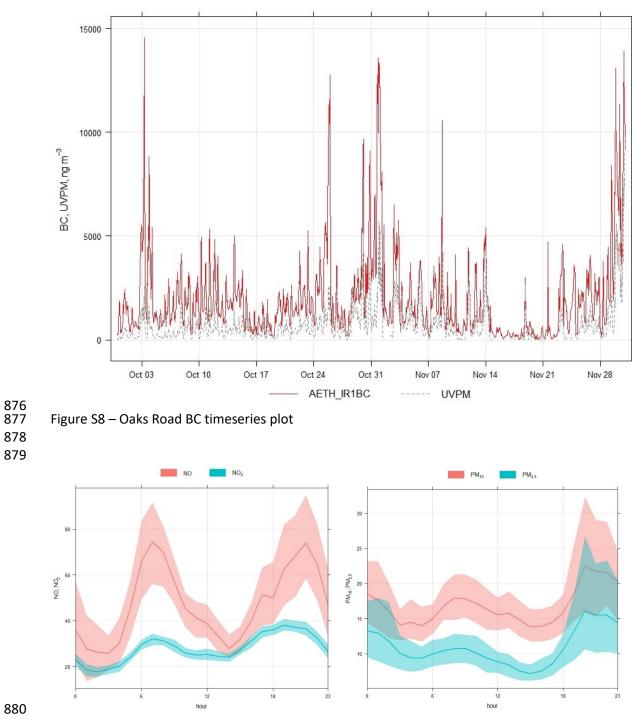
872 S6 – Oaks Road NOx timeseries plot

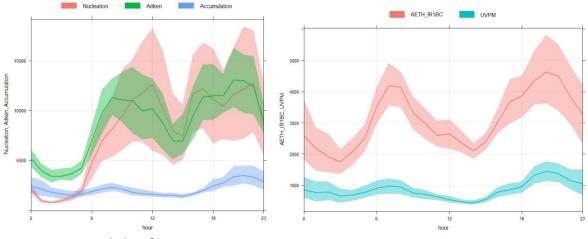




874 Figure S7 – Oaks Road PM timeseries plot

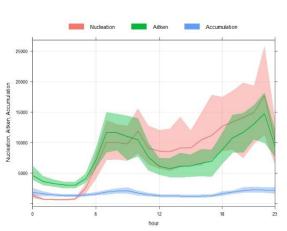
### Oaks Road Timeseries Plot



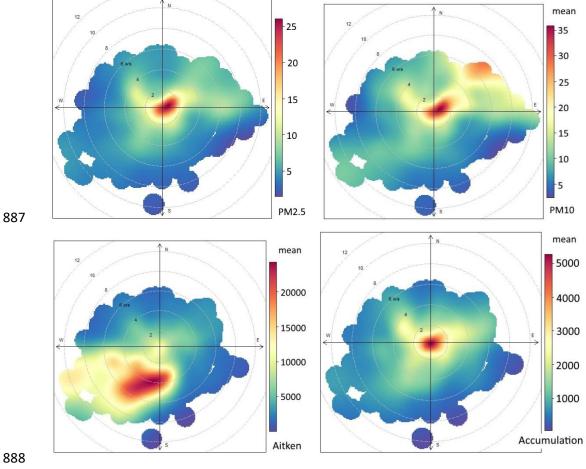


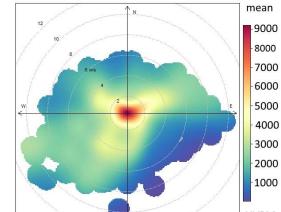
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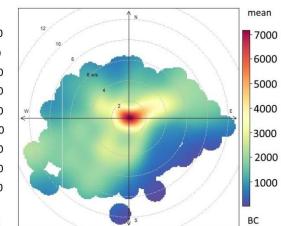
Figure S9 – Diurnal plots for measurements at LHR2

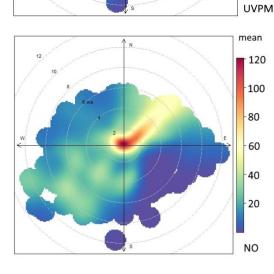


885 Figure S10 – Diurnal plots of Particle Number concentrations at Oaks Road 

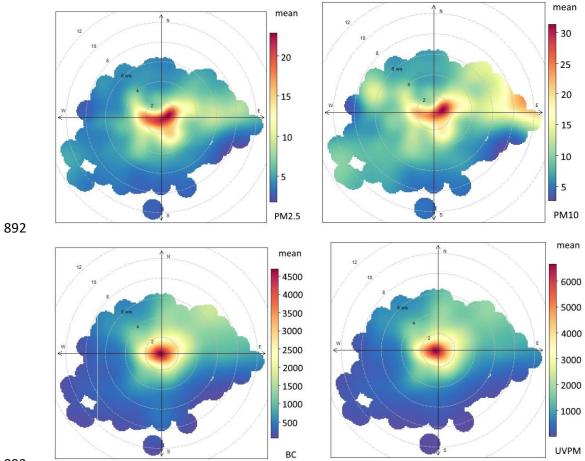


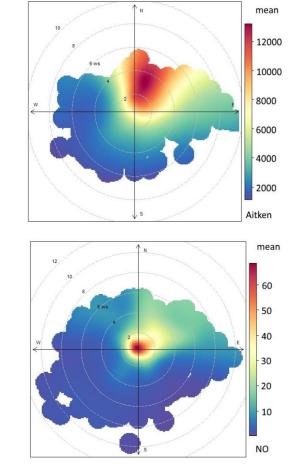






# 891 Figure S11 – Polar plots for LHR2 measurements





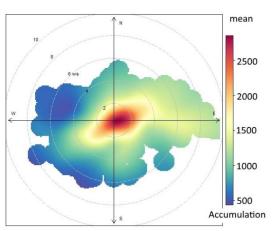




Figure S12 – Polar plots for Oaks Road measurements

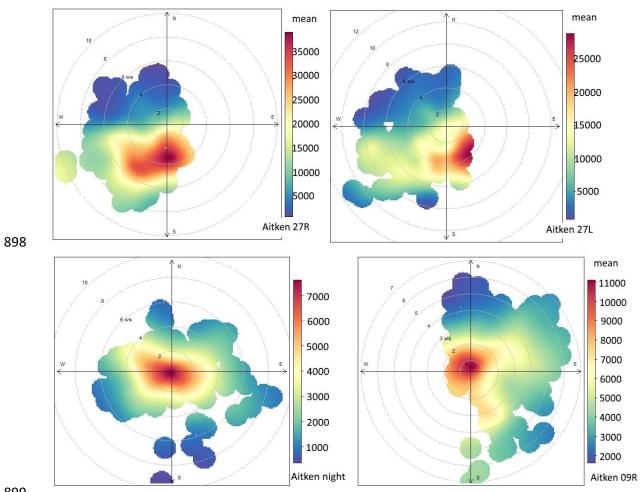
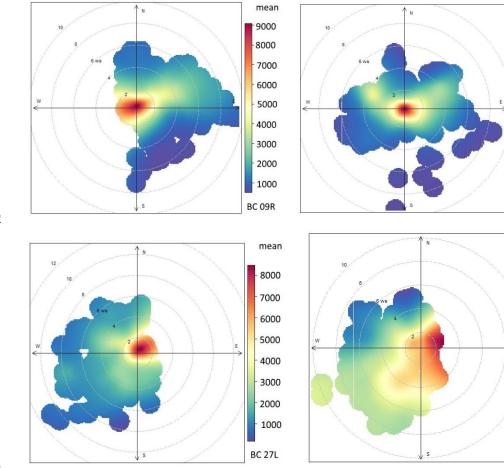


Figure SI13 – Aitken particle mode for LHR2, 27L, 27R, 09R and overnight modes



mean

BC Night

mean

BC 27R

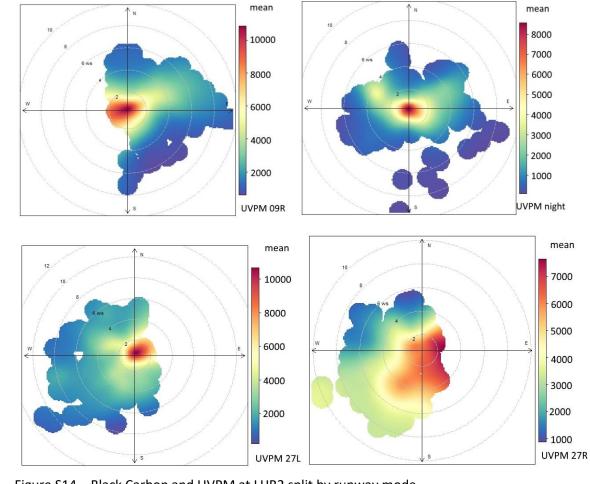
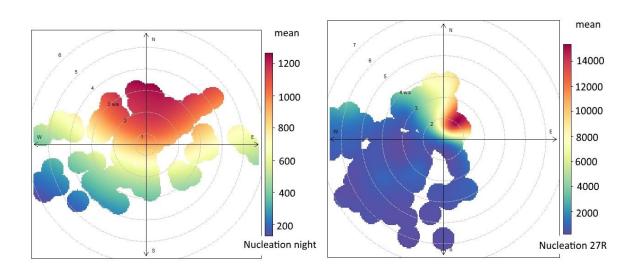




Figure S14 – Black Carbon and UVPM at LHR2 split by runway mode



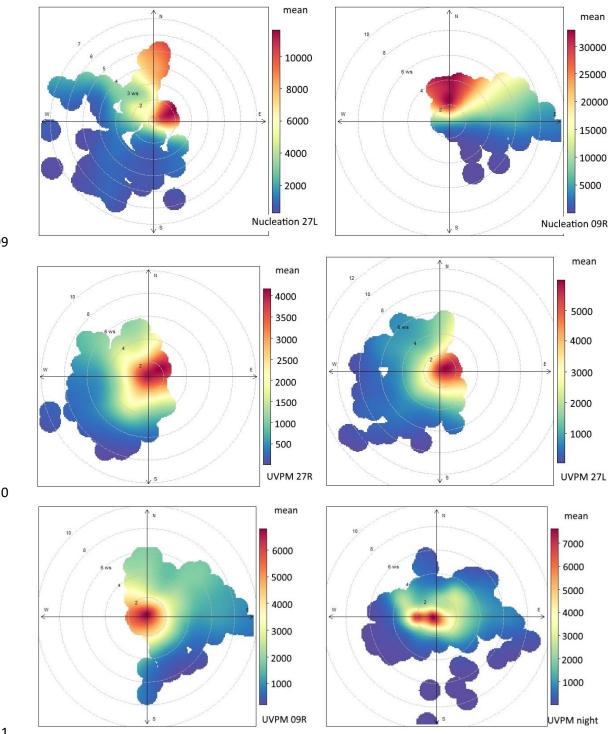


Figure S15 - Nucleation mode particles and UVPM at Oaks Road split by runway mode

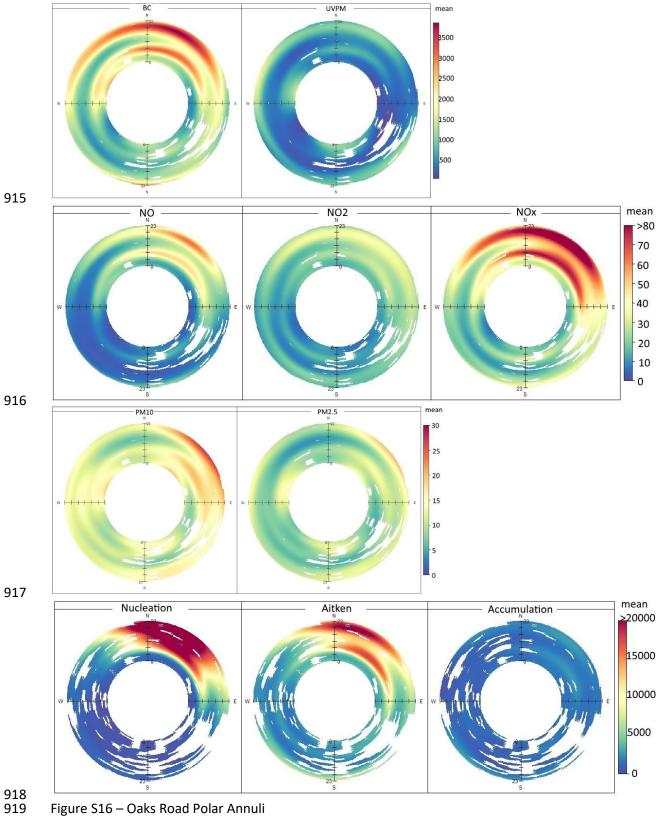




Figure S16 – Oaks Road Polar Annuli

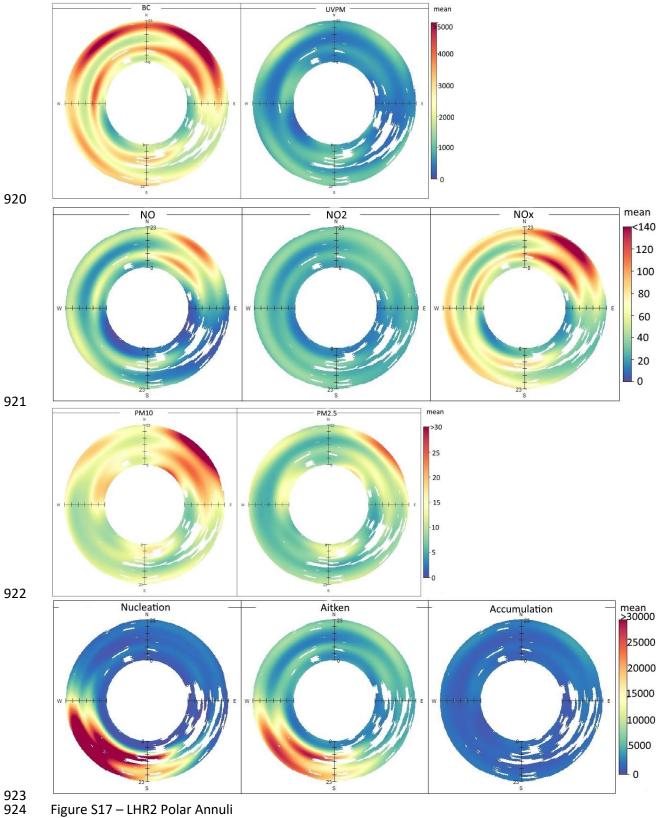
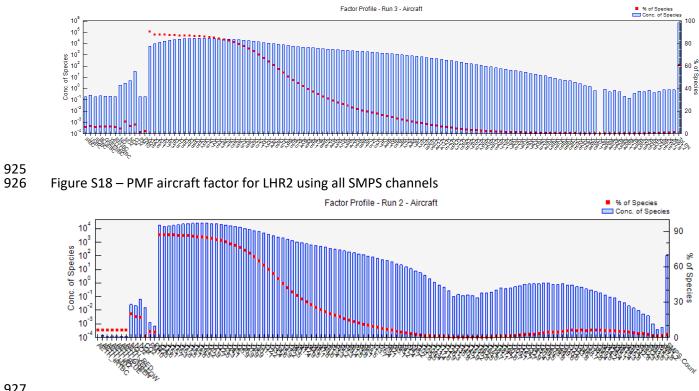
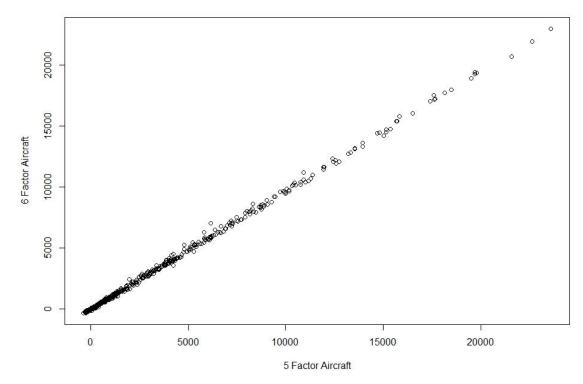


Figure S17 – LHR2 Polar Annuli



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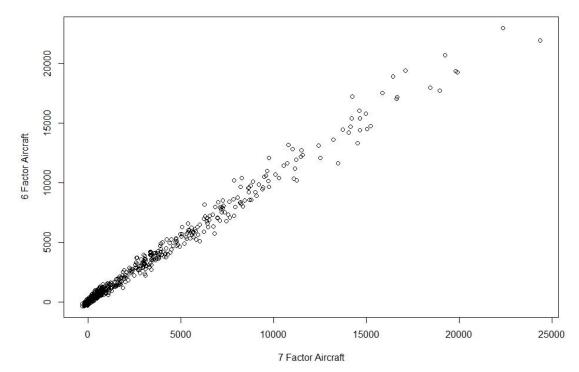
Figure S19 – PMF aircraft factor for Oaks Road using all SMPS channels



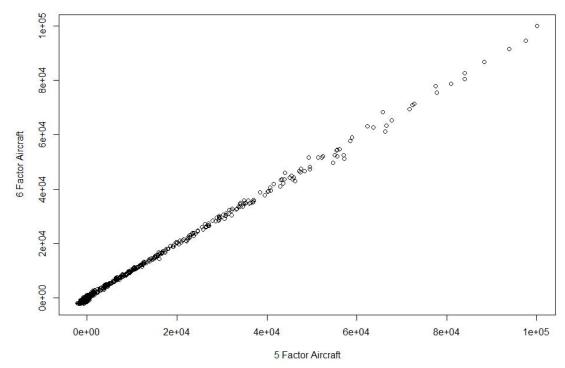
### LHR2 model correlation evaluation

929930 Figure S20 – Correlation between 5 and 6 factor solutions for Aircraft at LHR2

#### LHR2 model correlation evaluation



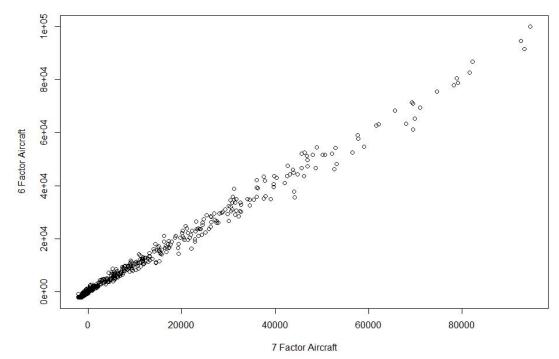
931
932 Figure S21 – Correlation between 6 and 7 factor solutions for Aircraft at LHR2



#### Oaks Road model correlation evaluation

933934 Figure S22 – Correlation between 5 and 6 factor solutions for Aircraft at Oaks Road

## Oaks Road model correlation evaluation



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936 Figure S23 – Correlation between 6 and 7 factor solutions for Aircraft at Oaks Road

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