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Evaluation of Ultrafine Particle Concentrations and Size Distributions at London Heathrow Airport

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ABSTRACT

A study to monitor UFP at Heathrow Airport 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. Measurements were made at two airport locations (called LHR2 and Oaks Road) at opposite ends of the airfield, to further understand the contribution of the airport to local air quality. Average concentrations showed that total particle number concentrations at the airport are typically lower than a traffic location and higher than an urban background location in London, matching the trends seen for NO_x, PM_{10}, $PM_{2.5}$ and BC pollutants. However, the size distribution of the submicrometre particles at the airport 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 larger mode of 30nm. This study demonstrated that measurements of particle number from within the airport perimeter are dominated by the 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. Nucleation mode particles are commonly associated with emissions from combustion processes. However, measurement of these particles at the airport are not strongly correlated with Black Carbon. There does appear to be some 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, LHR2. There is also modest association between nucleation mode particles and NO_2. The study showed that the classical air pollutants measured at Heathrow are very similar in concentration to typical urban environments in London and south east England, but particle numbers in the sub 30nm size range are markedly different to those measured in London.

1. INTRODUCTION

Heathrow Airport is the busiest two-runway airport in the world. In 2016, the airport handled over 75.7 million passengers and approximately 470,000 aircraft movements (https://www.heathrow.com/file_source/Company/Static/PDF/Investorcentre/Heathrow-(SP)-FY2016-results-release-(FINAL).pdf).

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_2, PM_{10} and $PM_{2.5}$. Black Carbon is measured at 2 of the 4 locations, while O_3 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.

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.

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.

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

Studies undertaken by, for example Durdina et al. (2014), Lobo et al. (2015), Abegglen et al. (2016), Turgut et al. (2015), and Vander Wal et al. (2016), measured emissions directly from the exhaust of aircraft. These largely focussed on non-volatile particles and showed that, generally, these particles are mostly carbon based and not significantly different in composition to other combustion sources.

As the exhaust plume emerges from the engine and interacts with the atmosphere, combustion products cool and can condense and/or interact with other components to form secondary aerosols. A study by Beyersdorf et al. (2014), looked at volatile and non-volatile UFP exhaust emissions with increasing distance from the source and found that as the exhaust plume cools and evolves, large quantities of very fine particles are detected.

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), 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.

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 Bezemer all showed that the finest particles emitted from aircraft remain in the atmosphere and can be transported over large distances downwind of an airport. Hudda and Fruin (2016) were able to 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 Airport
98 Schiphol .
99
100 Assessment of the research by Masiol and others made it clear that further robust investigation of
101 UFP measurements, and in particular the particle size distribution of aircraft emissions was
102 warranted. This paper presents the results from the measurement campaign, undertaken between
103 September and November 2016, to determine UFP concentrations and size distributions near the
104 airport.
105
106 **2. METHODS**
107 **2.1 Monitoring Locations**
108 This measurement campaign was designed to compare measurements of UFP at Heathrow against
109 measurements made at other measurement stations in the South East of the UK, and then to further
110 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:
112

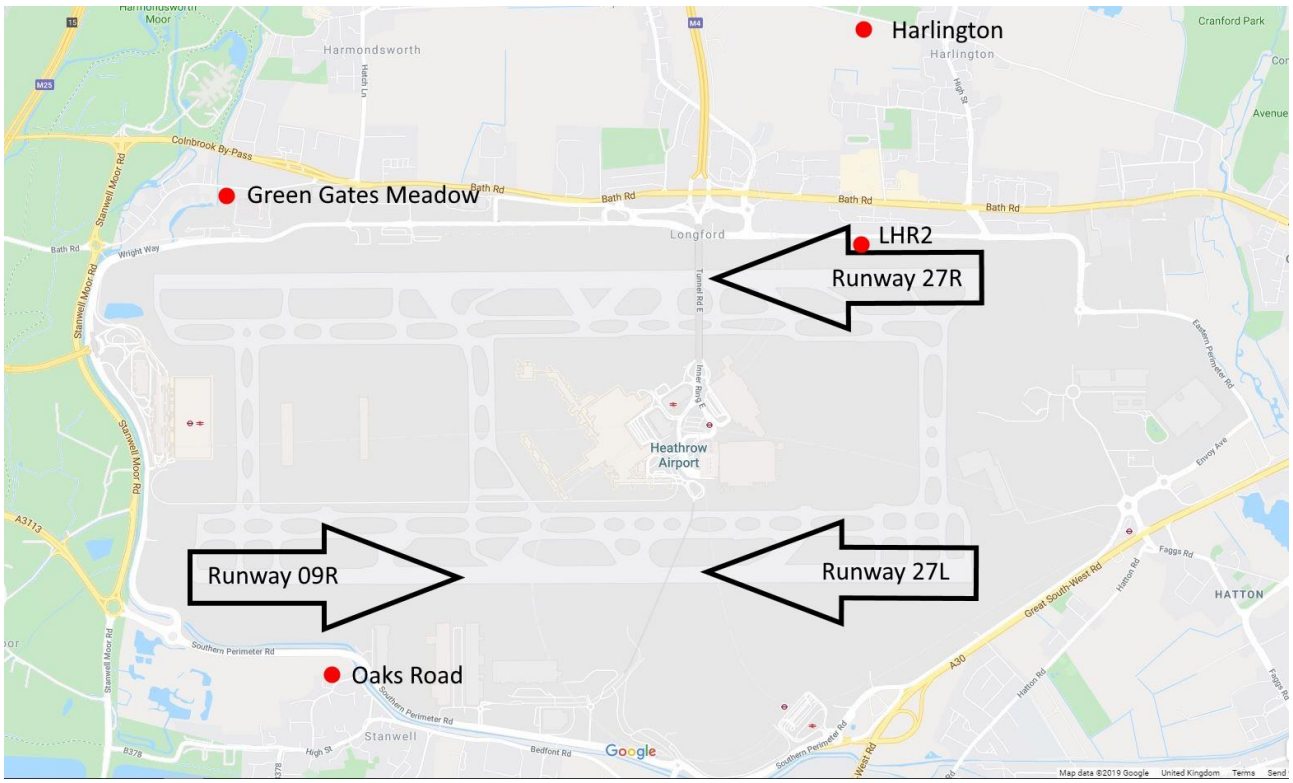


Figure 1. Locations of Heathrow monitoring stations. Runway 27R, Runway 27L and Runway 09R denote the three operating modes of the airport, indicating here the runway assigned for departing aircraft. Note that aircraft never depart in an easterly direction on the northern runway.

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.

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_{10}, $PM_{2.5}$, BC and NOx.

LHR2 – located airside in the north eastern corner of the airfield, 170m from the northern runway and less than 20m from the northern perimeter road. It has operated continuously since 1993, measuring PM_{10}, $PM_{2.5}$, BC, NOx and meteorology.

2.2 UFP Measurement Campaign

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

The following equipment was used:

- Butanol based TSI Model 3775 CPCs (TSI inc., MN, USA) to count particle numbers.
- At Oaks Road, TSI Model 3080 with long DMA (Model 3081) classifier and soft X-ray neutraliser. Automatic on-board software correction was enabled for diffusive losses and multiple charge. Analyser operation and data storage was managed on a laptop running AIM v9.0.0.0, which was used to control the operation of the TSI Model 3080/ Model 3775 setup.
- At LHR2, TSI Model 3082 with long DMA (Model 3081) classifier and soft X-ray neutraliser. Automatic on-board software correction was enabled for diffusive losses and multiple charge. Analyser operation and data storage controlled by the Model 3082 running AIM v10.1.0.6. Data was downloaded weekly from the 3082 to a USB stick for subsequent analysis.

The operating methodology of the TSI Scanning Mobility Particle Sizer (SMPS) and Condensation Particle Counter (CPC) has been extensively described in literature, for example by Wiedensohler et al. (2012) and Wiedensohler et al. (2018). The only difference from the recommendations of Wiedensohler et al. (2012) was the absence of a dryer.

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.

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

157 establish remote communication to the analysers. The monitoring stations were visited weekly to
158 ensure correct operation and take remedial action if required.
159
160 The Heathrow CPCs and SMPSs were calibrated before and after the monitoring campaign at the
161 ISO/IEC17025 accredited Ricardo Energy and Environment (REE) calibration laboratory in
162 Harwell, UK. The classifiers and CPC were calibrated using a Jing miniCAST model 6003 (Jing
163 Ltd, Zollikofen, Switzerland) soot generator, which creates particles using a controlled burn
164 propane flame. The results of these calibrations showed both CPCs were accurate to within 1% of
165 the reference device and the SMPSs were able to size particles within 1 size bin in the range 14.6nm
166 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 (https://uk-air.defra.gov.uk/interactive-map). 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:
173 • Butanol based TSI 3775 Condensation Particle Counters (CPC)
174 • TSI 3080 Scanning Mobility Particle Sizer (SMPS) with long DMA classifier and Kr-85
175 neutraliser source
176 • Nafion dryer
177 • Laptop running AIM v9.0.0.0
178 The SMPS / CPCs in the national network are also configured to sample in the range 14.6nm to
179 661.2nm. Sampling is also programmed to run for 3 minutes, sweeping up in size for 2 minutes 15
180 seconds, and returning down for the remaining 45 seconds. This will allow measurements between
181 the airport and national network analysers to be directly and robustly compared with each other.

The authors believe that this is the first time such a robust concurrent comparison has been made between UFP measured at airports and background locations.

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.

There were two main differences between measurements made at Heathrow and those made in the 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 to deploy. Comparison studies within CEN TC264 WG32 and ISO/TC24/SC4/WG12 (standards in development), show that the measurement differences between particles neutralised with soft X-rays and those neutralised with beta radiation are negligible, adding further confidence that the X-ray and beta radiation neutralisers behave in a reasonably similar manner. Additionally, the calibration of the Heathrow analysers at REE was undertaken using a Kr-85 neutraliser for the reference device. The close agreement of the Heathrow analysers, for both counting and sizing, reinforces the confidence that field measurements are valid.

2.4 Data Analysis

The plots and analysis undertaken in this paper make extensive use of the R and R Studio programs (R Foundation for Statistical Computing, Vienna, Austria, and R Studio Inc, MA, USA) and the OpenAir suite of analysis tools (Carslaw and Ropkins, (2012))

Unless specifically stated, particle number plots are aggregated into three size groups, defined here as:

- Nucleation (particles smaller than 25nm),
- Aitken (particles between 26 and 100nm)
- Accumulation (particles larger than 100nm)

Measurements from the black carbon aethalometers are reported here from two of the seven components:

- Black Carbon (BC) – the Particulate Matter concentration recorded from the attenuation of light by particles in the infra-red spectrum at 880nm
- Ultra Violet Particulate Matter (UVPM) – defined here as the additional particulate matter concentration recorded from the attenuation in the UV region of the spectrum. It is calculated from the difference between the concentration recorded at 370nm and the concentration recorded at 880nm using a wavelength-adjusted absorption coefficient. Some other studies have referred to this variable as "Brown Carbon" or Delta-C and interpreted it as a measure of wood smoke concentrations (e.g. Wang et al. (2011)):

$$UVPM = Conc_{ATT\,370} - Conc_{ATT\,880} \quad (1)$$

2.5 Measurement Quality Assurance and Quality Control

It is essential for the data collected in a measurement campaign to have clearly defined provenance. Without descriptions of methodology, stated levels of accuracy, precision, harmonisation and measurement uncertainty, it is extremely difficult to make meaningful comparisons between different datasets and research. This was explored in Stacey (2019), where it was clear that, historically, different UFP studies used a range of instrumentation, setups and calibration methodologies, meaning only qualitative comparisons between them was realistically possible. Wiedensohler et al. (2012) and Wiedensohler et al. (2018) emphasize the need for robust quality control and standardised measurement methodologies; the Heathrow study reported here uses quality assurance and quality control procedures that ensure consistency and comparability in UFP data collection between the Heathrow and national network datasets.

For measurements of NO_x, PM_{10}, $PM_{2.5}$, BC and meteorology, the measurements at Heathrow are managed, collected and processed following guidance described in https://uk-air.defra.gov.uk/assets/documents/reports/cat09/1902040953_All_Networks_QAQC_Document_2012__Issue2.pdf. Information about the analysers used at Heathrow is provided in the Supplemental Information, Tables S1 and S2.

3. RESULTS

3.1 Overall Summary

Timeseries data for the hourly measurements of particle number concentrations at LHR2 and Oaks Road are presented in Supplemental Information, Figures S1, S2. Measurements of NO_x, PM_{10}, $PM_{2.5}$ and BC are also fully reported (Figures S3 – S8) and accessible through the http://heathrowairwatch.org.uk webpages. Data from these analysers will be used to explore associations and differences to typical ambient environments, but not considered in detail.

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

Pollutant	Mean	Median	Standard deviation	Min-Max (15 min data)	Data capture %
NO, ppb	46.9	30.0	56.7	0 – 540	100
NO_2, ppb	27.7	27.9	11.7	1 – 84	100
PM_{10}, ug/m^3	16.8	12.4	17.2	0.7 – 346.5	100
$PM_{2.5}$, ug/m^3	10.9	7.2	14.4	0.4 – 288.3	100
BC, ug/m^3	3.11	2.30	2.79	0.08 - 28.08	100
UVPM, ug/m^3	0.84	0.49	0.96	0.03 – 11.41	100
Nucleation, dN/dlog Dp	7817	1871	15993	42 – 150000	87.6
Aitken, dN/dlog Dp	8638	5542	9704	93 – 107918	87.6
Accumulation, dN/dlog Dp	2088	1570	2110	70 – 30052	87.6
Total PN, #/cm^3	8911	4756	12014	394 – 118726	87.6

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

Pollutant	Mean	Median	Standard deviation	Min-Max (15 min data)	Data capture %
NO, ppb	23.1	10.3	35.1	0 – 328	99.8
NO_2, ppb	19.5	19.1	10.3	0 – 68	99.8
PM_{10}, ug/m^3	14.3	11.0	12.5	0.8 – 186.6	100
$PM_{2.5}$, ug/m^3	10.2	6.9	11.1	0.4 – 172.4	100
BC, ug/m^3	1.77	1.20	1.86	0.01 – 26.08	100
UVPM, ug/m^3	0.55	0.37	0.71	0.01 – 6.83	100
Nucleation, dN/dlog Dp	8476	2152	12064	0 – 86287	50.0
Aitken, dN/dlog Dp	7798	4723	8223	0 – 63372	50.0
Accumulation, dN/dlog Dp	1639	1370	1146	0 – 10280	50.0
Total PN, #/cm^3	7408	3948	8180	0 – 62124	50.0

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

Data for the first week of UFP measurements at LHR2 were rejected due to a software configuration error. 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 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 weekly calibration visits to the stations.

Examination of historic NO_x, PM and BC data at LHR2 and Oaks Road (available from http://www.heathrowairwatch.org.uk/reports) has shown that measured concentrations and profiles for NOx, PM mass and black carbon are not significantly different to those measured at nearby background and traffic monitoring stations operated by Local Authorities or the national network stations in London.

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.

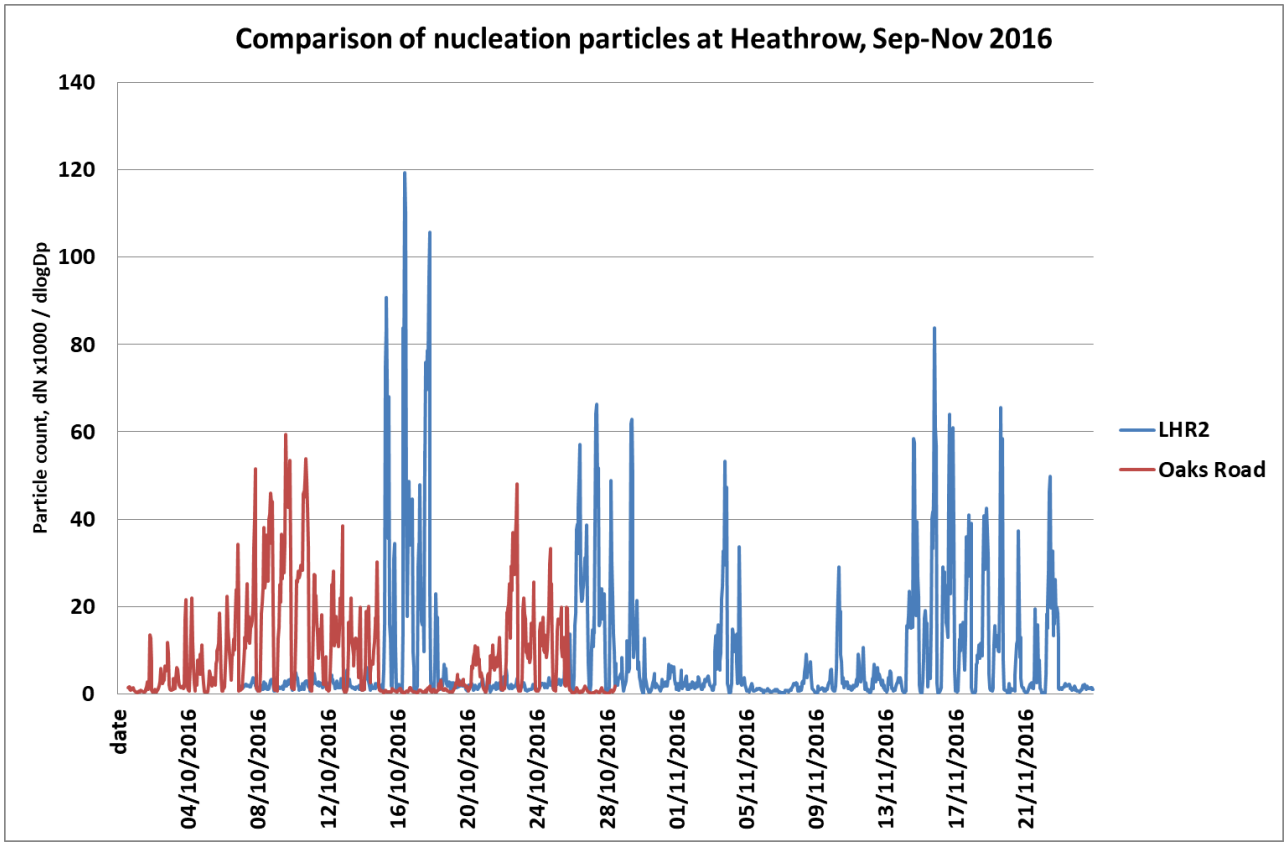


Figure 2. Time series of nucleation mode particles at the Heathrow Airport sampling sites, October and November 2016

290 It is clear from this plot that high concentrations of nucleation mode particles are only observed at
291 one location at a time, so wind direction is a critical factor in the presence and concentration of
292 these particles.
293
294 Examination of LHR2 and Oaks Road particle number diurnal data averaged for the entire survey
295 (presented in Figure S9 and S10) shows enhanced PN concentrations between 06:00 and 23:00,
296 coinciding with typical increased activity around the airport. The diurnal plots also show close
297 agreement with the two humped diurnal profiles of the NO_x and BC pollutants, suggesting that
298 these pollutants mostly share common sources, including road traffic and commercial / domestic
299 energy use. The PM_{10}, $PM_{2.5}$ and accumulation mode particle diurnal plots do not follow the exact
300 same pattern as NO_x, so likely originate from different sources to NO_x. The accumulation mode PN
301 appears to follow a similar trend to the PM_{10} and $PM_{2.5}$ diurnal profiles, but it is relatively flat and
302 significantly lower in number concentrations when compared to the Nucleation and Aitken mode
303 PN datasets. The diurnal plots for Nucleation and Aitken mode particles do not follow the trends
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**

307 Measurements of UFP were coincident at LHR2 and Oaks Road for the period 7 – 28th October
308 2016. As noted earlier, there are three measurement stations within the UK national monitoring
309 network (https://uk-air.defra.gov.uk/interactive-map) that measure UFP – these stations were also
310 all in operation during this time. A summary of average concentrations measured at all 5 sites is
311 presented in Table 3.
312

Pollutant	Marylebone Road	North Kensington	Chilbolton	LHR2	Oaks Road
NO, ppb	80.4	9.1	1.6	43.9	21.8
NO_2, ppb	39.2	18.9	8.4	27.5	20.7
PM_{10}, ug/m^3	21.6	17.1	13.4	15.9	13.8
$PM_{2.5}$, ug/m^3	12.8	11.1	7.3	9.5	9.4
BC, ug/m^3	3.787	0.912	0.620	2.901	1.792

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

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

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.

Airport PM_{10} and $PM_{2.5}$ concentrations are slightly lower than seen in London, but different measurement techniques are deployed, which may account for some of the differences. The Heathrow sites use Fidas 200 analysers, while TEOM1400/FDMS8500 units were deployed at the national network stations. There is ongoing work, in preparation for the UK Environment Agency by Ricardo, Bureau Veritas and Kings' College London, discussed at a number of seminars, for example http://www.scottishairquality.scot/assets/documents/reports/9_PM_analyser_replacement_Brian_Stacey.pdf, that suggests that there are differences in instrument signal performance that accounts for 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 any conclusions about observed differences.

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

339 protocols for the AE22 aethalometer published by Virkkula et al (2007) and Apte et al (2011) have
340 found that significant differences in ”corrected” concentrations are observed. It can therefore be
341 justifiably argued that neither method for attenuation correction can be guaranteed to give data
342 comparable to that produced by the AE33 aethalometer, where no correction for attenuation is
343 required.
344
345 Averaged particle number concentrations, calculated by summing all of the particle counts from the
346 SMPS analysers from 14.6nm to 661.2nm, shows that a similar trend is observed to other
347 pollutants: concentrations at the airport locations fall between the traffic and background
348 measurements made at the London stations.
349
350 On initial investigation of the measurement datasets therefore, the ambient air environment at
351 Heathrow appears reasonably similar to the rest of London. However, data from the SMPS
352 analysers also provides valuable information about the particle size distribution at all 5 locations. It
353 has already been demonstrated in earlier research that nucleation mode particles are strongly
354 associated with airport activity. The plot in Figure 3 shows the average particle size distribution at
355 each station for the period when all 5 SMPS were operational; the period between 7 and 28th
356 October 2016.
357
358

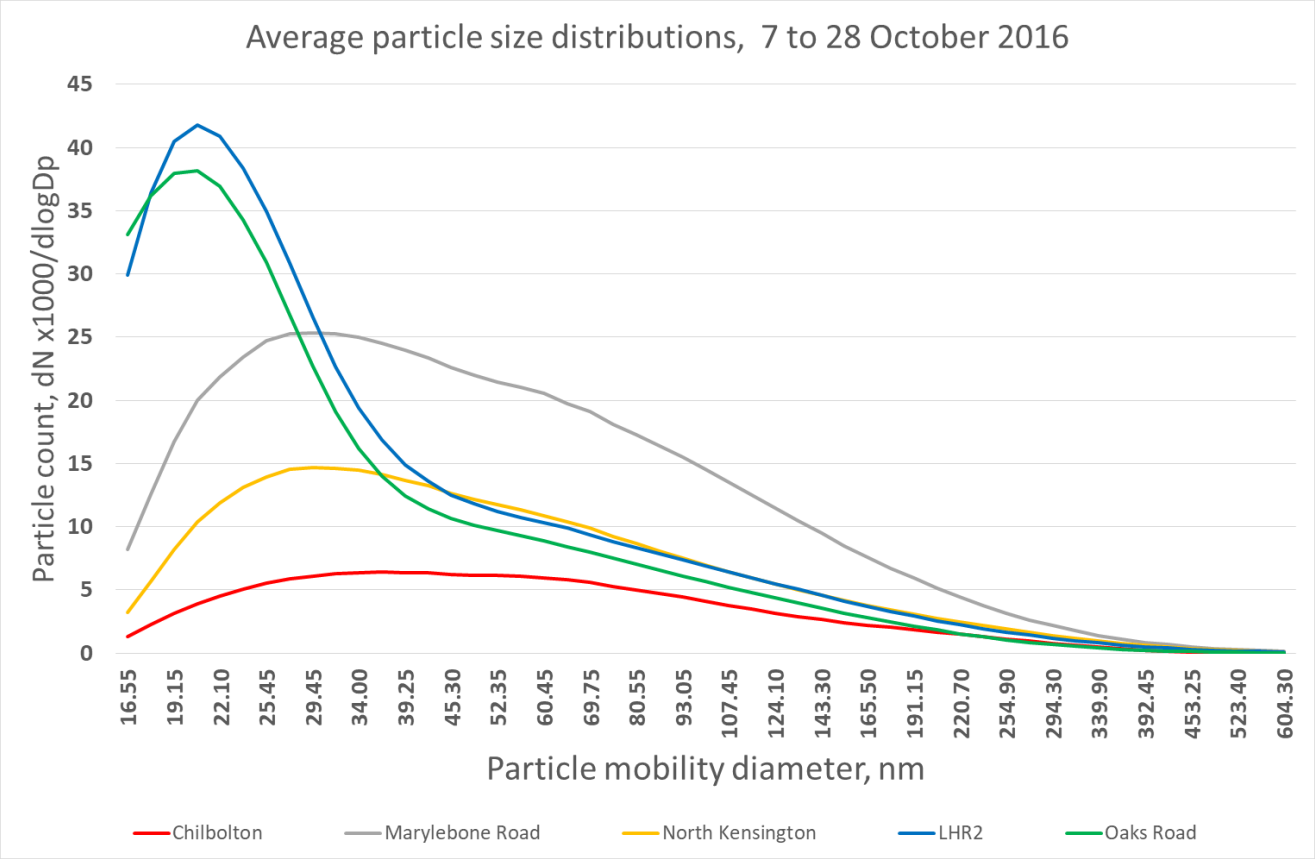


Figure 3. Comparison of particle size distributions at 5 locations

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. 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 Chilbolton, suggesting that these larger particles are more background in nature than the 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.

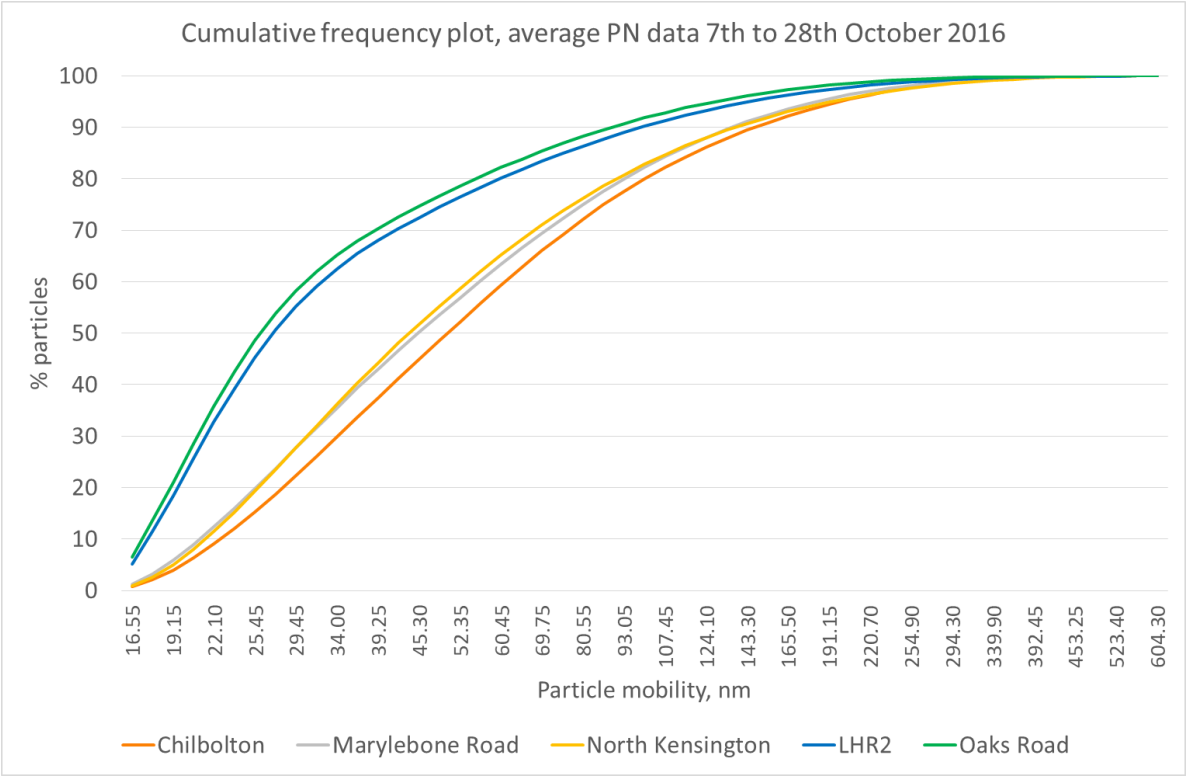


Figure 4. Cumulative particle size distributions at the five sites.

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.

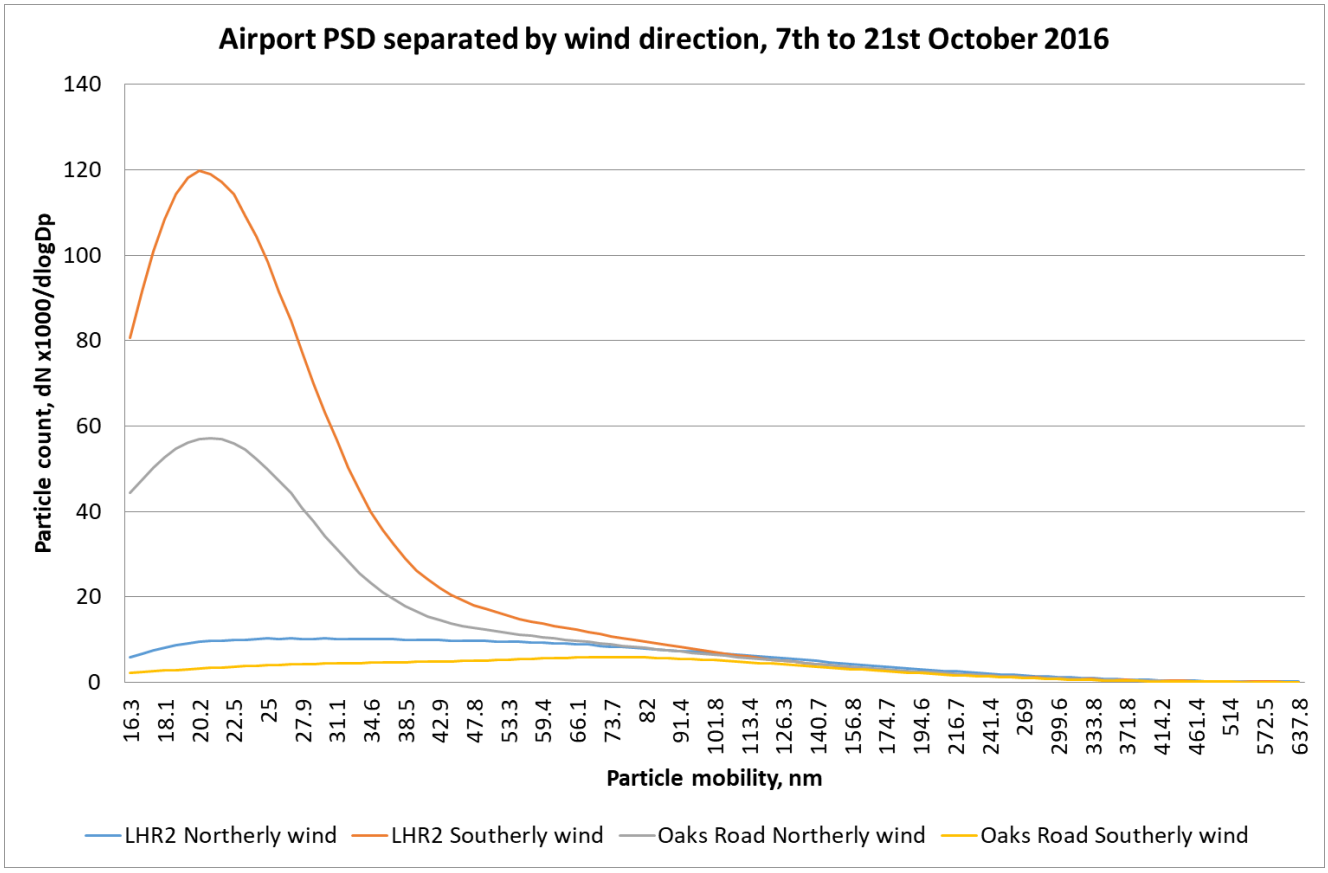


Figure 6. Airport PSD separated by wind direction for the Heathrow Airport sites. Note that the particle mobility axis differs slightly from earlier figures due to the need to align Heathrow particle size concentrations exactly with the data provided by the national network stations.

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

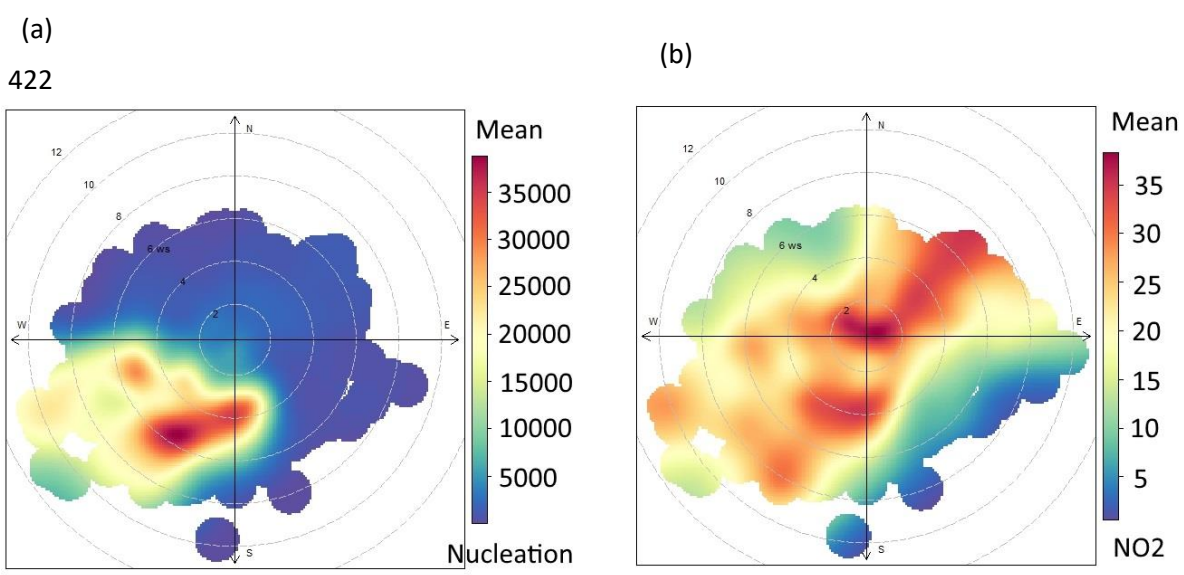
415 ### 3.3 Dependence of Airport Measurements on Meteorology

416 Meteorological measurements made at Heathrow allow for further analysis of the data using the

417 polarPlot function in OpenAir. The plots in Figure 7 examine the dependence of measurements on

418 wind speed and direction.

419



422

423

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

425

426

427 The plots for nucleation mode (Figure 7(a)) and Aitken mode (Figure S11) mode particles show a

428 very strong influence from the airport, to the south and west of the measurement station. The

429 nucleation mode particles plot shows very little influence from other directions, clearly pointing to

430 airport activities as the dominating source of these particles at this location.

431

432 NO_2 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 station are all seen to influence the polar plot.

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

Similar patterns are seen at Oaks Road (Figure 8).

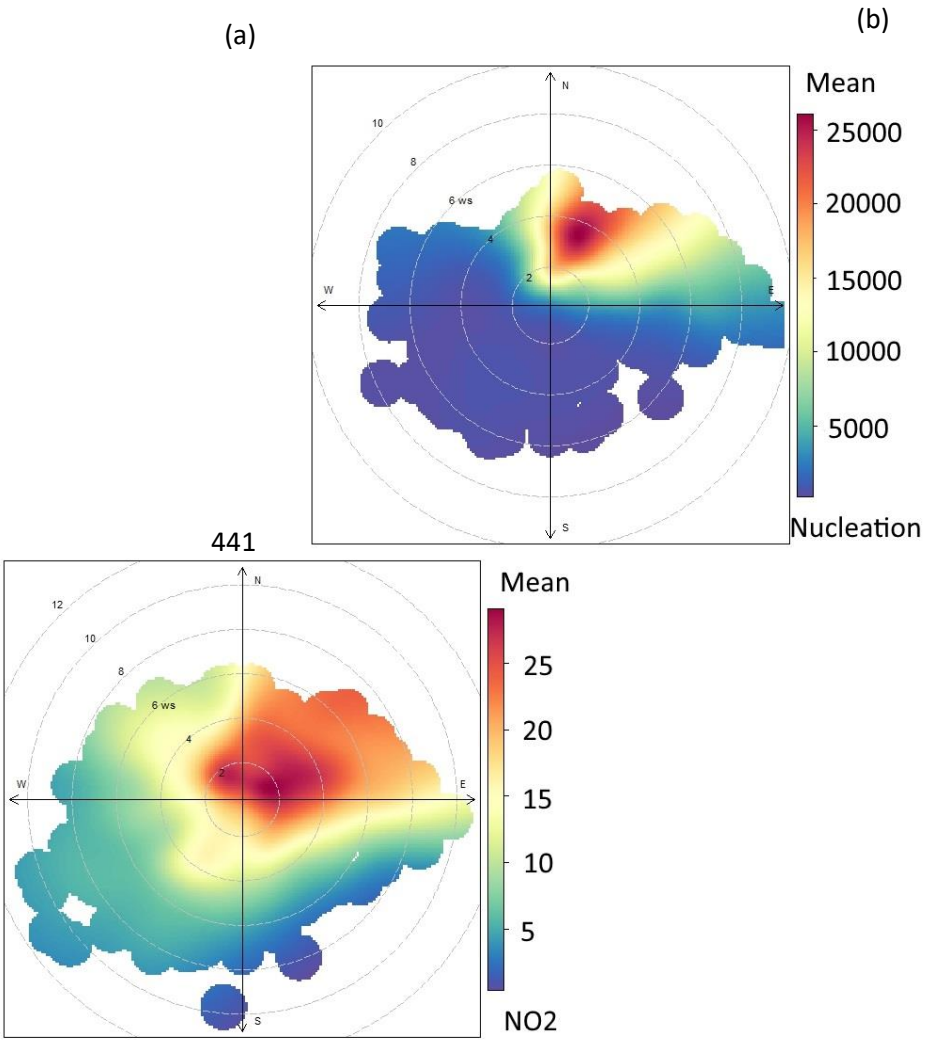


Figure 8. Polar plots for Oaks Road, (a) nucleation mode particles, (b) nitrogen dioxide

The plots show that high concentrations of nucleation mode particles, as well as NO_2, 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 **3.4 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.

Access to aircraft arrival and departure information, provided by Heathrow Airport Limited, allows 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.

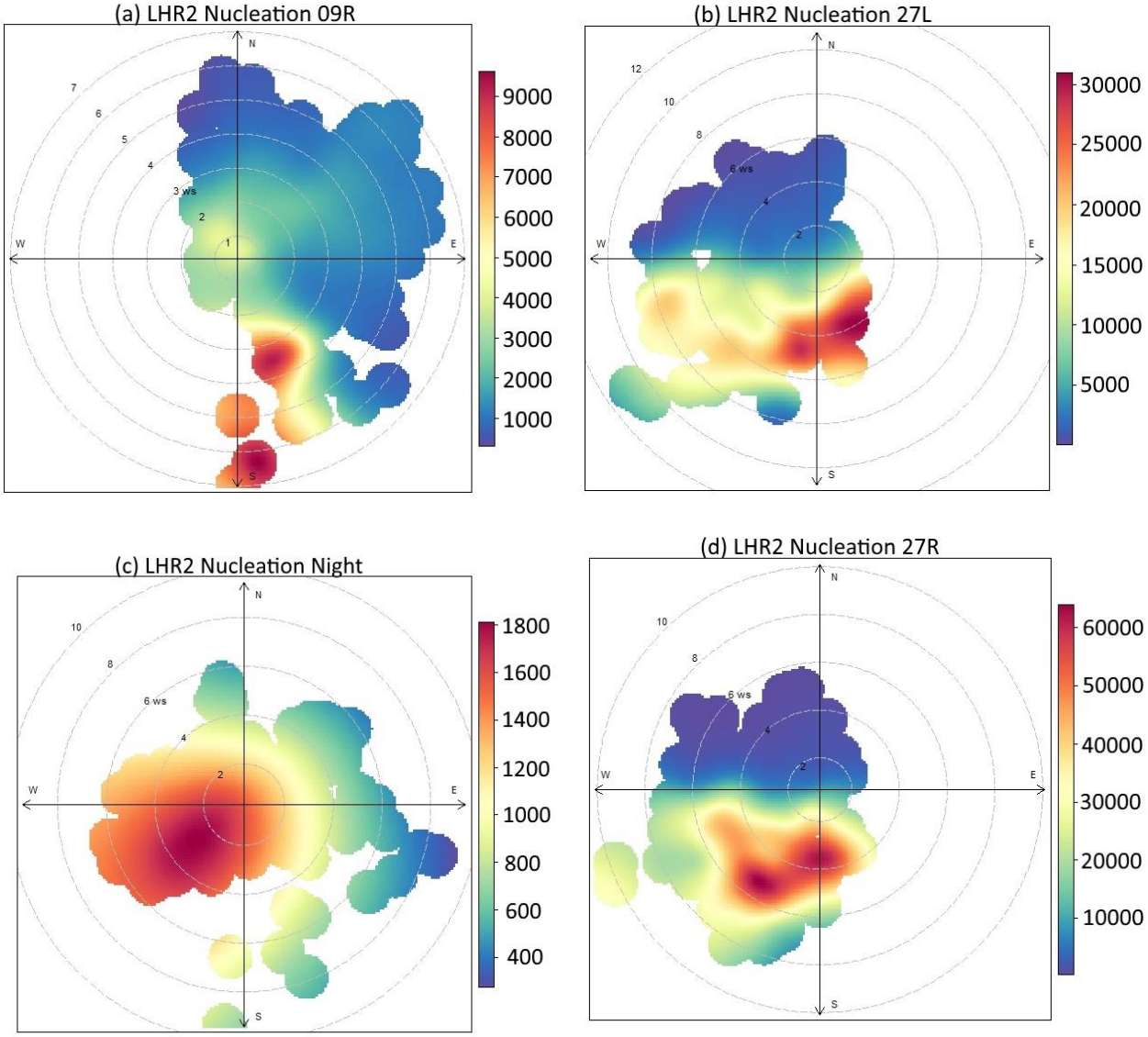


Figure 9. Polar plots of nucleation mode particles at LHR2, split by runway mode, (a) 09R, (b) 27L, (c) Night, (d) 27R. Each sub plot has different maximum concentrations defining the colour scales.

It is clear that nucleation mode particle number concentrations are highest when aircraft depart from 27R (closest to the monitoring station) and lowest when the airport is closed overnight. Nucleation 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 clear peak in nucleation mode particles can still be seen from the airfield, presumably from departing aircraft exhaust – arriving aircraft leave the runway before they are within 1km of the LHR2 monitoring station and are thus not expected to significantly influence measurements during easterly winds. Overnight concentrations of nucleation particles are generally comparatively very low, but still appear to be associated with winds from the airfield.

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.

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.

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

519 particles when aircraft are departing from runway 27R. A similar link is not obvious when aircraft
520 are landing on 27R, 09R or indeed any other aircraft operating modes at the airport, suggesting that
521 high thrust exhaust emissions may be associated with production of black carbon particles that
522 strongly attenuate UV light. In contrast (in Figure S15), UVPM at Oaks Road is dominated by
523 association with low wind speeds. There is some indication of a contribution from the direction of
524 the airport, but it is likely that a number of different sources contribute to measurements in this
525 residential location.
526
527 For Oaks Road, a similar picture emerges (plots presented in Figure S15). Highest concentrations
528 of nucleation mode particles are associated with aircraft departing from 09R, closest to the
529 monitoring station, but high concentrations of nucleation mode particles in other polar plot modes
530 clearly also originate from the airfield.
531
532 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**
536 The monitoring station at LHR2 is 170m from the centre of the northern runway. Under favourable
537 meteorology, plumes from aircraft departing and landing impact on the monitoring station, raising
538 the possibility that these plumes can be further analysed and characterised by, for example, aircraft
539 type, engine type, aircraft landing and aircraft departing.
540
541 On average (https://www.heathrow.com/file_source/Company/Static/PDF/Investorcentre/Heathrow-
542 (SP)-FY2016-results-release-(FINAL).pdf), an aircraft departs from the airport every 90 seconds
543 between 06:00 and 23:00 every day. The SMPS/CPC configuration at LHR2 was set to provide a
544 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.

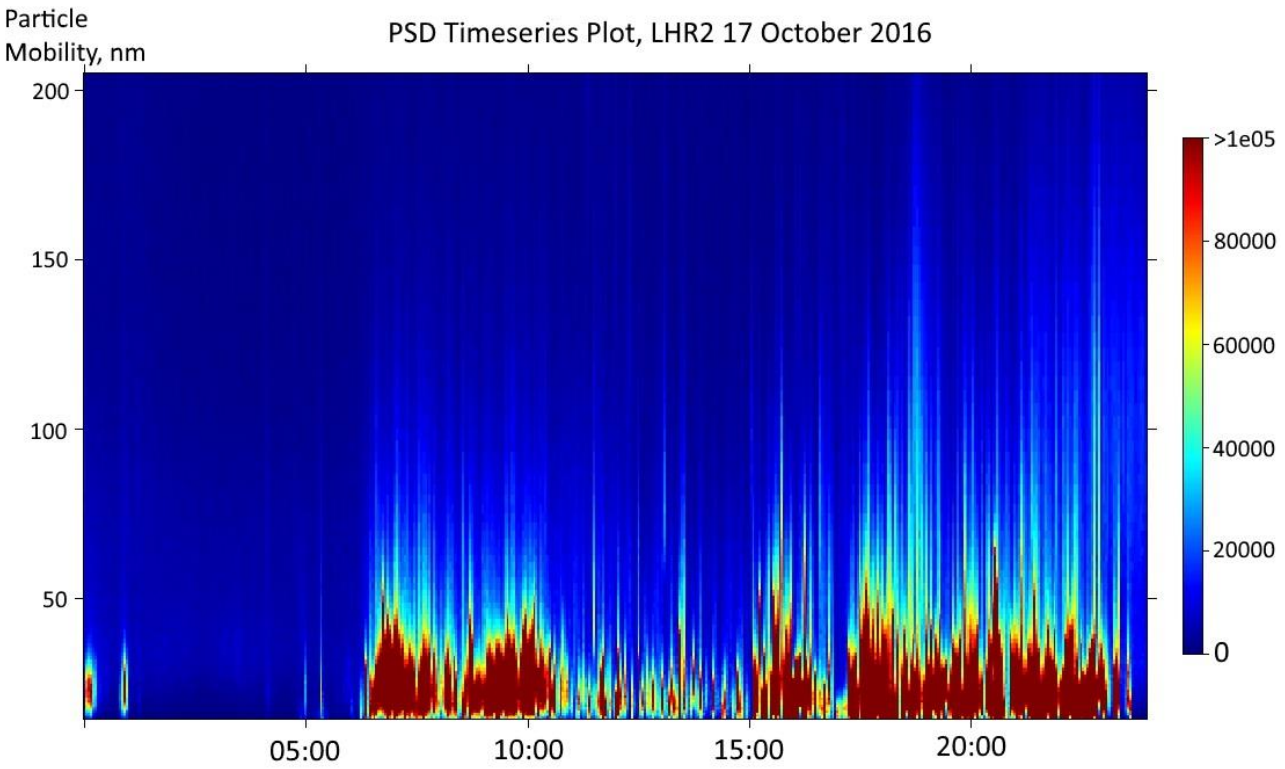


Figure 10. Particle Size Distribution plot, LHR2, 17 October

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

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.

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.

Data collected from this 2016 LHR2 and Oaks Road study were analysed using the PMF5 positive 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 USEPA's own guidance: https://www.epa.gov/sites/production/files/2015-02/documents/pmf_5.0_user_guide.pdf and 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.

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_2, NO_x, BC, UVPM, PM_{10}, and $PM_{2.5}$ were also included in the PMF runs.

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

The model was run for 3 to 10 factor scenarios, with strong relationships set for all pollutants except PM_{10} and $PM_{2.5}$. 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.

The model outputs were examined to check that all factors were unique and that factors had not been subdivided unnecessarily. There was no adjustment required for rotational ambiguity.

At both locations, 5 factors was identified as the optimal number, with factors identified as:

- Airport. Factor dominated by PN <50nm, comparatively low contribution from all other pollutants.
- Fresh road traffic. Factor dominated by high concentrations of NO. Minor contribution also from larger particles.
- Aged road traffic. Factor dominated by NO_2. PN from 30-80nm and PM_{10} also observed
- Biomass PM. Factor dominated by BC, UVPM and particles 80 – 250nm. NOx and $PM_{2.5}$ also observed.
- Background PM. Factor dominated by PM_{10}, $PM_{2.5}$ and particles 80 – 640nm. BC and UVPM also observed.

The plots in Figures 11 and 12 show the base factor profiles for LHR2 and Oaks Road, with factors labelled according to identified sources.

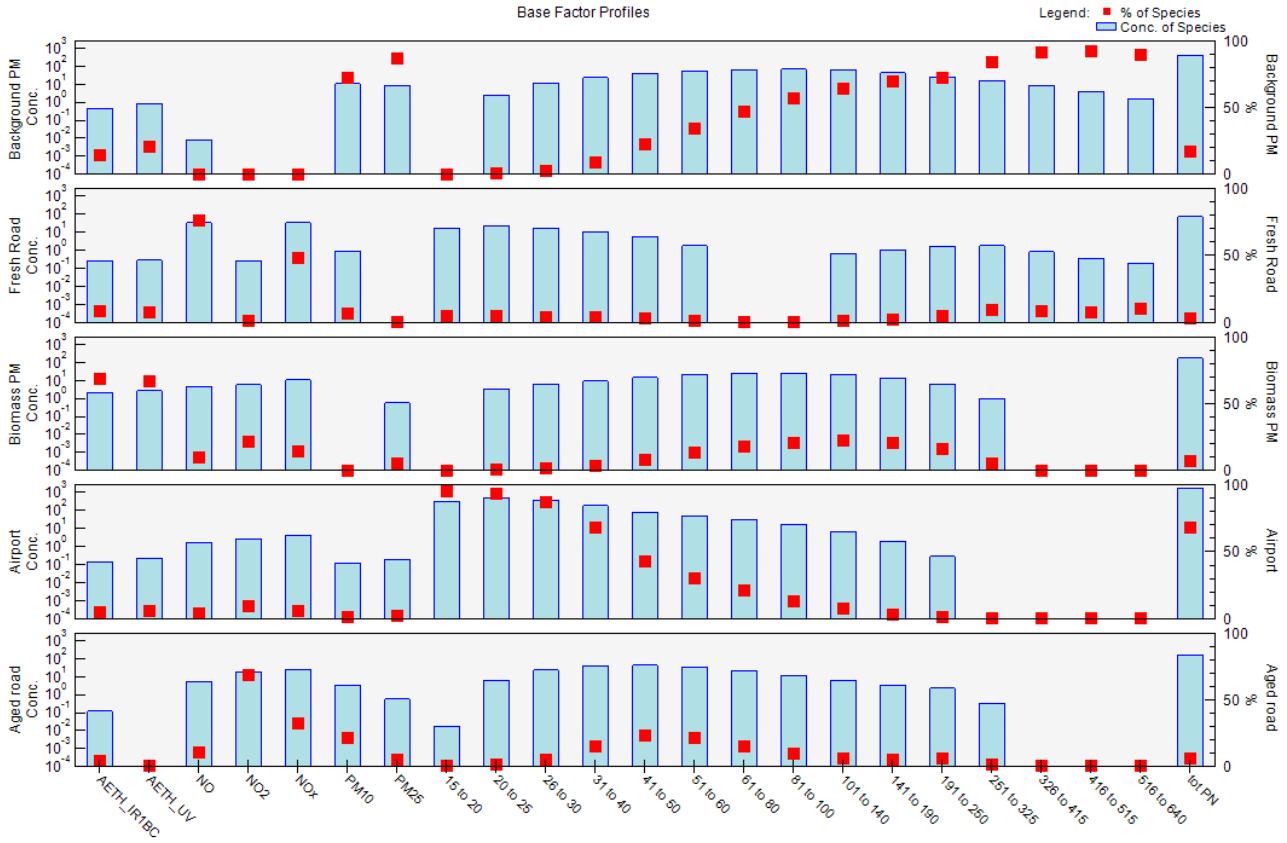


Figure 11. PMF Base Factor Profiles for LHR2 site

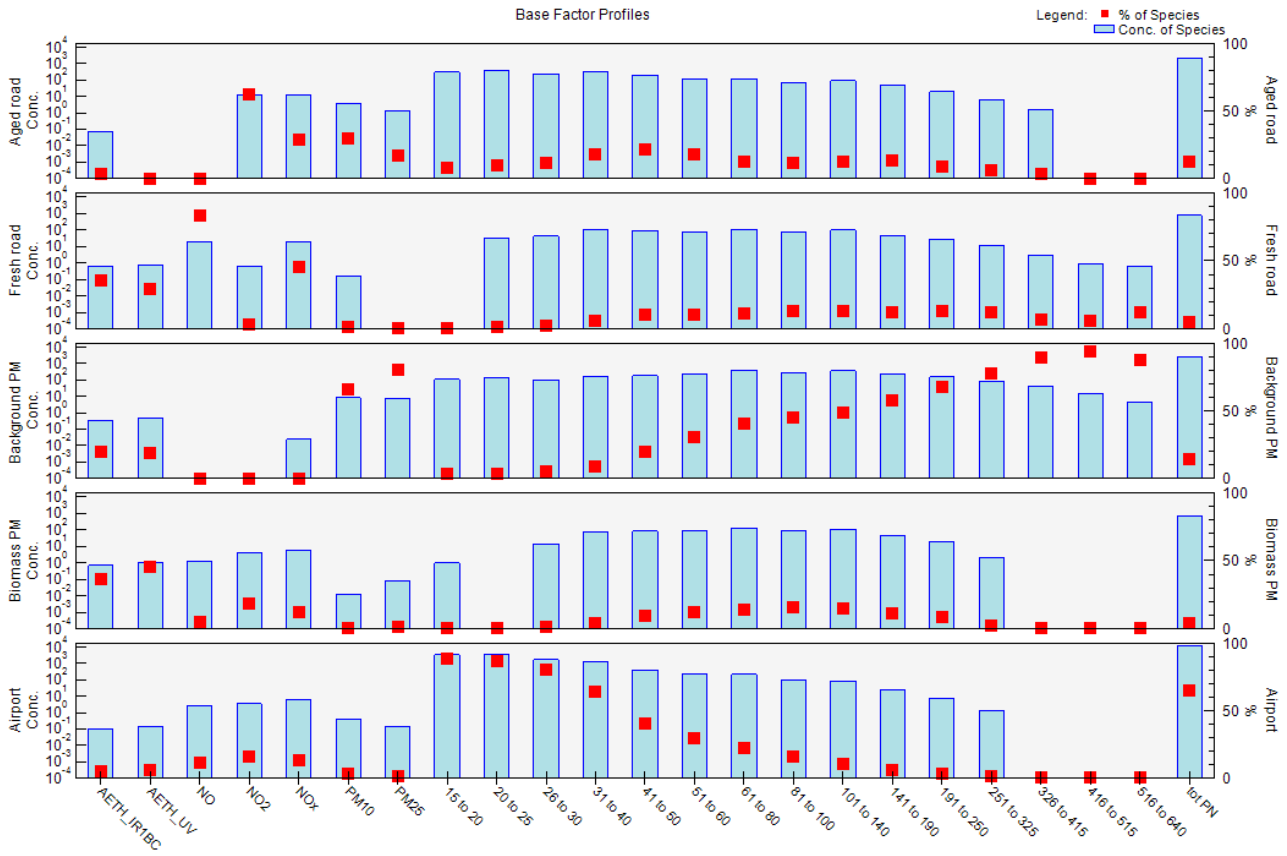


Figure 12. PMF Base Factor Profiles for the Oaks Road site

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_x, 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.

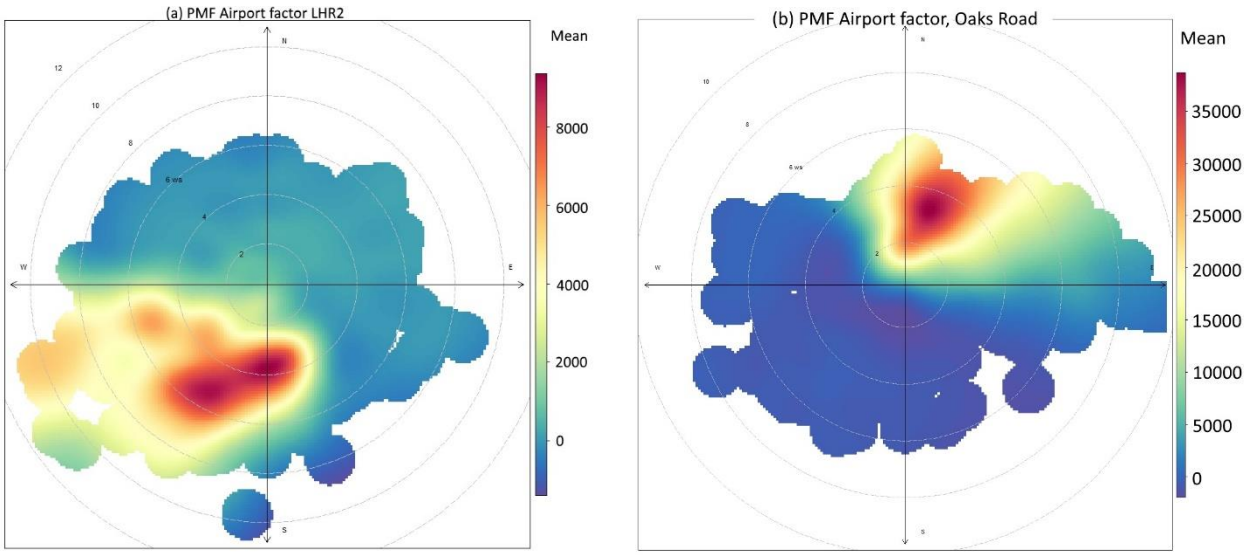


Figure 13. Polar plots of Aircraft PMF factors at (a) LHR2 and (b) Oaks Road

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.

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.

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.

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

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.

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.

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_2.

The Heathrow data were analysed using the USEPA PMF model to separate the contributions into 4 factors. A clear airport component was identified at both locations, where the largest proportion of the factor was associated with nucleation mode particles. Examination of these factor datasets in polar plots showed excellent agreement with the nucleation mode polar plots using data collected from the analysers.

DATA AVAILABILITY

Data supporting this publication are openly available from the UBIRA eData repository at https://doi.org/10.25500/edata.bham.00000349).

ACKNOWLEDGEMENTS

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SUPPLEMENTARY INFORMATION:

Evaluation of Ultrafine Particle Concentrations and Size Distributions at London Heathrow Airport.
Supplemental information.

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

Table S1 – Conventional instrumentation at LHR2 and Oaks Road

Pollutant	Accuracy	Limit of detection
NO	±14.0%	±2ppb
NO_2	±14.0%	±2ppb
PM_{10}	±7.5%	±3µg/m^3
$PM_{2.5}$	±9.3%	±3µg/m^3
BC	±15.4%	±0.1µg/m^3
Particle Number	20%	20 particles /cm^3

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

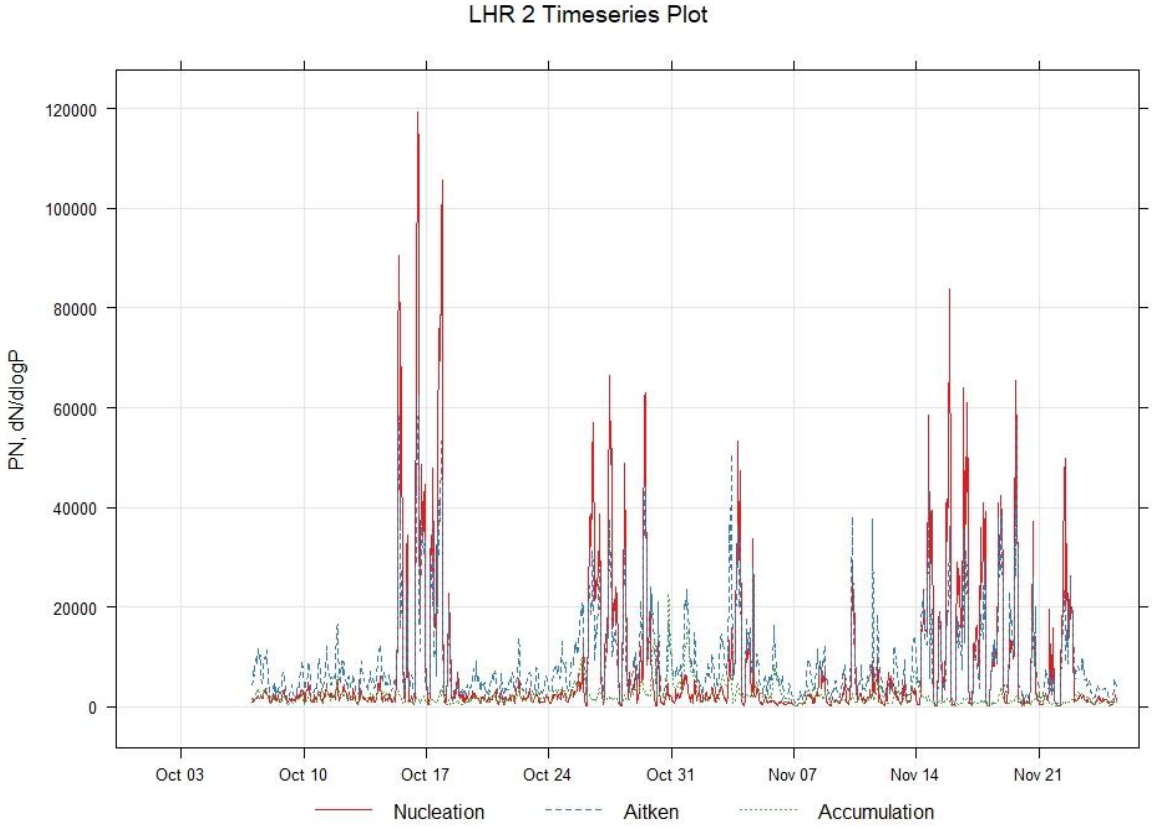


Figure S1 – LHR2 PN timeseries plot

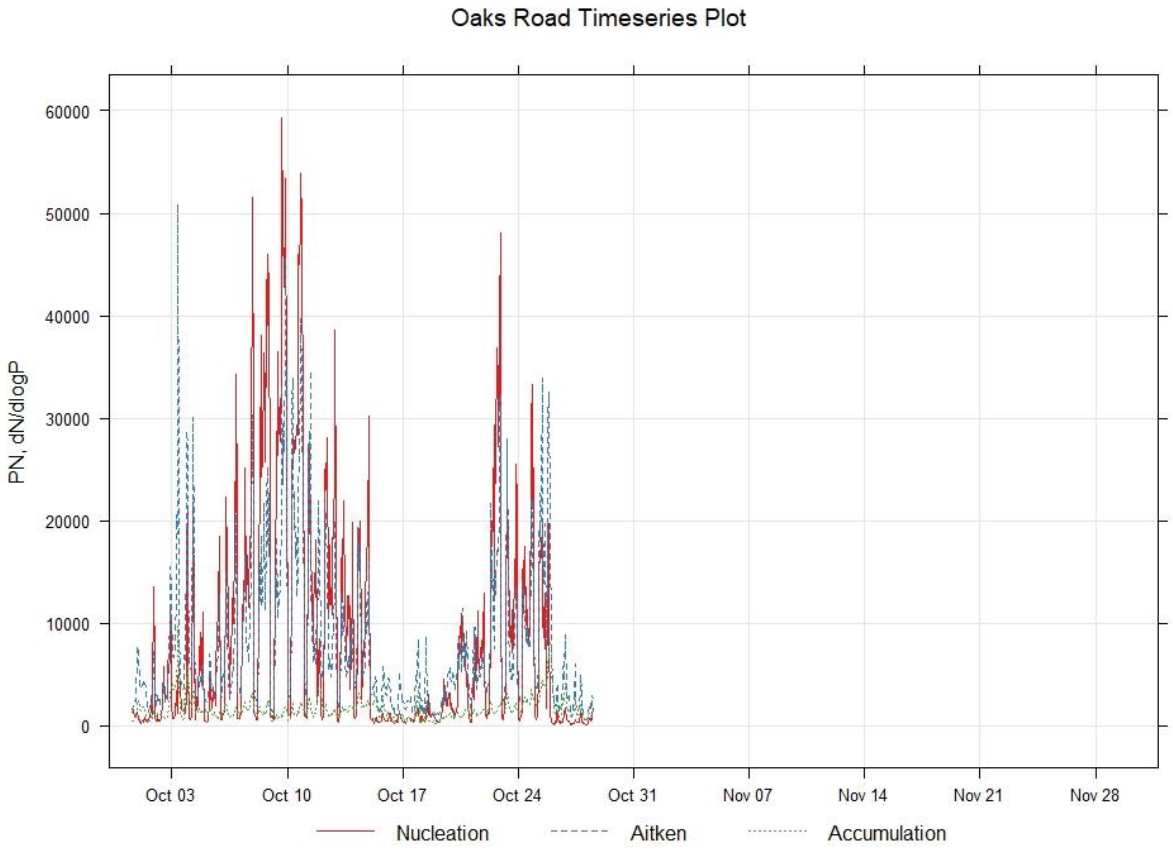


Figure S2 – Oaks Road PN timeseries plot

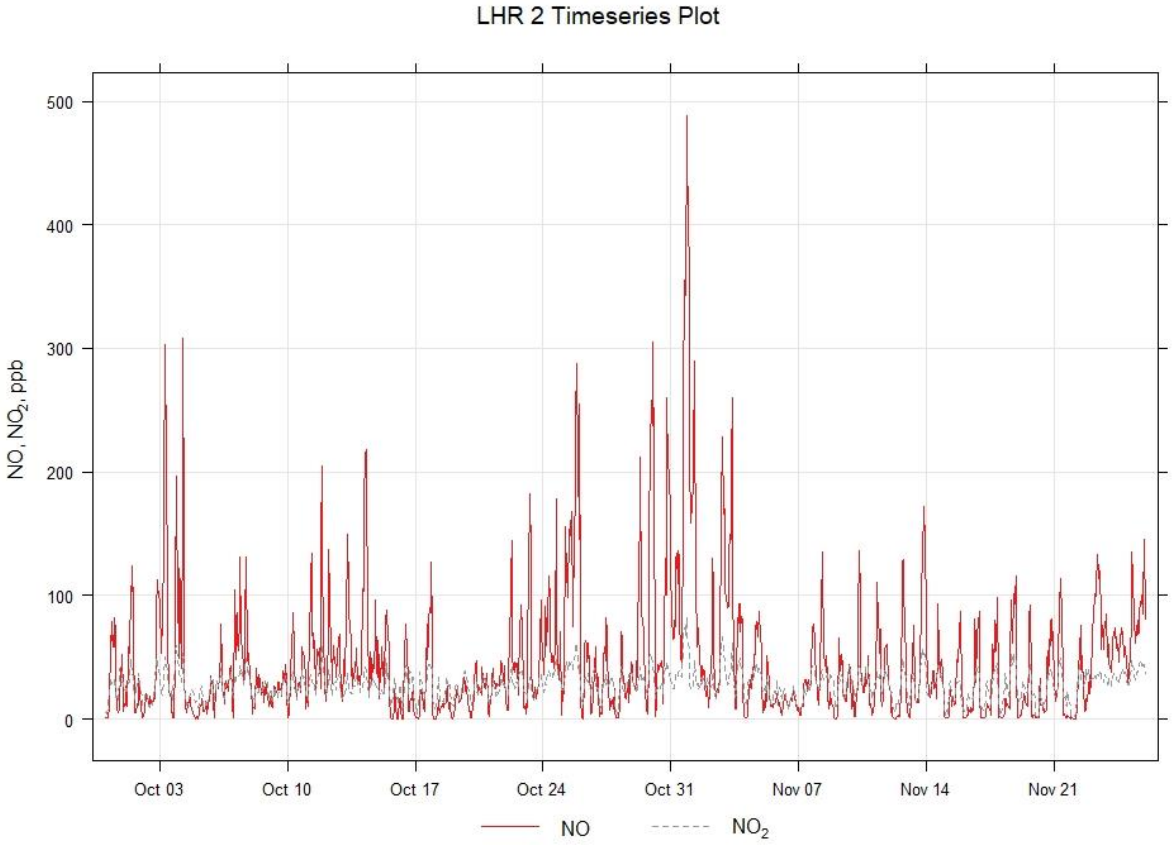
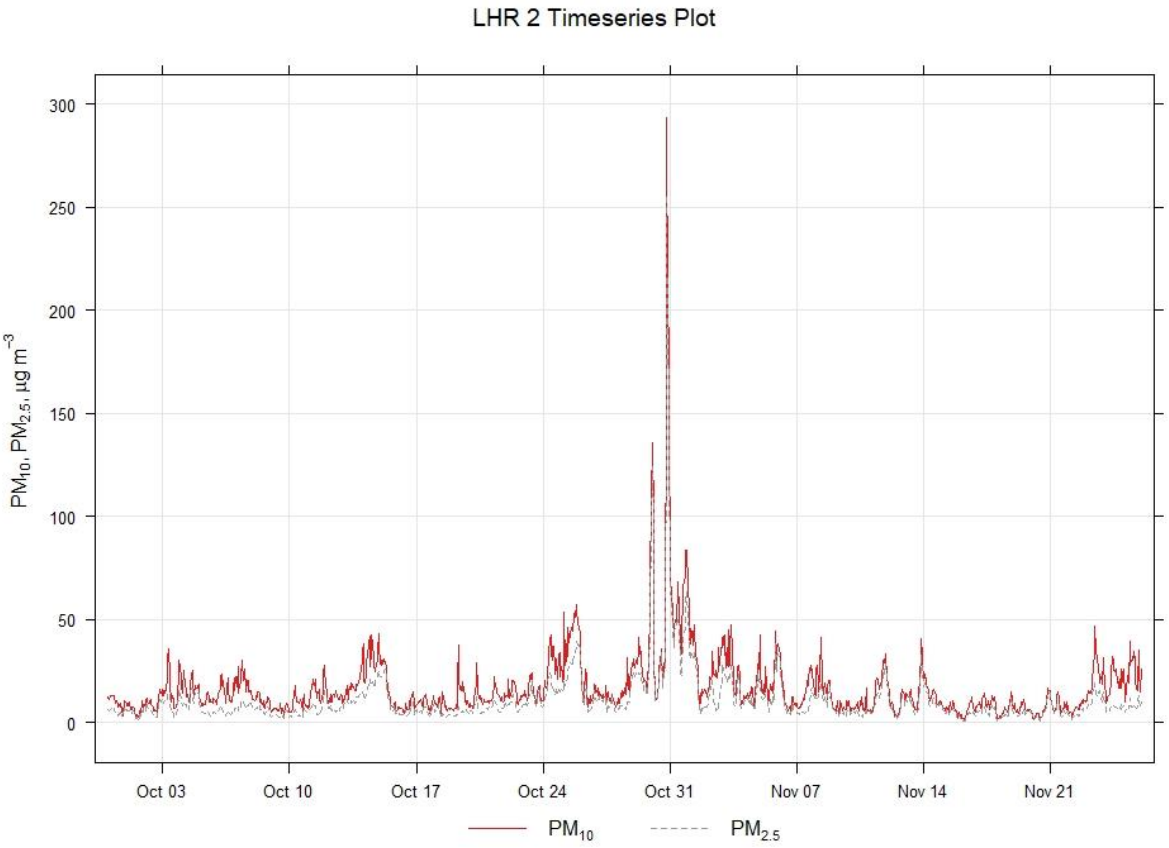
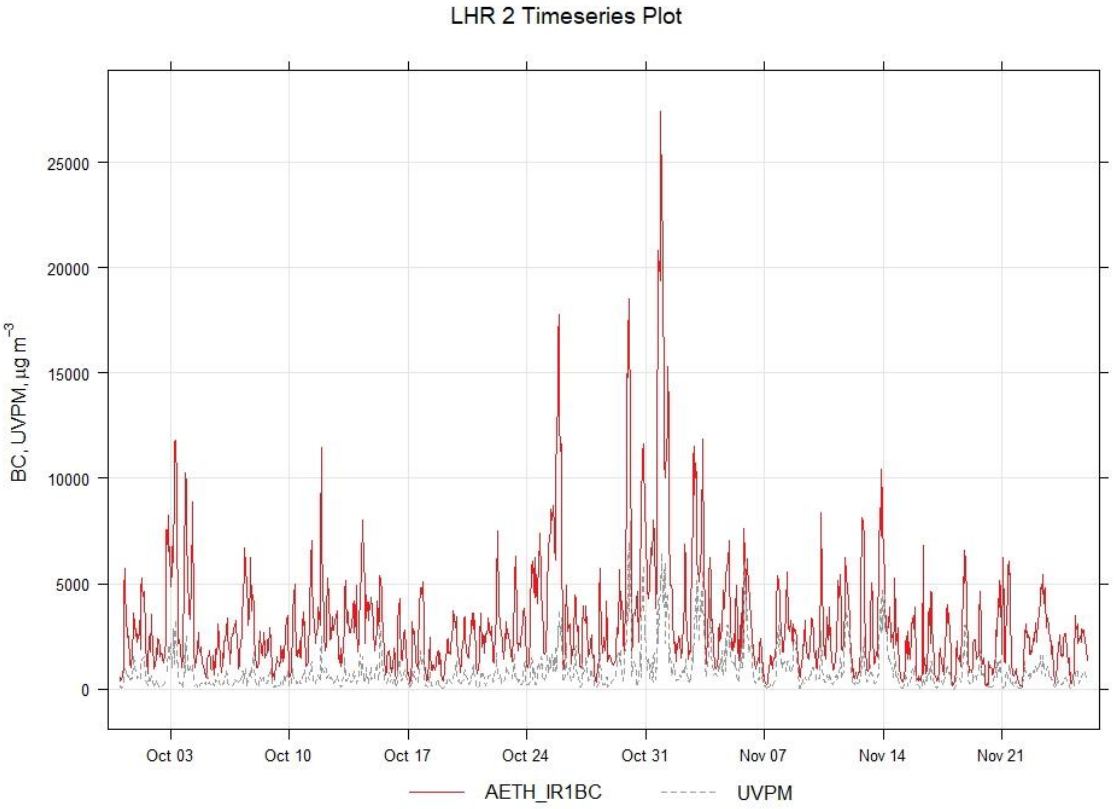


Figure S3 – LHR2 NOx timeseries plot



867
868 Figure S4 – LHR2 PM timeseries plot



869
870 Figure S5 – LHR2 BC timeseries plot

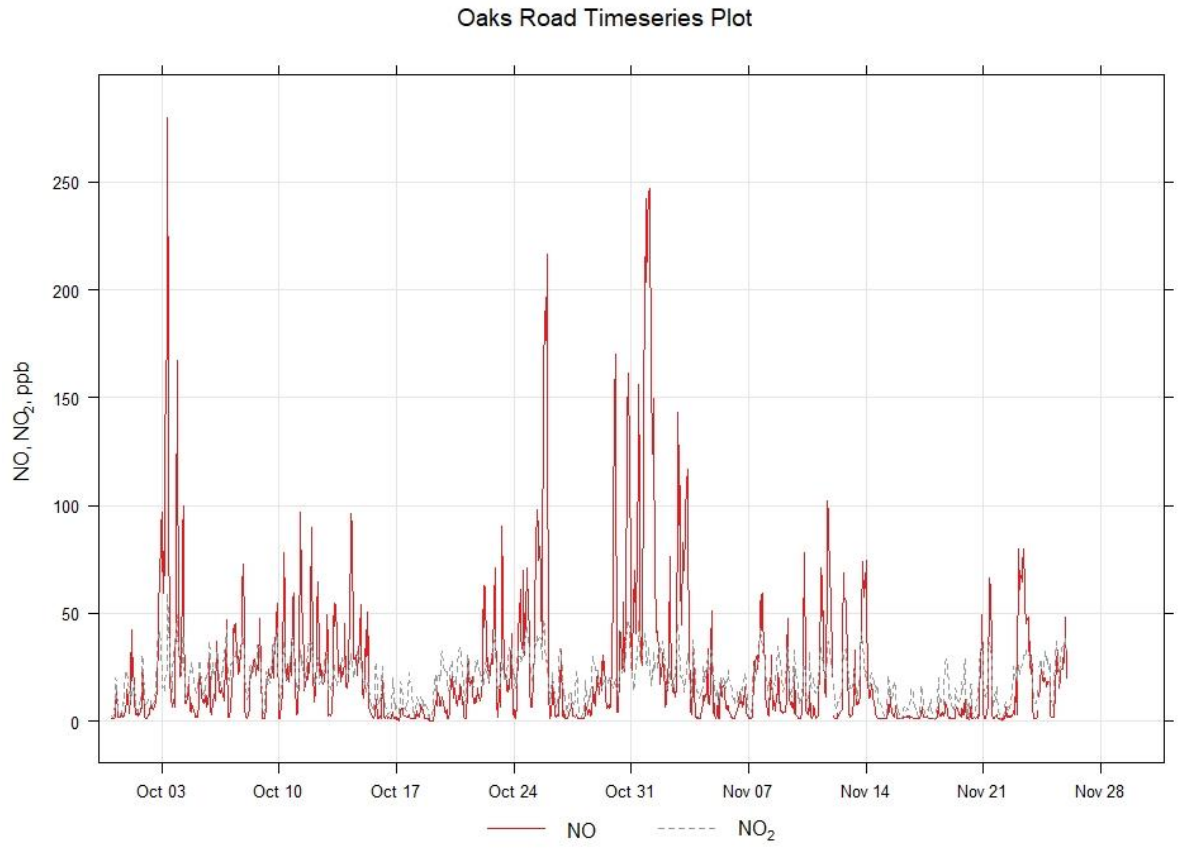


Figure S6 – Oaks Road NOx timeseries plot

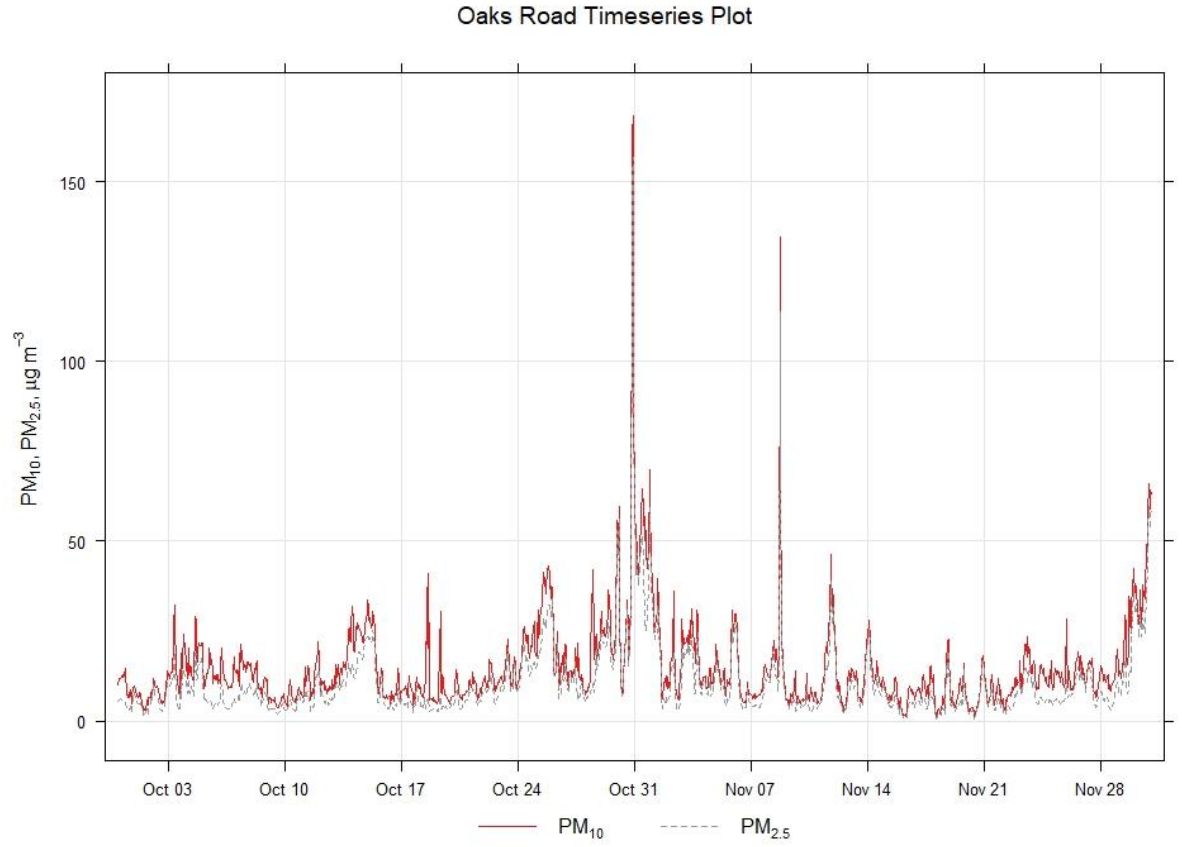


Figure S7 – Oaks Road PM timeseries plot

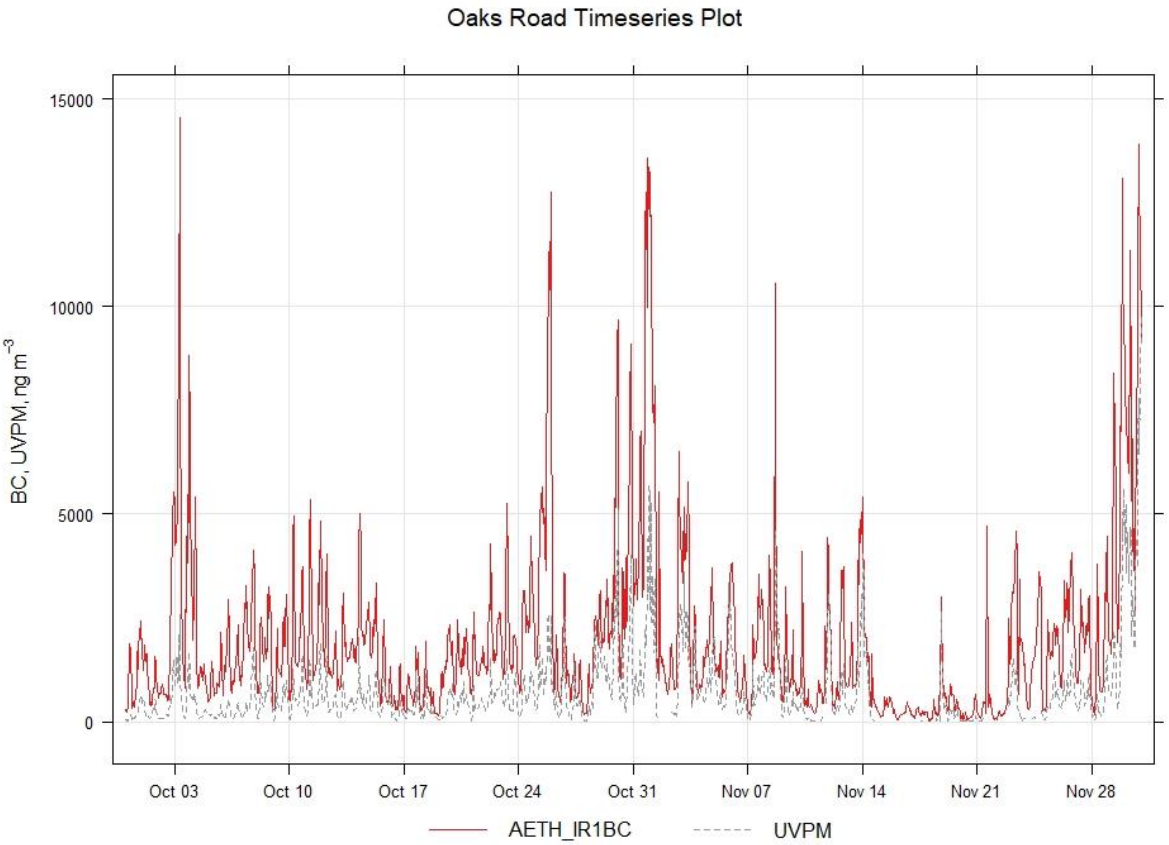
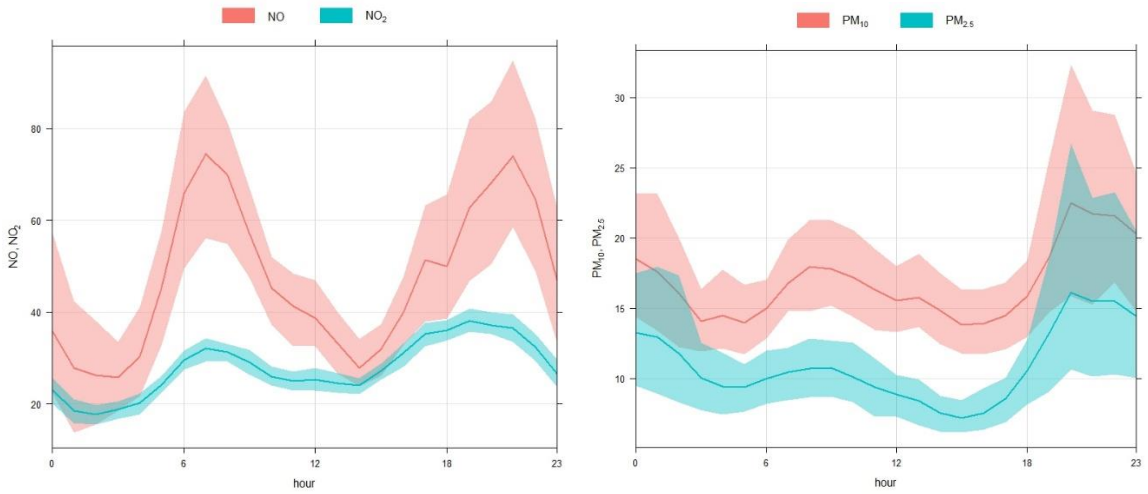


Figure S8 – Oaks Road BC timeseries plot



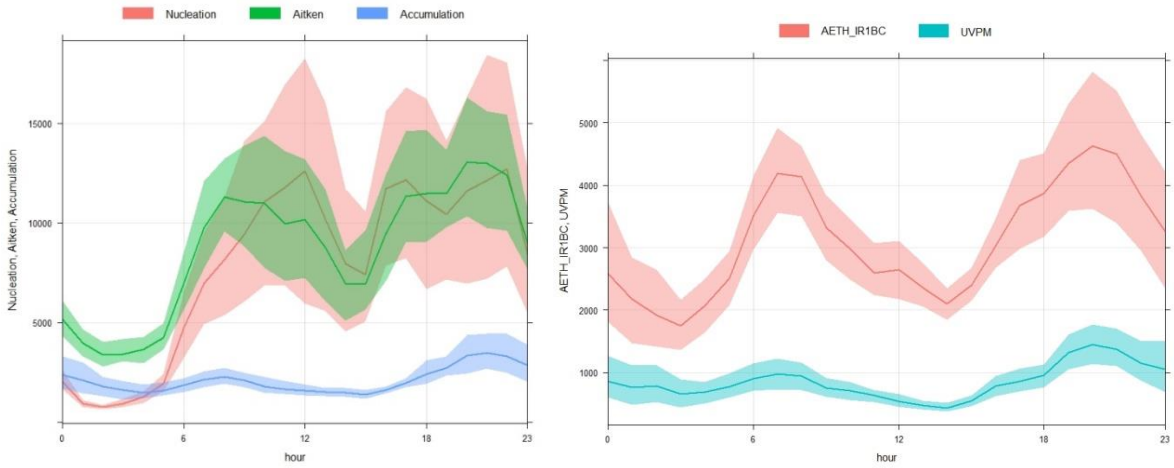


Figure S9 – Diurnal plots for measurements at LHR2

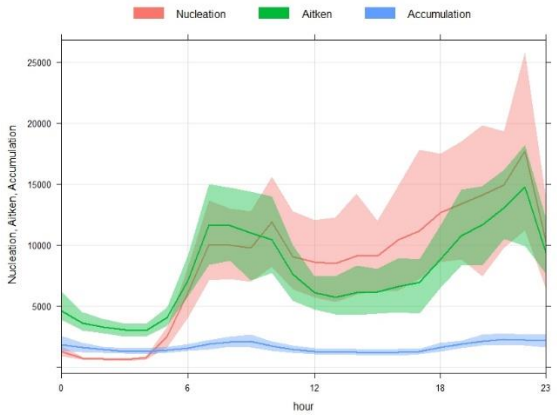


Figure S10 – Diurnal plots of Particle Number concentrations at Oaks Road

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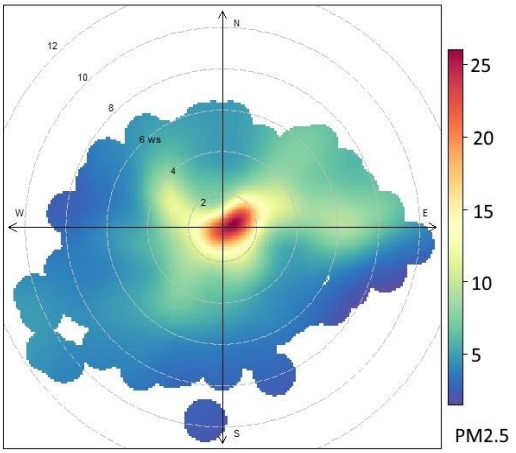

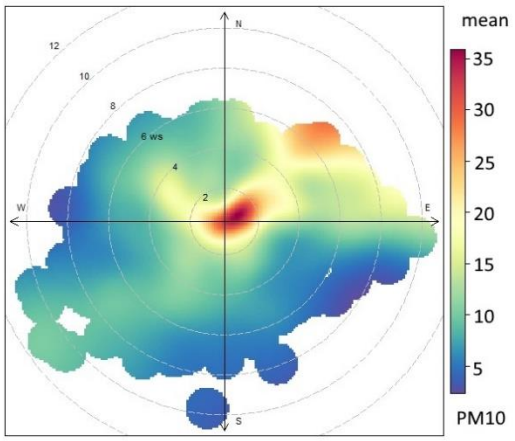

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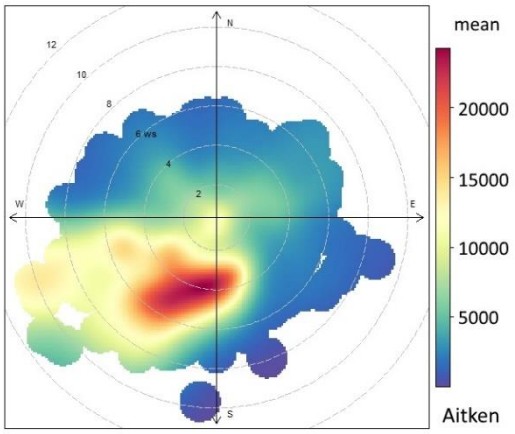

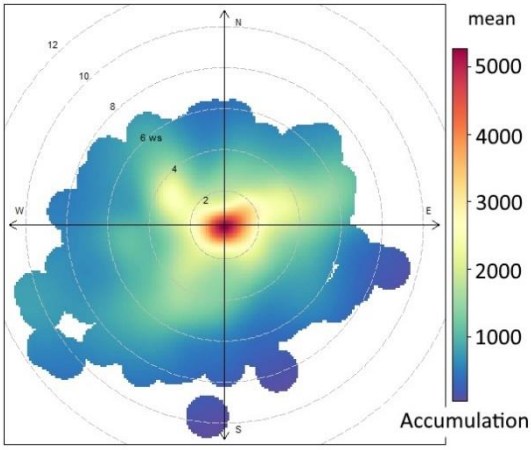

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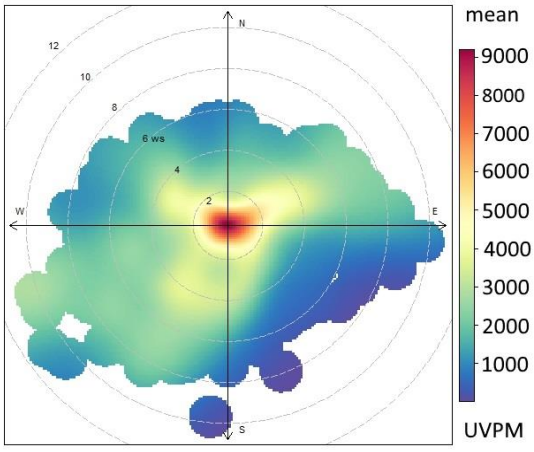

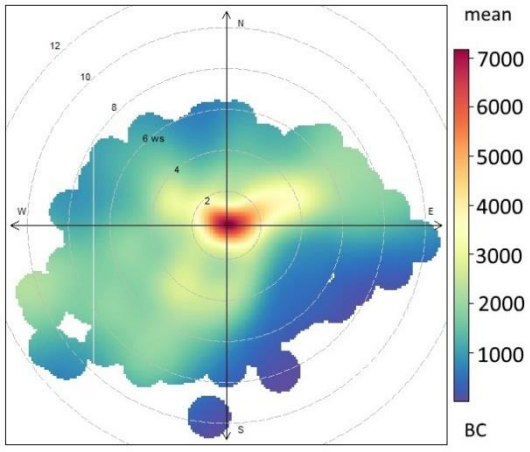

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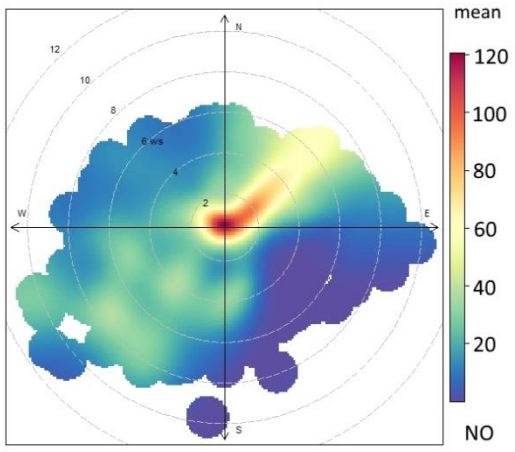
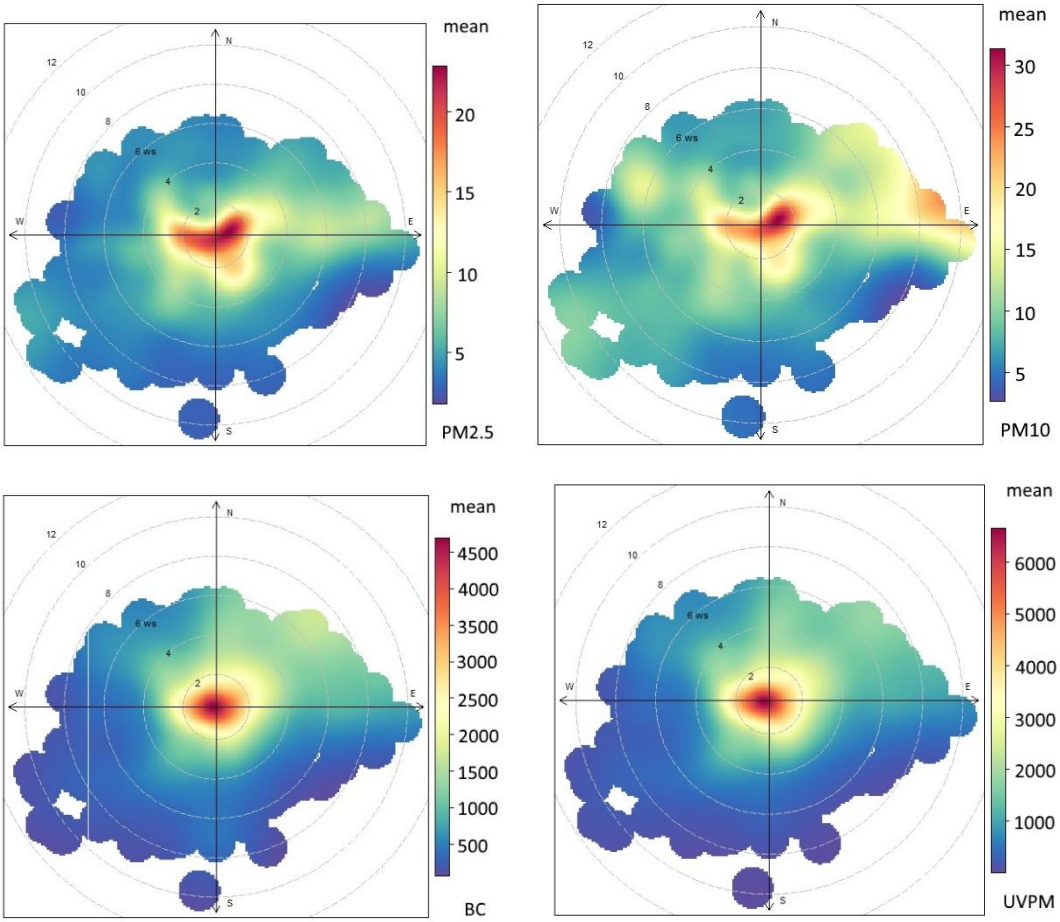


891 Figure S11 – Polar plots for LHR2 measurements



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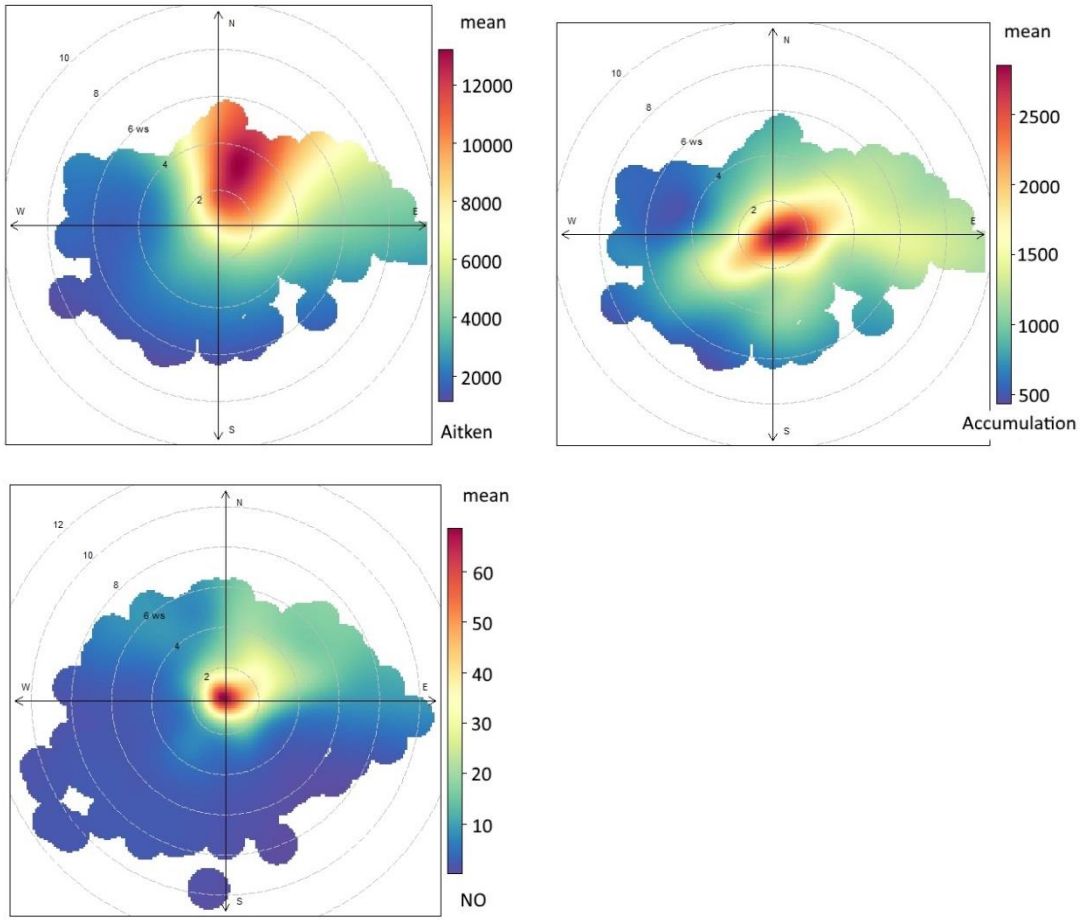
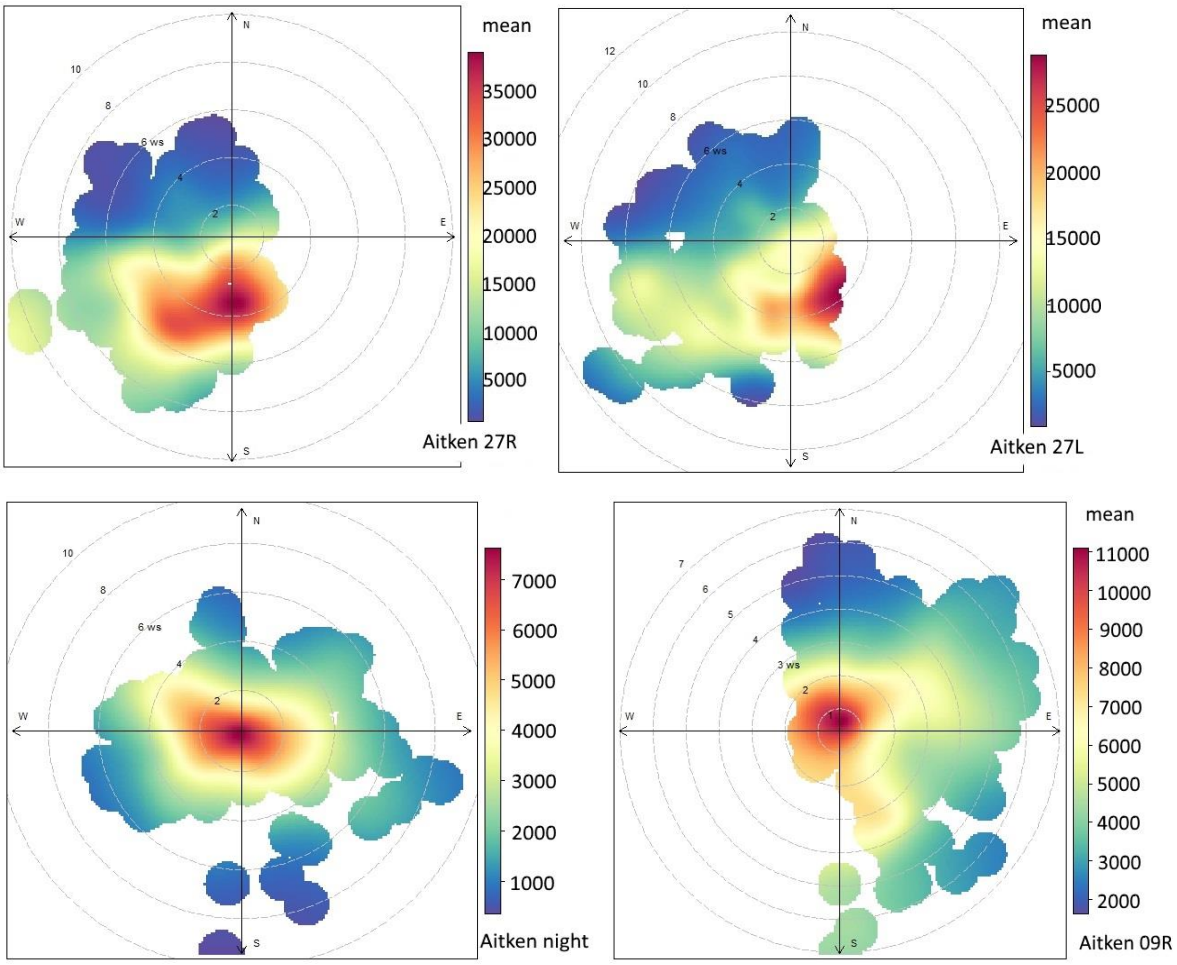


Figure S12 – Polar plots for Oaks Road measurements

898



899

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

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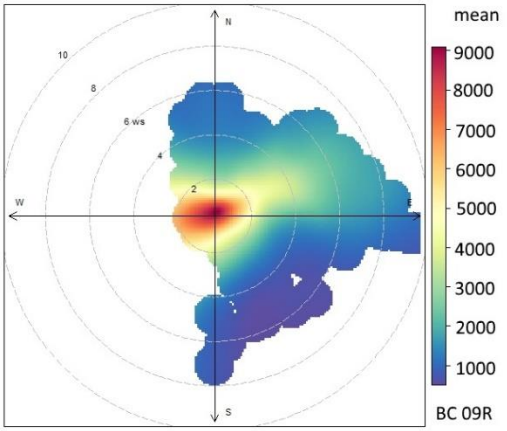
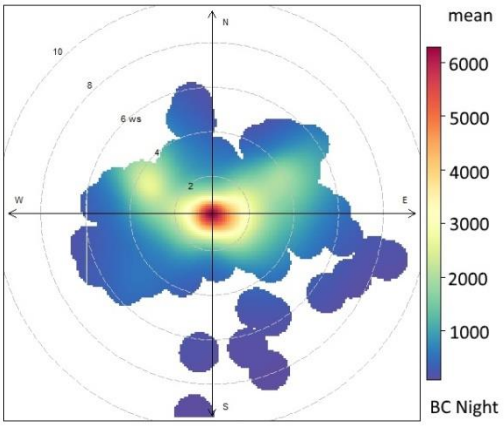
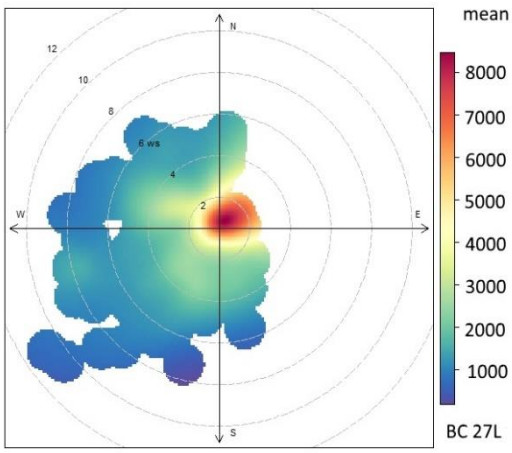
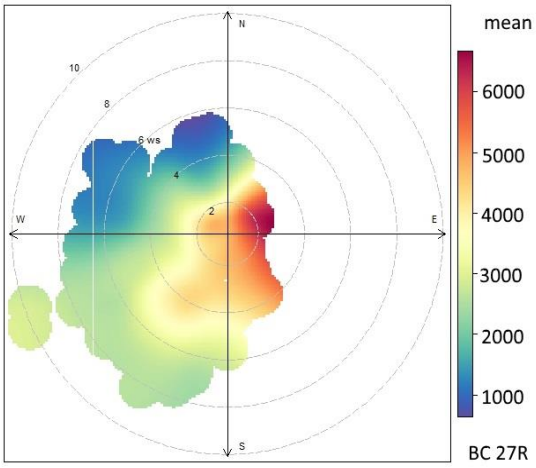
902


mean
9000
8000
7000
6000
5000
4000
3000
2000
1000
BC 09R


mean
6000
5000
4000
3000
2000
1000
BC Night

903


mean
8000
7000
6000
5000
4000
3000
2000
1000
BC 27L


mean
6000
5000
4000
3000
2000
1000
BC 27R

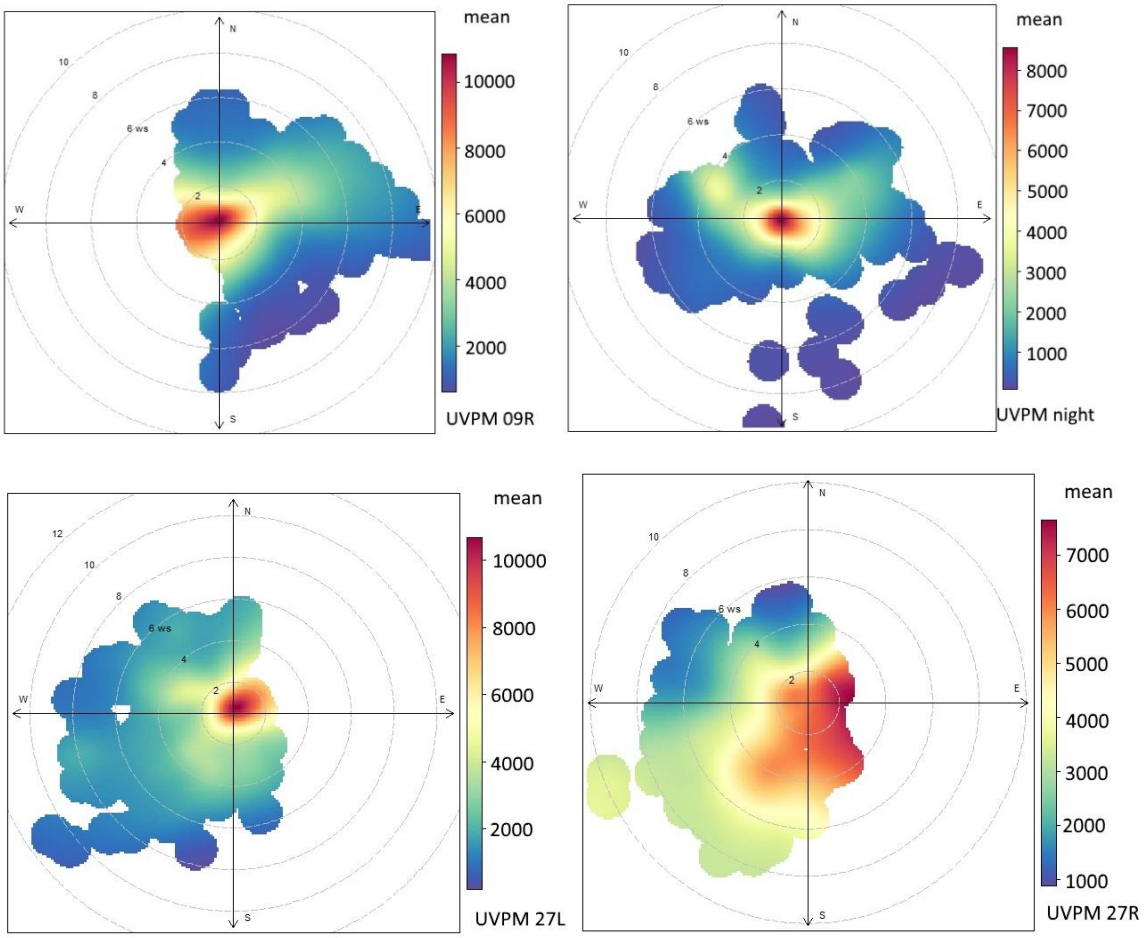
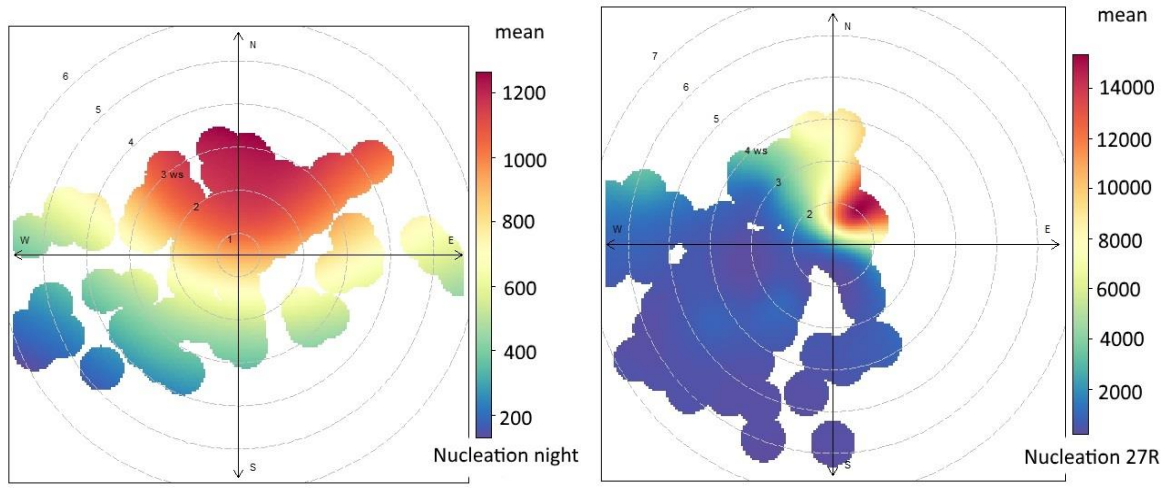
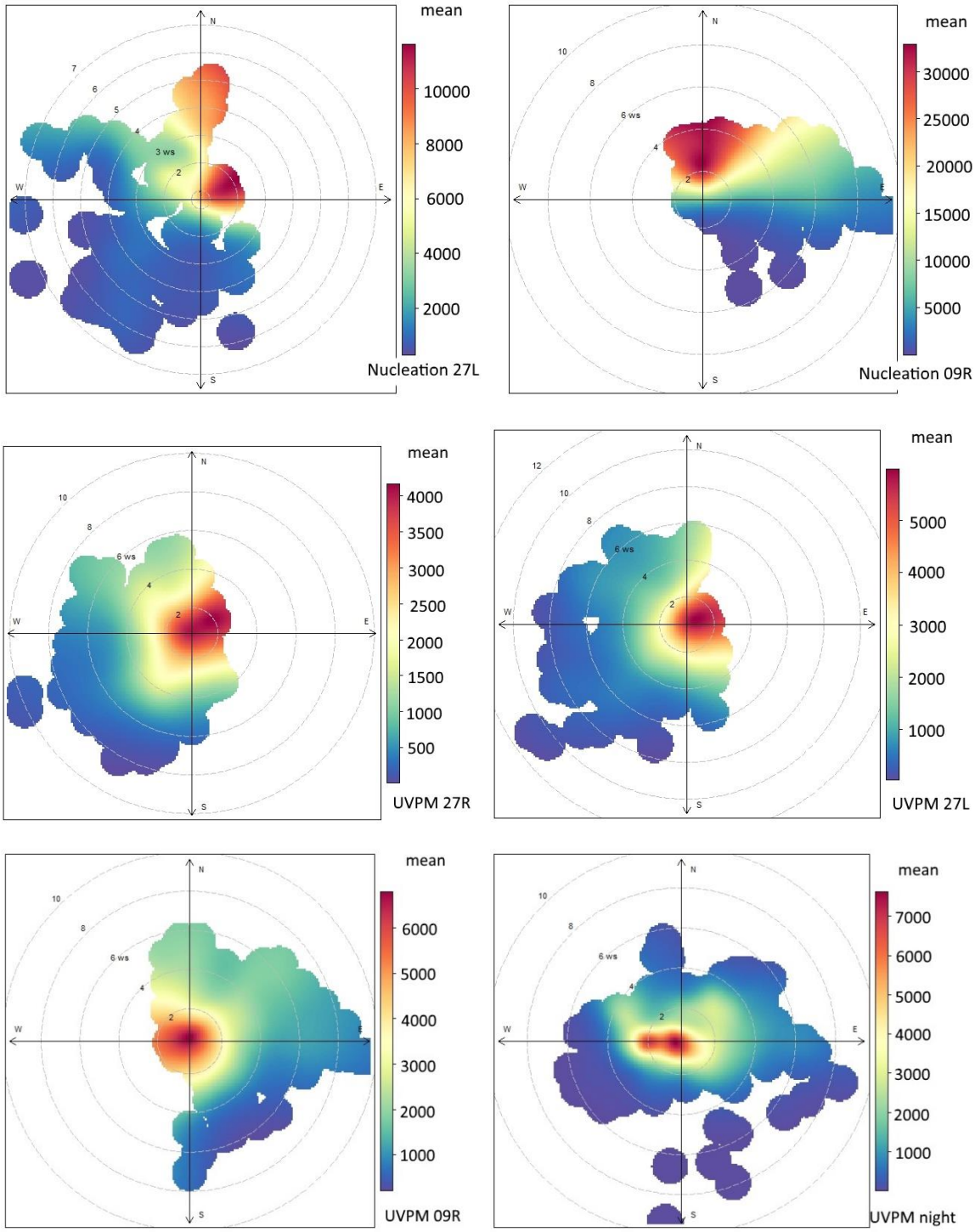


Figure S14 – Black Carbon and UVPM at LHR2 split by runway mode





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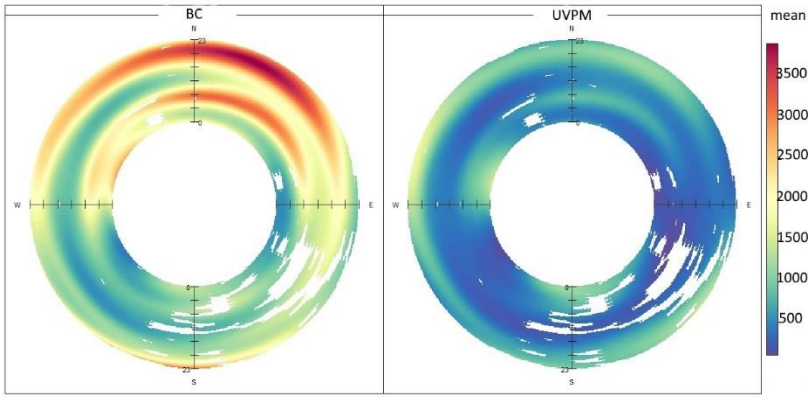
911

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

913

914



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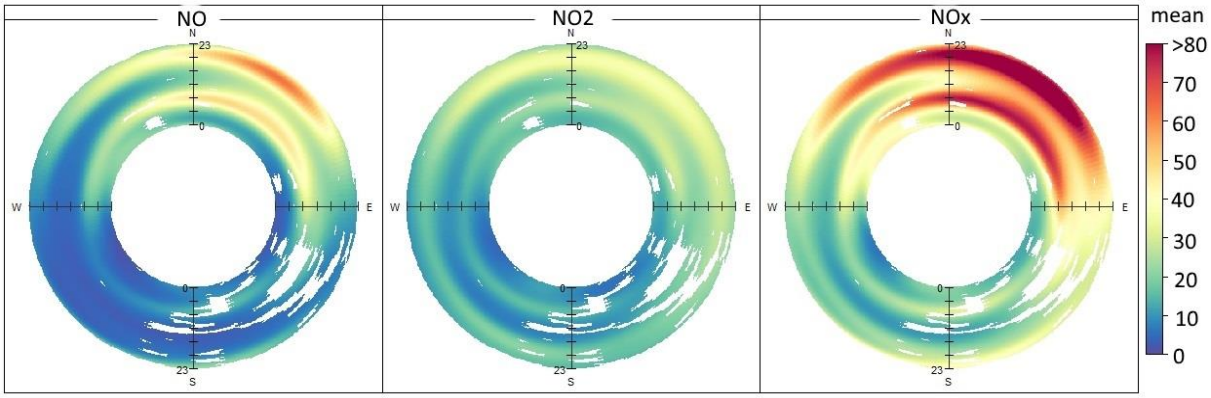

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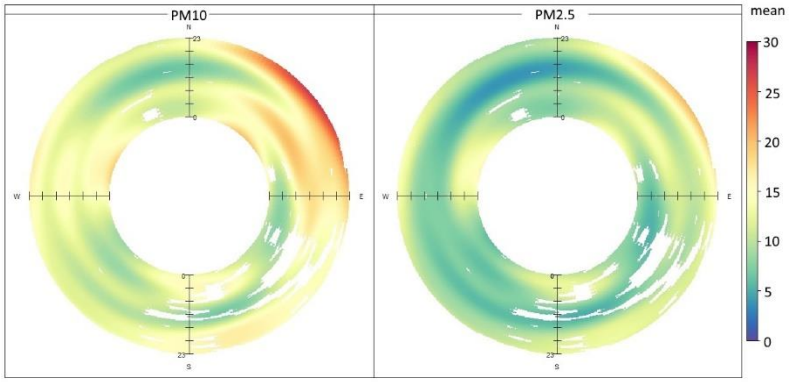

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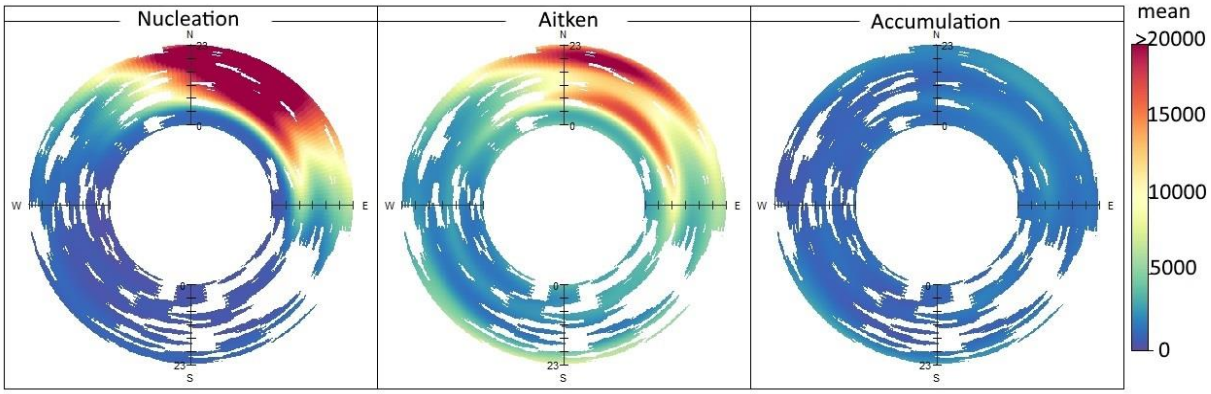
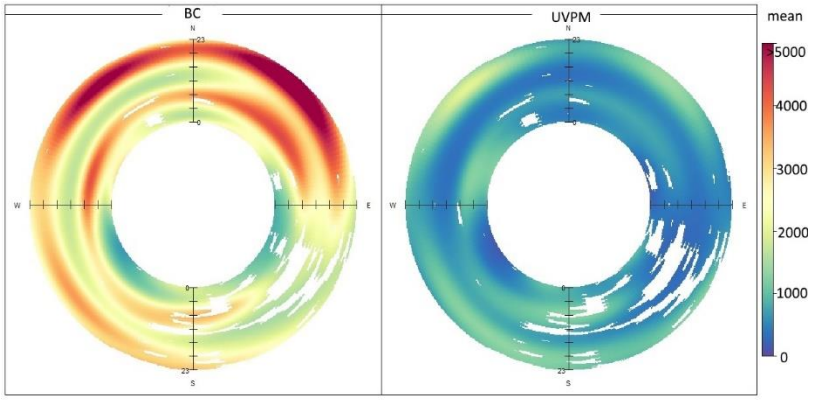


918

919 Figure S16 – Oaks Road Polar Annuli



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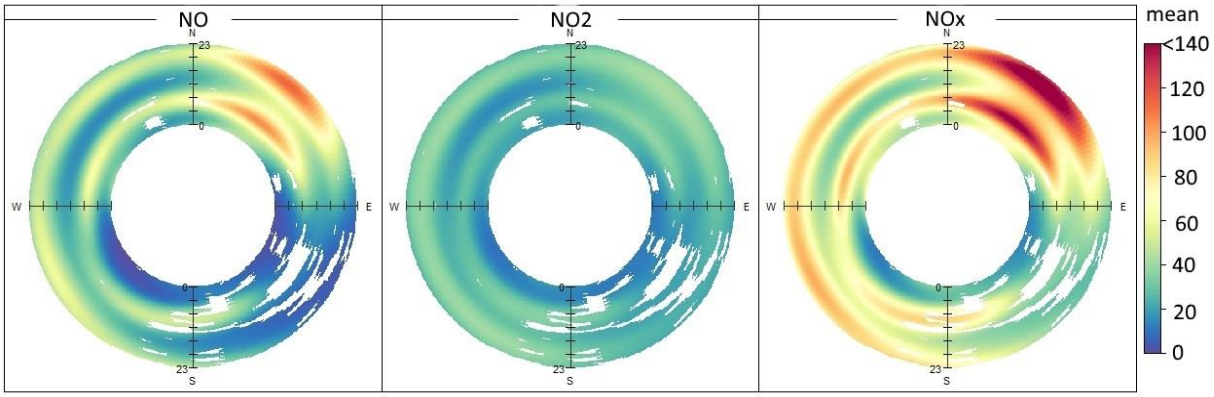

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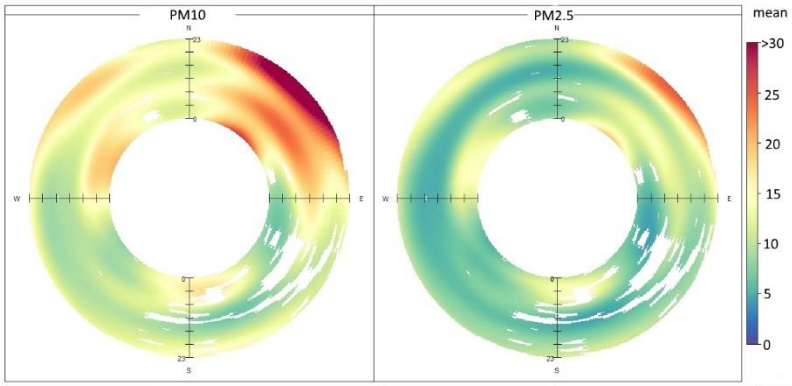

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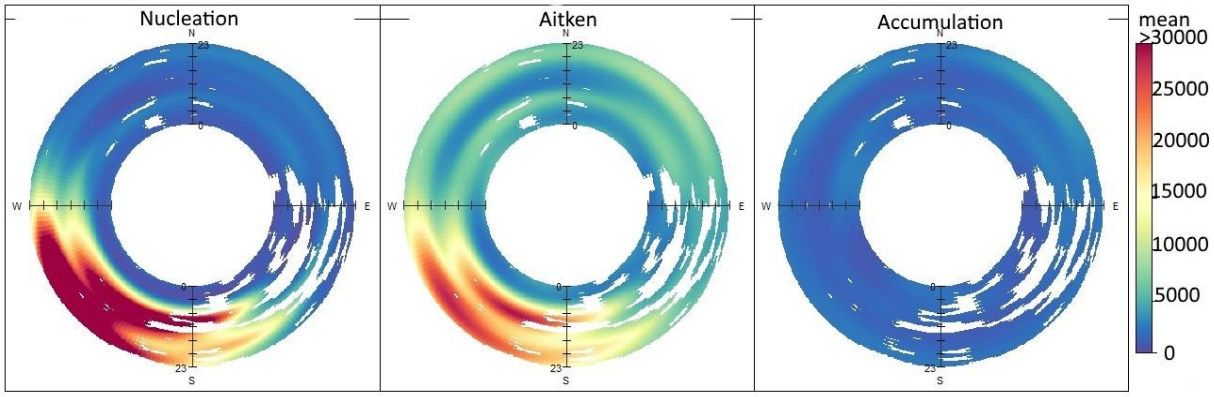
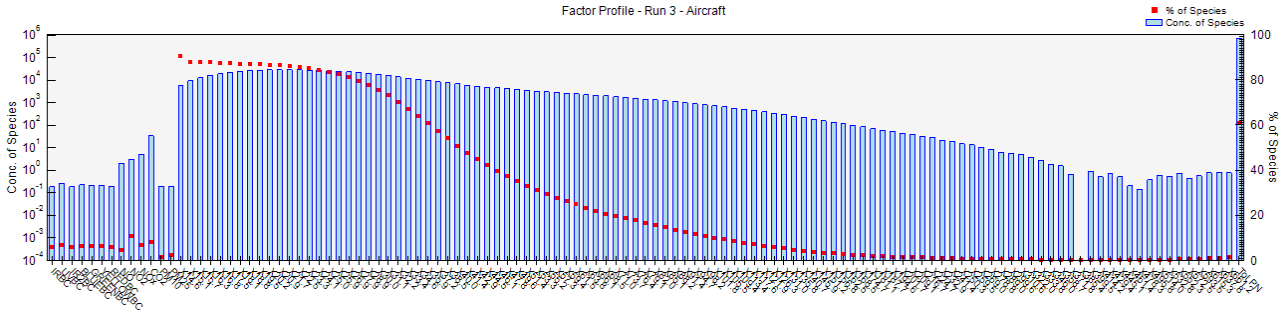
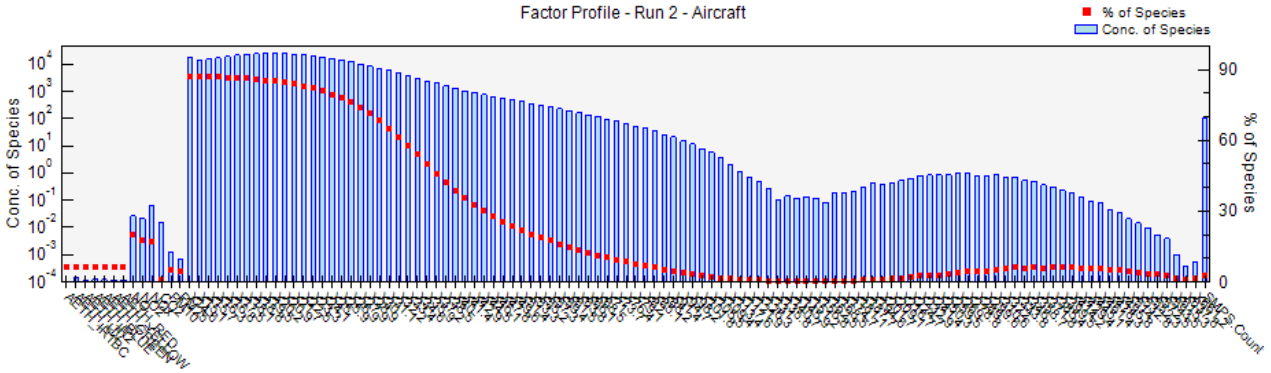


923

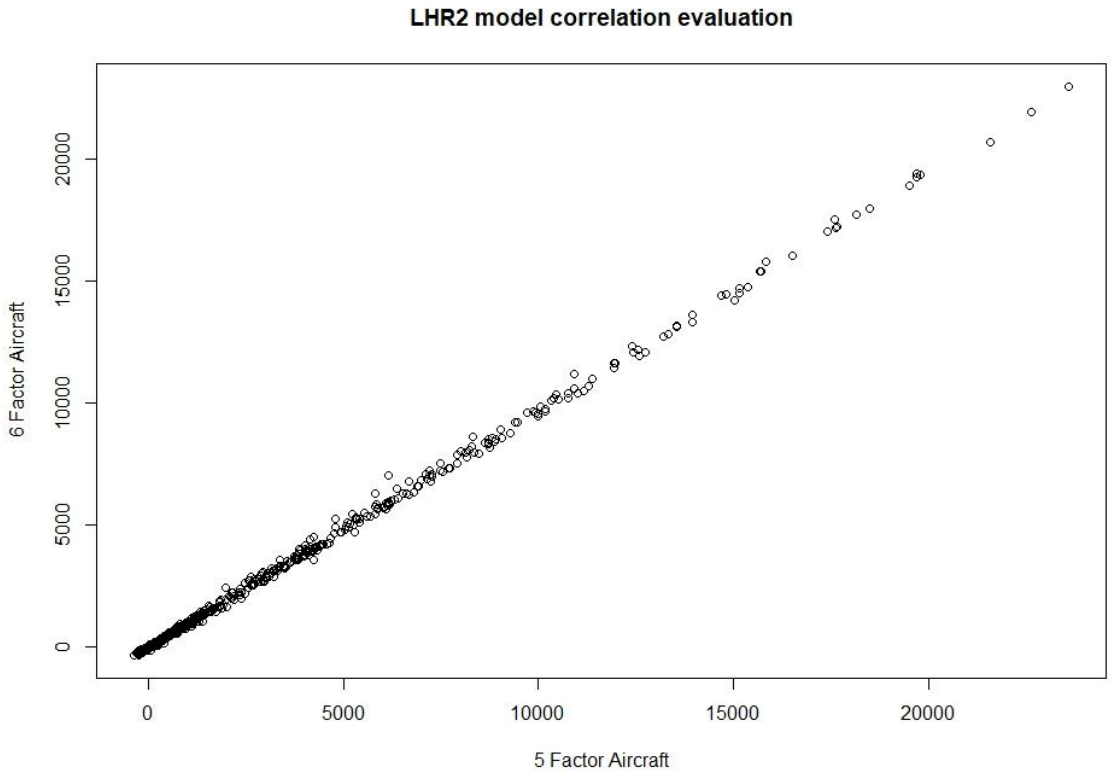
924 Figure S17 – LHR2 Polar Annuli



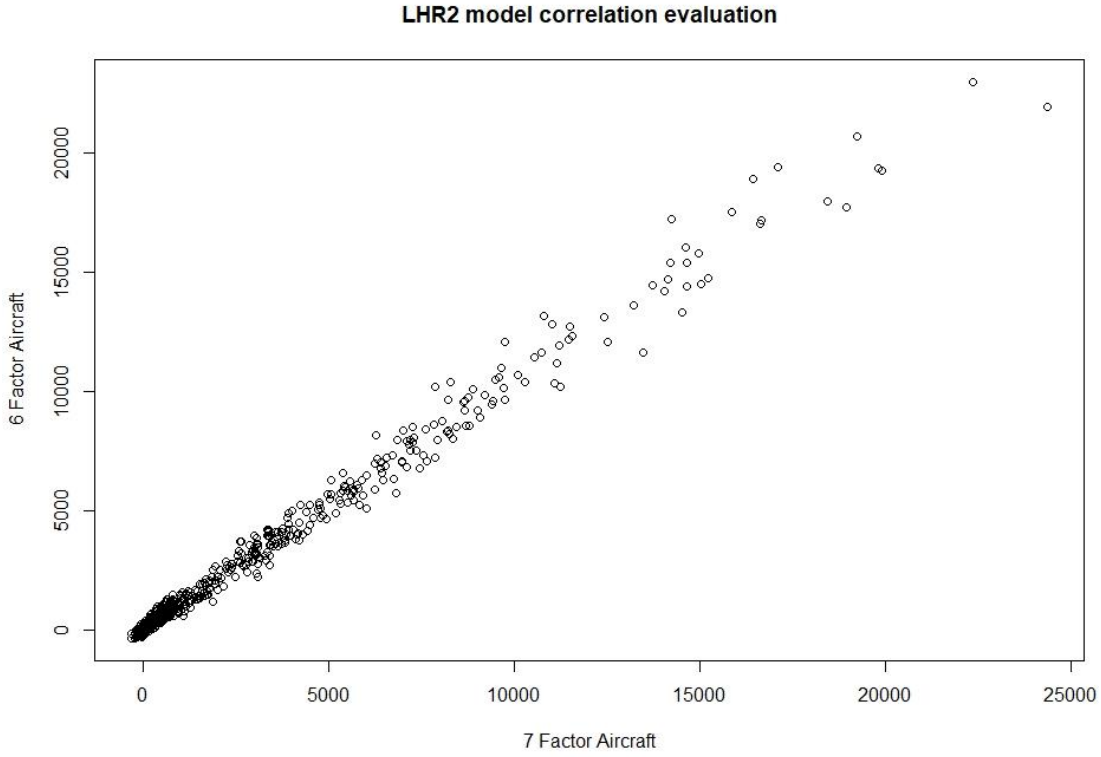
925
926 Figure S18 – PMF aircraft factor for LHR2 using all SMPS channels



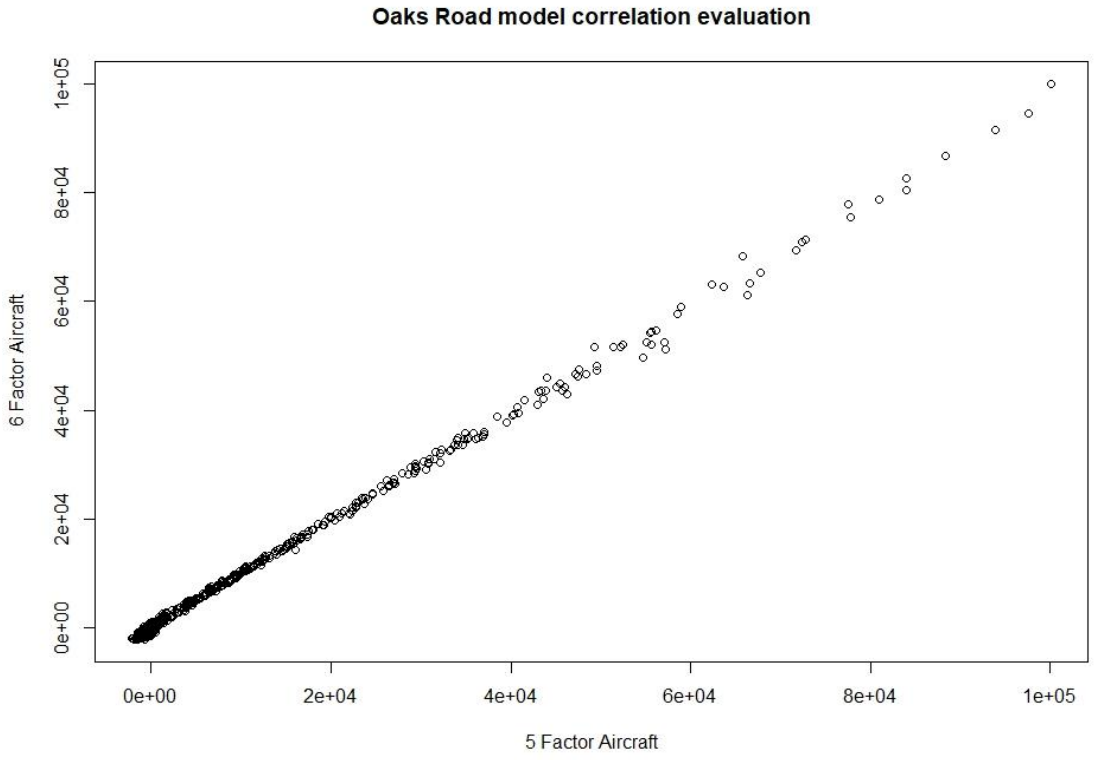
927
928 Figure S19 – PMF aircraft factor for Oaks Road using all SMPS channels



929
930 Figure S20 – Correlation between 5 and 6 factor solutions for Aircraft at LHR2



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



933
934 Figure S22 – Correlation between 5 and 6 factor solutions for Aircraft at Oaks Road

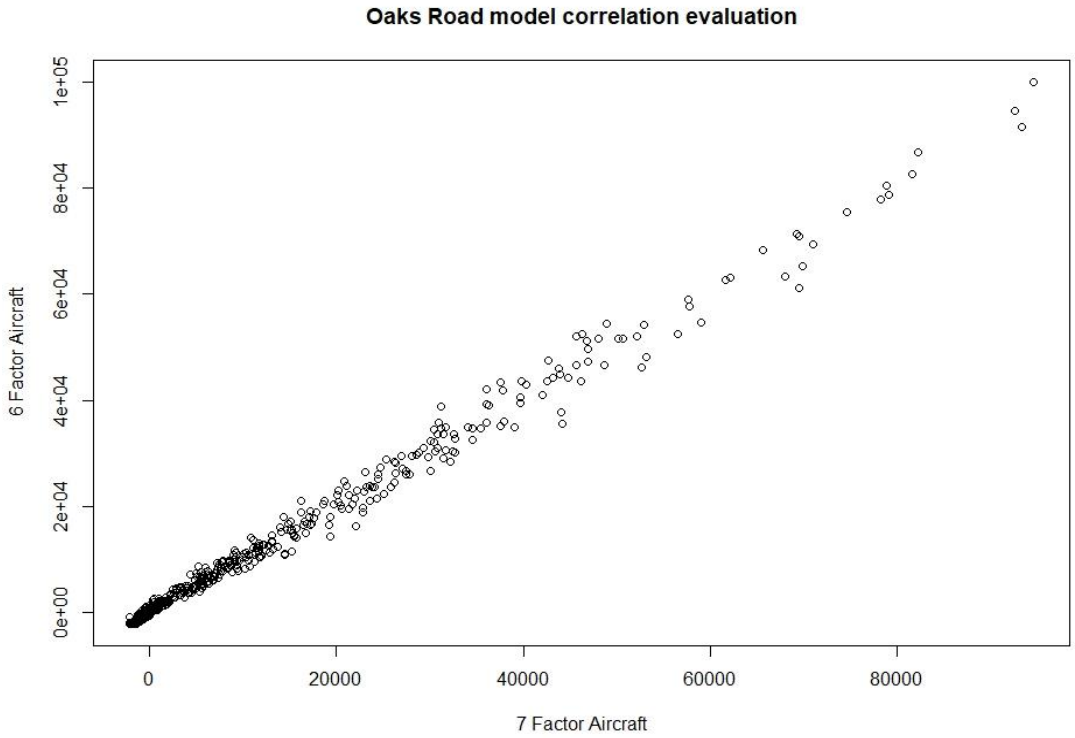


Figure S23 – Correlation between 6 and 7 factor solutions for Aircraft at Oaks Road