Characterising Ultrafine Particles at Heathrow Airport

by

Brian Stacey

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School of Geography, Earth and Environmental Sciences
University of Birmingham
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Abstract

Exposure to ultrafine particles (UFP) is increasingly associated with adverse health outcomes. However, measurement of UFP in the ambient environment is generally not widespread, which limits both understanding and data for detailed assessment of the health impact. Of particular interest is the impact of airport and aircraft activities on UFP concentrations, as there are no abatement strategies for emissions from aircraft engines and jet fuel typically has a relatively high sulphur content, which can contribute to UFP formation during combustion.

Three UFP measurement studies were undertaken between 2016 and 2019 to progressively understand the environment around Heathrow Airport:

- The study in 2016 was carefully arranged to put airport measurements into context with representative traffic, residential and rural measurements in the south of England. As far as possible, the analysers, configurations and QA/QC used at the airport was the same as the analysers used in the UK regulatory monitoring network. The results clearly show that local airport activity has a very significant effect on local concentrations. The size distribution of airport-related UFP was seen to be different to other locations; particles smaller than 30 nm were observed in far higher numbers at the airport. Departing aircraft were associated with higher UFP concentrations than arriving aircraft.
- The 2017 study was designed to just investigate particles smaller than 100 nm, at a faster time resolution than conventional analyser configurations. This study confirmed UFP measurements were greater in number concentration from departing aircraft and also from larger aircraft within that subset.
- The 2019 study made use of a very fast UFP analyser, accurate aircraft location data and meteorology to uniquely associate UFP measurements with individual aircraft. The data were used to calculate emission rates for each aircraft type, which were found to be much higher than stated literature values. This is almost certainly due to measurement of condensable particles not accounted for in the published literature data. Larger and older aircraft were associated with higher measurements and emission rates, not necessarily mitigated by carrying more passengers.

In 2021, the World Health Organization published guidance for recommended maximum hourly and daily exposure to UFP. The recorded measurements from all three studies exceeded these guide values; UFP exposure at large airports will be a key area of interest for future research and health assessments.

Acknowledgements

My journey to take up a Doctor of Philosophy programme, part-time, was largely unplanned and unexpected. After over 25 years as a consultant in the air pollution monitoring business, the challenge of a major research project, within my sphere of expertise and without the ties of delivering "just what the customer asked for, no more no less", seemed like a good idea. I can confirm it has been both challenging and rewarding and I'm very proud of the final result.

There are a few people and organisations that I want to recognise here. Firstly, my thanks to Heathrow Airport Limited for providing my tuition fees for the duration of the programme. Specifically, my gratitude to Andrew Chen and Tom Harrison at Heathrow for their support and permission to use their monitoring stations and datasets; without this, the programme could not have been undertaken.

I thank my supervisors, Roy Harrison and Francis Pope for their research direction and input throughout the research. Because of the distanced based nature of my studies, our monthly meetings have been my only real interaction with the University. I'm grateful for their friendship, insight, guidance and advice.

The acknowledgements in my first degree dissertation, awarded back in 1991, recognised the contribution of my family in allowing me to study, despite many hurdles along the way. 30 years on, the makeup of "The Family" has changed, some new, some no longer with us, and some who have become family without actually being related. Their support and companionship are my inspiration and they enrich my life every day.

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Abbreviations used in the thesis

The following terms, definitions and abbreviations are used throughout:

Term	Definition
Accumulation Particles	Particles with a diameter between 100 nm and 661 nm
ACTRIS	Aerosols, Clouds, and Trace gases Research Infra Structure
Aitken Particles	Particles with a diameter between 26 nm and 100 nm
AQEG	UK Air Quality Expert Group
CAEP	Committee on Aviation Environmental Protection
CEN	Comité Européen de Normalisation (European Committee for Standardisation)
CFD	Cumulative Frequency Distribution
CO2	Carbon Dioxide
CPC	Condensation Particle Counter
D50	Diameter at which particles are counted at 50% efficiency
DMA	Differential Mobility Analyser
EUSAAR	European Supersites for Atmospheric Aerosol Research
ICAO	International Civil Aviation Organization
ISO	International Standardization Organization
LDSA	Lung Deposited Surface Area
nm	nanometres
NOx, NO, NO_2	Oxides of nitrogen, nitric oxide, nitrogen dioxide
Nucleation Particles	Particles with a diameter smaller than 25nm
PAH	Polycyclic Aromatic Hydrocarbons
PM_{10} / $PM_{2.5}$	Particulate Matter with a mean aerodynamic diameter of 10 microns or 2.5 microns
PMF	Positive Matrix Factorization model
PN / PNC	Particle Number / Particle Number Concentration
PSD	Particle Size Distribution
QA/QC	Quality Assurance / Quality Control
SMPS	Scanning Mobility Particle Sizer
SO_2	Sulphur dioxide
TC / EC / OC / BC / UVPM	**Total Carbon** (total quantity of carbon in a sample) **Elemental Carbon** (fraction of non-volatile carbon according to a specified thermal-optical detection principle, often closely correlated with BC) **Organic Carbon** (fraction of volatilised/pyrolyzed carbon that is volatilised or pyrolyzed according to a specified thermal-optical principle) **Black Carbon** (fraction of carbon measured by attenuation of light at 880nm) **Ultra-Violet Particulate Matter** (fraction of carbon measured by attenuation of light at 380nm, often used as a marker for biomass burning)
TPM	Total Particle Mass
UFP	Ultra Fine Particles
USEPA	United States Environmental Protection Agency
V/NV	Volatile / Non-Volatile
VOC	Volatile Organic Compounds
WHO	World Health Organization
WMO-GAW	World Meteorological Organisation - Global Atmosphere Watch

Author's declaration

I declare that this thesis is a presentation of original work and I am the sole author. The thesis has not previously been presented for an award at this or any other University. All sources are acknowledged as References. This thesis is based on four peer reviewed papers (three published, one submitted for review), where I am either sole or lead author. The papers are listed below:

Stacey, B. Measurement of ultrafine particles at airports: A review. Atmos. Environ, 198, 463-477 (2019), https://doi.org/10.1016/j.atmosenv.2018.10.041

Stacey, B., Harrison, R. M. & Pope, F. Evaluation of ultrafine particle concentrations and size distributions at London Heathrow Airport. Atmos. Environ., 222, 117148 (2020), https://doi.org/10.1016/j.atmosenv.2019.117148

Stacey, B., Harrison, R. M. & Pope, F. D. Evaluation of Aircraft Emissions at London Heathrow Airport, Atmos.Environ, 254, 118226 (2021), https://doi.org/10.1016/j.atmosenv.2021.118226

Stacey, B, Harrison, R.M and Pope, F.D, Emissions of ultrafine particles from civil aircraft: (in preparation, submitted to ES&T April 2022)

These publications are used for Chapters 2, 3, 4 and 5 respectively. The review paper is updated at the end of Chapter 2, to account for relevant papers published or discovered subsequently. The text, figures and tables from the publications have been reformatted to fit the style of this thesis. Any changes from the published papers are listed at the end of each paper as an appendix. These clarifications are deemed small and do not warrant corrigenda to be issued to the journals.

1 Introduction

1.1 Background

Exposure to high concentrations of air pollutants is known to be strongly associated with adverse health impacts. In 2021, the World Health Organization (WHO) updated its guidance for recommended maximum exposures, presented in the table below:

Pollutant	Averaging Time	WHO AQG
$PM_{2.5}$, µg/m^3	Annual	5
	24-hour	15
PM_{10}, µg/m^3	Annual	15
	24-hour	45
O_3, µg/m^3	Peak season	60
	8-hour	100
NO_2, µg/m^3	1-hour	200
	Annual	10
	24-hour	25
SO_2, µg/m^3	10-minute	500
	24-hour	40
CO, µg/m^3	24-hour	4
	8-hour	10
	1-hour	35
	15-minute	100

Table 1.1 – WHO recommended Air Quality Guidelines (AQG), 2021

In addition to these guidelines, WHO included "good practice statements" for a number of other pollutants, including ultrafine particles (UFP). The guidance for UFP requires a Particle Number Count (PNC) and counting of all particles larger than 10 nanometres (nm, 10^{-9} m). The WHO classification of low and high concentrations is as follows:

UFP level description	Exceedance Criteria
Low PNC	< 1000 particles / cm^3 (24-hour)
High PNC	> 10,000 particles / cm^3 (24-hour)
High PNC	> 20,000 particles / cm^3 (1-hour mean)

Table 1.2 – WHO description of low and high ultrafine particle concentrations (2021)

1.2 Particles

It is difficult to achieve consensus for a universal definition of UFP. Throughout this thesis, particles smaller than 100 nm are termed UFP. They typically contribute very little to the conventional PM mass measurements (PM_{10} and $PM_{2.5}$, loosely described as particles smaller than 10 or 2.5 micrometres (microns, 10^{-6} m)), but dominate in terms of particle number concentrations. A typical profile for a number of different particle measurement metrics vs particle size is presented in the figure below:

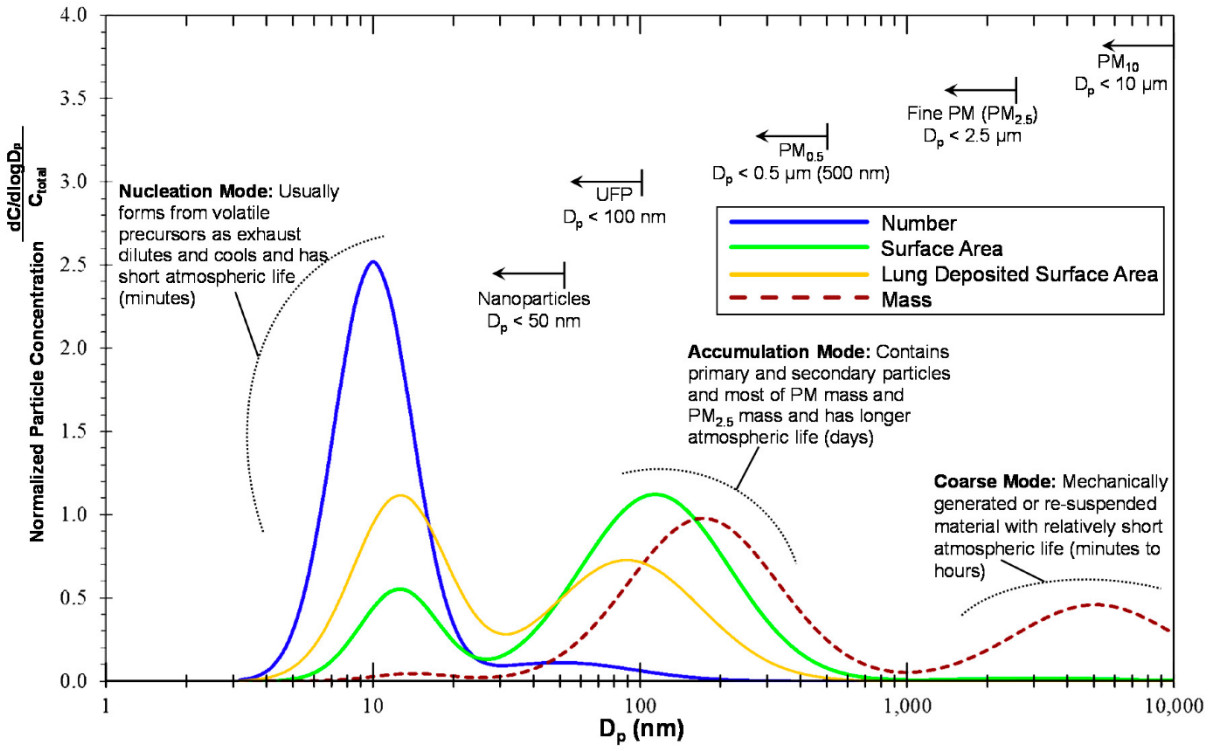

Figure 1 - Tri-modal particle size distributions using different particle metrics (number, surface area, lung deposited surface area, and mass). For this figure, Dp is the particle diameter, UFP are ultrafine particles, and PM stands for particulate matter. Reproduced from Baldauf et al (2016) (1)

As with the PM metrics, UFP is made up of a wide range of different components, including metals, elemental / organic / black carbon, secondary organic and inorganic aerosols.

Unlike conventional pollutants, measurement of UFP and PNC does not always follow accepted and harmonised conventions. There are many variables that need careful consideration:

- Particle size can be classified in a number of ways: aerodynamic diameter, differential particle mobility of charged particles, spectrometrically.
- Detection of particles can be undertaken in a number of ways: electrometer, condensation particle counting, optically.
- Particle shape, morphology and composition will vary depending on location, time of day, meteorology and season.
- Behaviour of particles during conditioning and measurement. Depending on the measurement technique, particles may need to have any existing electrical charge neutralised before further processing, and how the particles respond to these treatments is not always consistent between processes.
- The wide range of instrumentation available reports data in different ways: minimum and maximum detection sizes, sequential scanning of sizes, scan time, range of algorithms for deconvolving signals, reporting of data in different formats (PNC, sizing, surface area or LDSA).
- Conditioning protocols for particles prior to analysis. Drying, heating, catalytic stripping and dealing with charged particles all have a significant influence on measured concentrations.
- Calibration and Quality Control of measurements

All of the variables listed above combine in the final analysis of reported data. UFP are normally reported as number concentrations, rather than mass. So the ability of an instrument to detect and report particles at different sizes has a material effect on comparisons with other studies. An analyser that measures in the range 16 to 600 nm will report significantly different particle number concentrations to one that measures in the range 5 to 1000 nm. Additionally, there are no

commercially available instruments that only report UFP particle numbers. Where a calculation is possible, the user has to undertake this themselves.

As a result, it is often difficult to make comparisons between published research articles. There are mechanisms already in place for improved standardisation (2) and procedures for QC (3, 4), and these will allow relationships between the range of measurement techniques to be quantified and valid comparisons between datasets to be undertaken. The academic communities will need to adopt these procedures and document their calibration strategies if future research programmes are to feed robust data into health assessment studies.

1.3 UFP in the Airport Environment

In the UK, there are a small number of national network monitoring stations measuring UFP (5). These are focused on road traffic, urban background and rural locations and provide a reasonable picture of typical UFP size distributions and concentrations, at least in the south east of the country. In contrast, the environment close to airports is less well understood or documented. In 2012, Aarhus University in Denmark presented UFP data from Copenhagen Airport that found airport concentrations were three times higher than nearby background locations (6). Concerns were raised in Denmark that these higher concentrations could be linked to increased incidences of cancers in airport workers. There was evidence for and against this at the time (7, 8), but it was clear that more research and data was required.

Early studies suggested that airports are a significant source of UFP and can often be observed at large distances downwind (9, 10), but the precise nature of the UFP source remained a matter for speculation – airborne aircraft, aircraft on the ground, or a mix of both? Access to airports to closely measure emissions from airports is very difficult: of necessity many studies have been undertaken at some distance from airfields and aircraft.

The airport environment is a wide and complex mixture of emission sources, including:

- Aircraft:
    - Manoeuvring
    - Departing / Landing
    - Auxilliary Power Units
- Space heating
- Construction

- Ground support vehicles (fire appliances, tugs, snow clearance, fuel delivery, baggage,etc)
- Power generation (buildings and aircraft)
- Buildings
- Catering
- Passengers (cars, buses, etc)

Many of these emission sources will fall under established legislative and environmental drivers to either significantly reduce their carbon footprint, or eliminate them completely. These changes are already impacting in many sectors; improvements in mitigation strategies has seen dramatic reductions in their relative NOx and PM contributions to the UK emissions inventory:

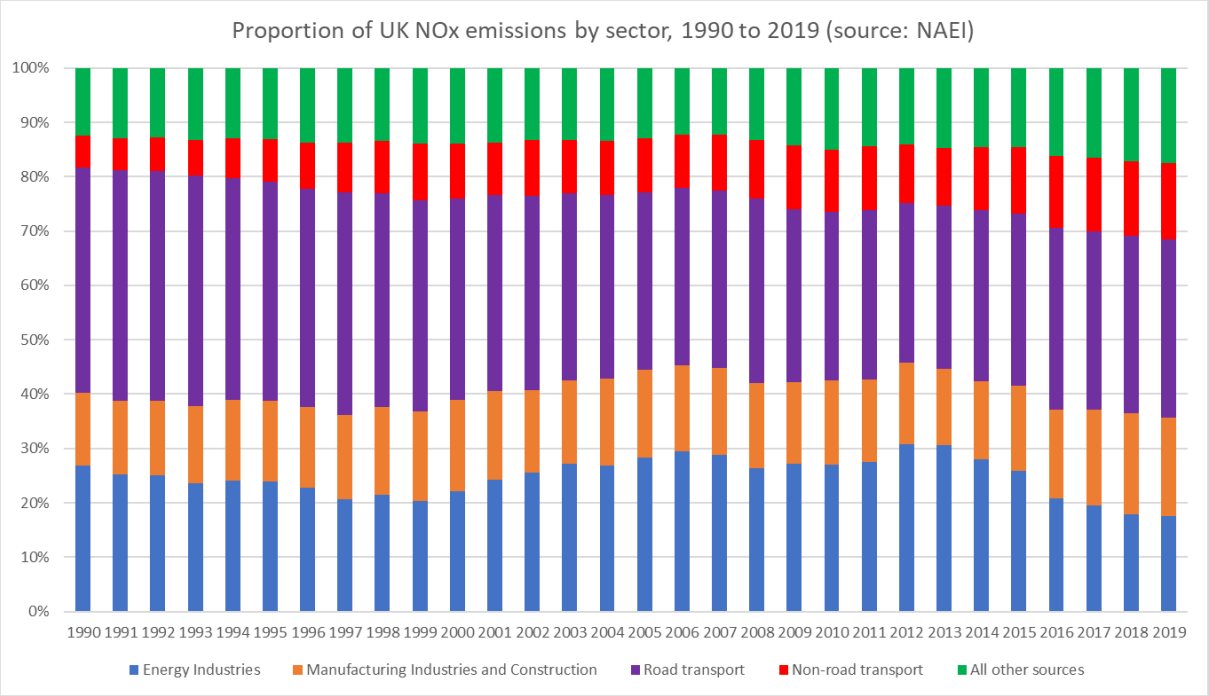

Figure 2 – NOx emissions contributions by sector, 1990-2019 (data from the National Atmospheric Emission Inventory (11))

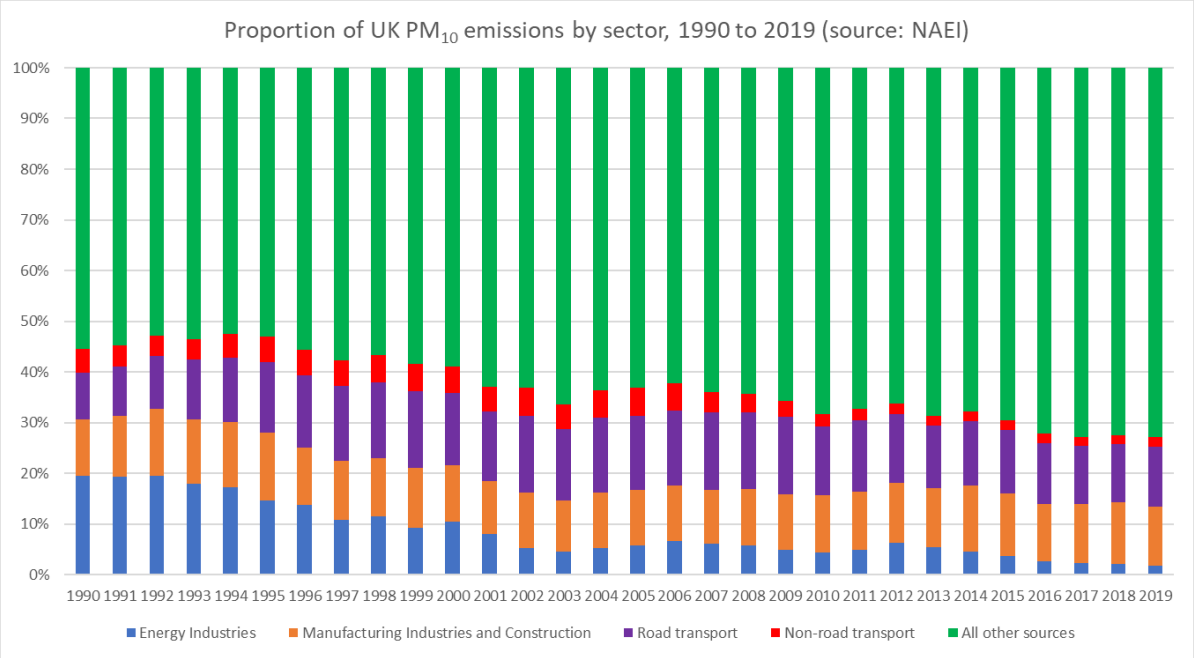

Figure 3 – PM_{10} emissions contributions by sector, 1990-2019 (data from the National Atmospheric Emission Inventory)

Figures 2 and 3 show how relative contributions per source has changed over the past 20 years.

For NOx, the combined energy and road traffic contribution has decreased from ~70% in 1990 to ~50% in 2019, with significant further reductions expected over the next 10 years due to changes in energy sources and uptake in electric vehicles. In contrast, the NOx contribution for non-road transport (Rail, Sea and Air) has shown an increase from 6% in 1990 to 14% in 2019. Overall, total UK emissions of NOx have reduced from ~ 2.98 million tonnes in 1990 to ~ 0.84 million tonnes in 2019.

For PM_{10}, combined energy and road traffic contributions have decreased from ~28% in 1990 to less than 14% in 2019, mostly driven by huge changes in fuel use in the energy industry. Non-road transport contributions have dropped from ~5% in 1990 to ~2% in 2019. PM_{10} emissions in 2019 are mostly driven by domestic heating (28%), industrial processes and solvents (30%) and agriculture (9%). Overall, UK emissions of PM_{10} have reduced from ~ 0.38 million tonnes in 1990 to ~ 0.17 million tonnes in 2019.

Reductions in emissions from airports are likely to follow road transport and energy emission profiles and timescales, at least in the processes and activities that are common to the aviation industry and the outside world. However, the timeframe for reducing emissions from the aircraft is more long term: the

aviation energy is committed to being Net Zero by 2050. This will be an incredibly challenging target to meet – while the supporting infrastructure can be upgraded to use clean energy, encourage the use of clean public transport and utilise zero emission ground support vehicles, the aircraft themselves will remain a significant emission source, especially for medium to long haul flights. It is likely, therefore, that the relative contribution of the aviation industry to emission inventory totals will increase in future.

For UFP in particular, aircraft are likely to become an increasingly significant contribution to emission inventories and impact on local air quality. Aircraft engines, as discussed in this thesis, may emit a very wide range of different particles into the atmosphere, including:

- soot,
- condensed volatile organic compounds from fuel combustion,
- volatilised/combusted lubrication oils,
- sulphur combustion products (e.g. sulphur dioxide, sulphate),
- metals,
- ceramics, etc

Aircraft operations dictate the quantity of these emissions. Fuel use is highest during take off, lower during landing and lower still for taxiing and use of the Auxilliary Power Unit. The combustion dynamics of the engines in these configurations will also impact on the nature of the emissions.

The quantities of these species emitted at airports is unlikely to undergo dramatic reductions in the short to medium term, at least until further regulation and fuel improvements are put in place. Assuming that the quantities of emissions from other major sources continue on a downward trajectory, airports will play an increasing role in the PM and UFP that populations are exposed to.

1.4 Philosophy

The investigation presented in this thesis follows a very structured "consultancy-based" approach.

1.4.1 Why the research is important

At the time that the research was planned (2016), understanding of the UFP climate close to airports was limited. The key areas where further knowledge is needed included the following topics:

- There is justifiable concern that exposure to high concentrations of fine particles has a detrimental effect on health. The evidence available at the time suggested that UFP

concentrations close to airports are higher than typical urban environments and the airports are the likely source, but the exact causes for this were not clear.

- The exposure of airport workers to high particle number concentrations is of concern, especially with the reported increased risk of cancer. By association, the exposure of population living and working close to airports will be of similar concern.
- Direct measurement of emissions from a selection of jet engines is available from a range of sources, but typically under very controlled conditions, rather than during routine operation, where operating practices may be significantly different. In addition, much of the emissions data samples are collected at high temperature, thereby specifically excluding particle formation during cooling and mixing of the exhaust plume.
- There was very little information about the particle size distribution of UFP close to airports, or the sources associated with the various size fractions. Similarly, little measurement data was available about how this particle size distribution was influenced by takeoff or landing operations. The operating strategy for flights at Heathrow Airport offered a unique opportunity to assess this.

1.4.2 How the research was undertaken

Once the need for further research was confirmed, a comprehensive monitoring campaign was devised to address the objectives:

- The relevant literature and data available in 2018 were examined. Chapter 2 documents the published literature associated with measurements of UFP at airports and measurement / emission studies from aircraft, and was published in 2019. The paper is updated at the end of Chapter 2 to include relevant publications from Jan 2019 to March 2022.
- A detailed methodology for the particle counting and sizing equipment used for the monitoring campaigns is presented in Chapter 3. The techniques and configurations are presented, together with a discussion about how differences in configuration and operation will have had a material impact on results and comparability between campaigns.

- A monitoring study at two locations around Heathrow Airport was planned and undertaken in 2016. To ensure direct comparability to national monitoring network stations, identical measurement techniques and QA/QC protocols were used throughout. Chapter 4 documents this measurement study, which was published in 2020.
- Building on the evidence from the 2016 dataset, a more intensive study was undertaken in 2017. Measurements were only undertaken at the airside monitoring station and the analyser was configured to operate a faster scan and over a narrower range of particle sizes. Meteorological data and aircraft movements were incorporated to quantify measurements from different aircraft types. Chapter 5 presents this measurement study, published in 2021.
- Following analysis of the 2016 and 2017 data, a study to collect very fast (1 second) UFP measurements, coupled with 1 second aircraft movements and 1 minute meteorology was undertaken in 2019. Data from this study was used to uniquely assign measurements to individual aircraft. From there, using rapid NOx measurements and NOx emissions data, the UFP emission rates for each aircraft type were calculated and compared with literature data, where available. Chapter 6 presents the results from this measurement study, which will be published in 2022.
- Chapter 7 collates all of the findings together in a conclusions section and provides a critical review of the studies, together with suggestions for future research.

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2. Literature Review

This Chapter presents work originally published in *Atmospheric Environment* on 1 Feb 2019. 10.1016/j.atmosenv.2018.10.041

Section 2.7 at the end of the Chapter reviews relevant airport and aircraft UFP papers published or identified after February 2019.

2.1 Abstract

Concern about the health impact of exposure to ultrafine particles has prompted a large number of research studies in the last twenty years. Attention focussed on conventional sources of pollution: vehicle emissions, generation of heat and power, as these are likely to be the most relevant sources of emission to which the general public are exposed. As a result, emissions from road vehicles are well characterised and regulated within Europe. In contrast, until relatively recently, little research into ultrafine particles had been specifically targeted at measurement of direct emissions from aircraft or their potential impact on the communities around airports. As a result, there are a number of gaps in our understanding of these emissions, behaviour in the atmosphere and the potential impact. Aircraft engines, especially the jet turbines used in commercial airliners, are known to emit large quantities of ultrafine particles under a wide range of operating modes, yet detailed information about the physical and chemical properties of these particles is poorly documented. This review aims to outline the background for the development of research studies, summarise the research and assessment of aircraft ultrafine particle emissions, and explore possible areas for future research in this area.

2.2 Introduction

Ultrafine particles (UFP) are a subset of particulate matter (PM) that are generally defined as smaller than 100 nanometres (100nm, 0.1 micron) in size. Typically, they can be carbonaceous, metallic or volatile/semi-volatile in nature, and predominantly formed in combustion processes. Kumar et al, 2014 (1) and Kumar et al, 2013 (2) discuss extensively the major likely sources of UFP in urban air, including sources unrelated to vehicle exhaust emissions such as cooking, heating and cigarette smoking.

Because of their extremely small size, there is concern about the possible health effects of UFP. As a result, population exposure to UFP has received increased attention in recent years. Conventional measurement of particle mass, using firstly the PM_{10} then the $PM_{2.5}$ sampling conventions, have allowed strong links between measured concentrations and impact on health to be established and permitted the setting of maximum exposure targets and limits globally (for example the European Directive on ambient air quality and cleaner air for Europe, 2008/50/EC (3) and the World Health Organisation air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide. Global update 2005 (4). These links, and specifically the role of road traffic in emissions of PM, has led to the enactment of European legislation to reduce emissions of particle mass and latterly control of UFP emissions from newly manufactured diesel engines, for example the European Community directives for light passenger and commercial vehicles 459/2012/EC (Euro 6) (5) and for large goods vehicles 582/2011/EC (Euro VI) (6). These two directives specifically focus on reducing emissions of particles, requiring strict target on emissions of both particles measured by mass and also by number of particles larger than 23nm.

Health studies, notably Donaldson et al, 2001 (7) have highlighted that, for example, exposure to particles smaller than 20nm can cause detectable inflammatory stress to respiration in rats, even at relatively low mass concentrations. Donaldson concludes from this, that size, numbers and the surface area of particles are more important than mass of particles deposited.

In contrast, the Health Effects Institute Perspectives 3, 2013 (8) concludes that there is insufficient evidence to directly attribute health impact to solely UFP – it maintains that it is not currently possible to decouple the effect of exposure to PM_{10} and $PM_{2.5}$ from UFP, and recommends further research.

There is increasing evidence that the particle size distribution of UFP is dependent upon many factors. Vu et al, 2015 (9) concluded that the combustion process, type of fuel burned, abatement strategies, secondary atmospheric processes and distance from source all play a major role in the resulting particle size distribution.

Much research has focussed on UFP emissions most closely associated with common human activities, e.g. road transport, cooking, heating, energy use and incineration, and yet collected evidence assessing the impact of these emissions remains largely ill-defined and occasionally contradictory.

Emissions of UFP from the aviation industry is a topic that is of increasing interest across the world. It is known that jet engines used in commercial aircraft are a significant source of UFP. For example, Westerdahl et al, 2008 (10), Hudda et al, 2014 (11), Hudda and Fruin, 2016 (12) and technical reports from Zurich, 2017 (13), Copenhagen, 2012 (14), Brussels, 2016 (15) and Schiphol, 2015 (16) airports all demonstrate that very high concentrations of UFP are observed close to aircraft emissions. These papers and reports also show that the particle size distribution profiles can be significantly different to those observed at typical urban locations, and may be observable for some distance downwind of the source, suggesting that airport and aircraft emissions and their impact should be considered in isolation from other sources.

Exposure to elevated UFP numbers in the workplace is recognised in some countries as a cause of occupational disease. Between 2008 and 2011, exposure to UFP for three workers at Copenhagen Airport was identified as a primary cause of them contracting bladder cancer, albeit from a variety of sources, not just aircraft. This resulted in a series of mitigation programmes to reduce emissions at the airport, including the use of electric vehicles, reduction in the use of diesel powered electricity supplies, and reduced aircraft engine operation whilst on the ground. However, despite this apparent clarity in Denmark, the available evidence still does not provide irrefutable links between exposure to UFP and impact on health. For example, RIVM, 2016 (17) could find no statistically significant risk of living close to Schiphol Airport, compared to the rest of the country and agreed with the HEI 2013 (8) report in concluding that more research is still needed to explore the relationship between UFP and health effect.

The intense scrutiny at Copenhagen Airport on air quality, and specifically for UFP, has triggered a number of research studies at other airports: Schiphol, Zurich and Brussels as noted earlier, but also Heathrow, Los Angeles International, Frankfurt, Paris CDG and Atlanta amongst others.

In addition to the possible health impact, emissions from jet aircraft, most recently from Delhaye et al (2017) (18) and Yu et al (2017) (19) have also found that the majority of particles exist primarily as soot, which has well documented effects on cloud formation and radiative climate forcing.

Throughout this review paper, an assessment of any quality assurance and quality control (QA/QC) of the data presented by researchers will be assessed. In order for links to UFP and medical effect to be

robustly established, the quality of measured data needs to be accurate, traceable and harmonised. Typically, this usually requires standard operating procedures, use of calibration reference materials and documented data management for processing and ratification. This type of philosophy is usually restricted to established national monitoring networks, where there are defined objectives for data quality, rather than the isolated research programmes undertaken in academia. As a result, any assessment of QA/QC applied in the studies listed in this review paper will be subjective and clearly depend on the extent to which the topic is discussed in each paper.

This review paper will explore the available texts that specifically focus on direct measurement of UFP emissions from aircraft and "ambient" measurements of UFP at airports. While the review will specifically focus on particles <100nm, the instrumentation used to measure these particles often provides useful information about particles larger than this. This value-added data will be retained for the reader to consider. The introductory discussion around health effect will not be presented further within this paper. Furthermore, research papers on the use of models and emission inventories are not the focus of this review paper and will not be discussed in detail.

Section 1 (Chapter 2.3) will examine the available evidence for the measurement of emissions from jet engines.

Section 2 (Chapter 2.4) will review the ambient measurement data collected from airports around the world.

Section 3 (Chapter 2.5) will examine the degree of agreement between texts and imagine a way forward for further research. Measurement standardisation and QA/QC will also be explored in this section.

2.3 (1 -) Emissions measurement of UFP from jet engines.

Jet engines do not lend themselves easily to direct exhaust emissions measurements. The quantities of exhaust gases, speed of the emissions plume, temperature and complex mixing processes all combine to make gathering meaningful data *at source* extremely difficult. The International Civil Aviation Organisation (ICAO), through its Committee on Aviation Environmental Protection (CAEP), is pursuing the adoption of a standard method for exhaust sampling and testing, based on the SAE

Aerospace Information Report 6241 from 2013 which documents such a procedure, published in 2013 (30).

Despite this difficulty, there are a few studies that successfully characterise UFP emissions from jet engines. Durdina et al (2014) (20), Abegglen et al (2015) (21), Lobo et al (2015) (22), Abegglen et al (2016) (23), Huang et al (2016) (24), and Delhaye et al (2017) (18) have all made assessments of jet exhaust emissions at a range of operating conditions, but specifically only targeted non volatile particle emissions, using heated sampling systems to keep the volatile species in the gas phase. Yu et al (2017) (19) conducted a survey to assess volatile and non volatile species within the exhaust, while Turgut et al (2015) (25) undertook a survey to assess concentrations of gases, which quantifies both organic and inorganic gases in the emissions. Beyersdorf et al (2014) (26) in a study that primarily explored the possibilities for alternative fuels undertook detailed assessment of exhaust composition at the source, 30 metres and 145m, to assess composition as the plume ages, cools and mixes.

A summary of the surveys is presented in tables 1 and 1a below. The tables present information about what pollutants were measured, including details of techniques used and any supplemental measurements, whether volatile and / or non volatile particles were measured during the studies, sampling distances from the source for the measurement of particles and whether any steps are documented to ensure robustly quality assured and quality controlled data were obtained.

Table 1 – characterisation of emissions from jet exhausts

Study	UFP	PSD	BC	Speciation	Multiple engine types	EC/OC	V/NV	QA/QC	Distance from exhaust
Durdina et al (2014) (20)	Y	Y	Y	N	N	Y	NV	in data handling and sample transport, but measurement not specified	At exit
Abegglen et al (2015) (23)	Y	Y	N	N	N	N	NV	sample transport, but measurement not specified	At exit
Lobo et al (2015) (22)	Y	Y	N	N	Y	N	V and NV by dil	sample transport, but measurement not specified	At exit & also downwind
Abegglen et al (2016) (23)	Y	Y	N	Y	Y	Y	NV	not stated	At exit
Huang et al (2016) (24)					Y		NV	not relevant	At exit and 30m
Delhaye et al (2017) (18)	Y	Y	Y	Y	N	Y	NV (for PM) V from gases	Considered for sampling and data processing	At exit
Yu et al (2017) (19)	Y	Y	Y	N	y	N	V and NV	sample transport, but measurement not specified	10 - 15m
Beyersdorf et al (2014) (26)	Y	Y	Y		N	N	V and NV	Considered for sampling and data processing	1m, 30m, 145m
Turgut et al (2015) (25)	N	N	N	N	N	N	Heated inlet	not stated	In engine
Vander Wal et al (2014) (27)	-	-	-	-	-	-	-		1m and 30m
Vander Wal et al (2016) (28)	-	-	-	-	-	-	-		1m and 10m

Key:
UFP – Ultra Fine Particles. Expressed as total particle count, no size differentiation
PSD – Particle Size Distribution. Number of particles within a specific size range
BC – Black Carbon
Speciation – Further analysis of particles collected, e.g. metals, organic compounds
EC/OC – Elemental Carbon and Organic Carbon.
V/NV – Volatile and Non Volatile UFP component. Separation usually achieved by a thermally controlled sampling system
QA/QC – Quality Assurance and Quality Control

Table 1a – Campaign measurement details

Study	UFP Techniques used	Particle sizes reported	Other pollutants measured	Range of PN size mode
Durdina et al (2014) (20)	AVL model 389 FMPS TSI 3091-DMA 3081-TSI 3776 SMPS TSI 3096-DMA 3081-TSI 3776 Cambustion DMS 500	Analyser dependent: range 15nm – 160nm	THC, CO, CO_2, NOx,	Focusses on mass and particle density, not number distribution
Abegglen et al (2015) (23)	SMPS TSI 3096-TSI 3081-TSI 3776 Cambustion CPMA, TSI 3776	Analyser dependent: range 7nm – 225nm	None	Thrust dependent 25 to 60nm (NV)
Lobo et al (2015) (22)	Cambustion DMS 500	10nm – 200nm	CO_2	Thrust dependent 10 to 50 nm (NV)
Abegglen et al (2016) (23)	ATOFMS TSI 3800-030 SMPS TSI 3936	30nm – 300nm (TOF-MS), 7nm – 250nm (SMPS)	ICP-MS	Thrust dependent 15 to 60 nm
Huang et al (2016) (24)	Not used	-	TEM	Not assessed
Delhaye et al (2017) (18)	SMPS TSI 3087-TSI 3085-TSI 3025 CPC Grimm 5.403 Cambustion DMS 500 Pegasor PPS-M	Analyser dependent: range 5nm – 250nm	MS, Struct. Dens. CO CO_2 HC NOx	Thrust dependent 20 to 60 nm
Yu et al (2017) (19)	AVL APC EEPS TSI 3090	Analyser dependent: range 6nm – 560nm	SP-AMS CO_2 CO HC	Turboprop: 40nm at all thrust settings, Turbofan: thrust dependent 10 - 40nm (both V/NV)
Beyersdorf et al (2014) (26)	CPC TSI 3775 EEPS TSI 3090 SMPS TSI 30xx-TSI 3776	Analyser dependent: range 4nm – 310nm	CO, CO_2, HC, SO_2, SO_4^{2-}, Organic	Varies with thrust and distance from engine: 1m 40-100nm 30m 15-100nm (bimodal) 145m 15-100nm (bimodal) (all V/NV)
Turgut et al (2015) (25)	Not used	-	THC, CO, CO_2, NOx	Not assessed
Vander Wal et al (2014) (27)	Not used	-	Optical	
Vander Wal et al (2016) (28)	Not used	-	Optical and chemical analyses	

Durdina et al (2014) (20) - The paper evaluates and corrects for losses of PM sample to the sampling systems and uses these corrections to evaluate emission indices. While the paper states that multiple pollutants were measured, most of the analysis focusses on particle density and integrated particle size distribution (IPSD), used by Liu et al (2009) (29) to evaluate UFP emissions from diesel engines, concluding that effective particle density (calculated using particle density and particle shape) is

lowest at lower thrust settings for all particle sizes, whereas at higher thrust settings, particle density was higher, suggesting that smaller particles are associated with high thrust and vice versa. It further confirms that there are higher concentrations of non volatile particles associated with lower thrust settings.

Abegglen et al (2015) (21) – The study mainly focusses on particle density, but concludes NV PSD at 65%+ thrust broadly peaks at 40-70nm. At <30% thrust, the peak count is much lower, and smaller particle sizes at 20-30nm. Analysis of the particle density at different thrust settings shows that lower thrust is associated with less dense particles, but also that particle density decreases with increasing particle size.

Lobo et al (2015) (27) – The study looked at measurements at different thrust settings for three different engine / aircraft at the exhaust. They also ran mobile surveys at the end of the active runways during the survey to examine real world measurements. Results show significant differences in PSD between engine types and thrust settings. The CF6 engine shows maximum particle numbers below 20nm for all thrust settings (highest particle numbers were seen to peak at both low and high thrust settings, reducing significantly for middle thrust settings). The JT8D engine shows two modes: small particles dominate at low thrust, larger particles at high thrust. Comparatively low particle numbers were measured at 30% thrust. PW2037 engine shows highest concentrations of the smallest particles at low thrust, (slightly) larger sizes and much fewer particle numbers at higher thrust, minimum particle numbers at 15% thrust. CF6 engines gave the highest particle number measurements of the three engine types (1.5x higher than the other engines). The end of runway studies identified plumes from over 300 individual exhausts (100-350m from the aircraft), allowing average PN Emission indices to be calculated for 11 different types of engine. JT8D engines gave lowest particle number counts, the remaining engine types were all reasonably similar (1-3 x 10^{17}). Most CF-type engines give lowest mass emitted per kg of fuel burned, and within a narrow range, other engines have a large range of outputs.

Abegglen et al (2016) (23) - The paper looks specifically at the composition of PM emitted directly from three different types of aircraft engine, using a TOF MS capable of analysing and sizing individual particles (vacuum aerodynamic diameter). The analysis is not quantitative however, as

multiple components in a single particle can compromise the results. TOF MS EC/OC/TC measurements have in the past been shown to agree with NIOSH method, but this is not assessed here. Depending on thrust and engine type, ~95-98% of the particles measured by TOF MS contain EC, metals mixed in with these particles, confirming that the particles are a diverse matrix of non-volatile components. Differences were identified in the metals detected from the three different engine types, this observation could be used to specifically identify engine during movement / take off. However, it was not possible for the study to successfully differentiate between metals generated by the exhaust and the typical atmospheric distribution of these metals in an urban environment. Only Cobalt and Zirconium might have potential to be used as aircraft tracers. EC/TC ratios for one engine type tested shows minimum of 0.96 at 30% thrust, approaching 1.00 for high thrust and similarly approaching 1.00 at 5% thrust (3% thrust shows a drop in the ratio, suggesting another change in combustion efficiency), but uncertainty in the EC/TC calculations could be a significant contribution to these observations. Unfortunately, while the TOF MS equipment used in this study was optimised to analyse particles in the range 30-300nm, assumptions made about particle shape and density of the analysed particles made it impossible to assess the particle size for smaller particles. The results from TOF-MS, while interesting, may be of limited value for the sub-100nm investigation of this review paper. The study used ICP-MS for analysis of fuel, oil and engine wear samples for comparison to TOF MS data.

Huang et al (2016) (24) – This paper looks at TEM assessment of non volatile particle emissions from different engines. Soot samples were collected at high thrust settings from 4 different engine types. The results were compared with a soot source from a Santoro type generator (Ethylene diffusion flame, which differs from most contemporary processes that use a propane-based soot generator). There was evidence that aircraft soot structure is much more compact in nature than the ethylene soot, which is more open and "stringy". Nevertheless, it was still possible to differentiate the morphology of the soot particles from the different engine types, most likely due to differing conditions during the growth process. The similarity between the different aircraft exhaust particles as they get significantly larger is probably as a result of the similar cooling conditions post combustion.

Delhaye et al (2017) (18) - Wind tunnel tests examined the symmetry of emissions profile 5cm from exhaust at 16 individual measurement points in an "X" shaped sampling configuration, for engine thrust settings between 30 and 100%. This differs from the multi hole "rake" normally used for these exercises. The shape of soot particles mirrors the fractal growth seen by Huang et al (2016) (24) – small, discreet clusters at low thrust, "stringier" agglomerations at increased thrust. ~97% of the particles analysed are carbon atoms, ~3% oxygen, ~0.1% sulphur. Traces of Ca, Ba and P were also identified in the analyses, probably due to lubricating oil. EC composition of PM dominates at high thrust (thrust settings 70+%), OC at lower thrust (thrust settings below 30%), contrasts with Abegglen et al (2016) (23) which suggested that content was always EC dominated. This observation may be due to differences in the analyses / engines used for the different studies. The authors suspect that condensation of volatile components in the sampling line as primary cause. But the observation agrees with, e.g. Timko et al (2014) (31). Analysis of the OC showed that it was correlated with PAH concentrations – possibly due to OC depositing on EC, as a result of lower combustion temperatures. A large variation between measurements of PN with different techniques was identified (2-3x range). The authors attributed this to differences in the penetration indices for the different instrument types and state that corrected values are in good agreement with each other. There are also differences for mean particle size, but less than 1.5X in all cases. All PSD results agree with other papers e.g. Beyersdorf et al (2014) (26) that suggest small particles at low thrust, larger at high thrust. Again PSD at higher thrust shows mean particles (measured with SMPS) between 30-80nm. DMS500 PSD profile is broader and stretches down to finer particles. The mixing of the exhaust at the exit appears to show highest PN concentrations at the middle of the plume. Concentrations appear to be up to 15% lower at the outside of the plume, suggesting that probe position in the exhaust is important for representative sampling to be collected for analysis.

Yu et al (2017) (19) - This paper looked at emissions of volatile and non volatile emissions from two engines; one turboprop and one turbofan. The International Civil Aviation Organisation (ICAO) Committee on Aviation Environmental Protection (CAEP) is developing a new standard for NV PM emission from aircraft, likely to be published and implemented by the end of 2018, but Yu et al used a method "representative of real world PM emissions", not the AIR6241, 2013 (30) methodology: they

used an unheated inlet using concentric N_2 dilution to prevent condensation, 1m from the exhaust. The study - conducted on a single day - confirm very little CO and unburned HC in either exhaust – less than 0.2% of the CO_2 concentrations. For the turbofan, maximum particle numbers were seen at less than 50% thrust, peaking at ~10nm particle size. Particle size distribution at higher thrust was seen to be slightly bimodal but far more dominated by 30-60nm particles and at much higher PN than the other mode. PSD at 50% thrust is a mix of 10nm and 30-60nm particles at similar numbers to lower thrust. Reported data are corrected for sampling efficiency: the authors used a model to correct for turbulent diffusion losses to the 40 metre long sampling system. But the smaller particles are not quantified in this study. Losses of <50nm particles were judged to be significant by the author - emission indices for the turbofan at different thrusts follow a similar U shaped profile to other studies i.e. highest at low and high thrusts, lower at middle settings. The study calculates that PM in turbofan exhaust emission at full thrust is ~89% NV BC, ~7% Vol HC, ~4% SO_3. As will be seen in later ambient measurement studies, the BC component of the exhaust is significantly reduced at bigger distances downwind from the source. For the turboprop engine, the air/fuel mix is much leaner, thus higher dilution and lower temperature of exhaust gases. PSD is largely the same throughout the thrust range of the turboprop, peaking at 30-50nm. No nucleation mode particles were seen in the plume. There was evidence that the volatile component of the exhaust is already coating the BC particles, presumably as a result of higher dilution and lower temperatures. At full thrust, turboprop emissions are ~70% NV BC, ~23% volatile HC and ~7% SO3. Absolute PN concentrations are about 10 times higher for turboprop over turbofan engines however.

Beyersdorf et al (2014) (26) - The authors undertook measurements of volatile and non volatile UFP, at distances of 1, 30 and 145m from exhaust, as part of a comparison study of a selection of alternative, low sulphur fuels. The examination of the combustion of the lower sulphur fuels showed significant reduction in UFP compared to JP-8. This observation is most likely due to the fact that the low sulphur fuels will create fewer oxidised sulphur particles to act as nucleation sites for cooling exhaust gases. Removal of the sulphurous compounds from aviation fuel presents a different range of challenges; for example, ensuring that the fuel does not cause the fuel line seals to perish and potentially introduce leaks. Further investigation of jet aircraft emissions when using alternative and

low sulphur fuels is warranted, but this is not strictly relevant for this review paper, which focusses on existing fuel and aircraft. An unheated sampling inlet was used. The authors found that virtually all fuel is converted to CO_2 during combustion at thrust settings above 20%, and in any case better than 96% at all thrust settings. At 1m from the exhaust, the calculated emission index for volume of particles at high thrusts peaks at ~60-100nm. As distance increases, particle size distribution profiles at highest thrust settings become bimodal; at 10-20nm and 60-150nm, while very low thrust settings show a huge increase in the volume of ~20nm particles. Most likely, this is due to condensation of volatile components in the exhaust as the plume mixes and cools as it moves downwind. Particle size distribution is certainly influenced by distance from the source and atmospheric processes.

Turgut et al (2015) (25) – This paper evaluated measurement of exhaust gases from eleven notionally identical jet engines at a range of different thrust settings. Whilst not directly connected with UFP measurements, it confirms results from Beyersdorf (2014) (26) that HC (and CO) is emitted in higher quantities at low thrust settings when compared to higher thrust settings. It also finds that measured data shows some deviation from ICAO emission indices, suggesting that real world operations may not accurately be reflected by the official literature.

Vander Wal et al (2014) (27) - Undertook a study of the soot emitted from a selection of different engines, analysing the particles using transmission electron microscopy (TEM) and X Ray photoelectron spectroscopy (XPS). Samples were collected at a range of different thrust settings. The study focussed on a single aircraft and found that thrust settings have a marked effect on particle size, shape and composition, low thrust settings being associated with smallest particle sizes. Using XPS, the particles were assessed to be predominantly organic carbon at low thrust and elemental carbon at higher thrust settings.

Vander Wal et al (2016) (28) - Followed on from the 2014 paper to examine the speciation and chemical composition of exhaust particles from four different engines. Examination of the material collected from the exhausts suggest that differences between chemical speciation can be used to characterise different engines, based on wear profiles and fuel additives used during operation. Differences in elemental carbon and organic carbon output at different thrust settings were again

observed during the tests. It was also observed that the non volatile PM component was actually hydrophilic in nature, suggesting that agglomeration of NOx and SOx components onto these particles was a likely mechanism for nucleation.

There have been some modelling studies, notably Koudis et al (2017) (32) that have explored the relationships between aircraft operating modes at ground level (including take off thrust settings) and exhaust emissions, which confirm that heavier aircraft, different engines and higher thrust settings are associated with modelled increases in NOx and BC emissions and suggest that lower thrust settings might improve emissions of these pollutants. There was no consideration of the impact on UFP emissions; from the measurement evidence above, it could be suggested that higher emissions of smaller particles could result from lower thrust settings. As noted earlier, research contributions from modelled data will not be extensively considered in this review paper.

Summary of engine measurements:

There has been reasonable effort in characterising the exhaust from jet aircraft, especially in recent years. Most of the research focusses on characterising the non-volatile component of the exhaust, as dictated by the sampling protocols and the requirement for heated inlets to eliminate condensation of the volatile / water present in the exhaust.

The literature seems to agree that:

- The vast majority of non volatile PM from aircraft exists as carbon particles, soot or organic carbon.

- At low thrust settings, organic carbon particles are more common, most likely as a result of the reduced engine temperature at these settings and the likelihood that volatile compounds condense on the carbon particles.

- Combustion in the engine is very efficient – research suggests that combustion of fuel is better than 95%, and close to 100% for any settings above idle.

- The particle size distribution research finds that finest particles, less than 30nm in diameter, are associated with low thrust settings. Larger particles, in the range 30-90nm are associated

with thrust settings from 35% and higher. Particle number concentrations follows a similar trend in thrust settings.

- A number of studies have shown that engine design (and fuel burned, in an alternative fuel experiment) is critical to the concentrations and sizes of particles created. Differences between turboprop and turbofan principles, as well as turbofan combustion chamber design all have an effect. It is possible, with sufficiently high resolution measurements to qualitatively differentiate between different engine types.

- Speciation of the exhaust components has not quantitatively succeeded in identifying key tracers unique to aircraft activity. The composition and proportions of aircraft exhaust components are too similar to typical human activity to adequately differentiate between them.

As noted earlier, a standard methodology for exhaust sampling has been agreed and a standard method is in preparation by CAEP, which will improve the comparability of measured data from future papers. One study has shown strong evidence that the plume composition changes dramatically as it travels from the engine. Mixing and cooling causes nucleation of volatile and sulphate compounds, which are present in large particle numbers as distance increases. Very high concentrations of particles smaller than 20nm are measured significantly downwind of the source.

While the non-volatile component of the plume at the exhaust has been the most researched area of investigation in the papers discussed above, the impact of the aging plume on the area around the airport is likely to be of more interest to the community.

The quality assurance and quality control protocols adopted for the above papers appears to focus on a few key aspects:

- Sampling design
- Preserving sample integrity (or correcting for losses)
- Protocols for post processing of data

None of the documents present any detail about the robustness or integrity of the actual measurement data. There is no discussion about the quality of the sample delivery systems, calibration of the

analysers or servicing and maintenance regimes used to ensure the reliable operation of the analysers for the studies. It can be strongly argued that the calibration of the measurement devices and sampling systems is the most important parameter of the entire study. A lack of characterisation of analysers before (during) and after the study, perhaps under an assumption that changes in calibration, drift and repeatability are all negligible, carries a very significant risk. While this lack of published detail might be acceptable for discreet studies, it brings several compromises when comparing papers:

- It means that comparability between studies becomes more difficult
- General trends between studies can only be implied, it would be extremely difficult to draw solid conclusions by comparing results from different researchers and institutions.
- The data are not likely to be sufficiently robust to feed into any heath effect studies

It would be extremely beneficial to future research if this QA/QC detail could be brought to subsequent studies and papers.

2.4 (2-) Ambient measurement of UFP at Airports

In some ways, measurement of UFP at large distances from aircraft is much less challenging than measurements at the exhaust of a jet turbine. There is no need to condition the sample inlet – the temperature of the plume is at prevailing conditions, residence time in the sampling system is minimal compared to the age of the plume, diffusive losses are easily calculated and minimised and the location of the measurement equipment is (usually) safe from the hazards of jet blast. However, the nature of the exhaust plume will have changed significantly in transporting from the exhaust to the measurement location: cooling, condensing, nucleation and potentially evaporation of water, volatile organics and sulphates will all contribute to this dynamic.

Compared to measurements of more traditional air pollutants, there are few measurements of ambient UFP. In the UK for example, there are 4 monitoring locations that routinely measure UFP (http://uk-air.defra.gov.uk). While there is not a shortage of peer-reviewed articles looking at air quality around airports, only a small selection of these measurement studies specifically focus on the impact of airport-related UFP.

Table 2 – Ambient measurements of UFP near airports

Study	UFP	PSD	BC	Others	QA/QC	Distance from aircraft	Duration of study
Fleuti et al 2017 – Zurich (13)	Y	Y	N	LDSA,	Yes, before and after survey	Varies – transects in survey	5 weeks
Ellermann et al 2011 – Copenhagen (33)	Y	Y	N	N	Not documented in English section of report	2 locations, 1 runway, 1 apron, plus roadside and background	2 months total
Ellermann et al 2012 – Copenhagen (14)	Y	Y		NOx, SO_2, $PM_{2.5}$, VOC, EC/OC, PAH	Not discussed	Varies – 3 to 15 locations	Between 4 weeks and 5 months
Fanning et al 2007 – LA International (LAX) (34)	Y	Y	Y	$PM_{2.5}$, BC, PAH, VOC, CO_2	Not discussed, except for remedial action after breakdowns	140m main, up to 600m for dispersion, plus regional comparison	2 studies, less than 1 week each
Hudda et al 2014 – LAX (11)	Y	N	Y	NOx, PAH	Not discussed	Varies – transects from 0-18km from airport	29 discreet measurements – 1 hour to 11 hours (5hrs max) duration, vary times of day
Hudda and Fruin 2016 – LAX (12)	Y	Y	N	LDSA	Cross referencing between instruments	Varies – transects from 0-18km from airport. Also spot measurements of PSD	12 discreet measurements – 3 to 10 hours (5hrs max) duration, vary times of day
Westerdahl et al 2007 – LAX (10)	Y	Y	Y	NOx, PAH	Not discussed	Varies – driving mobile monitoring around the perimeter	4 afternoons in April 2003
Masiol et al 2017 – Heathrow (35)	Y	Y	Y	NOx, PM_{10}, $PM_{2.5}$	Yes, before during and after surveys	1km from northern runway	2 x 1 month long surveys
Moller et al 2014 – Copenhagen (36)	Y	Y	N	-	Assessed before start of study	Personal exposure of 30 airside employees	8 days
Moore et al 2017 – LAX (37)	Y	Y	Y	CO_2 / H_2O	Manufacturer specs quoted. Some comparison and processing of different measurement techniques	400m from runway	2 days
Peters et al 2016 – Brussels (15)	Y	Y	Y	NOx, PM_{10}	Yes, all analysers cross referenced and normalised	Various – transect of 4 sites along the 07L/25R runway. 250m to 7km from the airport perimeter	2 months
Keuken et al 2015 – Schiphol (38)	Y	Y	Y		Yes, in set up and ongoing QC	2 stations, 7 and 40km from Schiphol airport	3 months, two different campaigns 2012 and 2014

Study	UFP	PSD	BC	Others	QA/QC	Distance from aircraft	Duration of study
Bezemer et al 2015 – Schiphol (16)	Y	-	N		Not discussed in English summary	10 stations around the airport	6 weeks, different systems
Costabile et 2015 – Rome (40)	Y	Y	Y		Wiedensohler et al 2012 used for PN analysers	2 stations, <1km and 5km from the airport	Two three month studies, summer and winter.
Stafoggia et al 2016 – Rome (39)	Y	N	N		Not discussed	1 station at far end of runway	2 weeks Summer, 2 weeks Winter
Shirmohammadi et al 2016 – LAX (41)	Y	Y	Y	$PM_{2.5}$ CO_2	Calibration stated, but no further detail	1 fixed, 1 mobile	6 x 6 hour surveys in summer 2016
Riley et al 2016 – LAX and Atlanta (42)	Y	Y	Y	NO_2	Calibration of analysers, but up to 2 years before study	Various – mobile transects in both areas	2 weeks at each, constrained to 12-19:00
Masiol et al 2016 – Venice (44)	Y	Y	Y	APS, NOx, $PM_{2.5}$, CO	Yes, factory service and calibration, referenced with other analysers	1 location, at south western edge of airport	6 weeks
Hudda et al 2018 – Boston (45)	N	Y	N		Yes, factory service and calibration, referenced with other analysers	Multiple locations examining indoor and outdoor air	6 weeks
Ren et al 2018 – Tianjin indoor terminal (46)	Y	Y		$PM_{2.5}$, CO_2	Factory calibrated, intercompared for surveys	20 locations inside the terminal building and one outside	3x 2 day monitoring campaigns
Hu et al 2009 – Santa Monica (47)	Y	Y	Y	$PM_{2.5}$, PAH, CO, NOx, CO_2	Calibration stated, but no further details	Mobile surveys at 5 locations	4 days, up to 6 hours per day, rotating locations
ACI Europe 2012 (48)							Summarising known work at airports until 2012
ACI Europe 2018 (49)							Updates relevant work at airports until 2018

Table 2a – Campaign measurement details

Study	UFP Techniques used	Particle sizes reported	Maximum Particle number concentration ranges (mode)	Range of PN size mode
Fleuti et al 2017 – Zurich (13)	DiSCmini SMPS TSI 3080-3775 (1 minute resolution)	DiSCmini – mode size SMPS 6.4 to 200nm	from ~$1x10^4$ (no aircraft) to ~$2x10^5$ p/cm^3 (aircraft)	from 50nm (no aircraft) down to ~12nm (aircraft)
Ellermann et al 2011 – Copenhagen (33)	DMPS (2 minute resolution)	6nm to 700nm	from $4x10^3$ (background) to $5.5x10^4$ p/cm^3 (on airport)	from 70nm (background) to ~20nm (on airport)
Ellermann et al 2012 – Copenhagen (14)	DMPS (2 minute resolution)	6nm to 700 nm	Total PN from $3x10^3$ (background) to $4x10^4$ p/cm^3 (on airport)	Aggregated into 6-40nm bin. Airport PN is ~88% particles <40nm, background is ~42% particles <40nm
Fanning et al 2007 – LA International (LAX) (34)	SMPS TSI 3080-3025 (2 minute resolution)	6nm to 290nm	$1x10^4$ (background) to ~$2x10^6$ dN/dlogDp (on airport)	80nm (background) to ~15nm (on airport)
Hudda et al 2014 – LAX (11)	CPC TSI 3007 (1Hz resolution)	Total PN	Up to $7x10^4$ p/cm^3 reported but study focussed on increased loading to urban traffic locations. Aircraft contribution 2-8 times "normal" concentrations downwind of the airport	n/a
Hudda and Fruin 2016 – LAX (12)	CPC TSI 3007 (1Hz resolution) SMPS TSI NanoScan 3910 (1 minute resolution) DiSCmini (1Hz resolution)	Total (CPC) 10nm to 420nm (SMPS) Mode and total PN (DiSCmini)	from $2x10^3$ p/cm^3 at background locations, up to ~$1x10^5$ p/cm^3 directly down wind of the airport. Smallest particles associated with airport emissions	60-100nm (coastal) 25-60nm (urban) 10-15nm (downwind of airport)
Westerdahl et al 2007 – LAX (10)	SMPS TSI 3080-3007 (1 minute resolution) SMPS TSI 3080-3022A (1 minute resolution) EAD TSI 3070A (10 second resolution)	16nm to 600nm 5nm to 153nm Particle length	from $1x10^4$ at coastal site up to $1x10^7$ dN/dlogDp at end of runway	80nm at coastal site and ~20nm and ~80nm modes at end of runway (20nm dominates)
Masiol et al 2017 – Heathrow (35)	SMPS TSI 3080-3775 (5 minute resolution)	14nm to 673nm	$8x10^4$ dN/dlogDp (daytime) to $3X10^4$ dN/dlogDp (night)	15-20nm. Some evidence that mode might be smaller in summer
Moller et al 2014 – Copenhagen (36)	Philips NanoTracer (16 second resolution)	Mode and total PN	$5X10^3$ p/cm^3 (background), up to $5x10^5$ (apron)	27nm (smallest mode landside) to 17nm (smallest mode airside)
Moore et al 2017 – LAX (37)	CPC TSI 3022A (1 second resolution) EEPS TSI 3090) 1 second resolution	Total PN 6nm to 575nm	Individual aircraft characterisation, compiled as emission indices	10-20nm, evidence of second mode at ~100nm

Study	UFP Techniques used	Particle sizes reported	Maximum Particle number concentration ranges (mode)	Range of PN size mode
Peters et al 2016 – Brussels (15)	SMPS TSI 3936L76 (5 minute resolution) SMPS IfT custom-TSI 3772 5 minute resolution)	10nm to 300nm	$4x10^4$ p/cm^3 (rural) to $3x10^5$ p/cm^3 closest to airport. All stations underneath flight/glide paths	10-20nm. Evidence of second mode at 30-50nm for station closest to Brussels
Keuken et al 2015 – Schiphol (38)	SMPS TSI 3034 (3 minute resolution)	10nm to 480nm	Averaged - $2x10^4$ dN/dlogDp (background) to $8x10^4$ dN/dlogDp (airport)	50-60nm background to ~20nm (airport)
Bezemer et al 2015 – Schiphol (16)	SMPS TSI 3031 EPC TSI 3783 SMPS Grimm CPC TSI 3775 CPC TSI 3022 DiSCmini Philips NanoTracer	Not stated in document – only total PN is reported	Median concentrations around airport: $2x10^4$ to $5x10^4$ p/cm^3	n/a
Costabile et al 2015 – Rome (40)	CPC TSI 3022A BC Ecotech mod.Aurora 3000 / Radiance PSAP SMPS – not stated	Not explicitly stated – data for Nucleation, Aitken, Accumulation 1 and Accumulation 2 particles reported, corresponding to 4 mean sizes between 20-700nm. BC particles of 60-200nm investigated for optical properties	<20nm particles up to $2x10^6$ p/cm^3	n/a
Stafoggia et al 2016 – Rome (39)	CPC TSI3022A (1Hz resolution)	n/a	up to $3x10^6$ p/cm^3, median ~$3x10^4$ p/cm^3	n/a
Shirmohammadi et al 2016 – LAX (42)	DiSCmini	Mode range 7nm to 500nm	$1x10^5$ p/cm^3 (roadside) to ~$5x10^5$ p/cm^3 (airport)	30-40nm (roadside) to 15nm to 20nm (airport)
Riley et al 2016 – LAX and Atlanta (42)	Grimm NanoCheck 1.320 P-Trak 8525 CPC TSI 3007	25nm to 400nm Total PN >50nm Total PN >10nm	LAX median ~$7x10^4$ p/cm^3, ATL median ~$2x10^4$ p/cm^3	LAX ~30nm (airport), 40nm (freeway / background) ATL ~40nm (airport), 60-100nm (freeway / background)
Masiol et al 2016 – Venice (44)	SMPS TSI 3080-3022A (5 minute resolution) APS TSI 3321	14nm to 673nm 500nm to 19,800nm	Up to $6x10^4$ dN/dlogDp (daytime), $8x10^3$ (overnight)	<20nm (daytime), ~50nm (overnight)
Hudda et al 2018 – Boston (45)	CPC TSI 3083 (30 or 60 second resolution)	Total PN	Up to $5x10^4$ p/cm^3 for winds associated with airport, 80% lower PN for winds from opposite direction.	n/a
Ren et al 2018 – Tianjin indoor terminal (46)	CPC TSI 3007 (1 second resolution) AMS MSP ASM1500 (1 minute resolution)	Total PN 15nm to 1000nm	Terminal $8x10^4$ p/cm^3, Apron $1.2x10^5$ p/cm^3, Background $2x10^4$ p/cm^3	Terminal ~30nm and 100nm. Possible 3rd mode <15nm Background 100nm

Study	UFP Techniques used	Particle sizes reported	Maximum Particle number concentration ranges (mode)	Range of PN size mode
Hu et al 2009 – Santa Monica (47)	FMPS TSI 3091 (10 second resolution) CPC TSI 3007 (10 second resolution)	6nm to 560nm Total PN	$3x10^4$ p/cm^3 at 75m from airport to $1x10^6$ p/cm^3 closest to airport	11nm for departing aircraft plumes, 22nm for diesel HGV

Fleuti et al (2017) (13) undertook an extensive survey across two transects and 11 locations at Zurich Airport. They used cross referenced and calibrated Miniature Diffusion Size Classifiers (MiniDiSC) at all sites, together with a central TSI SMPS/CPC set up at a single location. The survey was a 5 week campaign from April to June 2016. The MiniDiSCs were configured to measure particles in the 10 - 300nm range, and report particle number and average size every second. The SMPS/CPC was configured to scan every minute in the range 6.4 - 217 nm. Some quality control was applied to the MiniDiSC instruments: cross referencing at the start of the campaign showed that when the analysers were challenged with measuring 20nm particles, agreement was within 11% for measurements of diameter and particle number, while this worsened slightly to 20% after 5 weeks. The authors conclude that the analysers were operating satisfactorily for the survey, suggesting that inter analyser variability was typically 10%. At 20nm, this would suggest that the MiniDiSCs were able to size particles to within an accuracy of ±2nm. The study shows that average particle size decreases during the day, while total particle number increases, coinciding with aircraft activity. The report found that measured concentrations were dependent on meteorology; highest concentrations were seen when the measurement stations were directly downwind of aircraft activities. The transects also demonstrated that highest concentrations were seen in the centre of the airport, apparently dropping off significantly at a distance of 2.5km from the centre of the airport. The study attempted to assign peak events to individual aircraft, but found significant challenges in achieving robust correlation between aircraft and peaks. When bulked to hourly averages, the correlation between aircraft movements and concentrations was much better. The study also examined the influence of road vehicles on the measurements at one location, where aircraft and vehicles were in close proximity to each other. Use of polar plots examining particle number and mean particle diameter show clear differences: larger particles are associated with road vehicles, higher PN and smaller particles with aircraft.

Ellermann et al (2011) (33) undertook a 2 month long survey of UFP at Copenhagen airport at three different locations, primarily to assess exposure of airside workers and to place this into context for "normal" ambient environments. The initial study report, expanded below, compared measurements at the apron near the terminals, a runway location, 2 roadside locations and a background site. Data from one of the roadside sites is not concurrent with the other 4 datasets, so any comparisons should be made with caution. Measurements at the terminal apron, which is downwind from all aircraft activity, and most likely to represent the highest area of impact, were highest of all stations for particles sized between 10 and 40nm. For particles larger than 40nm, the number measurements at the apron are similar to one of the roadside locations and lower than the other (not concurrent) roadside location. Peak particle number size at the apron and runway sites is ~15nm, compared to the roadside maximum at ~30-50nm. Average apron particle numbers between 6-40nm are ~4 times higher than the runway station, ~2.5 times higher than the (non concurrent) roadside station and ~8 times higher than the other roadside station. The diurnal variation of the particles shows the 6-40nm fraction at the apron station is considerably higher than all of the other comparators, and coincides exactly with aircraft activity at the airport. Diurnal data from 6-40nm at the runway site is very similar to the other stations, suggesting that this station was not unduly affected by aircraft activity during the survey.

Ellermann et al (2012) (14) produced a second technical report for Copenhagen Airport, specifically to examine the air quality in the airside working environment. A wide range of pollutants were measured, providing an ability to infer source apportionment to the measured concentrations. Concentrations of most "conventional" pollutants (for example NOx, $PM_{2.5}$, PAH, VOC, EC/OC) were found to be lower than those observed at a nearby busy road location, while concentrations of UFP were seen to be up to three times higher in the airside environment. Two campaigns were conducted, spring and autumn 2010. Three fixed locations were used, all permanent stations with varying levels of monitoring in normal operation. During the campaigns, UFP was measured at all locations (usually just one station at a time). Separate campaigns for $PM_{2.5}$ NOx and VOCs were undertaken at a larger number of stations, using "simple and relatively low-cost instruments", though these are not described or the quality control of these data discussed in the report. Particles smaller than 40nm were found to make up 85-90% of the PN composition at the apron, while particles in the range 40-700nm are

comparable in distribution and number to the nearby roadside location, over the two surveys. Supplemental surveys, using hand-held NO and PN devices (again not described in the text), show that many different processes can influence local exposure. Auxilliary Power Units, main engines, service vehicles and snow removing processes were all seen to contribute to measured concentrations. A supplemental study to explore the relationship between SO_2 and PN showed reasonable correlation, suggesting that jet exhaust particles have some association with the sulphur in jet fuel. A study of the composition of $PM_{2.5}$ sampled suggested that there is a source of OC at the airport, when compared to EC concentrations.

Fanning et al (2007) (34) produced a report exploring PN measurements 140m from the southern runways at Los Angeles International airport (LAX) and compared this data to seven background / community sites in the area. In addition, 15nm particles were measured downwind of the airport, to assess how the concentrations varied with distance from the source. For the SMPS data, scans undertaken every 2 minutes, from 6.15 to 225nm (3081 DMA) at the closest station to the aircraft. This station was also used on a total of five days to sample at high temporal resolution for a narrower selection of particle sizes. Average particle size distributions for both campaigns shows highest particle numbers are associated with 10-20nm particles at the airport, contrasting with 60-100nm particles seen at the nearby background location. There is some evidence that the summer campaign had higher average concentrations of particles smaller than 15nm, compared to the winter campaign. The winter campaign, on average, observed more particles larger than 15nm, compared to the summer campaign. The background station does not show a peak in the 10-30nm range at all. Examination of the high resolution CPC data was able to identify peaks with aircraft activity, both take off and landing. Take off peaks were much higher than peaks from landing aircraft. Similar observations were possible from the SMPS/CPC when it was set up to examine single size particles at high time resolution. The short BC surveys showed that concentrations at the airport were significantly higher than the background location ($14\mu g/m^3$ vs $\sim 1\mu g/m^3$). $PM_{2.5}$ concentrations at the airport location were also higher than the background station, though the PAH components of the $PM_{2.5}$ at the airport were not statistically different to the PAH in $PM_{2.5}$ measured at the background station. The downwind dispersion studies (2 hours of measurements on 4 different days) looked at concentrations of 15nm

particles and BC at up to 600m from the two southern runways 25R and 25L (25R is primarily used for take off, 25L for landing). The studies showed highest concentrations associated with take off. Individual aircraft take off events can still be observed ~300m from the runway, although the magnitude of the peaks does not always match, indicating that dispersion plays a major role in the measurement study. Concentrations of 15nm particles fall slowly over 600m: over 60% of the particles measured at the blast fence are still measured at 600m. A similar dropoff was not as evident for 25L measurements of 15nm particles, although measured concentrations of 15nm particles and BC were significantly lower than 25R. BC concentrations under the landing aircraft decreased with distance to the airport, suggesting that a different emission mechanism is at work for landing aircraft. The survey confirmed that 15nm particles are emitted in significant quantities from both landing and departing aircraft, but a full particle size distribution analysis for both modes of operation was not performed. Measurements of UFP at the nearby, and immediately downwind, locations showed very similar PSD profiles to those seen at the airport; peaks in average particle concentrations between 10 and 20nm.

Hudda et al (2014) (11) undertook a series of mobile monitoring campaigns to assess PN concentrations downwind of LAX. Measurements of PN were made on transects at increasing distances from the airport. The vertical and horizontal grid structure of the street layout at Los Angeles lends itself well to this methodology. Measurements were coordinated with westerly winds, when airport impact on the community would be expected to be highest. In order to eliminate local interferences, a rolling 5^{th} percentile, taken from 1 second data and averaged over 30 seconds, was used to report PN for the transects. The study identified that the 5^{th} percentile PN concentrations are significantly higher in the areas downwind of the airport, dropping off rapidly once the vehicle drives either north or south, out of the downwind impact zone. The increase in the 5^{th} percentile PN concentration can be seen over large distances, even 18km from the eastern boundary of the airport. The study also observed concentrations when the winds were blowing from other directions. While transects were less structured compared to the east of the city, a similar trend in the PN data is observed. The study authors suggest that plume drifting downwind from aircraft activity is very well mixed by the time the air mass has travelled 5-10 km; no identification of individual aircraft was

observed in the data. Examination of other pollutants (at least on one survey day) showed similarities of BC and NOx to the PN 5th percentile measurements, though some differences are observable, suggesting additional sources for these events. The study concludes that large areas of Los Angeles east of the airport are impacted by elevated baseline concentrations of PN as a result of airport activities.

Hudda and Fruin (2016) (12) undertook additional measurements at LAX, broadly repeating the earlier study, augmenting it with PSD and LDSA information at a selection of fixed sites as well as mobile locations. The study observes that the highest PN concentrations are associated with the smallest diameter particles, and the smallest particles are highest in the areas in a tight envelope downwind of the airport. The survey was able to observe the averaged UFP plume of landing aircraft up to 3km from the airport perimeter, confirming work by Graham et al (2006) that enhanced vertical mixing of the exhaust plume is possible. Two modes were observed in the particle size data associated with airport operations: 15-40nm and 115-150nm. The overwhelming majority of particles downwind of the airport were smaller than 40nm, while size distribution from other wind directions were seen to be more dominated by larger particles. The author suggests that the increased PN concentrations at large distances from the airport are caused by vortices causing the plumes from landing aircraft to have an increased impact at ground level. Examination of the mobile LDSA data confirmed that surface area followed a similar pattern to the PN concentrations, though the magnitude of the differences was lower.

Westerdahl et al (2007) (10) undertook a series of short campaigns over 4 afternoons to examine concentrations of UFP, NOx, BC and PAH around LAX in 2003. The study found that PN was highest on the busy highway roads, inside the tunnel under the southern runways and also downwind of the airport. The study highlighted differences in averaged PSD between coastal and exhaust related environments, though there was some similarity between all emission airmasses, in PSD profiles at least, if not in magnitude. All of the combustion related data show bimodal PSD; with peaks at 10-40nm and 80-150nm. Apart from a single event, the study does not attempt to differentiate PSD between different sources, rather it just reports the average of the four days in each location. The

study was one of the first to speculate that UFP emissions from aircraft may have a significant impact on local communities.

Masiol et al (2017) (35) conducted two monitoring campaigns 1km downwind of Heathrow airport during warm and cold periods. Concentrations of particles <30nm were found to be significantly higher than nearby urban background environments. The average PSD profiles in winter and summer showed differences in magnitude and peak particle size, with higher concentrations and larger particles seen in winter, as a likely consequence of increased condensation of semi volatile particles more likely in cooler weather. Diurnal plots of the nucleation PN data correlate well with airport activity. Cluster analysis of the PSD data, limited to 5 clusters, revealed potential influences from London and surrounding roads, as well as from the airport and aircraft. Winter clusters were mostly associated with SW winds, an artefact of this being the dominating wind direction in the winter survey. PMF analysis of the data revealed 6 different potential factors, with the top three factors in each season being responsible for over 80% of the PN count, at particle sizes <50nm. In both cases, approximately a third of the PN was attributed to the airport, though this accounted for less than 2% of the particle volume. A regional nucleation episode recorded in the warm season showed particle growth clearly during the afternoon and evening, likely associated with regional nucleation as the winds were not from the airport at this time. Evidence suggests that long range transport of larger sized PN from mainland Europe also has a significant effect on measured concentrations at Heathrow. PM and BC in the area appears to be more dominated by contributions from London, rather than from the airport itself. The clustering study suggests that airport appears to be a significant potential source of NO_2.

Moller et al (2014) (36) carried out mobile measurements of UFP using portable devices to assess exposure of airside workers at Copenhagen airport. 30 workers in 5 different job functions were assessed over 8 days. Measurements were linked to GPS sensors, to allow accurate mapping of concentrations and sizes to locations. The data show clearly that exposure is highest for workers who are on the airport aprons and lowest for landside operations, with exposure for the remaining three groups statistically indistinguishable from each other. Apron workers were apparently exposed to higher average concentrations than were recorded from stationary measurements undertaken at the

airport in 2012 ($37x10^3$ vs $29x10^3$ particles/cc), though different measurement techniques were used for these studies.

Moore et al (2017) (37) reports on a study of 275 engine takeoff plumes, recorded from departing aircraft at LAX over 2 days in May 2014. Measurement was on the northern departing runway, other studies have operated at the longer southern runways. Detailed aircraft and engine meta data for 29 different configurations was collected by observers and correlated manually with peaks in PN, BC and CO_2. This allowed averaged emission numbers to be collected for each engine type, relating to PN, NV PN, BC and particle volume, together with an analysis of the various PSDs for each of the plumes. There is clear variation in the volume PSDs from different aircraft, but this is not decoupled or clustered further in this paper. The number PSDs show remarkably similar trends throughout the size range of the EEPS analyser, suggesting the combustion processes are also similar for the engines sampled.

Peters et al (2016) (15) reported on a 2 month assessment of UFP concentrations at 4 stations along a transect of the northern runway at Brussels airport. The survey focusses mainly on particles in the 10-20nm size range, and references everything to the rural station 7km downwind and to the northeast of the airport. This may have an impact of the magnitude of the ratio analyses: aircraft will predominantly be landing by approaching from the NE and the prevailing wind will bring the airport airmass along the transect, so the rural station will inevitably be impacted to a certain extent by this. The Dutch language section of the report suggests that aircraft are at an altitude of 420m at 7km from the runway, 90m altitude at 750m. Results show a drop off in particle number concentrations as distance from the airport increases. The proportion of finest particles compared to the total particle count also decreases with increasing distance from the airport. Polar plots of the 10-20nm particles at the 4 stations showed that highest concentrations were associated with winds from the airport, though at the southwesternmost locations, there is a clear influence from Brussels itself, and these stations are in close proximity to urbanised areas in any case. The very extensive (in Dutch) report concludes that meteorology plays a significant role in the measured data. Additionally, BC, NOx and PM_{10} concentrations at the airport are comparable to other measurements across Brussels.

Keuken et al (2015) (38) presented measurement data from 2 stations in The Netherlands that investigated the impact of Schiphol Airport on communities at some distance from the airport. Two stations, at 7km and 40 km distance from Schiphol were equipped with UFP and BC analysers on 2 separate campaigns. The measurements showed that 10-25nm and 25-100nm particles could be attributed to airport activity, even at 40km distance from the airport. Diurnal assessment of UFP at the closest monitoring station showed close correlation with periods of aircraft activity, reducing significantly during the "quiet" hours. The PSDs from the monitoring stations were examined to identify differences in wind direction. When winds were associated with Schiphol, the closest station saw maximum concentrations of particles at 15-20nm. Overnight, the peak PN size was 50-80nm. For the distant station, when the winds were from Schiphol, a clear difference in peak particle size can be seen compared to other directions, 20-30nm as opposed to 40-50nm from other directions. For all datasets, the larger particle size profiles (larger than ~150nm), the profiles are reasonably similar. Airmasses associated with Schiphol at the nearest station show elevated concentrations of all particle sizes below ~150nm. Examination of the BC measurements showed no increased concentrations could be related to airport activities, suggesting that BC concentrations are dominated by other contributions.

Bezemer et al (2015) (16), (report in Dutch) led a collaboration of 4 teams in the Netherlands to assess concentrations of UFP around Schiphol airport. 10 stations, at varying distances from the airport, were used in the assessment. A range of different measurement techniques were used, from conventional CPC/SMPS through to MiniDiSC and Aerasense portable devices. Measured total concentrations, when combined with meteorological measurements, showed clear influences from airport sources and activities. A dispersion model was also used to investigate spatial distribution of PN, but the agreement between the model and actual measurements was only "reasonable" at half of the stations. Information on the Particle Size Distribution at the stations is not reported within the document.

Costabile et al (2015) (40) undertook a pair of monitoring campaigns at Ciampino Airport and a nearby background location with CPC, SMPS and BC analysers. Measurements were collected in winter and summer, specifically to explore the nature of BC particles in the fine and ultrafine PM

fractions. The survey found that for particles larger than ~90nm, the properties of particles are similar at background and airport locations, further supporting findings from other studies that the finest particles are specific to the airport environment. There was also little correlation between diurnal BC and total PN concentrations at either location.

Stafoggia et al (2016) (39) undertook a pair of two week measurement campaigns at Ciampino Airport in Rome using a CPC analyser. Measurements were conducted in Summer and winter, collecting 1 minute averages for analysis. The studies had mixed success; because the sampling station was at the end of the runway, aircraft were either at an elevated altitude after take off, or had manoeuvred away from the runway upon landing. As a result, not all aircraft emissions were observed by the instrumentation. However, analysis of the (weighted) measurement data by polar plot showed a clear influence of the airport on measured concentrations. The polar plots of the CPC data do not show any influence from the proximity of the roads. The weighting algorithm used to visualise the additional burden of the airport clearly influences all the other data. This work followed on from the study by Costabile et al (2015) (40). Both the 2016 and 2015 papers identified that higher UFP concentrations were associated with winds from the airport.

Shirmohammadi et al (2016) (41) undertook a short series of mobile and fixed monitoring, primarily to assess the emission rates of PN and BC from aircraft at LAX. The study used 5th percentile data to assess the background for the area, but reports actual measured data from the surveys as well. The study found that average daily particle number emissions were 11 times higher from the airport than the three nearby freeways and actual measured concentrations were over 4 times higher than those measured on the freeways. Average particle size at the airport was 20nm, contrasting with the freeways, where the average particle size was typically greater than 35nm. Calculation of emission factors for PN and BC showed no significant differences between take off and landing, but $PM_{2.5}$ emissions appeared to be slightly higher for landing aircraft. The paper states that 175 individual aircraft plumes (95 take off, 80 landing) were identified during the study, but no data or further investigations are presented.

Riley et al (2016) (42) conducted mobile (stationary) measurements of UFP at a number of transect locations around LAX and Atlanta airports. The study measured concentrations on freeways and

background locations at distances of up to 30km from the airports, and additionally focused on approach transects much closer to the airports. The investigation used 5th percentile measurements to assess background concentrations. Significant differences in the concentration profiles were identified at the two airports. Particle counts for <25nm particles were higher at LAX than on the freeways, but the reverse was true at Atlanta. However, particle sizing investigations showed that the mean particle size (less than 20 nm) was an effective tool for differentiating aircraft from other emission sources. The data (together with data from prior research by Kinsey et al (2009) (43)) have been used to calculate emission indices for a selection of aircraft and assumed thrust settings and compared against published papers for Petrol and Diesel fuels. In this comparison, PN to BC ratios for aircraft are typically an order of magnitude higher than road vehicles. The report concludes that measuring 25nm particles and BC and investigating the ratio between these data can usefully aid in the apportionment of data and emissions to specific aircraft sources.

Masiol et al (2016) (44), conducted a study at Venice airport to investigate source apportionment of PM measurements. CPC/SMPS and APS analysers were deployed for 6 weeks at the southern edge of the airfield and the data analysed using k-means clustering and positive matrix factorisation techniques (PMF). Measurement of UFP follow profiles seen at other airports, mirroring aircraft movements and finest particles association. But simple polar plots were not able to isolate airport contribution from other sources. 5 cluster components were used for k-means clustering analysis, 6 factors for PMF. These techniques were able to separate the many different contributions at this complex location and identify the airport and aircraft components of the measurement data, yielding results that bear good comparison to other studies where measurements are dominated by airport contribution.

Hudda et al (2018) (45) undertook a study at multiple (consecutive) locations in the vicinity of Boston airport. The study looked at particle numbers, both inside residences and ambient air to examine the effect of emissions from the airport and aircraft on the local communities. The study was augmented with a test of the effectiveness of HEPA filtration to reduce indoor particle numbers. All unfiltered measurements showed a reasonable correlation of elevated PN concentrations associated with winds from the airport, with indoor air measurements only slightly lower than the ambient data. HEPA filtration of indoor air was seen to reduce PN concentrations by over 65% compared to

unfiltered air. Temporal variation of the PN concentrations correlated with hours of peak airport activity. The survey was not specifically targeted at assessing airport exposure. Disaggregation of other contributions e.g. heating, cooking (though smoking was specifically eliminated), was not considered in this study and the lack of concurrent measurement data meant that reliable assessment of the relative contributions from highways and the airport was not performed.

Ren et al (2018) (46) undertook a study to investigate the effect of emissions at Tianjin airport inside the terminal building, to assess exposure of passengers. Measurement of PN and $PM_{2.5}$ was undertaken at 20 different sampling points, sequentially, and compared to measurements at a fixed outside location. Measurements of PN were made at a single location in the terminal. The study found that indoor PN concentrations were 85-90% of those measured outside, suggesting that the air exchange had little effect in removing UFP. Comparison of the particle size distribution with a "normal" city background PSD showed that PN concentrations inside the terminal were both higher and showed a peak at 20-30nm, not seen at the background location. The report concludes that passengers and employees at the airport are exposed to significantly higher PN concentrations compared to background environments and that the air handling systems at the terminal do not adequately remove particles from the air delivered to the terminal building.

Hu et al (2009) (47) undertook a short measurement survey at Santa Monica airport, using mobile measurements over four days. The study looked at the impact of departing aircraft downwind of the airport on the local communities, at distances of up to 900m from the airport perimeter. The results confirm observations found in other studies: UFP emissions from aircraft have a smaller median size than emissions from road vehicles. The study found that airport emissions can be measured ~700m directly downwind of the airport and 250m perpendicular. The study explored the possibility to identify individual aircraft plumes. There was some evidence of correlation, especially for larger, powerful aircraft, but not every departing aircraft was detected. From the data presented, UFP and BC showed good correlation for plumes associated with departing large aircraft.

Airports Council International Europe (ACI) (2012) (48) has published a position document, setting out the understanding and relevant measurements undertaken at the time of the report. The report was produced as a direct response to studies and conclusions from monitoring at Copenhagen

airport in 2010. At the time, the report concluded that the available evidence suggested that UFP number measurements were higher at airports compared to other activities away from airports, and supported the need for further long term studies. The report summarised a range of measurement studies undertaken at Zurich (by the Swiss Federal Office for Civil Aviation (FOCA) in 2008), Rome (Carvallo et al, 2006, reported above), Copenhagen (Ellermann et al, 2010 and 2012, reported earlier), Zurich Ground Power Units (GPU) exhausts, Heathrow (2012), Stockholm (2012), LAX (Reported above), Santa Monica (reported above). The ACI report identifies that particle size distribution of UFP differs by source: it confirms observations from other studies that jet exhaust particles are smaller than combustion of diesel or petrol. The report opens discussion about the need for regulation, setting of exposure limits and standardisation of measurement techniques, in both occupational and ambient exposure contexts.

Airports Council International Europe (ACI) (2018) (49) has prepared an updated document on ultrafine particles, building on the 2012 document with evidence gathered in the following 6 year period. The report provides excellent background information about formation and characteristics of UFP, emissions and impacts from the aviation industry, and results from measurement studies undertaken at airports since 2012. The report summarises work undertaken at Copenhagen (health impact and additional monitoring), Schiphol (reported above), Heathrow (not yet peer reviewed or published elsewhere), Brussels (reported above), Zurich (reported above), Frankfurt (not yet published elsewhere) and Berlin (not yet published elsewhere). Measurements from Berlin made measurements with a CPC to measure total particle number within the airport complex, while Frankfurt used CPCs at two locations beyond the airport perimeter to measure total particle number. Both these new and updated measurements (reported above) reinforce the observations that the size of particles emitted from aircraft are much smaller than typical road vehicles and emitted in much larger numbers. Investigation of the data with polar plots confirms that the finest particles are associated with wind directions from airports, and that these may travel significant distances downwind and still be observed. Initial health studies from Copenhagen suggest that the risk of exposure to UFP at the airport is no worse than the risk of exposure in a “normal” environment, but accepts that more research is required.

Summary of ambient measurements around airports:

There have been a number of studies of ambient air measurements, increasingly in the last few years, that attempt to identify the magnitude of the UFP contribution of aircraft close to airports. Much of the research has been conducted using mobile systems, either driving equipment to different fixed locations or making measurements whilst following defined routes downwind of the airport sources. All of the studies have measured both volatile and non volatile UFP components; no conditioning of the sample was undertaken for any of the studies reported here.

The literature agrees that:

- Particle numbers close to airports are significantly higher than locations distant and upwind from airports
- Aircraft emissions are dominated by extremely fine particles, 10-20nm in size. This contrasts with locations away from airports, where particles are typically significantly larger.
- The airport related PSD profile is different to traditional road traffic, which has peak PSD from 30-50nm. PSD profiles at background locations peak at 60-100nm.
- Aircraft emissions can readily be identified in polar plot analyses, by virtue of the much smaller particles emitted at airports. This appears to hold true even at considerable distance from the source.

The surveys identify a number of challenges:

- Identification of individual aircraft plumes, while possible, is not 100% successful
- Assessment of PM speciation shows that airport related emissions are very similar in composition to normal urban emissions.
- The measurements and conclusions drawn in individual studies may not be directly intercomparable. Distinct differences between operating and sampling methodologies, instrumentation, configuration will all play a critical role in the value of comparison.

- Quality Assurance and Quality Control is often not well described in any of the literature. The robustness of the measurements, especially when considering the use of multiple devices or mobile measurements staggered over many days, is critical to deriving any reliable conclusions.

- Some conclusions are drawn from research that occasionally represents a very small sample size, collected over a few days or even hours.

- While many of the studies have measured pollutants other than UFP, relationships between UFP and these pollutants are not extensively discussed.

2.5 (3 -) Considering research undertaken to date and direction for future investigations

The review of published papers and literature described above represents the majority of relevant work over the past 20 years. It covers studies that measure directly at the exhaust of jet engines, examination of how the exhaust behaves as it travels away from the source, and finally the impact of the aircraft exhaust on the communities around airports.

This review has focussed on published papers on measurement of UFP from aircraft which, despite more than 20 years of active research, is still a field lacking in some aspects of understanding of the measurements and impacts of emissions from commercial jet aircraft. A general summary of the key findings is presented below:

- Particle number concentrations can be significantly higher immediately downwind of airport locations than at typical urban traffic and background locations.

- Jet engines are a significant source of UFP. The particle size distribution of ambient air near airports appears to be different to typical urban environments – Airport UFP numbers seem to be concentrated in particles smaller than 20nm, in contrast to UFP in ambient air near to road traffic vehicles, which seems to be dominated by larger particles ranging from 30-50nm. The observation of large numbers of the smallest particles at airports compared to

non-airport environments is repeated through the studies and could be considered as a valuable marker for the presence of an airport.

- The vast majority of non-volatile particles emitted from jet engines are carbonaceous.
- The smallest non-volatile particles are associated with emissions from lower thrust settings, though the precise nature of the size and composition varies across engine types.
- The vast majority of the near- or in-exhaust studies have considered just the non-volatile component of the PM emission.
- Measurement of UFP at larger distances downwind is strongly influenced by physical and chemical processes acting on the plume. Cooling, mixing, meteorology, condensation, evaporation and reaction will all play a major role in what is measured as distance from the plume source increases. Most of the "ambient" measurement studies measure both volatile and non-volatile particles combined.
- UFP, both in terms of the finest particle sizes and increase particle number, can be detected as originating from the direction of airports and aircraft many kilometres from the airport. There is evidence to suggest that emissions from landing aircraft can also impact at ground level as a result of downward vortices from the wings on approach.
- Highest concentrations of UFP are observed in airside locations downwind of aircraft activities, presenting potential risk to the work force in these areas.
- Measurements are highly dependent on temporal and spatial variability. It has been possible, under favourable measurement conditions, to collect data from individual aircraft passing a measurement station.
- Limited assessment of data collected to date in this review suggests that there are no obvious unique links between concentrations of black carbon or PM and UFP particle numbers at airports. It is therefore unlikely that measurements of BC or PM alone can be

used to uniquely identify aircraft activity over typical urban emissions. At this stage UFP, and specifically particles smaller than 20nm appear to be unique in originating solely from the airport environment.

There are still however, significant challenges to our understanding of UFP emissions from aircraft. These are summarised below:

- The size, composition and transformational behaviour of particles as they are transported downwind of the exhaust needs further study. There is a complex and not fully understood mechanism around formation of carbonaceous particles, volatile and semi-volatile components, water vapour and gases in the exhaust plume as it ages that merits further detailed investigation. There is already some work that investigates the behaviour and composition of exhaust plumes from road traffic as they emerge, cool, mix and interact with ambient air; e.g. Dall'Osto et al (2011) (50), Harrison et al (2015) (51) for road traffic, and also Beyersdorf et al (2014) (26) in the study of alternative jet fuels. But there would be enormous benefit from improved understanding of UFP formation and extinction mechanisms.

- The role of sulphur in jet fuel appears to be critical to UFP formation from aircraft. Jet fuel typically contains between 600-900 ppm S by volume, compared to less than 10ppm for road transport fuels sold in Europe. Development of lower sulphur aviation fuels could drastically reduce the amount of UFP emitted from jet aircraft, but much more research about the total combined effect of these changes in formulation and any associated engineering requirements would need to be fully explored.

- Characterisation of jet exhaust to account for other emissions is not well documented in the literature. For example, how the relationships between UFP, EC, OC, BC, NOx, SOx, VOC, CO and CO_2 vary between engine types and differing thrust settings.

- The impact of individual aircraft on the surrounding neighbourhoods is not well understood. For example, do heavier aircraft have a larger impact on local air quality than smaller

aircraft? Is there a difference in measured emissions when taking off or landing? Is it possible to operate measurement equipment at a fast enough sampling time to be able to identify and characterise individual aircraft plumes?

- Almost all of the studies published to date use different methodologies to measure UFP, making detailed comparison between them extremely difficult. Even where nominally identical techniques are used, for example Condensation Particle Counters, the size ranges reported in studies are often different, making comparison of total particle counts largely meaningless.

- The quality of the data collected from a number of surveys is at best, undocumented. Some publications clearly describe simple protocols to characterise instruments, or intercompare them where multiple devices are used simultaneously, for example as part of an occupational exposure assessment, but others have no indication of any QA/QC measures used to ensure collection of robust data. Some of the surveys use factory calibrations of analysers that are 2 years old, with no indication of the analyser's history of operation, calibration or maintenance during this time.

- The data collected to date is generally not of a high enough quality, or with sufficient commonality between studies to be used meaningfully in any health impact assessments.

- Many studies draw conclusions from very short term monitoring programmes; often just a few days, during daylight hours. There is considerable risk that these conclusions may not present a true reflection of the intended campaign objectives.

- Emission factors for PN concentrations have been derived from the studies reported above for a limited range of individual engine types and thrust settings. These are obtained under necessarily controlled conditions and derived from a very limited dataset. It is likely that these factors may not accurately reflect either how the engines are used in normal operation,

or actual measured concentrations compared to modelled data using these factors. This is an area where more research and data would help to refine the factors and improve models.

For future research studies, there are a number of areas where improvements could be implemented to enhance data quality:

- Standardisation of measurement methods. A standard method exists for counting particles and a technical specification is in development for particle sizing.
- Robust comparisons between standard methods and alternative techniques.
- Presentation of data that proves comparability between devices used for concurrent monitoring studies, e.g. in personal exposure or mobile surveys.
- Detail of all calibration work related to the studies, ideally before, during and after measurements are collected, together with assessment and quantification of losses to the sampling systems.

Demonstration that monitoring follows documented, traceable procedures is essential if the research is to be robustly inter-compared or used to assess health impact. It is likely that the research documented above has had a reasonable level of QC applied to the measurements, but this work is largely undocumented and should not be assumed.

A large body of ongoing work has been undertaken to harmonise and standardise measurements of UFP, at least for statutory monitoring purposes. The European Committee for Standardisation (CEN) and the International Standards Organisation (ISO) have an agreed technical specification for particle counting (CEN TS 16976:2016), based on butanol based particle counting, and are currently developing a technical specification for particle sizing, based on the scanning mobility principle. Similarly, the EU has led a number of key projects: the EUSAAR project (European Supersites for Atmospheric Aerosol Research), which ran between 2006 and 2011 http://www.eusaar.net/ It established a network of high quality measurement stations throughout Europe, which could be

accessed by research communities for data and training purposes. EUSAAR was developed into the ACTRIS project (Aerosols, Clouds, and Trace gases Research Infra Structure), which continues this philosophy and includes contribution from The World Meteorological Organisation - Global Atmospheric Watch (WMO-GAW). ACTRIS play an active role in the development of the technical specifications being developed by CEN.

Wiedensohler et al have been instrumental in developing procedures, harmonisation techniques and development of the technical specifications described above. Wiedensohler et al (2012) (52) presents comprehensive information about the constituent parts of a CPC / SMPS instrumentation and then documents how to process the outputs and account for factors that require correction, such as humidity, temperature, pressure, diffusive losses, etc. It further reports on extensive comparison campaigns from a range of participants who followed these recommended procedures throughout the campaigns. The testing revealed that, most applicably to the airport / aircraft environment, harmonisation of measurements for both sizing and particle counts was excellent across all participants. This information has been used to arrive at a series of recommendations to feed into the development of technical specifications for operation of particle measurement devices. Interestingly, one of the recommendations is for research papers to provide access to the raw count data, without any processing post collection. It would be extremely useful to be able to compare raw outputs from measurement campaigns.

Wiedensohler et al (2018) (53) revisited this theme and proposed a calibration hierarchy for the performance assessment of field instruments. If the recommendations from Wiedensohler et al from 2012 and 2018 are rigorously followed, the data from research studies will be robust, harmonised and comparable.

UK Air Quality Expert Group (2018) (AQEG) (54) published a report, describing the known sources, measurement techniques, ambient concentrations and composition, modelling studies and available control mechanisms from studies published up to the end of 2017. It concludes that, in particular, emissions of UFP from airports is poorly understood, requiring further investigation. It additionally recommends an expansion of permanent ambient measurement stations across the UK; as noted earlier in this review paper, there are three permanent SMPS instruments deployed in the UK.

It is very important to note that there is no mandatory requirement to follow any of the recommendations from EUSAAR/ACTRIS, WHO-GAW, Wiedensohler et al or AQEG. This currently makes comparison of individual research campaigns very difficult. For example, none of the papers reviewed in this document cite Wiedensohler et al (2012) (52) in their literature, placing further uncertainty on the data quality. Future research campaigns should at least document whether the philosophies described in the Wiedensohler et al (2012) (52) paper have been considered; at present, it can only be assumed that this is not the case.

There are a large number of future research exploration studies that can be considered, including:

- Further characterisation of ambient PSD from landing and departing aircraft.
- Further detailed chemical and physical speciation studies of jet exhaust.
- Further correlation studies to assess relationship of UFP with other pollutants.
- Exploration of UFP measurements from different aircraft / engines / departure / arrival.
- Further detailed assessment of how PSD changes with increasing distance from source.
- Correlation studies between UFP, PSD and health.

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2.7 Appendix update

A number of amendments have been made to the published paper. These are deemed to be minor in nature and do not require the original papers to be updated. The changes are as follows:

Section	Amendment
2.3	Durdina et al (2014) – provided clarification and description of effective particle density
2.3	Abegglen et al (2015) – deleted speculation about fractal particle growth
2.3	Abegglen et al (2016) – added clarification for EC/TC and particle density
2.4	ACI (2012) – Minor clarification of text

2.8 Literature update

Since publication of this paper in 2019, the amount of UFP monitoring studies around airports has increased dramatically. In addition to the monitoring undertaken as the three Chapters presented in this thesis, the following key studies have added to our understanding of UFP around airports:

- Fushimi et al (2019) (a) identified significant quantities of lubrication oil present as UFP in jet exhaust, confirmed later by Ungeheuer et al (2021) (j) using detailed speciation studies of UFP using filters from a cascade impactor.
- Bendsten et al (2019) (b) identified that the toxicity of jet exhaust particles was comparable to diesel exhaust particles in a study of exposure in mice. Similar findings and increased risk factors were observed in human studies by He at al (2020) (h), Moller et al (2020) (i), Selley et al (2021) (k), and Wu et al (2021) (o) while no significant observable effects on humans were noted by Andersen et al (2021) (m).
- Jones et al (2012) (c) identified that a dramatic change in urban UFP concentrations in London coincided with a reduction in the concentrations of sulphur in diesel and petrol fuels. While this research was undertaken prior to 2019, it was not considered in the original, airport specific literature review. However, the findings of Jones et al around the measured impact of removing sulphur from road transport fuels and the similarity of historic data to the current situation around airports adds to the evidence that sulphur in aviation fuel is a likely significant contributor to measured UFP at airports.

- Pirhadi et al (2020) (d) quantified the contribution (using Positive Matrix Factorization) of airport and aircraft activities on PNC measurements around Schiphol Airport, finding that departures contribute more than arrivals, in turn more than ground operations. Differences in mode particle size for each activity were also determined in this analysis.
- Yu et al (2019) (e) assessed the impact of UFP and PM at a number of locations around Los Angeles, finding that Airport activities contributed up to 6% of $PM_{0.1}$ and up to 10% of the UFP measurements.
- Hudda et al (2020) (f) undertook a measurement campaign at a residential property underneath the flight path at Boston airport to examine mitigation strategies for reducing exposure to aircraft UFP indoors. It found that (at least for the type of house tested) that UFP concentrations inside the building were still at least 75% of those measured outdoors, indicating a significant risk when winds were from the airport or aircraft operation trajectories.
- Lammers et al (2020) (g) found that healthy individuals exposed to aircraft UFP experienced reduced lung function after exposure.
- Jasinski et al (2021) (l) explored the effect on UFP emissions from turbofan engines of blending alternative fuels with Jet-A1, confirming that significant particle number reductions were seen with blended fuels.
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3. Methodologies

3.1 Introduction

This chapter presents, in detail, the physical setup, configuration and processing of the UFP instrumentation used for the 2016, 2017 and 2019 monitoring campaigns described in Chapters 4, 5 and 6. This will include a discussion about the differences in the data collected, arising from the measurement systems and configurations used in the different years. This will in part duplicate, but also supplement the methodology discussions in the published literature.

3.2 Monitoring station inlet designs

The sampling system is probably the most critical part of the entire measurement chain. The smallest of changes, for example not using grounded and electrically conductive inlet tubing, could result in significant sample losses and invalidate any attempt to compare measurements at different locations. For the most part, the configuration used at the Heathrow monitoring stations mirrored the setups at the national monitoring stations:

- Stainless steel and conductive tubing used throughout
- $PM_{2.5}$ sample inlet, at a height of 0.5 m to 0.75 m above the roof of the monitoring enclosure
- Sample maintained at ambient temperature, no heating
- Total sample inlet length of 2 m to the classifier inlet
- Smallest possible length of connecting tubing between classifier and counter
- Diffusive loss and multiple charge correction enabled
- All flow rates, impactors, scan times, size ranges and other configurations identical
- Air conditioned monitoring enclosure, regulated to ~22 ºC

There were a few differences:

- No dryer in either of the Heathrow configurations. It is likely that the lack of a dryer will result in both losses and formation of new particles. It was considered that these mechanisms would at least partially offset, and this is explored in more detail in Chapter 4.
- Setup at LHR2 used a more modern instrumentation set up

For the 2017 study, only one site was operated at the airport, with a significantly different set of flow rates, size range, impactor nozzle diameter and scan times. These changes to operating parameters had a significant effect on the results, noted and discussed in the published papers.

For the 2019 study, a completely different measurement technique was used. This allowed for the collection of high resolution, full size range scans, which meant that genuine identification of individual aircraft plumes could be identified and classified. The sampling setup was basically identical to the system used for the previous studies: 2m of conductive tubing, $PM_{2.5}$ sample inlet, ambient temperature sampling, no dryer.

3.3 Instrumentation used

3.3.1 Condensation Particle Counter (CPC)

For the 2016 and 2017 surveys, Butanol based TSI Model 3775 and 3776 CPCs were deployed to count particle numbers. The CPCs count the number of particles in air, increasing the apparent size of the smallest, normally undetectable, particles by condensing butanol vapour onto them before measurement. A basic schematic of the sample and measurement principle is shown below:

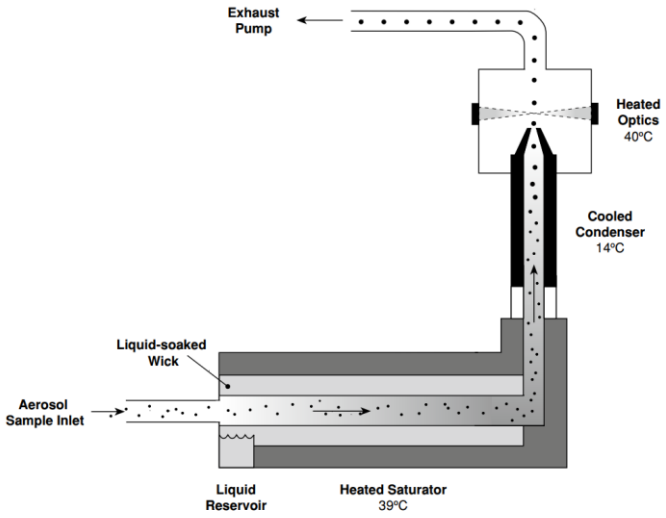

Figure 1 – CPC schematic, reproduced from TSI CPC operator manual

The only significant difference in the operation of the 3775 and 3776 CPCs is in their capability to detect the smallest particles. The 3775 CPC, used in 2016, is able to detect 7 nm particles at 50% efficiency (d_{50}), while the 3776 has a d_{50} of 2.5 nm. In practice, this will make no difference to the

survey data quality – the size ranges of particles measured were significantly higher than the minimum size capability of the CPC used in both surveys.

3.3.2 Scanning Mobility Particle Sizer (SMPS) / Differential Mobility Analyser (DMA)

For 2016, a At Oaks Road, the CPC was connected to a TSI Model 3080 with long DMA (Model 3081) classifier was deployed at Oaks Road, while a TSI Model 3082 with long DMA (Model 3081) classifier was deployed at LHR2. Both systems used a soft X-ray neutraliser. Automatic on-board software correction was enabled for diffusive losses and multiple charge. The Oaks Road 3080 used AIM v9.0.0.0 controlling software, the LHR2 3082 used AIM v10.1.0.6.

A basic schematic of the SMPS operation is presented below:

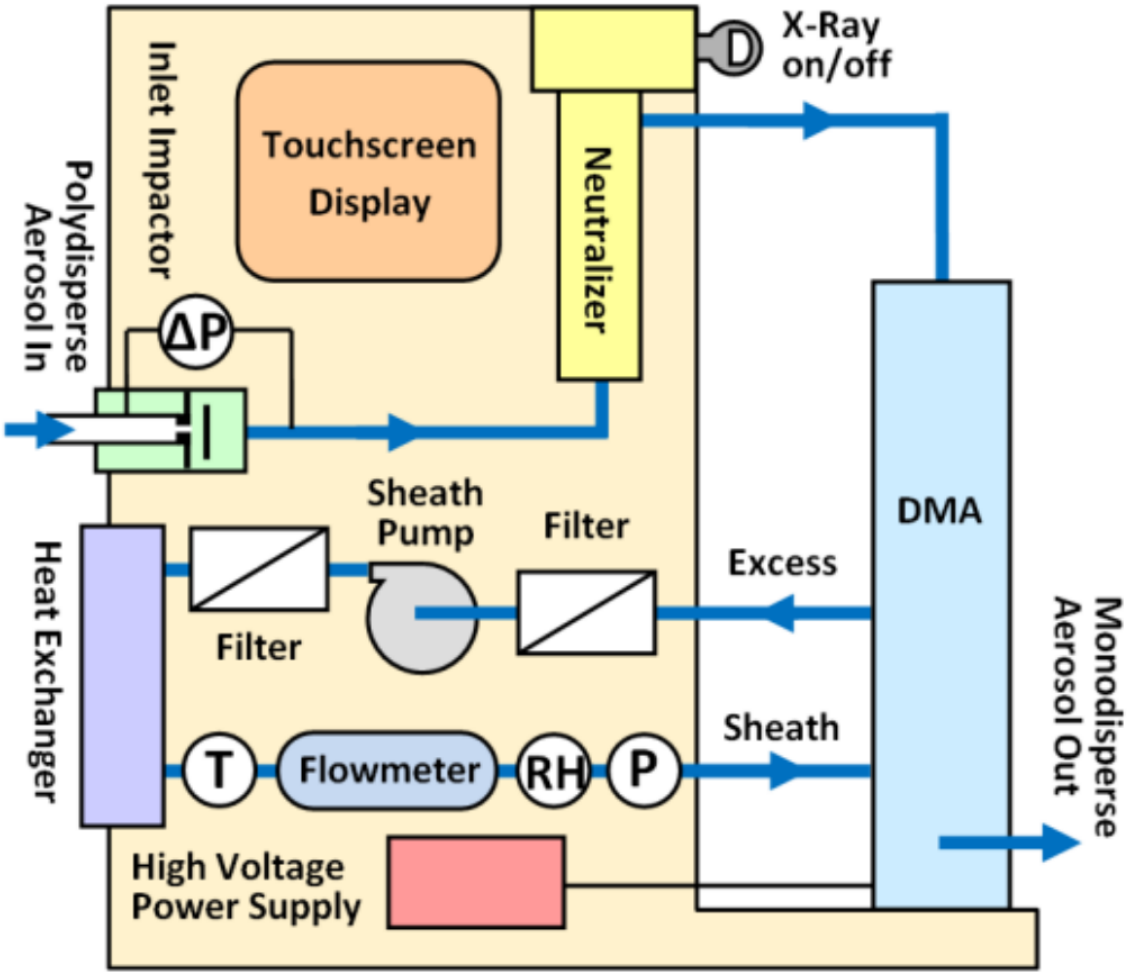

Figure 2 – Model 3082 schematic, reproduced from TSI operator manual

Particles enter the analyser and any charged particles are immediately neutralised, in this case, by using an X-Ray source. Other versions of the analyser use a beta radiation source, but airport

regulations prohibited the use of a radioactive source. Once the particles are neutralised, they are sized in the DMA classifier. The classifier makes use of a gradually varying potential difference to selectively size emerging particles for measurement by the CPC. A schematic of the DMA is presented below:

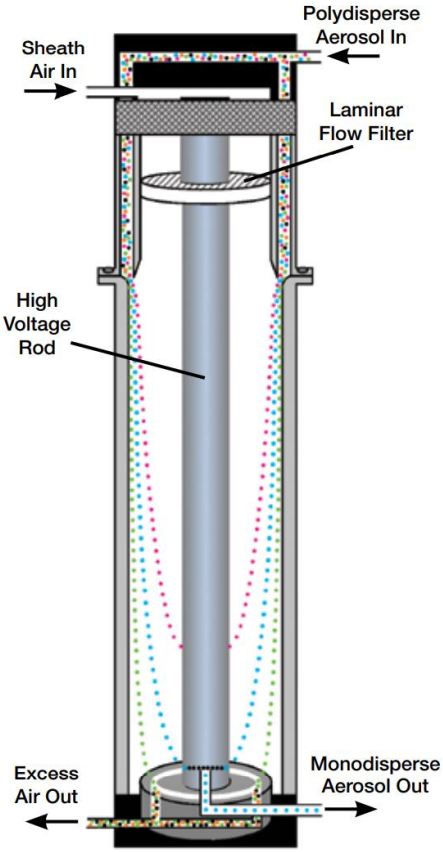

Figure 3 – Model 3081 DMA schematic, reproduced from TSI operator manual

The SMPS software is configurable to compensate for the behaviour of particles in the analyser. The two most important configurations are:

- **Diffusion correction**. Enabling this function allows the software to estimate the losses of particles of different sizes to the walls of the sampling system. As discussed earlier, smaller particles are susceptible to diffusive losses, and especially so at lower flow rates.
- **Multiple charge correction.** Enabling this function allows the software to estimate the number of particles that may carry multiple charges. It is possible for larger particles to be more charged than smaller particles, which would allow them to behave more like small

particles in the charged environment of the DMA. It is undesirable for these particles to be included in the counts for smaller particle sizes.

3.3.3 SMPS configuration differences

The exact configuration of the SMPS has a marked effect on the performance of the DMA. For the 2016 survey, the size range, flow rates and scan times were all configured to be identical to the setup in the national monitoring network, to allow direct robust comparisons between datasets.

The software versions used were specific to the instrument used. There is some evidence that shows that the AIM10 particle calculation reports significantly lower concentrations for <50 nm particles compared to AIM9. However, conversations with TSI were clear that the two software versions were created for their specific SMPS systems (AIM9 for Model 3080, AIM10 for Model 3082) and should not be interchanged. Checking the timeseries data, basic statistics and comparison with the national network datasets (all using AIM9) did not reveal any obvious underread of the LHR2 data. Additionally, a check of the 2016 data was undertaken for the Oaks Road and LHR2 datasets, processing both datasets using both AIM9 and AIM10; this also showed no significant differences in the total particle count suggesting, at least for the UFP measurements at Heathrow, that the two different software versions had minimal impact on the final datasets.

For the 2017 study, the 3082/3081 SMPS was configured to report faster scans and a narrower size range. This meant that the flow rates through the DMA were 5 times higher than the flow rates used in 2016. The table below shows how this difference affects particle diffusion in the DMA:

Particle size, nm	Thermal velocity, cm/s	Diffusion distance (cm) @ 300 cc/min (2016 data)	Diffusion distance (cm) @ 1500 cc/min (2017 data)
10	350.7	162.3611	32.1154
20	124	57.4074	11.3553
30	67.5	31.2500	6.1813
50	31.4	14.5370	2.8755
80	15.5	7.1759	1.4194
100	11.1	5.1389	1.0165
150	6	2.7778	0.5495
200	3.9	1.8056	0.3571
250	2.8	1.2963	0.2564
300	2.1	0.9722	0.1923
400	1.4	0.6481	0.1282
500	1	0.4630	0.0916
600	0.8	0.3704	0.0733
700	0.6	0.2778	0.0549

Table 1 – Dependence of particle diffusion on particle size and flow rate

The table clearly shows that the smallest particles can diffuse much larger distances at low flow rates compared to in higher flow rates and larger particles. Specifically, for the studies reported in the thesis, this goes some way to explain why higher particle concentrations were reported in 2017 compared to the 2016. This further emphasises the importance of knowing and reporting the exact configuration details of the instruments used to measure and size UFP.

3.3.4 Cambustion DMS500

For the 2019 study, a completely different analyser was used in order to capture measurement data at 1 second intervals, reporting data for the entire size range simultaneously. The Cambustion DMS500 uses a series of differently charged electrometers to calculate particle size and concentration. A schematic of the instrument is provided below:

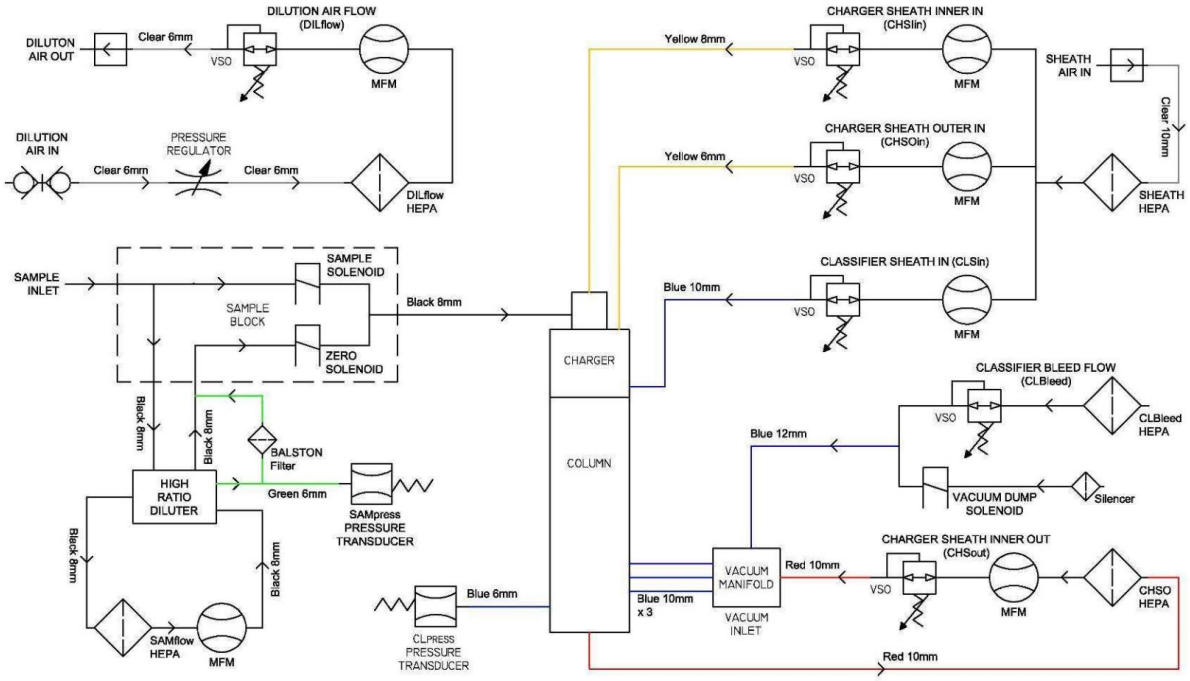

Figure 4 – DMS500 schematic – adapted from Cambustion operator manual

The analyser draws in sample air, optionally heated, optionally diluted, depending upon the nature of the source, before passing it through a unipolar corona charger. The sample then passes through the series of differently charged electrometers, where they are separated and the signals processed to calculate size and concentration values. A schematic of the electrometer assembly is presented below:

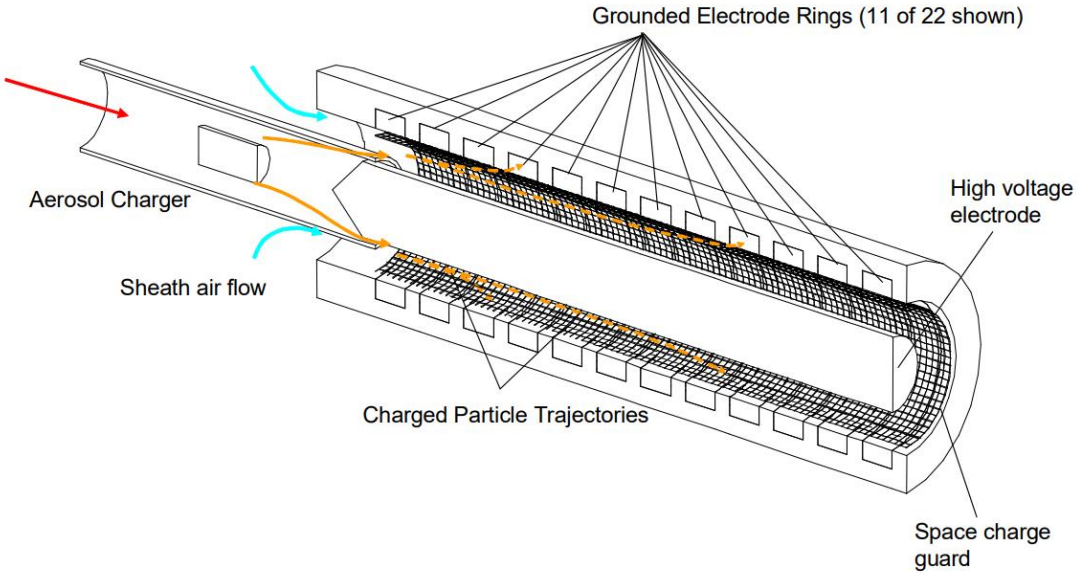

Figure 5 – DMS500 electrometer schematic – adapted from Cambustion operator manual

Correct and documented configuration of the DMS500 is just as critical as seen for the SMPS. The analyser was configured to sample at 8 litres per minute, sampling at ambient temperature with no

dilution. Sampling was set to 1Hz, with a Gasoline Direct Injection engine combustion inversion algorithm for outputting measurement data. These parameters were set up and agreed in advance with Cambustion and the Ricardo automotive group, as the most likely best configuration for the airport environment. The different particle composition and size distribution profiles for aircraft may have an impact on the robustness of the choice of correction algorithm, but the assumptions made represent the best assessment currently available and will not compromise the "between-aircraft-comparability" of the thesis investigation.

3.4 Calibration

A common thread that runs through all three surveys is robust calibration. All analysers were calibrated at the Ricardo Energy and Environment ISO 17025 accredited calibration laboratory before and after each survey.

The calibration was conducted using soot particles generated from a Jing MiniCAST particle generator. These particles were transformed into monodisperse particles using a tunable, calibrated TSI 3081 DMA, before being isokinetically split to feed a calibrated CPC and the candidate SMPS or DMS500 analyser. The soot output was adjustable for size (15 to 300nm) and concentration ($1x10^3$ to $5x10^5$ #/cm^3), to provide information about performance across a wide size and concentration range, certainly within the normal expected range in an ambient environment.

While the calibration gives confidence in the capabilities of the analysers with monodisperse soot particles, it is not possible to quantify the performance in a polydisperse, multiple species, physically and chemically changing environment, where particles will also not be a controlled shape. In addition, the calculation algorithms used to deconvolve raw data signals to produce measurement datasets are not necessarily fully appropriate for the airport environment. These compromises limit comparability between surveys, but the analysis within each discreet survey remain robust.

To date, there is very little published data available that reviews the comparability of data from different analyser types and the use of different deconvolution algorithms. Studies currently underway, for example the AVIATOR H2020 project (1), will address some of these questions, but this is clearly an area where more research would be beneficial.

3.5 Data Ratification

Data processing from conventional analysers (NOx, BC, PM_{10}, $PM_{2.5}$ is reasonably straightforward, and extensively documented in the UK-AIR QA/QC manual (2). For the UFP data, philosophies documented by Wiedensohler et al (3,4) were used throughout, regardless of the measurement technique or configuration used for the campaigns. The philosophies employed are summarised below:

- The analysers were calibrated before the measurement campaign. Where appropriate, the analyser was adjusted to agree with the traceable standard, or the calibration results recorded and earmarked for adjustment of data post-collection
- The analysers were operated as close as possible to national network configuration, both physically for inlets and in configuration, where comparability of datasets was important.
- The analysers were calibrated after the measurement campaigns. In the event of any significant difference between these calibration results, this would have allowed for a ramped adjustment of the measurement datasets, following the UK-AIR principles of data ratification. In all cases, such data processing was not required.
- Raw data was scrutinised and any anomalies removed. Zero tests, maintenance visits, data lower than detection limits and any spurious poor quality data was rejected from the datasets before further analysis was undertaken.

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4. UFP at Heathrow Airport, 2016

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https://doi.org/10.1016/j.atmosenv.2019.117148

4.1 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 from the airport are not strongly correlated with Black Carbon, but there does appear to be some correlation with nucleation mode particles and UV active BC (brown carbon) particles 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.

4.2 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) (22) 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) (11), Fleuti et al. (2017) (12), Hudda and Fruin (2016) (14), Masiol et al. (2017) (17) and Peters et al. (2016) (18), 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) (7) and Health Effects Institute (2013) (13) 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) (10) 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) (8), Lobo et al. (2015) (16), Abegglen et al. (2016) (1), Turgut et al. (2015) (24), and Vander Wal et al. (2016) (25), 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) (3), 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) (9), Fanning et al. (2007) (11), Hudda and Fruin (2016) (14), Westerdahl et al. (2008), Peters et al. (2016) (18), Keuken et al. (2015) (15), Bezemer et al. (2015) (4), Riley et al. (2016) (19), Fleuti et al. (2017) (12), and Shirmohammadi et al. (2017) (21), have all shown that high PN concentrations can be seen close to airports.

Masiol et al. (2017) (17) 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) (14) were able to detect airport related UFP emissions 18 km from Los Angeles International Airport, LAX , while the Keuken et al. (2015) (15) research detected airport related UFP over 40 km from Amsterdam Airport Schiphol .

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

4.3 Methods

4.3.1 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.

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

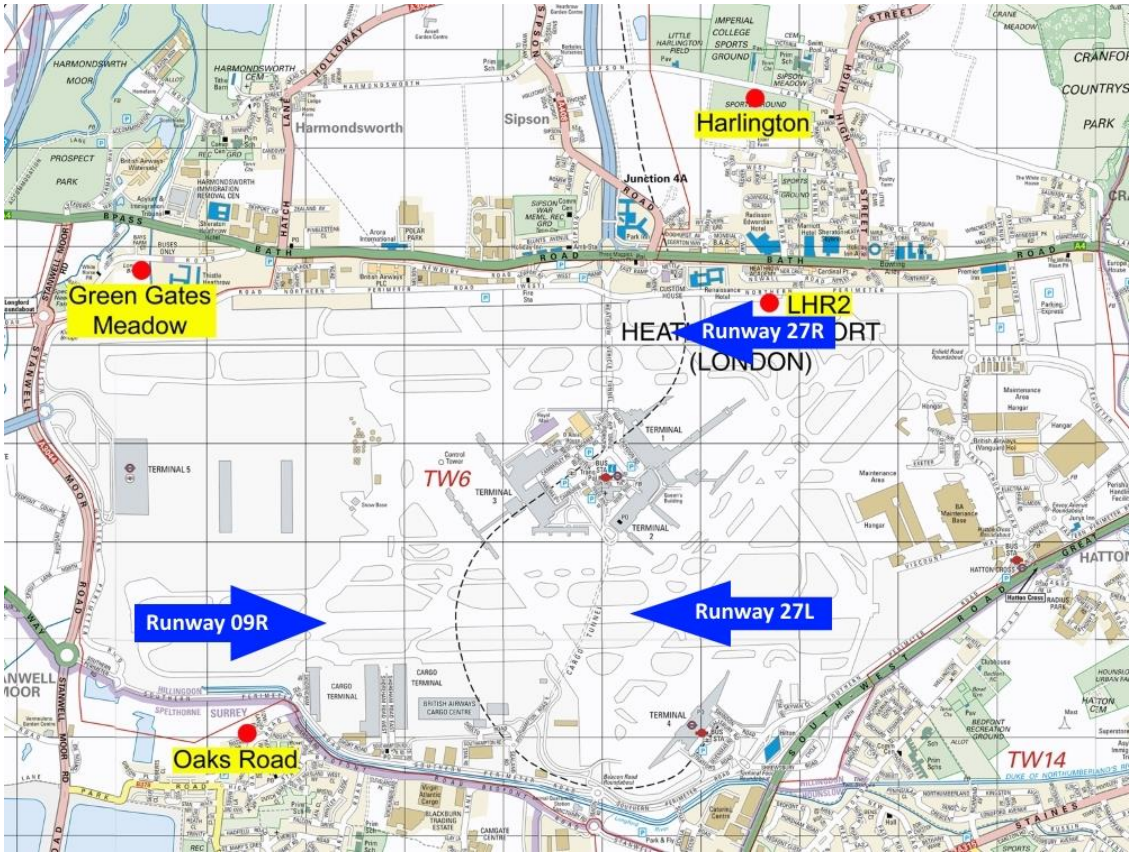

Figure 1. Locations of Heathrow monitoring stations. Runway 27R, Runway 27L and Runway 09R denote the three operating modes of the airport, indicating here the runway assigned for departing aircraft. Note that aircraft never depart in an easterly direction on the northern runway.

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.

4.3.2 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 was controlled via a PC connected to the Model 3082 running AIM v10.1.0.6. Data was downloaded weekly from the PC 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) (29) and Wiedensohler et al. (2018) (30). 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 establish remote communication to the analysers. The monitoring stations were visited weekly to ensure correct operation and take remedial action if required.

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.

4.3.3 Differences between Heathrow and National Monitoring UFP analyser setup

The configuration of the Heathrow analysers matched, as far as possible, the configurations used in the UK Particle Number monitoring network (https://uk-air.defra.gov.uk/interactive-map). This network is managed by Kings' College London, while operation and QA/QC is provided by the 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
- Nafion dryer
- 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. 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 (23)) have shown that relative humidity contributes little to increased particle size even for hygroscopic particles smaller than 50nm.
- The lack of a dryer may also contribute to two further potential scenarios for differences between measurements:

  - Particle losses in the dryer at the national network stations, due to increased surface area and residence time.

  - Formation of new particles within the dryer due to a pressure reduction in the dryer and increased residence time

  It is likely that losses and gains will partially offset each other, but there will be considerable variation, largely dependent upon the particle size, composition and

conditions. At this stage, the potential for differences due to dryer presence or not is noted.

- 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.

4.3.4 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) (5))

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 between 100 and 661nm. Accumulation mode particles can be up to ~2500nm in size, but will be present in very small concentrations compared to the smaller particle sizes.)

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 Delta-C and interpreted it as a measure of wood smoke concentrations (e.g. Wang et al. (2011) (27)):

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

4.3.5 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) (22), 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) (29) and Wiedensohler et al. (2018) (30) emphasise 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.

4.4 Results

4.4.1 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, #/cm^3	7817	1871	15993	42 – 150000	87.6
Aitken, #/cm^3	8638	5542	9704	93 – 107918	87.6
Accumulation, #/cm^3	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, #/cm^3	8476	2152	12064	0 – 86287	50.0
Aitken, #/cm^3	7798	4723	8223	0 – 63372	50.0
Accumulation, #/cm^3	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.

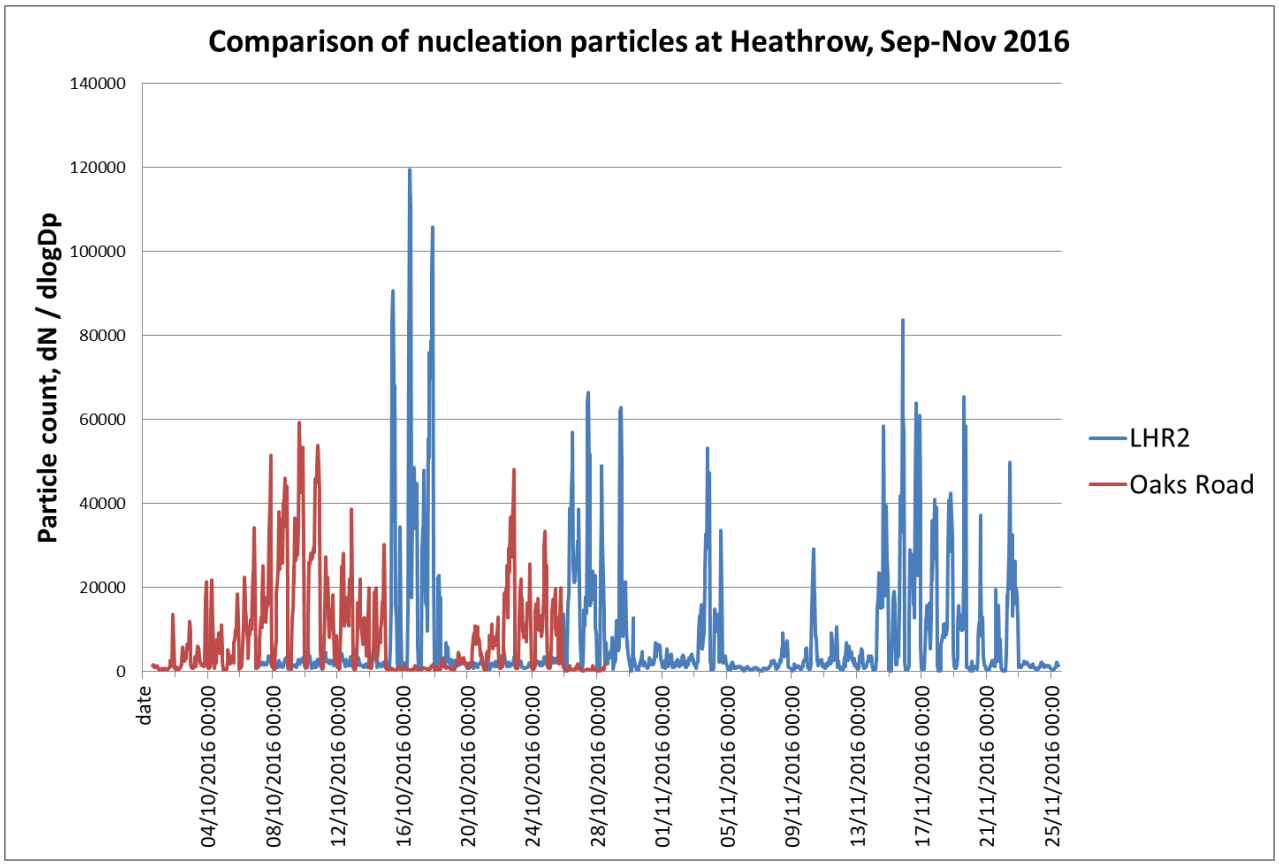

Figure 2. Time series of nucleation mode particles at the Heathrow Airport sampling sites, October and November 2016

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.

Examination of LHR2 and Oaks Road particle number diurnal data averaged for the entire survey (presented in Figure S9 and S10) shows enhanced PN concentrations between 06:00 and 23:00, coinciding with typical increased activity around the airport. The diurnal plots also show close agreement with the two humped diurnal profiles of the NO_x and BC pollutants, suggesting that these pollutants mostly share common sources, including road traffic and commercial / domestic energy use. The PM_{10}, $PM_{2.5}$ 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 appears to follow a similar trend to the

PM_{10} and $PM_{2.5}$ diurnal profiles, but it is relatively flat and significantly lower in number concentrations when compared to the Nucleation and Aitken mode PN datasets. The diurnal plots for Nucleation and Aitken mode particles do not follow the trends for the other pollutants, further confirming that they are not associated with the same sources.

4.4.2 Results in Context with Other Monitoring Data

Measurements of UFP were coincident at LHR2 and Oaks Road for the period 7 – 28th 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 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 protocols for the AE22 aethalometer published by Virkkula et al (2007) (26) and Apte et al (2011) (2) 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.

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.

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 28th October 2016.

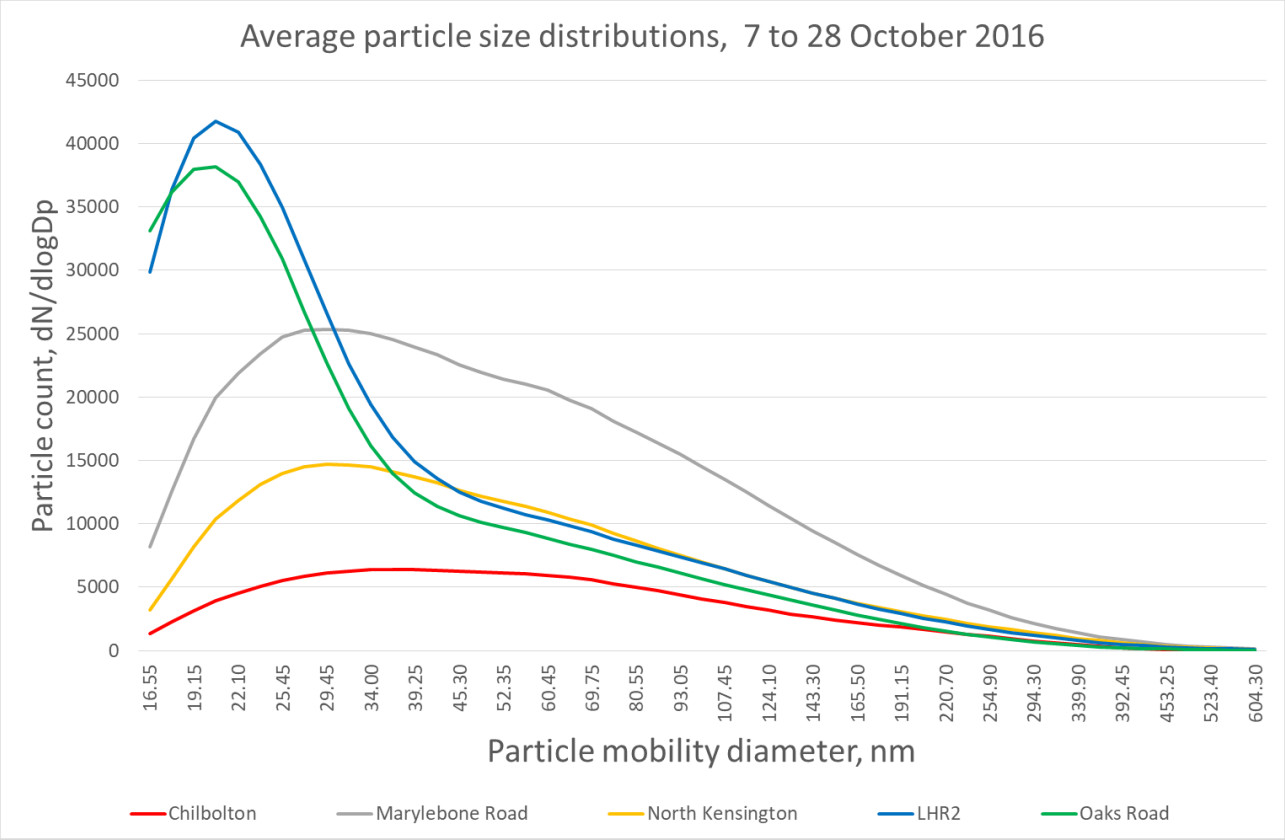

Figure 3. Comparison of particle size distributions at 5 locations

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.

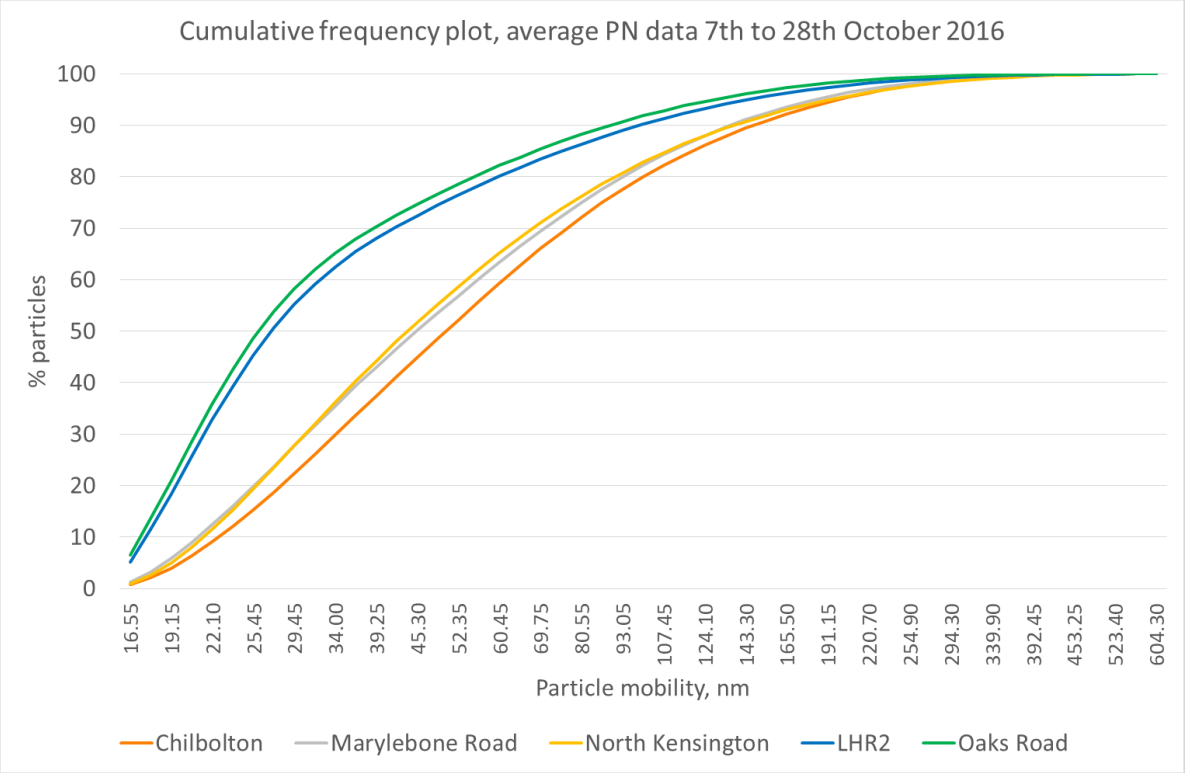

Figure 4. Cumulative particle size distributions at the five sites.

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.

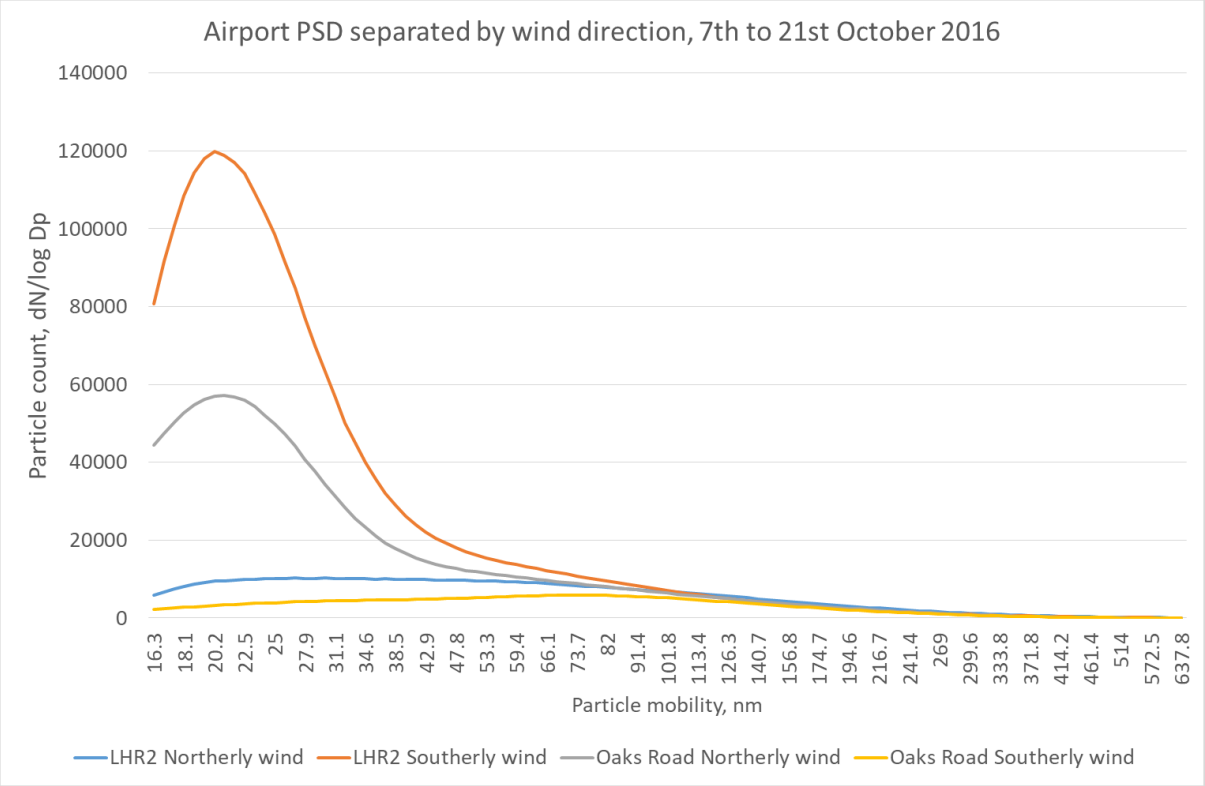

Figure 6. Airport PSD separated by wind direction for the Heathrow Airport sites

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.

4.4.3 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.

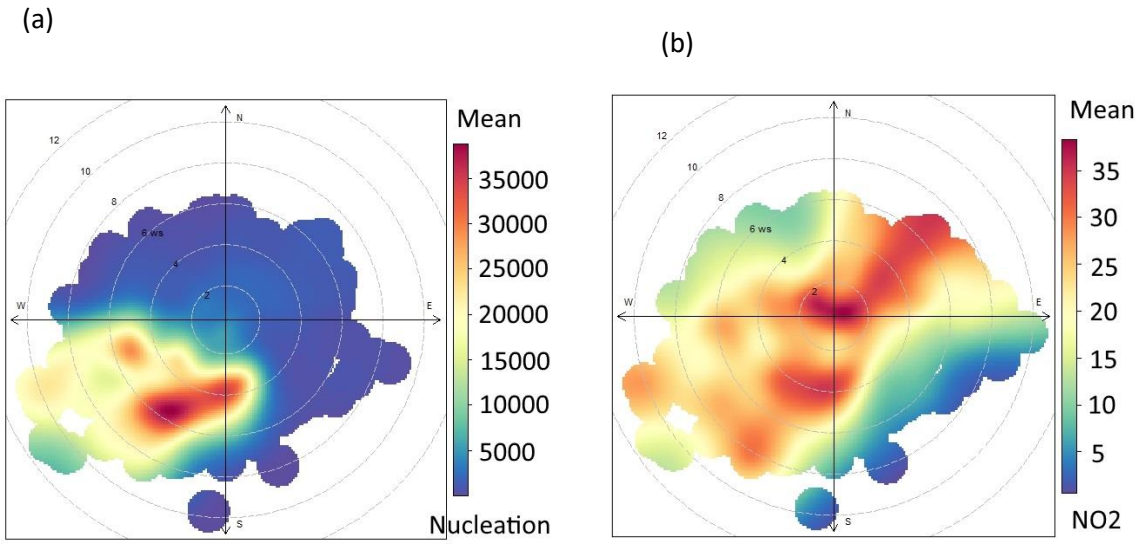

Figure 7. Polar plots for LHR2 data, (a) nucleation mode particles (dN/dlogDp), (b) nitrogen dioxide (ppb)

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.

NO_2 at LHR2 (Figure 7(b)) is strongly associated with south west and north east wind directions, 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).

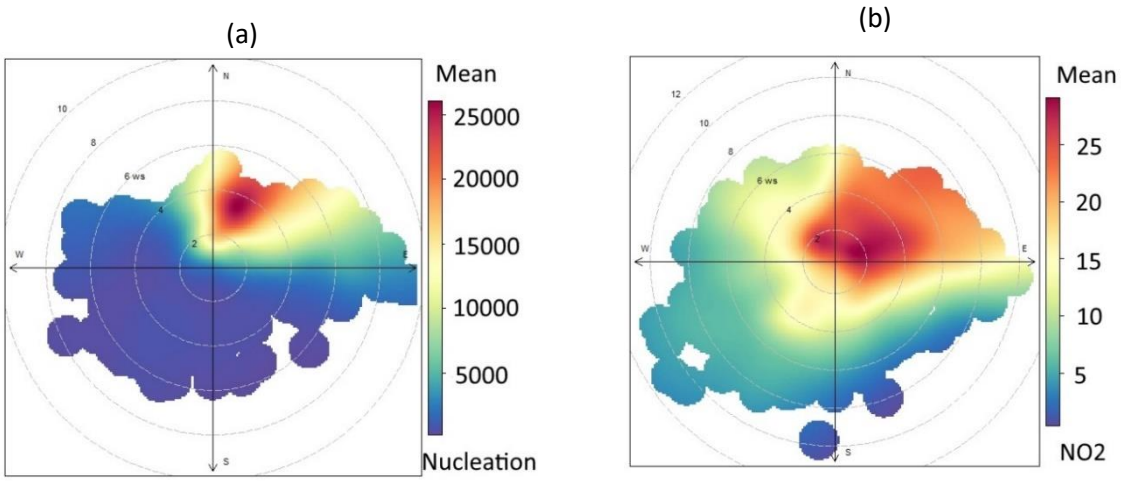

Figure 8. Polar plots for Oaks Road, (a) nucleation mode particles (dN/dlogDp), (b) nitrogen dioxide (ppb)

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 trend to nucleation mode particles, but are associated with a slightly wider range of wind directions than the nucleation mode. It is clear from the two monitoring station datasets that nucleation mode particles are predominantly associated with winds from the airport, suggesting that the airport is by far the major source of emissions of these particles.

In contrast, all other pollutants (Figure S12) are strongly influenced by low wind speeds, indicating local sources, and the background environment as significant contributors. PM_{10} and $PM_{2.5}$ appear to originate largely from the same common sources, and PM mass sources appear to be mostly independent from the other pollutants.

4.4.4 Dependence of Measurements on Airport Operation

The two runway configuration at Heathrow allows the airport to operate in a number of modes. When winds are easterly, aircraft exclusively depart from Runway 09R, the southernmost runway, and generally arrive on 09L, the northerly runway.

When winds are westerly, the airport typically operates a shift-based departure system, departing on one runway for half the day, and the other runway for the remainder. Landing occurs on the other runway during these times.

These operating modes are primarily chosen for practicality. For westerly departures, spreading the distribution of landings and departures on Runways 27R and 27L equalises the wear and tear on the landing zones on each runway, reducing the amount of maintenance required. For easterly departures, the taxiways approaching the thresholds of Runway 09L are not suitable for modern aircraft. This means that departures in this mode are exclusively from Runway 09R.

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.

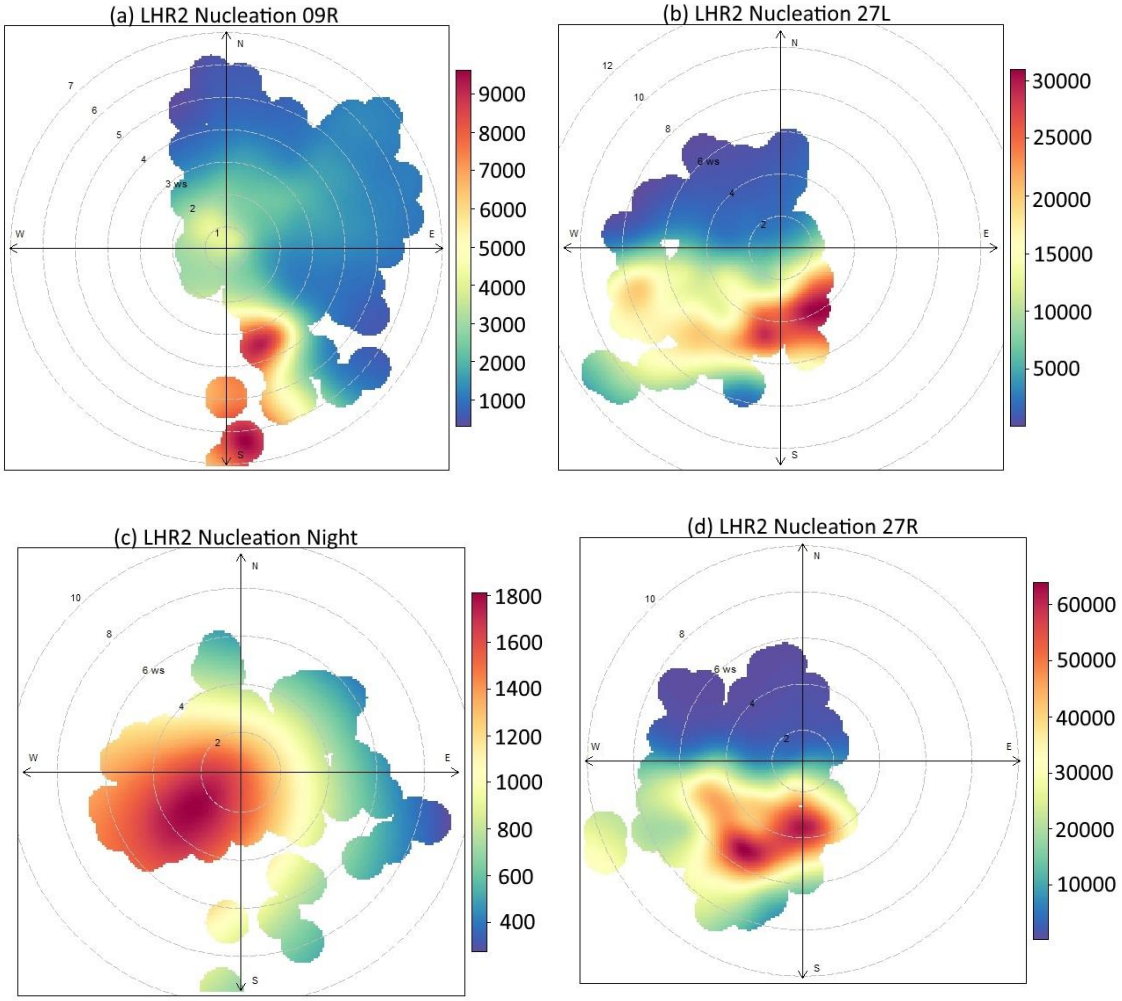

Figure 9. Polar plots of nucleation mode particles at LHR2, split by runway mode, (a) 09R, (b) 27L, (c) Night, (d) 27R. Each sub plot has different maximum concentrations defining the colour scales.

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) (6), 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 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.

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.

Polar annuli for all pollutants at both sites are presented in Figures S16 and S17. These plots further reinforce the directional and diurnal nature of emissions around the airport

4.4.5 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.

On average (https://www.heathrow.com/file_source/Company/Static/PDF/Investorcentre/Heathrow-(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.

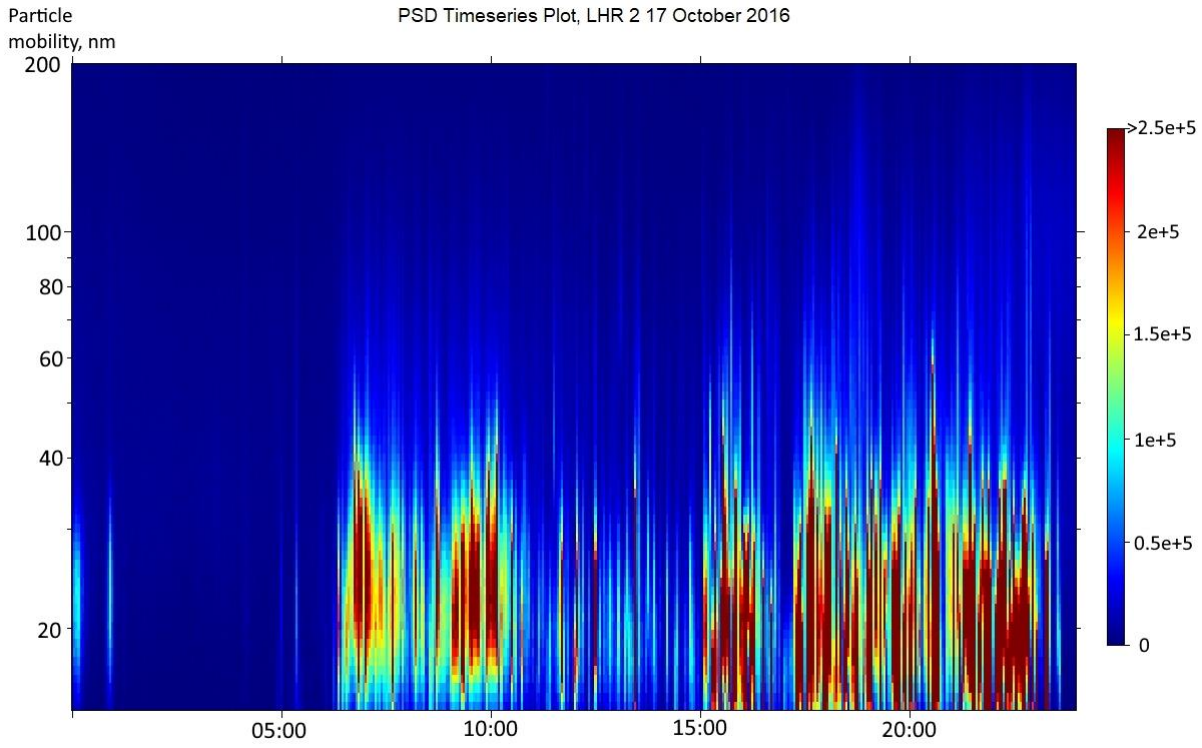

Figure 10. Particle Size Distribution plot, LHR2, 17 October

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.

4.4.6 Comparison with Receptor Modelling Outputs

Masiol et al. (2017) (17) 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) (17) 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) (20), Masiol et al. (2017) (17), 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. Investigation of rotational ambiguity was not considered. The qualitative relative contributions for each factor, and comparison of the polar plots of the airport PMF factor with the measurement data, were the only outputs used from the PMF.

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.

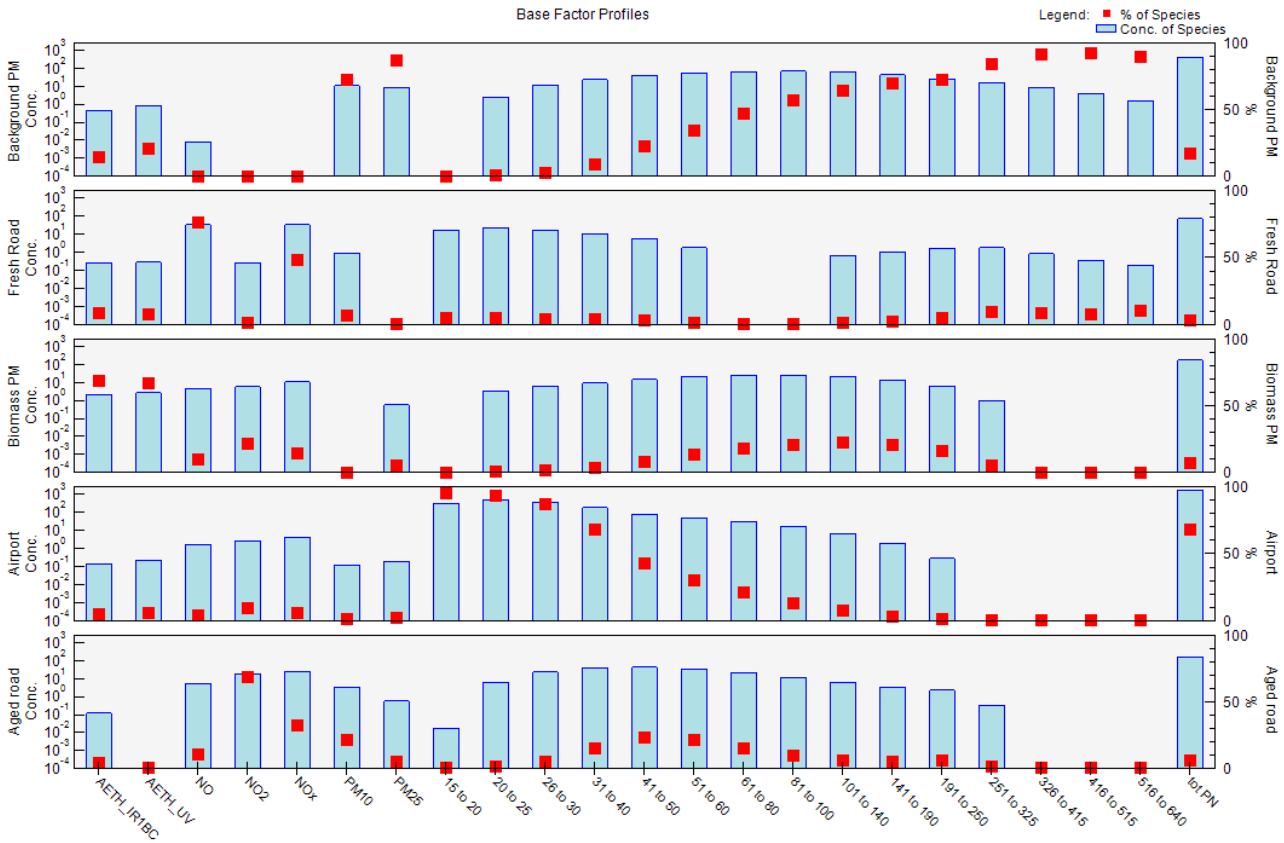

Figure 11. PMF Base Factor Profiles for LHR2 site

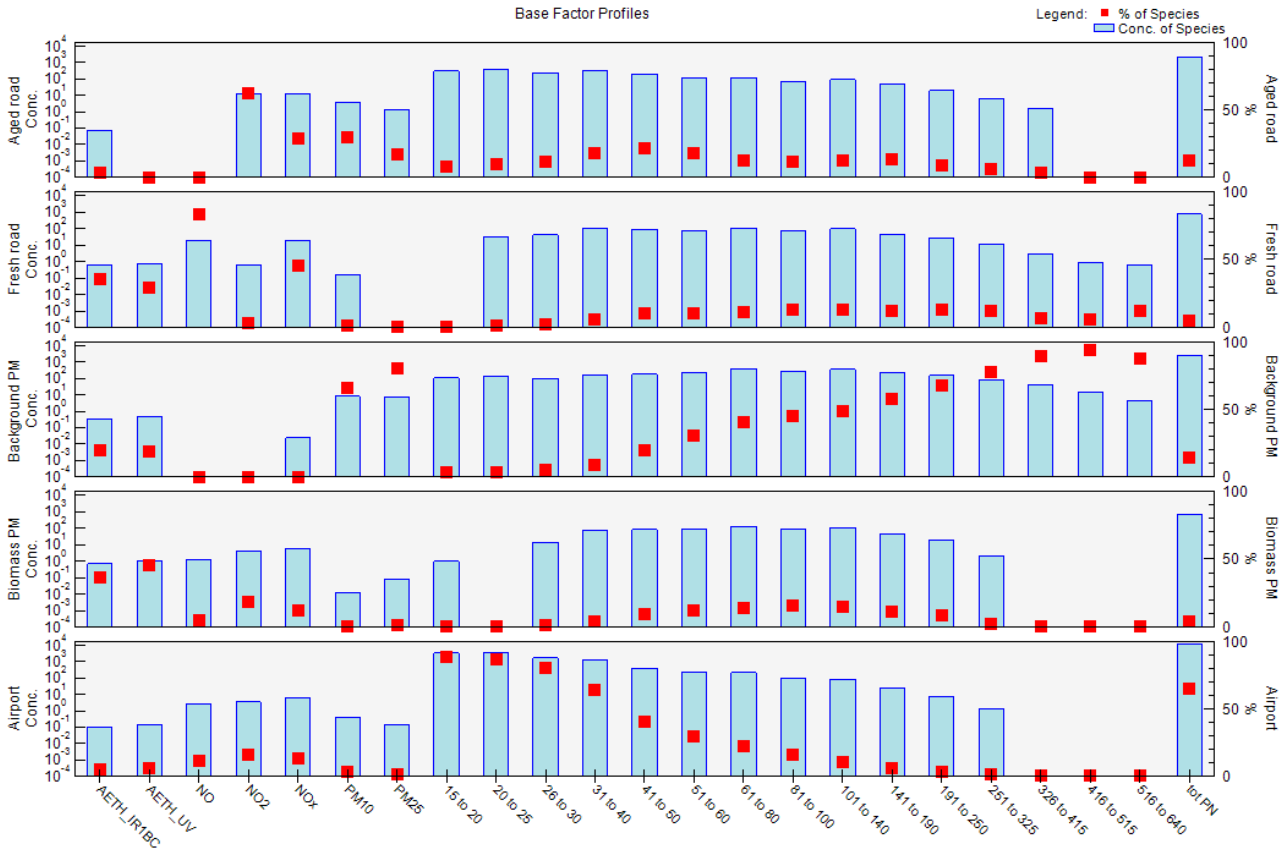

Figure 12. PMF Base Factor Profiles for the Oaks Road site

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.

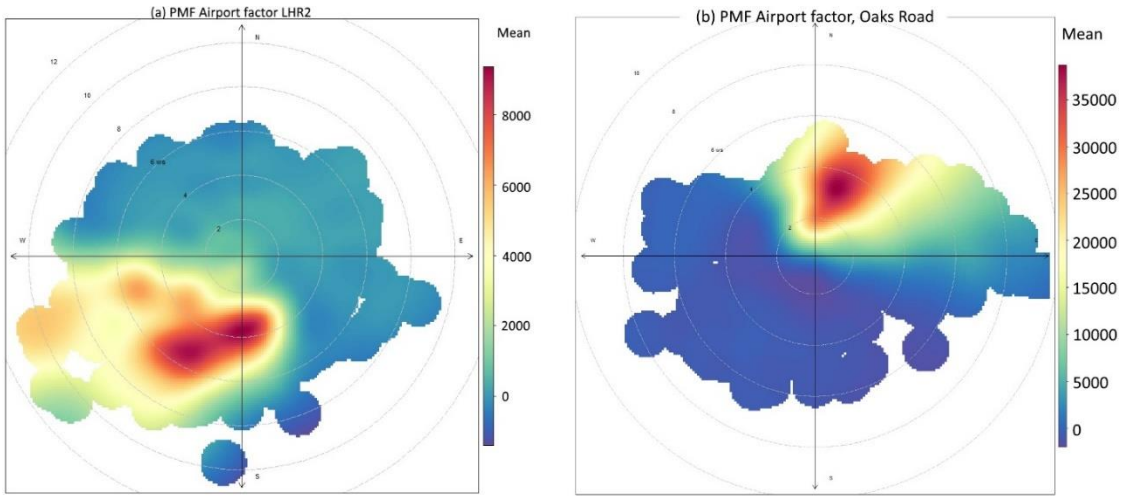

Figure 13. Polar plots of Aircraft PMF factors at (a) LHR2 and (b) Oaks Road

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.

4.5 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 5 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.

4.6 Data Availability

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

4.7 Acknowledgements

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4.9 Appendix update

A number of amendments have been made to the published paper. These are deemed to be minor in nature and do not require the original papers to be updated. The changes are as follows:

Section	Amendment
4.3.2	Minor grammatical clarity
4.3.3	Discussed the potential impacts on measurements of not having dryers at the airport locations, compared to the national network
4.4.1	Corrected the measurement units in Tables 1 and 2
4.4.5	Improved the clarity of Figure 10
4.4.6	Added a statement explaining that rotational ambiguity was not considered

4.10 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.

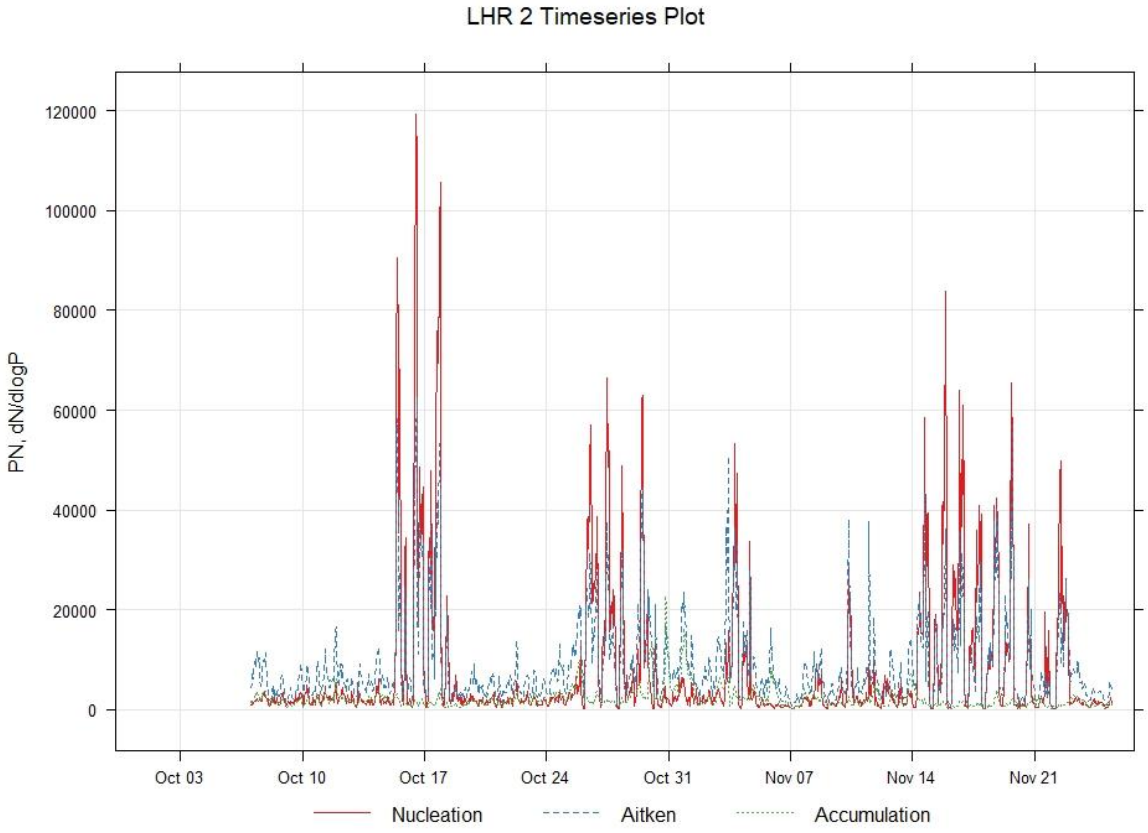

Figure S1 – LHR2 PN timeseries plot

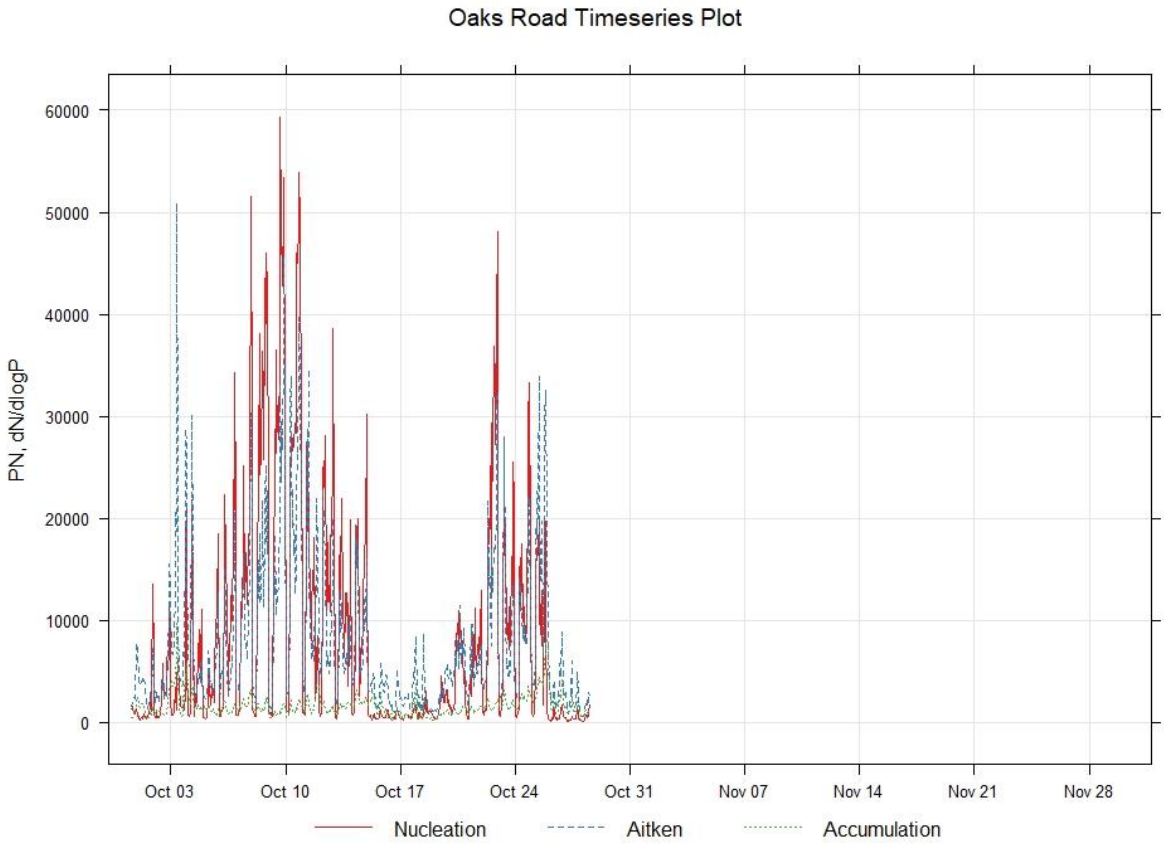

Figure S2 – Oaks Road PN timeseries plot

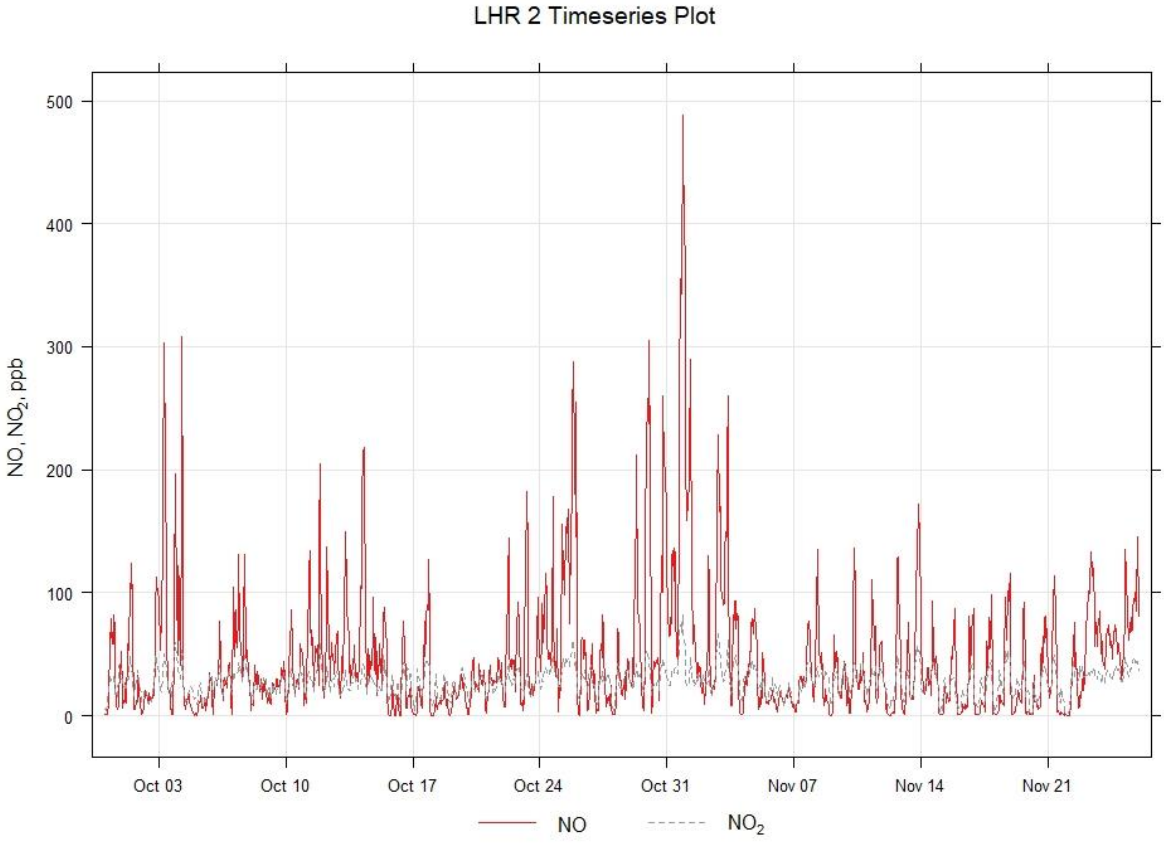

Figure S3 – LHR2 NOx timeseries plot

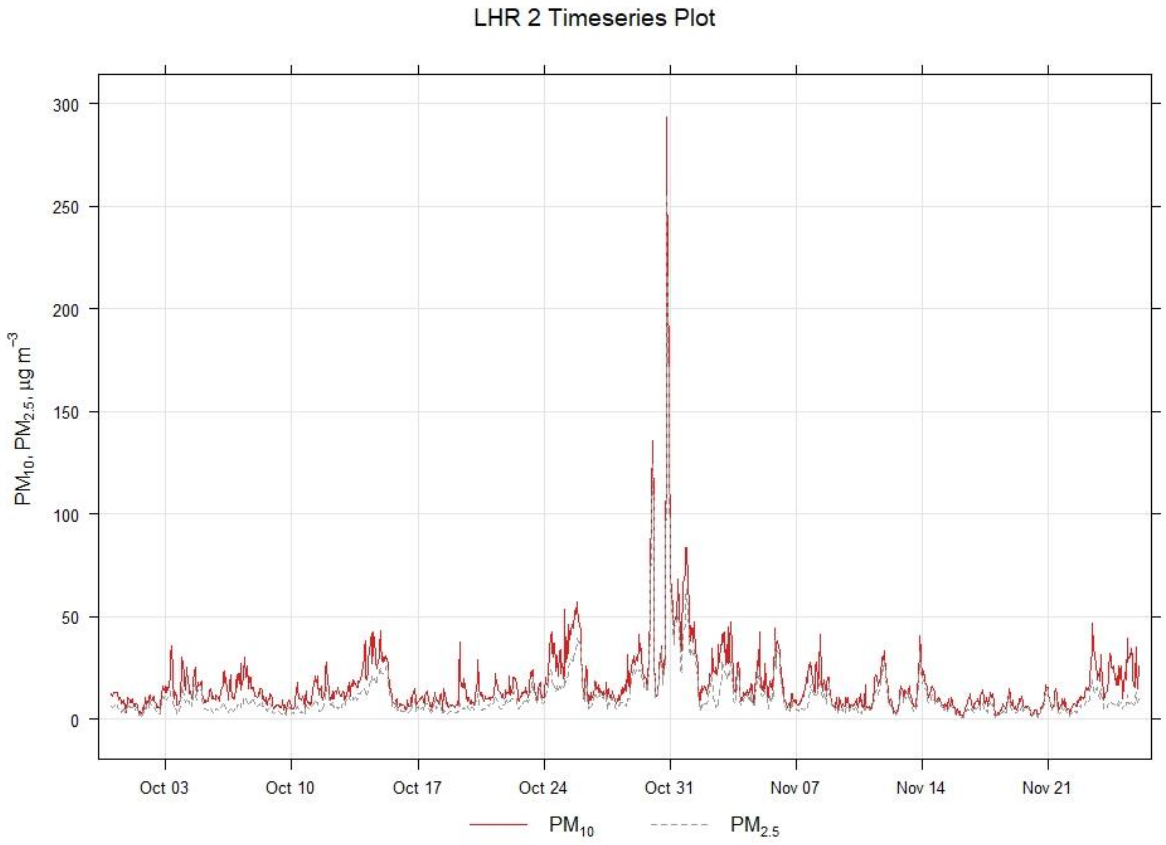

Figure S4 – LHR2 PM timeseries plot

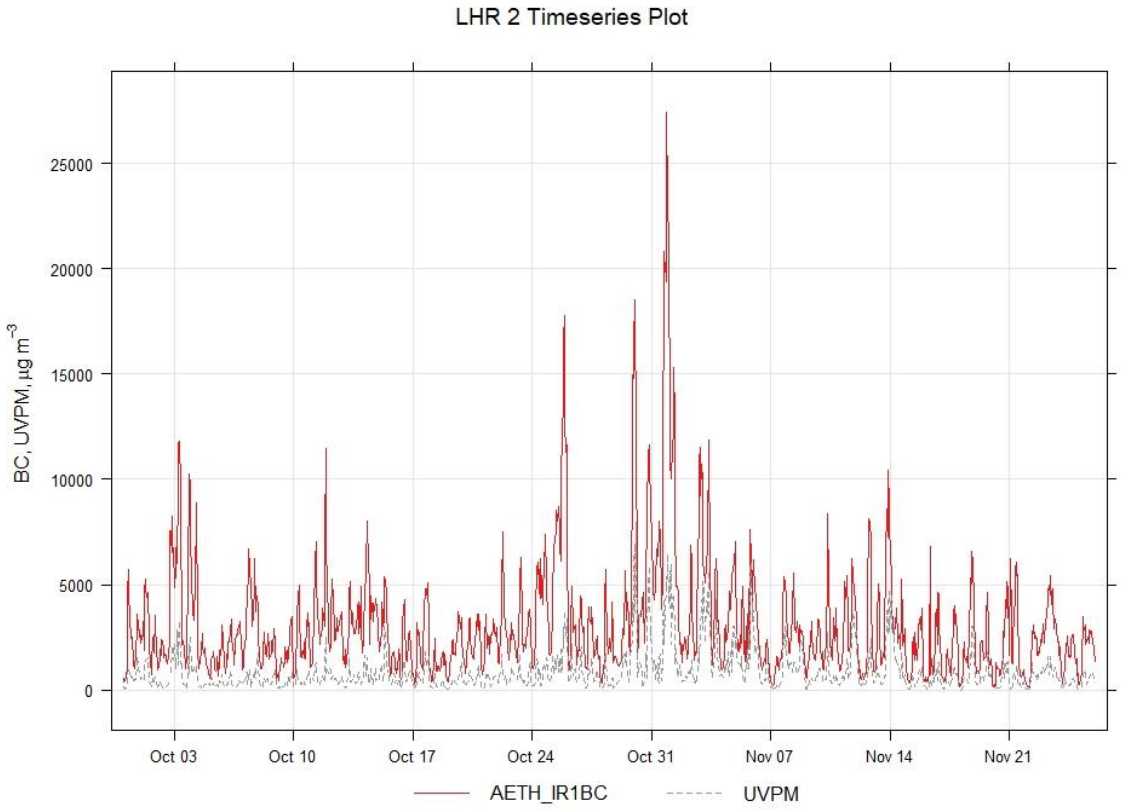

Figure S5 – LHR2 BC timeseries plot

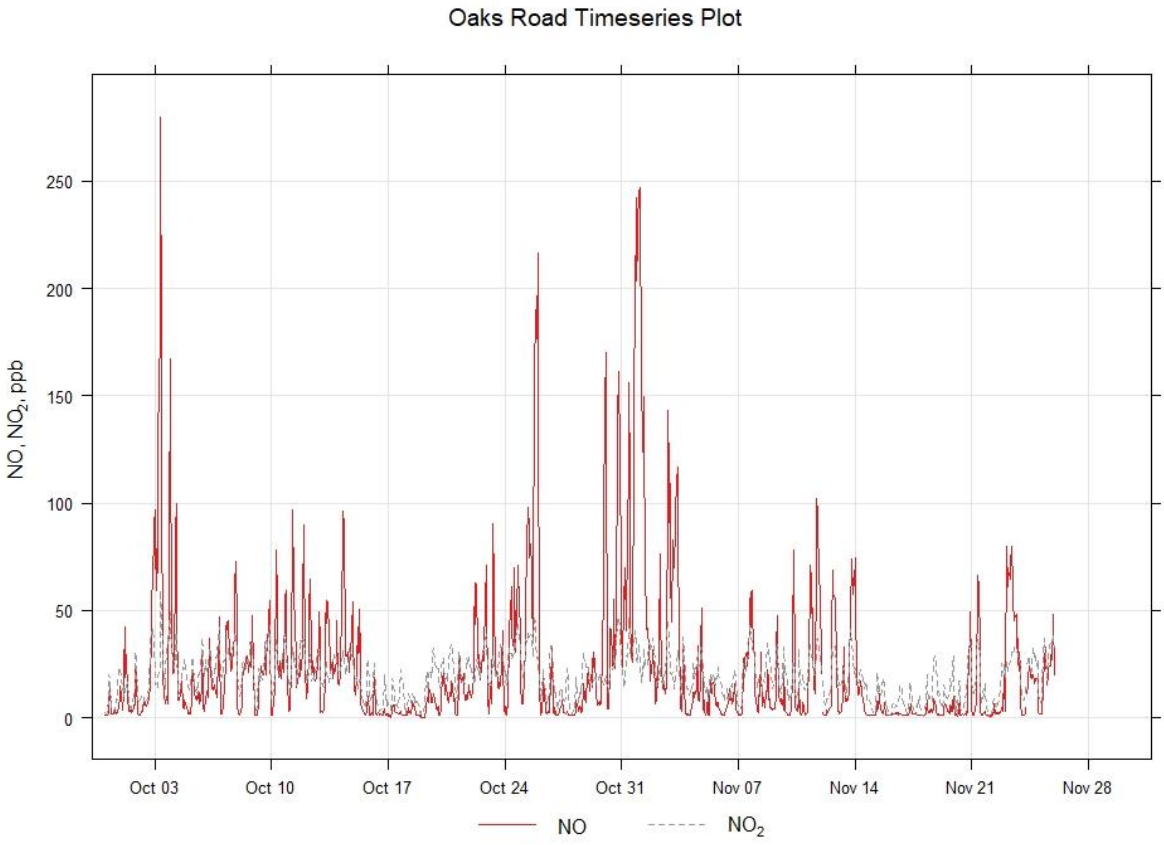

Figure S6 – Oaks Road NOx timeseries plot

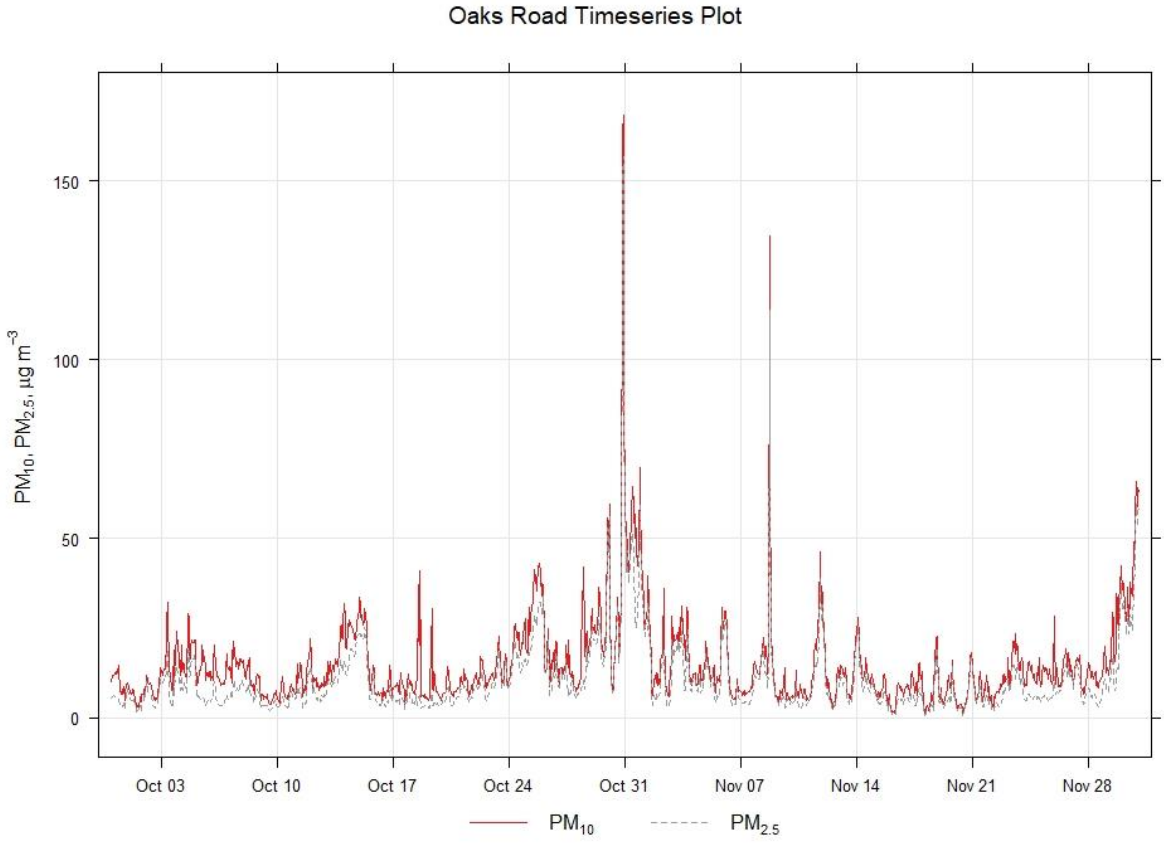

Figure S7 – Oaks Road PM timeseries plot

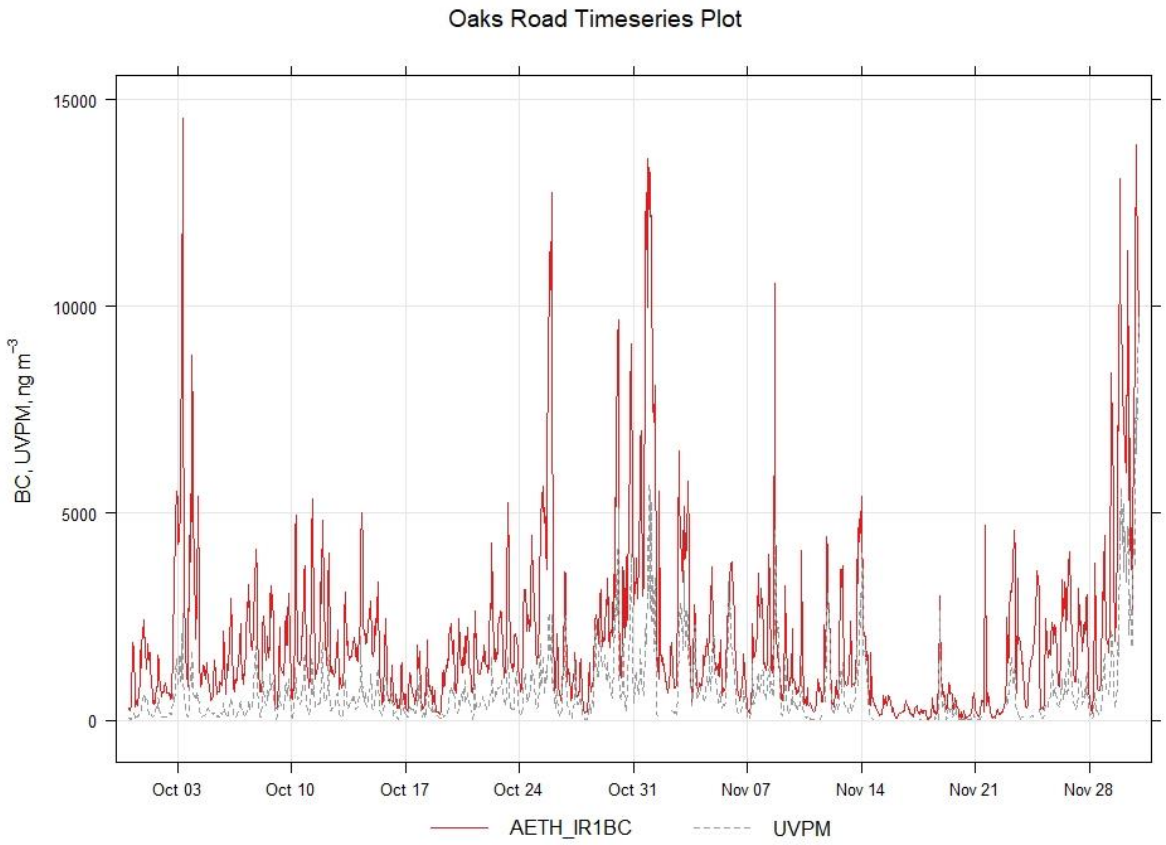

Figure S8 – Oaks Road BC timeseries plot

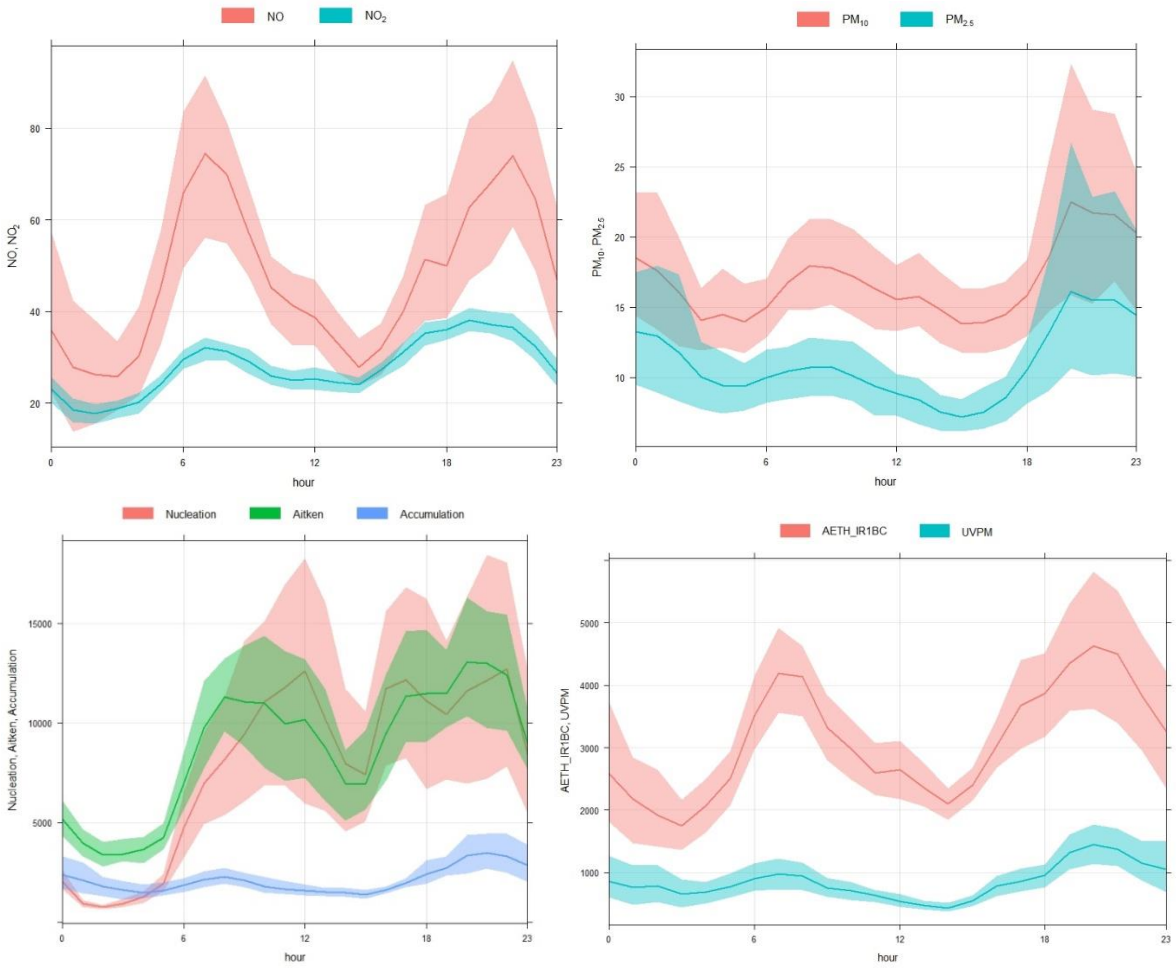

Figure S9 – Diurnal plots for measurements at LHR2

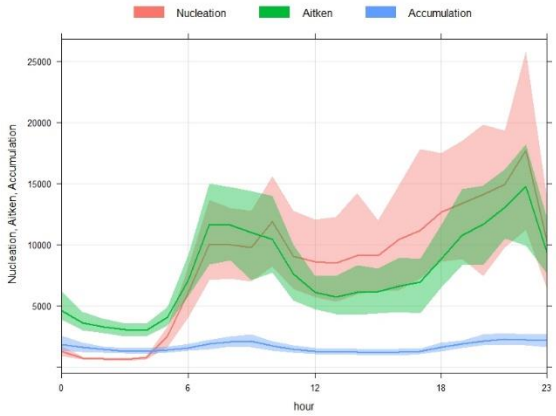

Figure S10 – Diurnal plots of Particle Number concentrations at Oaks Road

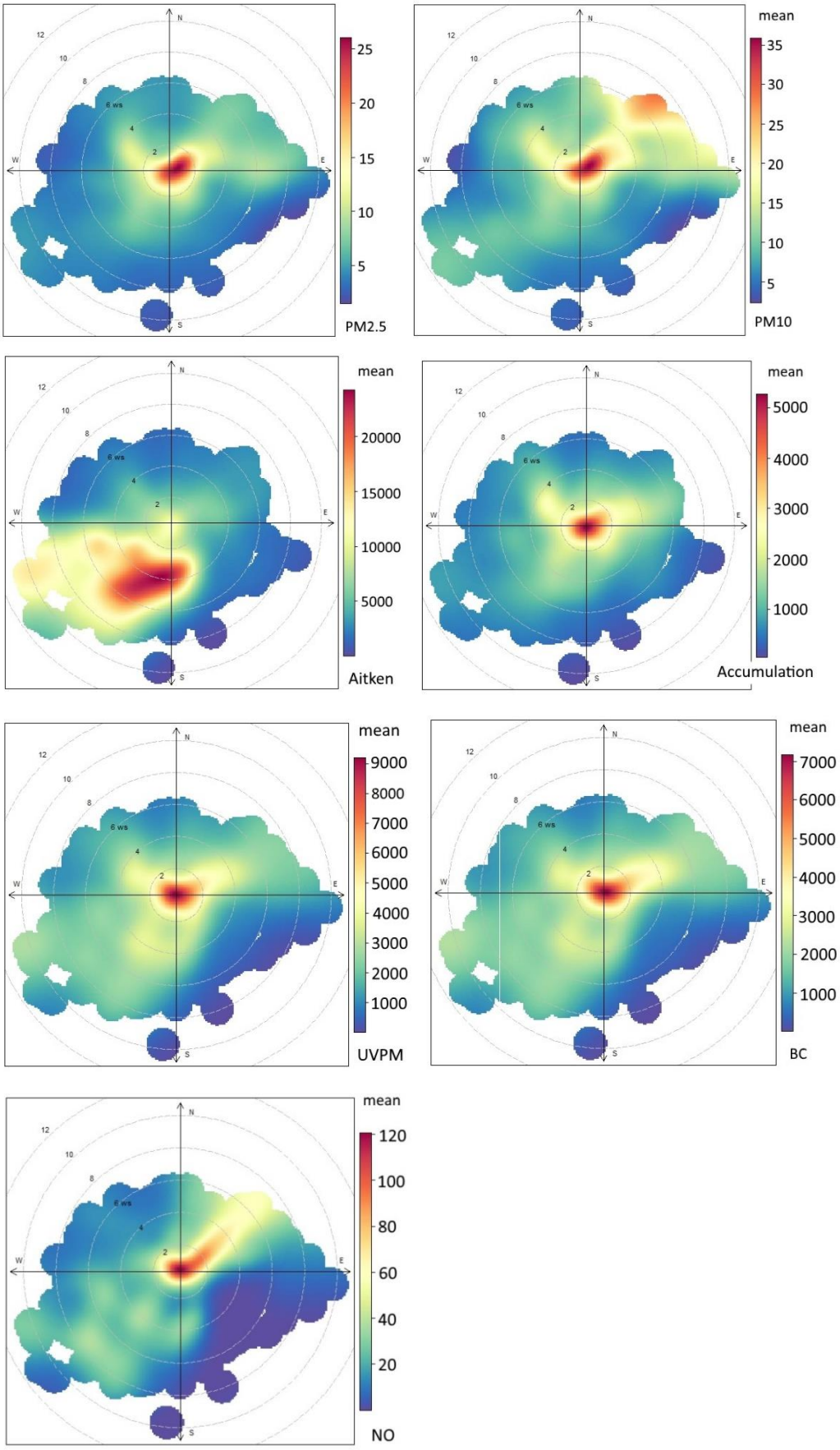

Figure S11 – Polar plots for LHR2 measurements

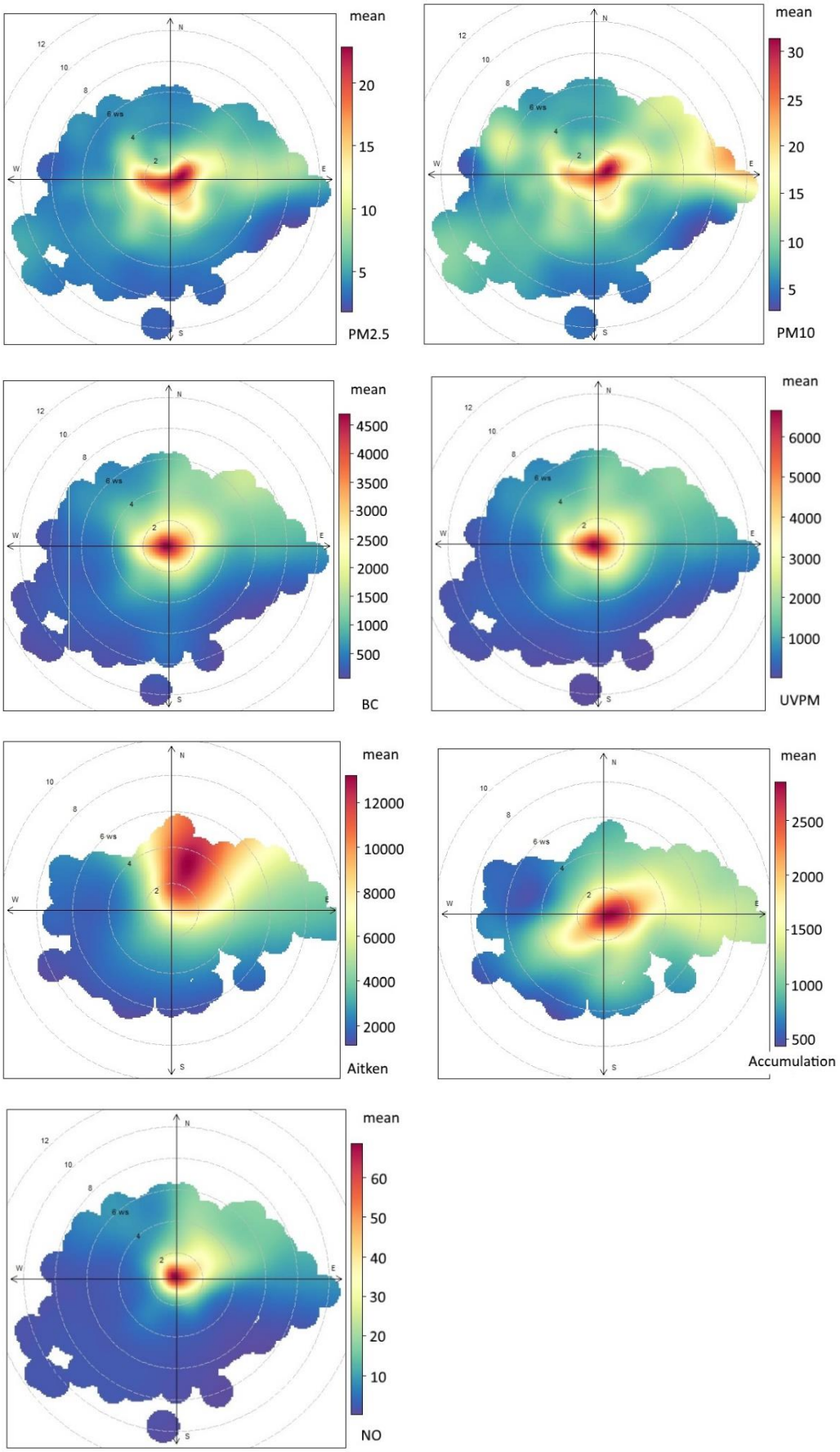

Figure S12 – Polar plots for Oaks Road measurements

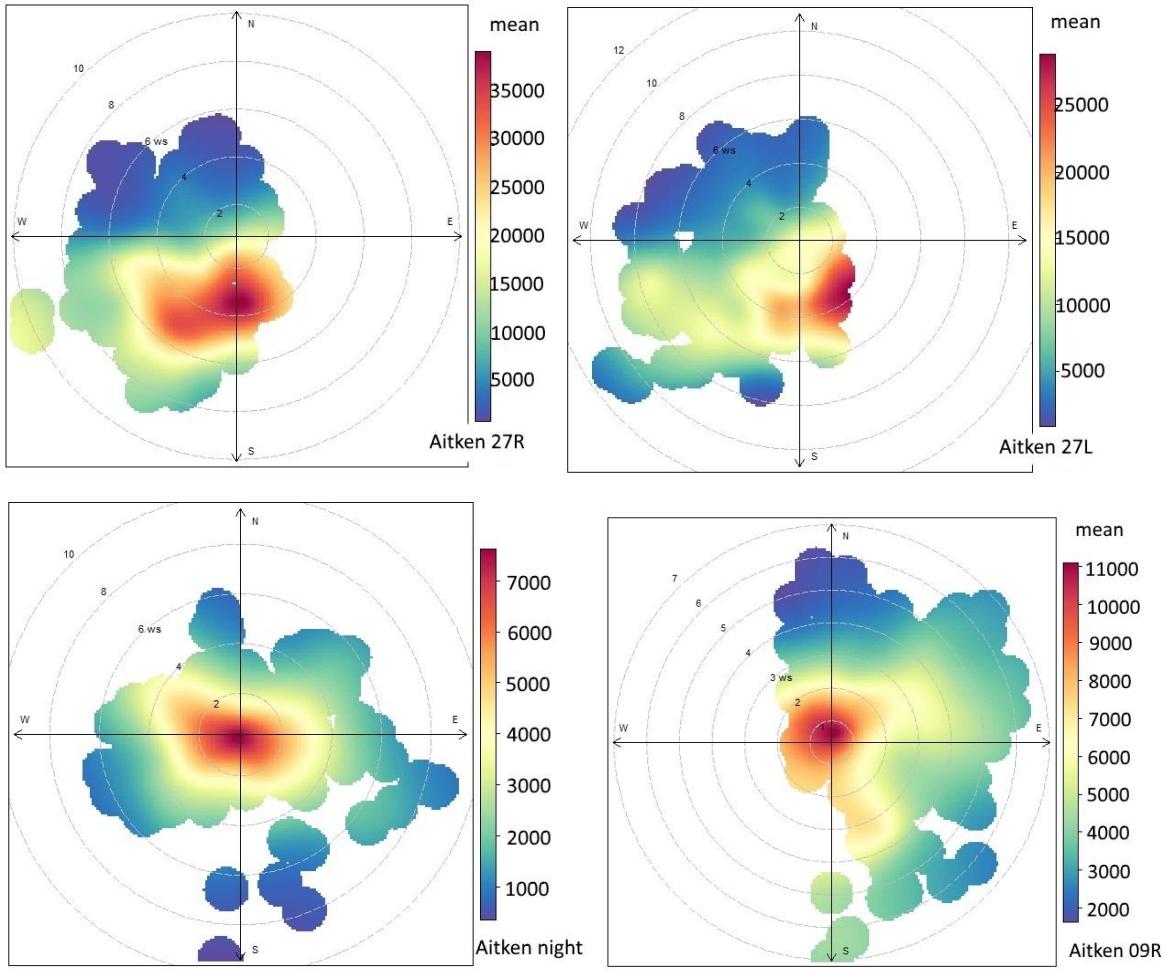

Figure S13 – Aitken particle mode for LHR2, 27L, 27R, 09R and overnight modes

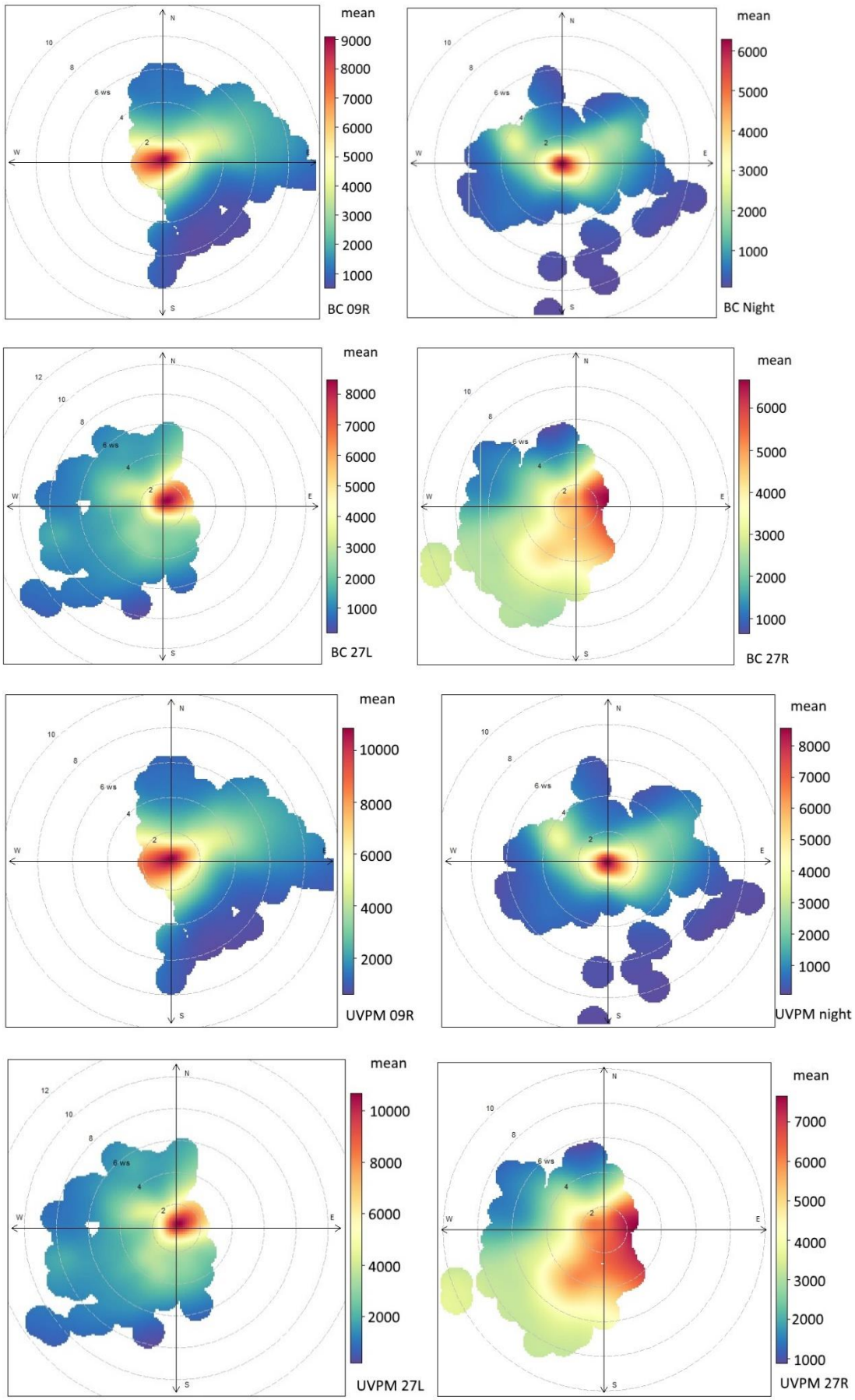

Figure S14 – Black Carbon and UVPM at LHR2 split by runway mode

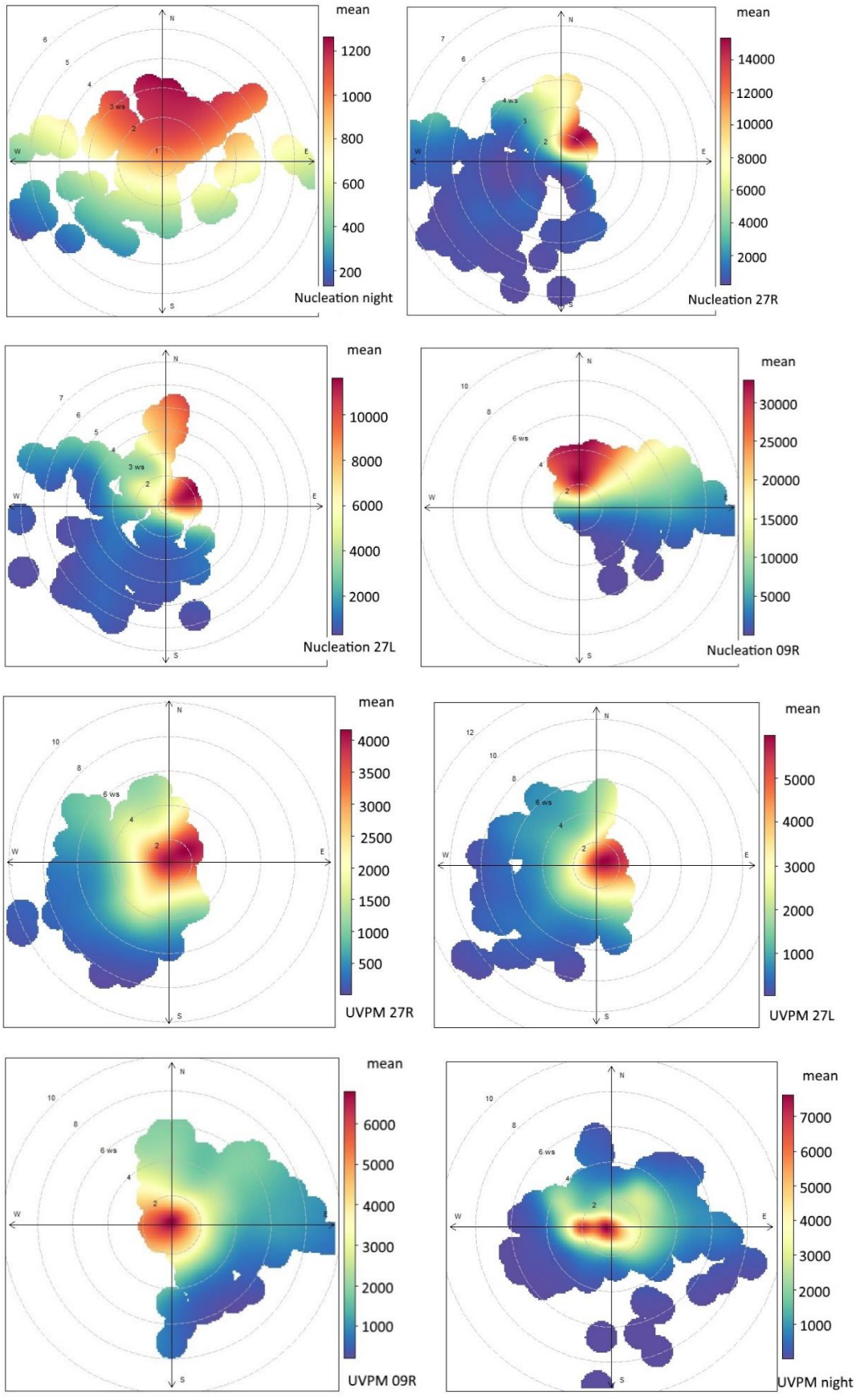

Figure S15 - Nucleation mode particles and UVPM at Oaks Road split by runway mode

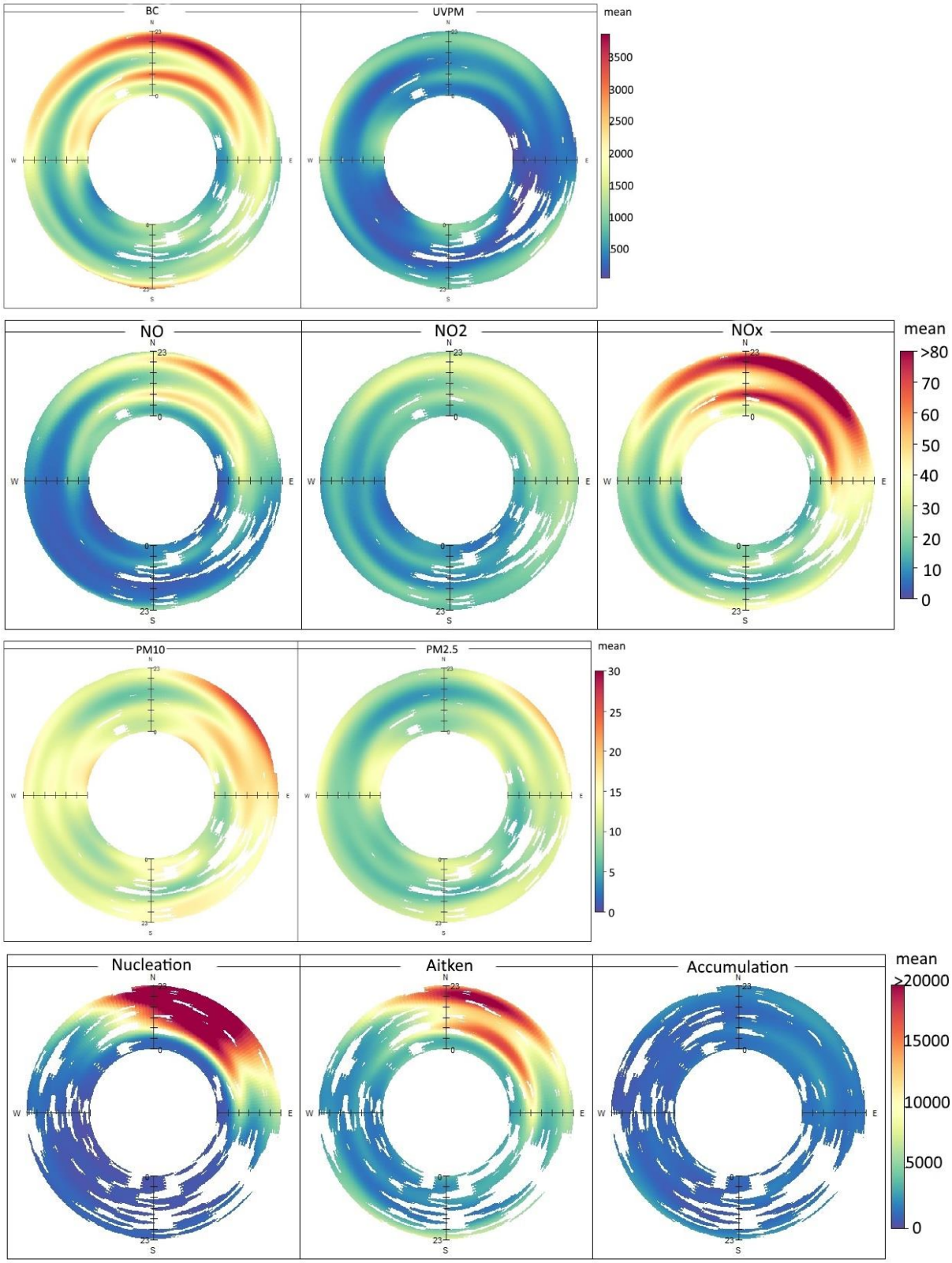

Figure S16 – Oaks Road Polar Annuli

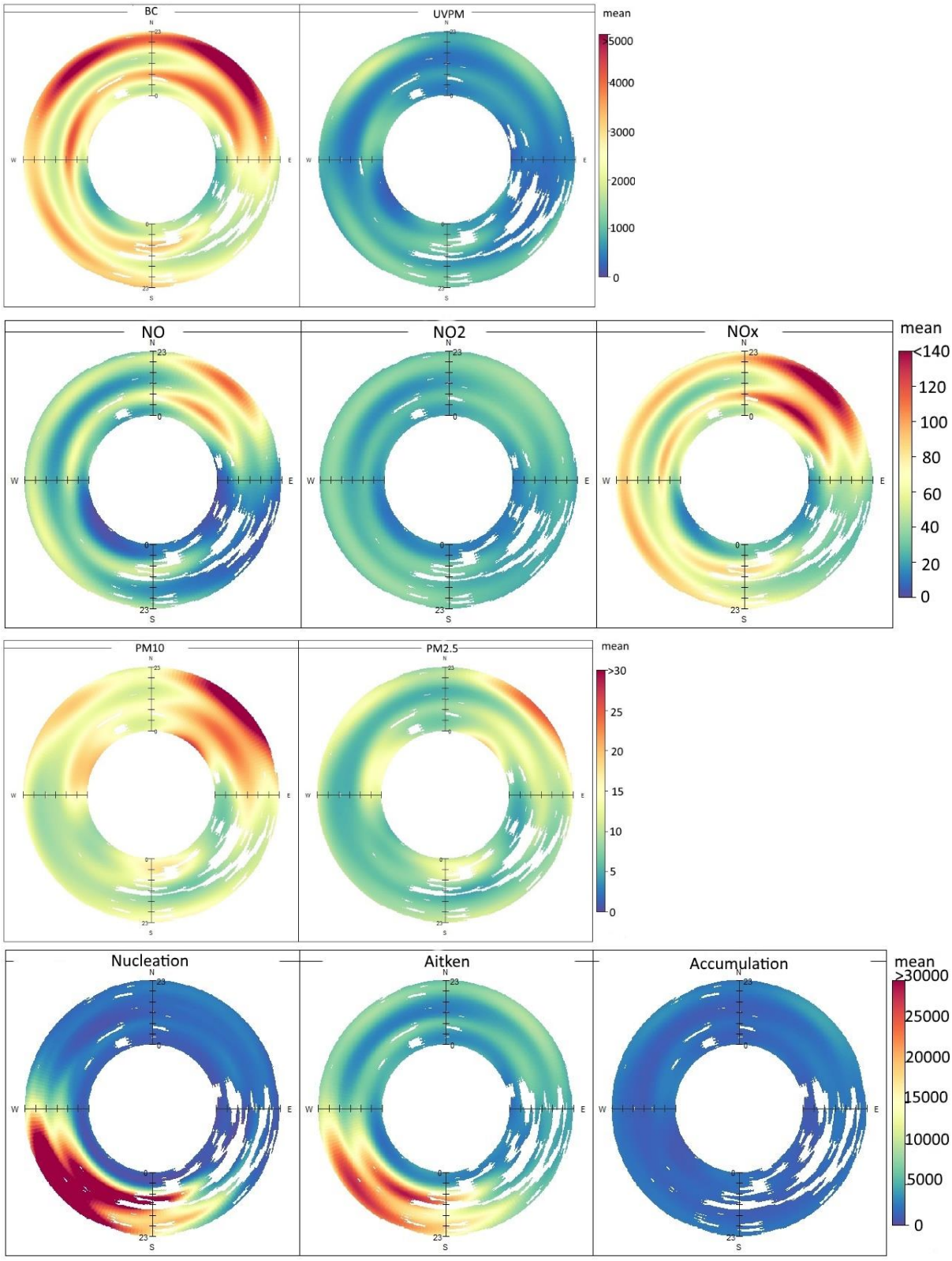

Figure S17 – LHR2 Polar Annuli

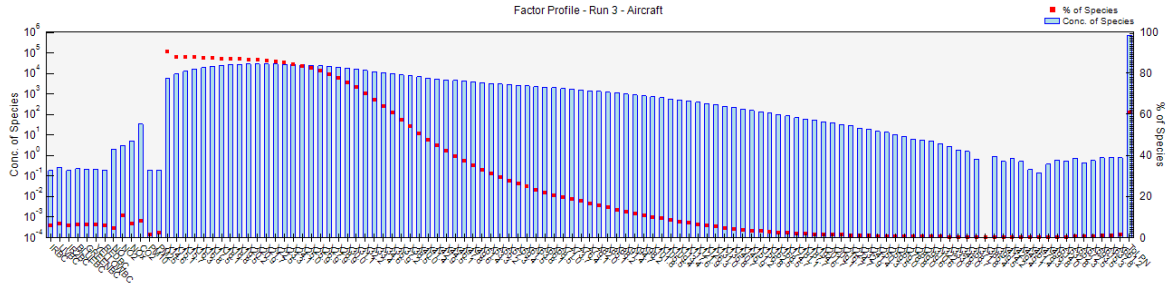

Figure S18 – PMF aircraft factor for LHR2 using all SMPS channels

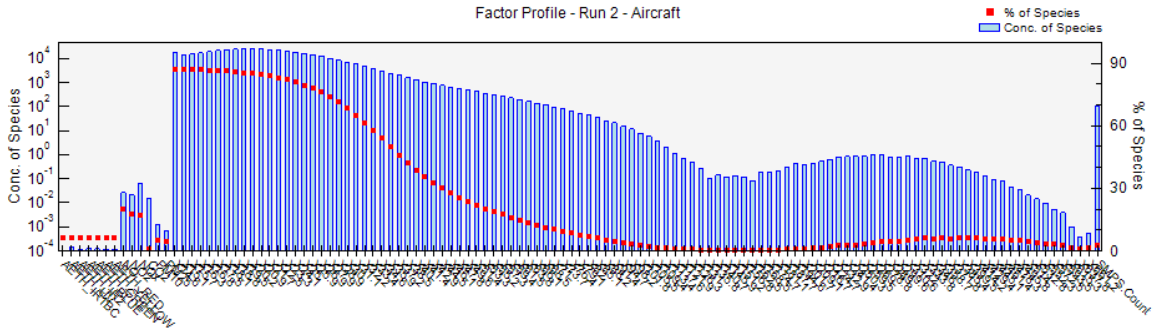

Figure S19 – PMF aircraft factor for Oaks Road using all SMPS channels

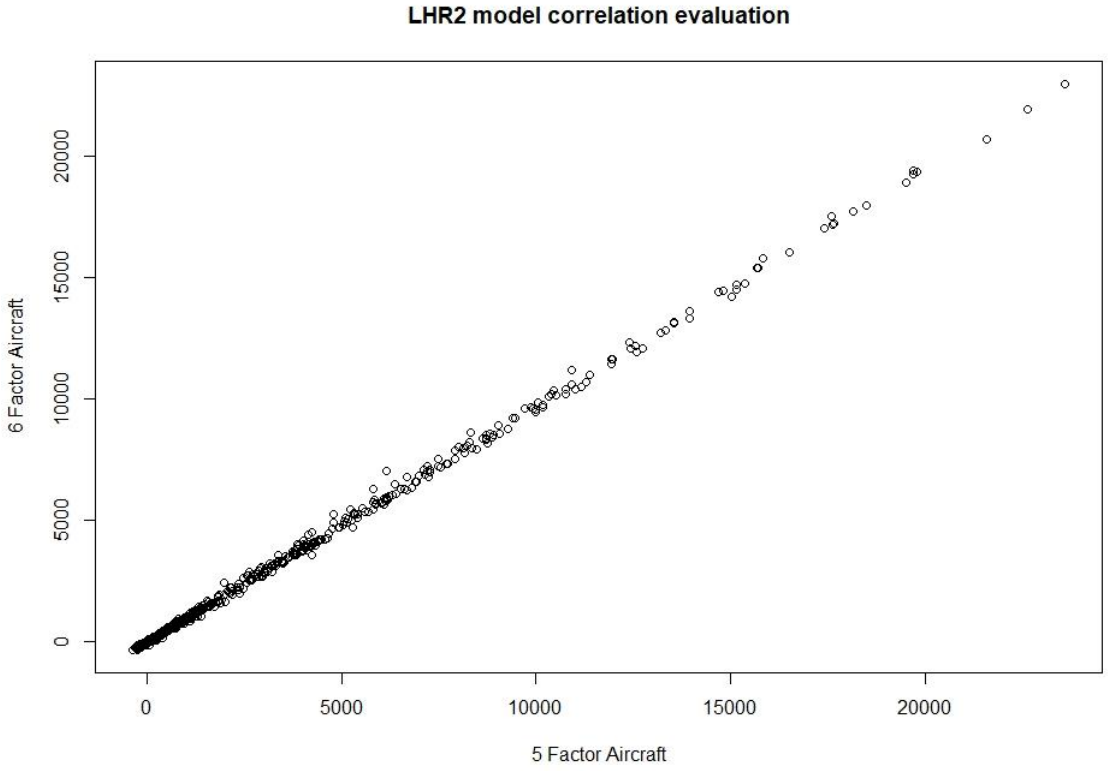

Figure S20 – Correlation between 5 and 6 factor solutions for Aircraft at LHR2

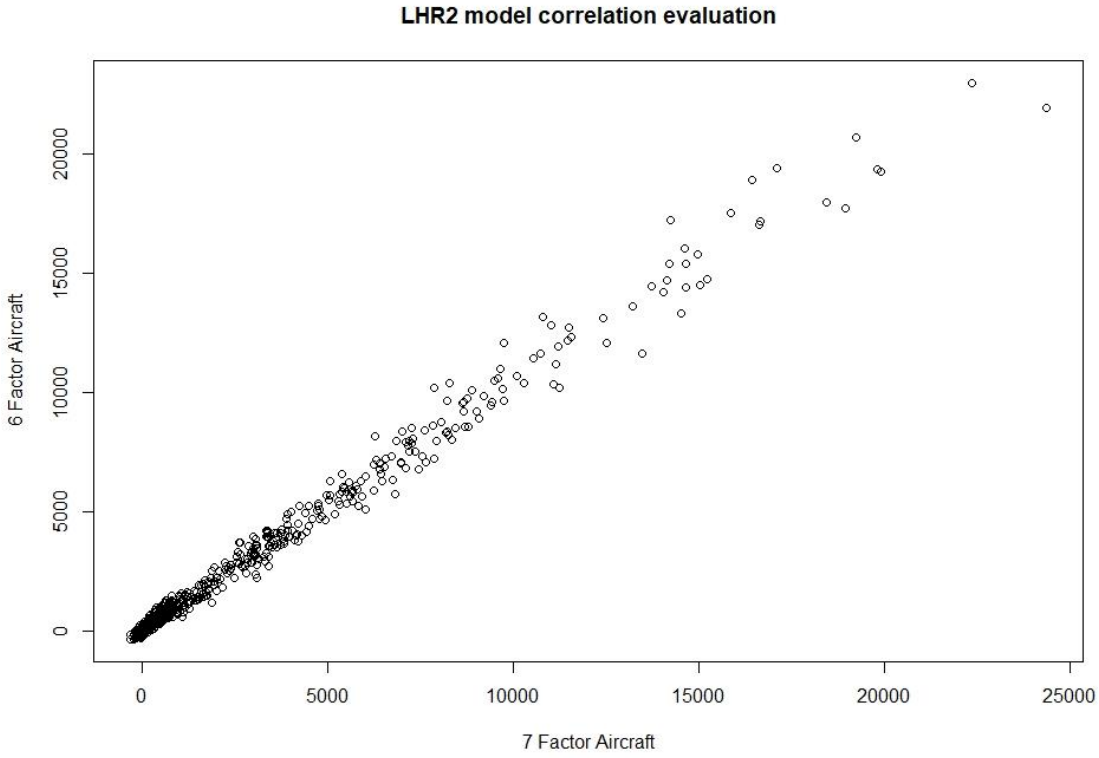

Figure S21 – Correlation between 6 and 7 factor solutions for Aircraft at LHR2

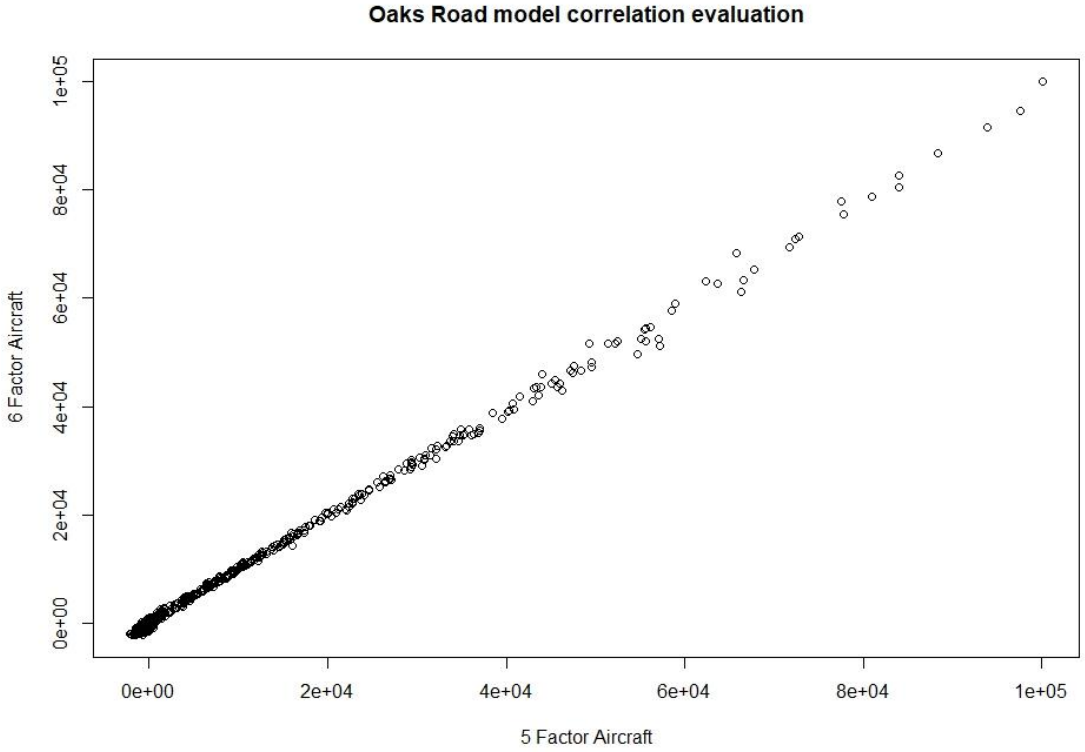

Figure S22 – Correlation between 5 and 6 factor solutions for Aircraft at Oaks Road

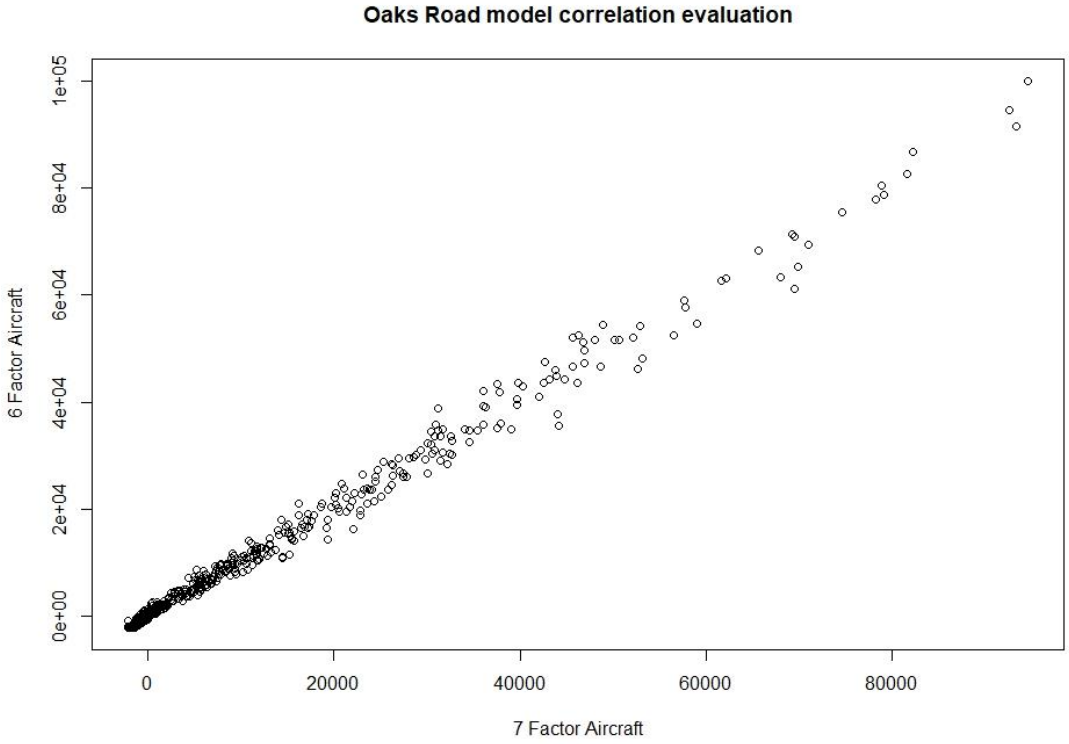

Figure S23 – Correlation between 6 and 7 factor solutions for Aircraft at Oaks Road

5. UFP at Heathrow Airport, 2017

This Chapter presents work originally published in *Atmospheric Environment* on 1 June 2021.

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5.1 Abstract

A study to monitor Ultrafine Particles (UFP) at Heathrow Airport was undertaken in the autumn of 2017. The campaign followed on from a similar study in 2016, which put UFP at the airport into context with nearby measurements. The objective of the 2017 study was to undertake UFP monitoring at higher time resolution (60 second scans) and in a narrower particle size range (6 to 100 nm). High resolution data from the NOx, PM and Black Carbon analysers on site was also collected during the survey. Measurements were made at the runway station, LHR2 to attempt to characterise individual aircraft using the runway. Nucleation mode particles are again seen to predominantly originate from the airport, with highest concentrations associated with departing aircraft. While there is some correlation of nucleation particles with NOx and BC, these pollutants, together with PM mass and Aitken mode particles, also show strong associations with winds from off-airport directions. There is some evidence that BC emissions from landing aircraft are enriched in UV-active BC (UVPM), most likely as a result of tyre abrasion upon landing. Comparison of UFP measurements with the 2016 survey was not possible because of the differences in configuration of the SMPS for the two surveys. This observation demonstrates the importance of documenting SMPS configuration, to determine if comparison between published data is possible. Analysis of the 1 minute measurement data with associated aircraft departure information was used to group the data by aircraft type. Larger aircraft departing from the runway recorded higher measurements of nucleation particles and NOx compared to smaller aircraft, while emissions of BC, UVPM and NO_2 appear to be dependent upon the age of the engine design, rather than the size of the aircraft.

5.2 Introduction

Heathrow Airport is the busiest two-runway airport in the world. In 2017, the airport handled over 78.0 million passengers and approximately 471,000 aircraft movements (https://www.heathrow.com/content/dam/heathrow/web/common/documents/company/investor/reports-and-presentations/financial-results/2017/2017-FY-Heathrow-SP-results-release.pdf). The airport is located in a complex environment: bounded by the M25 and M4 motorways on two sides, and by the outskirts of London on a third side.

The history of AQ measurements at Heathrow Airport, together with review of UFP at airports and the results of our UFP study at Heathrow Airport in 2016 are extensively discussed in Stacey (2019) (20) and Stacey et al (2020) (21)

An increasing amount of research has been undertaken close to airports, to better understand the nature of ultrafine particles (UFP) emitted from aircraft. The literature review by Stacey (2019) (20) collected the most relevant literature at the time into a single document. Prior research undertaken and referenced in this review, together with a research study of UFP measurements undertaken at Heathrow Airport in 2016 by Stacey et al (2020) (21), informs the direction of research and analysis throughout this paper. More recently studies by, for example Henry et al (2019) (11), Lopes et al (2019) (14), Bousiotis et al (2019) (3) and Rivas et al (2020) (18) have supported the work of others that UFP from airports and aircraft can be observed many kilometres downwind of an airport. Fushimi et al (2019) (9) found that a significant proportion of UFP measured at Narita Airport consisted of unburned jet lubrication oil.

Similarly, the impact of UFP on health has been increasingly studied in recent years. Bendtsten et al (2019) (2) reported that the UFP sampled at two airports in Denmark is comparable in toxicity to UFP from diesel exhaust. Habre et al (2019) (10) found observable health impacts in sensitive receptors downwind of Los Angeles International Airport (LAX),

while Wing et al (2020) (25) also identified a link between exposure to aircraft-related UFP and pre-term birth in the region of LAX

For the first time, a panel of experts (Morawska et al, White paper, 2019 (16)) has put forward a proposal to regulate exposure to concentrations of UFP. In terms of mitigation, both Morawska et al (2019) (16) and de Jesus et al (2019) (5) found that reducing emissions of $PM_{2.5}$ was not likely to have any significant effect on measured concentrations of UFP.

The Stacey et al (2020) (21) study showed that UFP concentrations at Heathrow in 2016 were clearly influenced by aircraft activity and wind direction. The smallest particles were associated with winds from the airfield, and the particle size distribution of the airport-derived airmass was clearly different to typical urban roadside, urban background and rural distributions. The study focussed on ensuring comparability with the reference monitoring stations, which report measurements every three minutes. At this time resolution, it is not possible to use the data to identify individual aircraft, which depart or arrive on average every 90 seconds at Heathrow.

A follow-up campaign was therefore devised to measure UFP, and where possible the other pollutants at the monitoring station, at a faster time resolution to evaluate individual aircraft emissions and the relationships between aircraft, UFP and other pollutants. This paper builds on the 2016 report and presents the results of the 2017 study.

5.3 Methods

5.3.1 Monitoring Location

This measurement campaign was designed to undertake rapid measurements of UFP and the conventional pollutants at Heathrow to further explore the local nature of these pollutants in the context of aircraft movements at the airport.

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

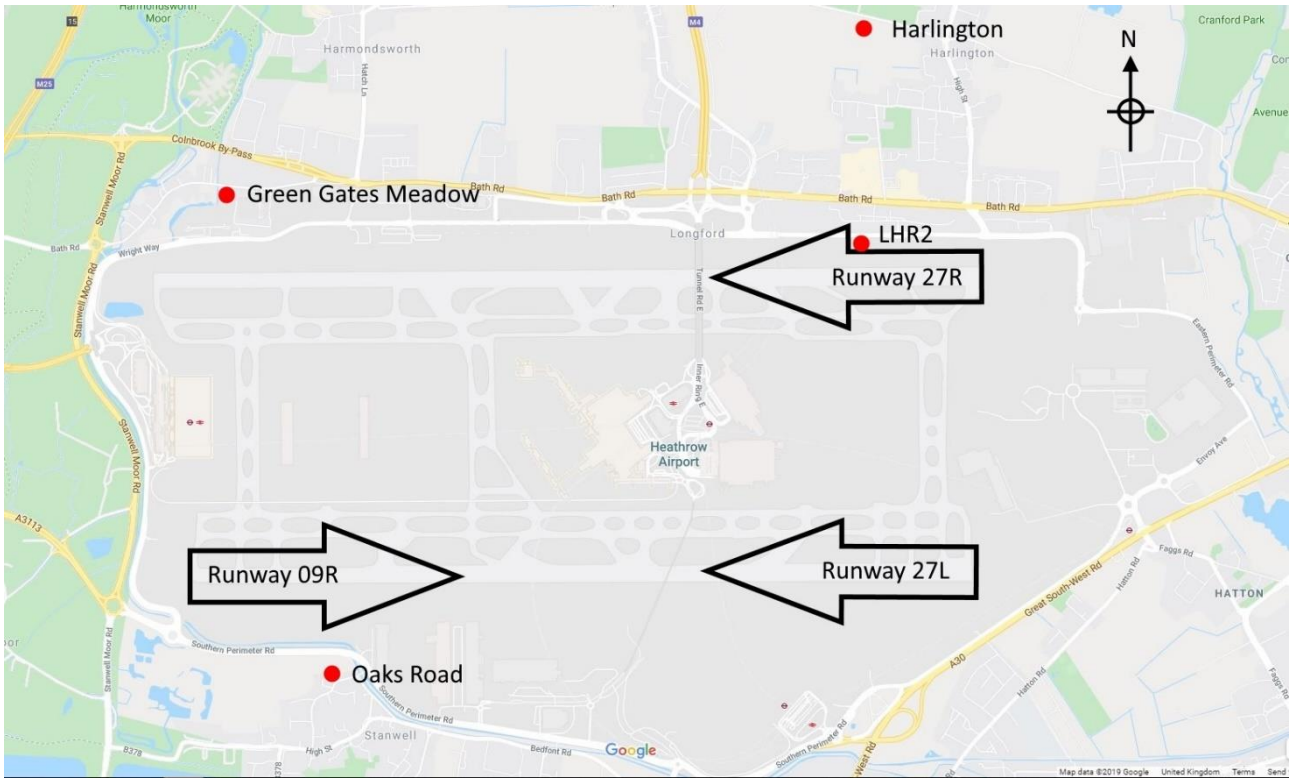

Figure 1. Locations of Heathrow monitoring stations. Runway 27R, Runway 27L and Runway 09R denote the three operating modes of the airport, indicating here the runway assigned for departing aircraft. Note that aircraft never depart in an easterly direction on the northern runway.

Because of the dominant south-westerly nature of the winds in the UK, the LHR2 monitoring station is ideally positioned to measure aircraft exhaust plumes. This location was also one of the two monitoring stations used in 2016 and fully described in Stacey et al (2020) (21).

5.3.2 UFP Measurement Campaign

Measurement of UFP at the LHR2 monitoring station was undertaken between 4th October and 7th November 2017.

The following equipment was used:

- Butanol based TSI Model 3776 CPC (TSI inc., MN, USA) to count particle numbers (the 3776 is more effective at detecting smaller particles than previous TSI CPCs - D50 2.5nm).

- 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 was controlled by a PC connected to the Model 3082 running AIM v10.2.0.11. Data was downloaded weekly from the PC 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) (24) and Wiedensohler et al. (2018) (23). The only difference from the recommendations of Wiedensohler et al. (2012) (24) was the absence of a dryer. This is considered advantageous due to minimising diffusive losses of particles while having little effect upon the size distribution of largely hydrophobic nanoparticles subject to a significant Kelvin effect.

The SMPS instrument was configured to sample in the range 6.38nm to 98.2nm, 64 channels per decade. Sampling was programmed to run for 1 minute, sweeping up in size for 45 seconds, and returning down for the remaining 15 seconds.

The instrument was 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, remote communication to the analysers was not undertaken. The monitoring station was visited weekly to ensure correct operation and take remedial action if required.

Calibration of the CPC and SMPS followed identical procedures and used facilities described in Stacey et al (2020) (21) but within the narrower particle size range used for the 2017 survey.

The 6-100nm configuration of the SMPS in 2017 differs significantly from the setup used at Heathrow in 2016 by Stacey et al (2020) (21) and in the UK National Particles network. Both

the Heathrow 2016 and National Network configurations are described in the Stacey et al (2020) (21) paper and are not documented further here. Comparisons of the 2016 and 2017 datasets will be explored in the results, but will be significantly influenced by the differences in configurations used in 2016 and 2017 and, to an extent, the differing meteorology.
The other analysers deployed at LHR2 are described fully in Stacey et al (2020) (21), but were additionally configured to collect 1 minute average data.

5.3.3 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) (4))
In accordance with the processes defined in Stacey et al (2020) (21) for the 2016 datasets, Nucleation particles are defined as particles smaller than 25 nm, Aitken particles are defined as particles between 26 and 100 nm.
Particle number concentrations are reported in units of particles / cm^3, and are calculated from individual size bin data from the SMPS, with no decade adjustment applied.
Measurements from the black carbon aethalometers are reported using identical procedures as reported in Stacey et al (2020) (21).

5.3.4 Measurement Quality Assurance and Quality Control

Processing of the data was undertaken using the same QA/QC procedures described in Stacey et al (2020) (21). While the Heathrow study UFP data reported here uses the same quality assurance and quality control procedures used for the national network datasets and the 2016 study, the differences in configurations in 2017 (including flow rates, size ranges, sample

time, software), will have a significant impact on the ability to make direct comparison between the two surveys. These differences will be discussed later.

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 these analysers is also provided in the Supplemental Information, Tables S1 and S2.

5.4 Results

5.4.1 Overall Summary

Timeseries data for the hourly measurements at LHR2 are presented in Figure 2 below. One minute data for all pollutants are available in the DOI, and are presented graphically in Supplemental Information, Figure S1. Hourly averaged measurements of NO_x, PM_{10}, $PM_{2.5}$ and BC are also accessible through the http://heathrowairwatch.org.uk webpages. The 1 minute averaged data from these analysers will be used to explore associations and differences to typical ambient environments.

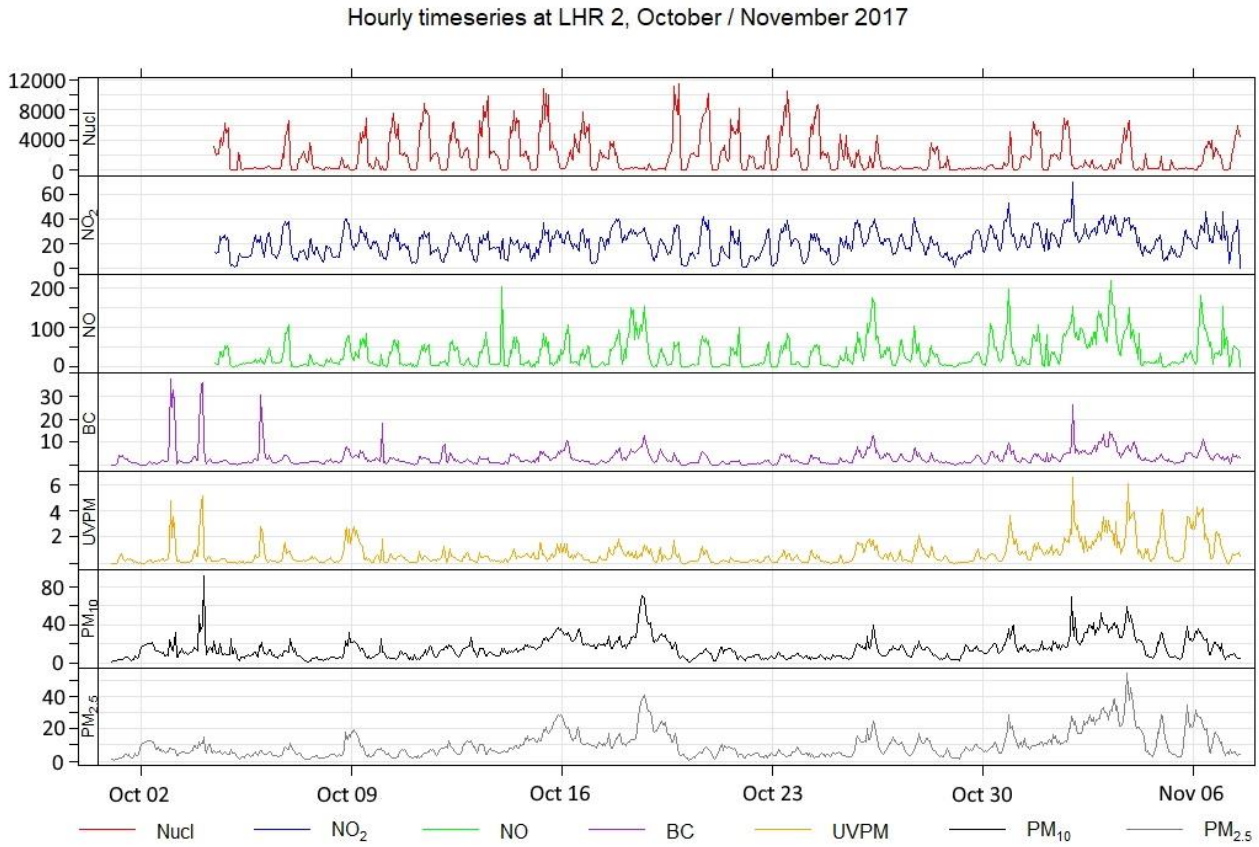

Figure 2 – Hourly timeseries data at LHR2, October and November 2017. Reported concentrations are ppb for NO and NO_2, ug/m^3 for PM_{10}, $PM_{2.5}$, BC and UVPM, and particles/cm^3 for nucleation particles (labelled Nucl on the plot).

It can be seen from Figure 2 that the nucleation concentrations show poor visual agreement with NO_2 and BC (confirmed with regression analysis - all data r^2 0.2 to 0.3), but the relationship for nucleation particles is poor with PM_{10} and $PM_{2.5}$ (all data r^2 less than 0.01). Concentrations of nucleation particles clearly undergo a diurnal cycle and, as observed in 2016 (Stacey et al (2020) (21)), increase coincident with periods when aircraft are active. The average particle size distribution for the 2017 survey reaches a maximum number concentration at 12.2 nm.

Diurnal plots are presented in the SI (SI figs S26 to S33). Concentrations of NOx, Nucleation and Aitken particles follow the expected diurnal profiles, where highest concentrations are experienced between 06:00 and 21:00. In contrast, diurnal concentrations of PM, BC and UVPM are highest between the hours of 18:00 and 02:00, coinciding with traditional periods

of domestic heating, and the increase in the evening due to the road traffic rush hour. The diurnal profiles for PM follow the typical profile of regional monitoring (presented in the SI), suggesting that airport measurements are strongly influenced by off-airport airmasses.

5.4.2 Dependence of Airport Measurements on Meteorology

The meteorology for the 2017 survey was dominated by south westerly winds, ideal for assessing the contribution of the airport and aircraft emissions at the LHR2 monitoring station. The wind rose plot for the survey is presented in the SI, figure S34

Polar plots of the hourly average data are presented in the SI, figures S2 to S10. As with the 2016 survey, nucleation particles are almost exclusively associated with winds from the airport. Aitken particles are strongly associated with winds from the airport, but there is also contribution from easterly and northerly wind directions. NOx, PM_{10} and $PM_{2.5}$ polar plots are very similar to those seen at other monitoring stations across the south east of England, and mostly not from the direction of the airport, reflecting the diverse sources of these pollutants in the UK. The polar plots for BC and UVPM show some influence from the airport, but also when winds are low and immediately west of the station. There was a construction depot around the monitoring station during the survey. It was used as a parking facility for the construction plant, vehicles were only active between 23:00 and 04:00. As a result, it is likely that this vehicle activity has influenced the overnight measurement data. For the purposes of aircraft analysis, this period is not included in analysis in any case. Imagery of the construction depot is presented in the SI, figure S11.

5.4.3 Relationship Between Pollutants

Following exact time synchronisation of all measurement datasets, bivariate regression analysis was undertaken using the polarPlot function in openAir. This analysis was used to identify which wind directions were associated with the closest correlation between pollutants. These plots are provided in the SI, figures S12 to S25.

The plots show very strong (r^2 greater than 0.8) correlation between Nucleation and Aitken particles from the direction of the airport, much weaker when winds are from northerly directions. The weak correlation (r^2 less than 0.4) between Nucleation and Aitken particles from the north clearly indicates that nucleation particles mostly originate from the airfield. Nucleation particles from the airport are also closely correlated with BC (r^2 above 0.8). Nucleation particles from the airport show some correlation with UVPM and NOx (r^2 between 0.5 and 0.9) , but correlation is weak (r^2 less than 0.4) for Nucleation particles with either PM_{10} or $PM_{2.5}$.

PM_{10} and $PM_{2.5}$ correlation with NOx is mostly weak (r^2 less than 0.5) for most wind directions, though there are clusters of good correlation (r^2 above 0.8) to the north and one to the south that could be associated with the nearby runway. PM_{10} and $PM_{2.5}$ correlations with UVPM (r^2 above 0.8) are strongly associated with some airport wind directions and speeds, as well as from directions north of the monitoring station, suggesting a multitude of sources contributing to PM and UVPM in the area. The correlation immediately to the south closely mirrors the PM/NOx correlation, further suggesting the influence of the runway as a contributor to local measurements.

NOx and BC correlation is good to strong (r^2 between 0.6 and 1.0) for most wind directions except for the NW sector. Correlation between NOx and UVPM is strong (r^2 above 0.8) for wind directions associated with the airport.

5.4.4 Dependence of Measurements on Airport Operation

As previously stated, Heathrow operate the two runways in a rotating system when aircraft depart and arrive in a westerly direction. During any typical day, aircraft land on runway 27R for half the day, before swapping and landing on runway 27L. Aircraft depart on the other runway, allowing complete independence of departure and arrival schedules. From a

monitoring perspective, this is very useful, because it raises the possibility to assess emissions from departing and arriving aircraft. It needs to be remembered that the measurements at the monitoring station will be impacted by cooling, dilution and interaction with other sources, but this is mitigated to some extent by the proximity of the station to the runway and the absence of any other sources between the aircraft and the monitoring station. In addition, especially for gaseous and mass-based PM measurements, while the background concentrations will contribute to the reported measurements, their contributions are not removed from the datasets. This has been considered during the analysis. Aircraft movement information for the 2017 survey was again provided by Heathrow Airport Limited.

The 2016 survey found that average concentrations of Nucleation particles was highest when aircraft were departing closest to the monitoring station.

The table below provides average concentrations measured at the airport in October / November 2017 in various operating modes:

Pollutant / Operation	Overall (902 hours)	Depart 27R (320 hours)	Depart 27L (308 hours)	Depart 09R (45 hours)	Overnight (229 hours)
Nucleation, # / cm^3	1813	3625	1328	422	141
Aitken , # / cm^3	205	317	191	127	81
BC, $\mu g/m^3$	3.22	3.61	2.34	4.47	3.61
UVPM, $\mu g/m^3$	0.71	0.85	0.49	0.58	0.83
PM_{10}, $\mu g/m^3$	15.1	14.4	13.5	28.4	15.5
$PM_{2.5}$, $\mu g/m^3$	10.0	9.7	8.9	18.6	10.2
NOx, ppb	52.9	76.5	39.2	71.5	33.4
NO, ppb	32.6	50.1	20.6	48.8	20.1
NO_2, ppb	20.3	26.4	18.7	22.7	13.2

Table 1 – average concentrations in different airport operating modes.

The table clearly shows that:

- Highest particle numbers are associated with aircraft departing from runway 27R, closest to the monitoring station. On average, Nucleation particle concentrations are 3 times higher than those seen for aircraft landing on runway 27R (departing on 27L), ~8.5 times higher than operations in easterly winds (departing on 09R) and 25 times higher than when the airport is closed overnight.
- For Aitken particles, the differences are less marked: when aircraft are departing on 27R, average concentrations are 1.7 times higher than departures on 27L, 2.5 times higher than departures on 09R and 3.9 times higher than overnight concentrations. Additionally, concentrations of Aitken particles show a significant baseline that appears to be independent of airport operating mode, confirming that emissions of Aitken particles from the airport are produced in far smaller quantities when compared to emissions of Nucleation particles.
- The proportion of Nucleation and Aitken particles in the total changes dramatically during Airport closed hours. During the day, Nucleation particles make up 70% of the average particle number concentration. Between the hours of 23:00 and 05:00, combined with much reduced total concentrations, Nucleation particles only make up approximately 40% of the total particle count. This contrasts with the day/night Nucleation component at Marylebone Road (approx. 80%, regardless of the time of day), and North Kensington (22% day, 16% night), which further highlights the difference of the environment close to the Airport.
- PM concentrations are highest during easterly winds (departing on 09R). This is certainly the influence of longer range transport of PM from London and beyond.

- BC concentrations are also highest during easterly winds, but average concentrations are higher for aircraft departing on 27R compared to landing on 27R. This is also true for UVPM.
- UVPM concentrations are elevated overnight, confirming that domestic heating is a likely additional source in the area.
- Average NOx, NO and NO_2 concentrations are all higher when aircraft depart on 27R compared to landing on 27R (departing on 27L). But high average NOx and NO concentrations are also recorded during easterly winds, reflecting the influence of emissions from London on these local measurements.

The observation that departing aircraft emit higher numbers of UFP compared to arriving aircraft appears to contrast with work by other researchers, eg. Hudda and Fruin (2016) (12), Shirmohammadi et al (2017) (19), which suggest that arriving aircraft have a significant effect on UFP concentrations directly under the flight path. However, other studies, including Keuken et al (2015) (13) have shown that elevated UFP concentrations can be attributed to airports even 40km from the airport and not under flight paths. It is therefore possible that ground level dispersion of UFP emissions from aircraft movements has not yet been fully considered in modelled and measured approaches to the assessment of UFP from aircraft and further investigation of the possible impact mechanisms is warranted.

5.4.5 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.

On average, an aircraft departs from the airport every 90 seconds between 06:00 and 23:00 every day.

(https://www.heathrow.com/file_source/Company/Static/PDF/Investorcentre/Heathrow-(SP)-FY2016-results-release-(FINAL).pdf) The SMPS/CPC configuration at LHR2 was set to provide a full particle size sweep every minute, allowing the possibility to investigate whether to uniquely assign a single measurement to an individual aircraft. Some structure in the PN measurements can be observed which bears excellent correlation to the runway operations. The plot in Figure 3 presents the stacked timeseries collected on 20 October.

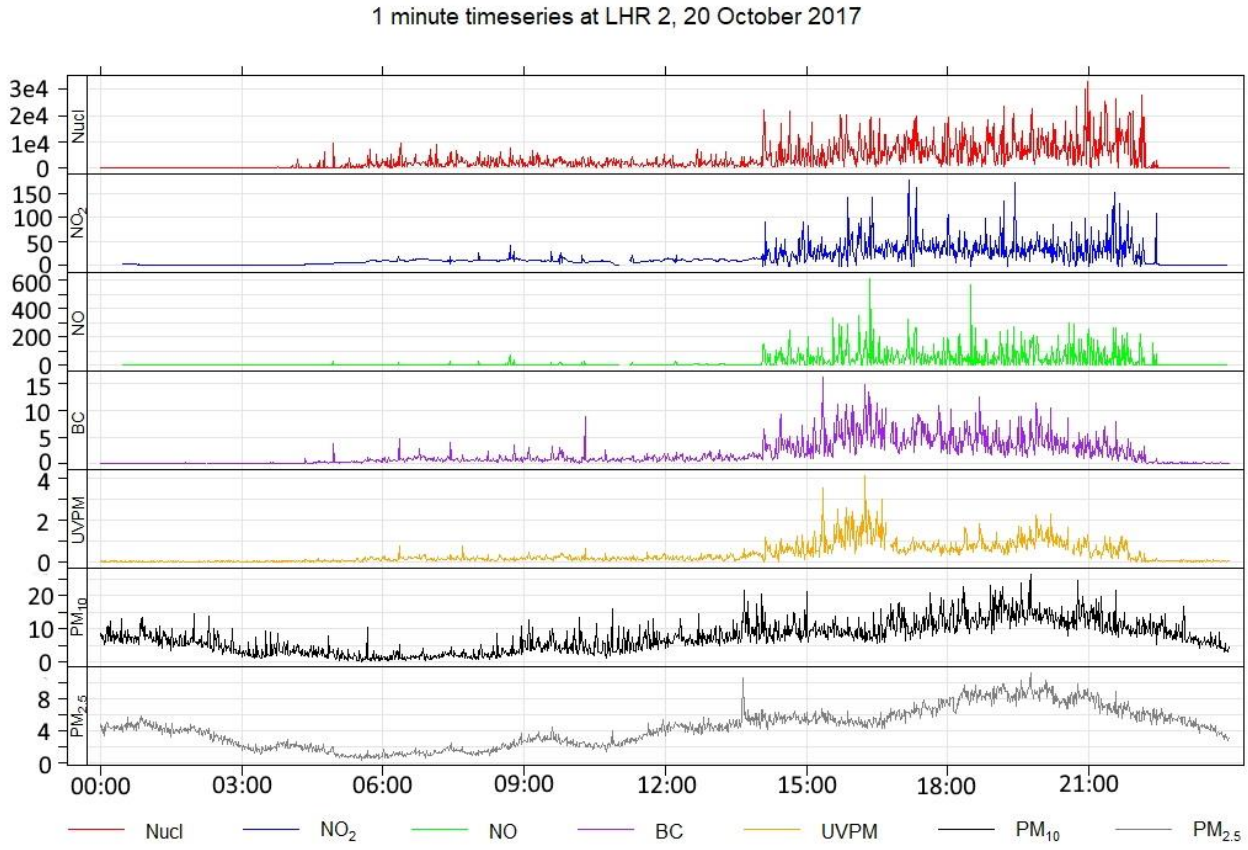

Figure 3. Stack timeseries plot, LHR2, 20 October. Nucl in #/cm^3, NO and NO_2 in ppb, BC, UVPM, PM_{10} and $PM_{2.5}$ in µg/m^3

On this day, aircraft were landing on 27R during the morning period, and departing from 27R in the afternoon period. The elevated Nucleation particle count during departures is very clear in this plot and mirrors the observations seen in the 2016 survey. The plot also shows very clear correlation of NOx, BC and UVPM with particle number concentrations, lower for arriving aircraft and higher for departing aircraft – as would be expected for the different thrust settings in these two modes of engine operation.

Correlation of the above pollutants is less obvious for PM_{10} and $PM_{2.5}$, which do not follow the abrupt change in scale when the aircraft operating mode changes. The level of detail seen in the 1 minute data allows some unexpected observations to be made. The plot in Figure 4 shows the stacked timeseries for 16 October:

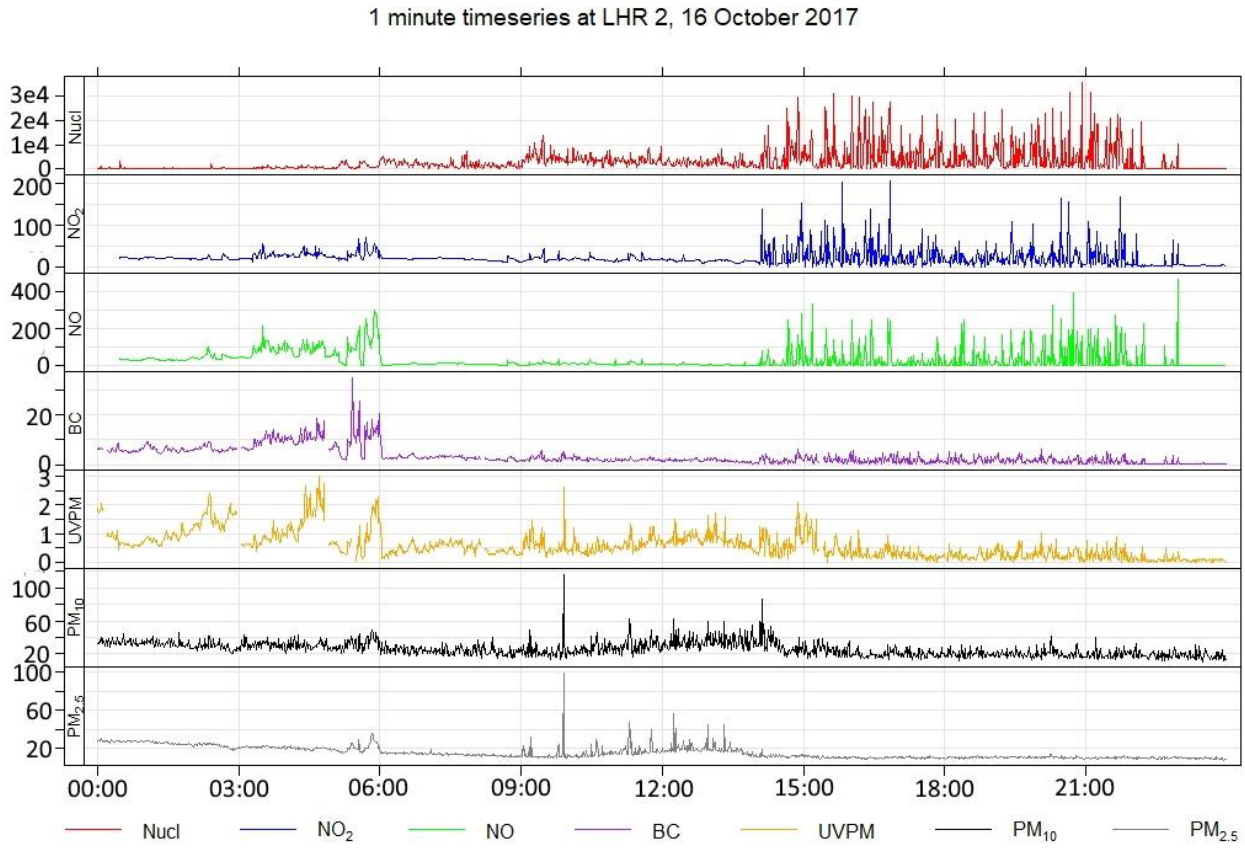

Figure 4. Stack timeseries plot, LHR2, 16 October. Nucl in #/cm^3, NO and NO_2 in ppb, BC, UVPM, PM_{10} and $PM_{2.5}$ in µg/m^3

Aircraft movements on 16 October followed the same pattern as 20 October and the trends between pollutants is, by and large, similar. Closer inspection reveals some subtle differences:

- PM_{10} and $PM_{2.5}$ concentrations are higher during the morning arrival mode than the afternoon departure mode.
- BC and UVPM concentrations do not follow each other at all throughout the airport operating day. Prior to 06:00, the agreement between them is reasonable, though the

effect of non-aircraft sources (e.g. overnight domestic heating) is observable in the data. UVPM and BC relationships change on the two days, indicating that the sources might be different: concentrations are higher on the 16th (averages 0.65 µg/m^3 16th, 0.24 µg/m^3 20th) between 09:00 and 15:00 (during arrivals), compared to concentrations after 15:00 (averages 0.28 µg/m^3 16th, 0.78 µg/m^3 20th). BC concentrations on the 20th between 09:00 and 15:00 (averages 1.97 µg/m^3 16th, 1.38 µg/m^3 20th) are lower than measurements after 15:00 (averages 1.64 µg/m^3 16th, 3.98 µg/m^3 20th). On examination of the meteorology between 09:00 and 15:00, recorded wind directions were between 170 and 220 degrees. This is the sector where air sampling captures the point where the majority of aircraft touchdown on the runway, leading to the possibility that tyre smoke from landing aircraft was transported from the runway and measured at the station during this period. Tyre smoke from landing aircraft is a blue-grey colour and likely to be in the fine particle range, as the tyres are subjected to great stress from the acceleration and weight of the aircraft. The correlation between $PM_{2.5}$ and UVPM, together with the absence of correlation with NOx and Nucleation particles, associated with exhaust emissions, further supports this observation. Examination of the UVPM/BC ratios on 16th and 20th October (Figure S41) shows key differences during the afternoons, further supporting the likelihood of different sources.

- Winds from 0:00 to 06:00 originated from the north east and east of the monitoring station, suggesting off-airport emissions contributed to the elevated levels of NOx, BC and UVPM during this period. The NOx, BC and UVPM measurements at LHR2 are very similar to measurements made at other monitoring stations in the area.

The SMPS/CPC setup provides detailed information about the PSD every minute. This detail is not necessary for analysis, as the breakdown into nucleation and Aitken particle number

concentrations demonstrates how the particle size distribution is dominated by fine particles. For completeness, an animation of the 1 minute PSD data from 20 October is provided in SI Animation S1. This animation clearly shows three distinct modes:

- Period when aircraft are not operating (0:00 to 06:00 and 23:00 to 0:00)
- Period where aircraft are landing (06:00 to 14:00)
- Period where aircraft are departing (14:00 to 23:00)

The animation also shows just how dependent the measurements are on aircraft movements. There are many periods of both high emissions, associated with aircraft, and relatively "quiet" periods, coinciding with reduced aircraft activity. This is the first time that we are aware of that airport UFP measurements have been reported in this way, clearly illustrating the nature and effect of the aircraft activity.

5.4.6 Correlation of UFP with aircraft movements

As part of normal airport operation, Heathrow Airport Limited keep a log of all aircraft ground movements. Records of aircraft type, time of departure or arrival and the relevant runway used were provided at 1 minute resolution. This allows analysis of Nucleation mode particles to be closely associated with exhaust plumes by tying together aircraft location, wind speed and direction, time taken for the plume to arrive at the measurement station and the associated pollution data. By knowing what aircraft is being measured, clustering of Nucleation particle concentrations by aircraft type is also possible.

For the purposes of this investigation, only aircraft departing on 27R were examined, and only when winds were from the 105 to 265 degree sector – i.e. when the exhaust plume would be transported to the monitoring station. Reviewing the timeseries data for the entire survey, there were a selection of days when concentrations of Nucleation particles were highest, providing the strongest potential to assign peak concentrations to individual aircraft. The

comparison was therefore further restricted to include only departures on 9-16, 19-21, 23 and 31 October 2017.

During this time, 5127 aircraft departed from Runway 27R, clustered into the following groups:

Aircraft type	Number of aircraft	Number of aircraft successfully identified	Percentage of total successfully identified
Airbus A31x / A32x series	2408	1188	49%
Airbus A33x series	191	113	59%
Airbus A34x series	72	52	72%
Airbus A35x series	42	21	50%
Airbus A380 series	315	200	63%
Boeing 737 series	137	69	50%
Boeing 747 series	308	202	66%
Boeing 757 series	39	15	38%
Boeing 767 series	307	196	64%
Boeing 777 series	732	477	66%
Boeing 787 series	442	274	62%
Others	134	70	52%
Total	5127	2877	56%

Table 2 – Departing aircraft on Runway 27R, separated by type, on selected days in October 2017. "successfully identified" represents the number of aircraft where measured nucleation concentrations were elevated above the prevailing background concentrations at the expected arrival time of the plume at the monitoring station.

Initial review of the assignment of peaks revealed that a large number of departures were poorly identified by the analysers. A higher proportion of heavier aircraft, with an expected higher fuel use during takeoff, are successfully identified when compared to lighter aircraft (for example 72% of all A340 aircraft were identified, vs 49% of all A31x/A32x). By way of example of the problem of identification, Figure 5 shows Nucleation particle concentrations over a one hour period on one day.

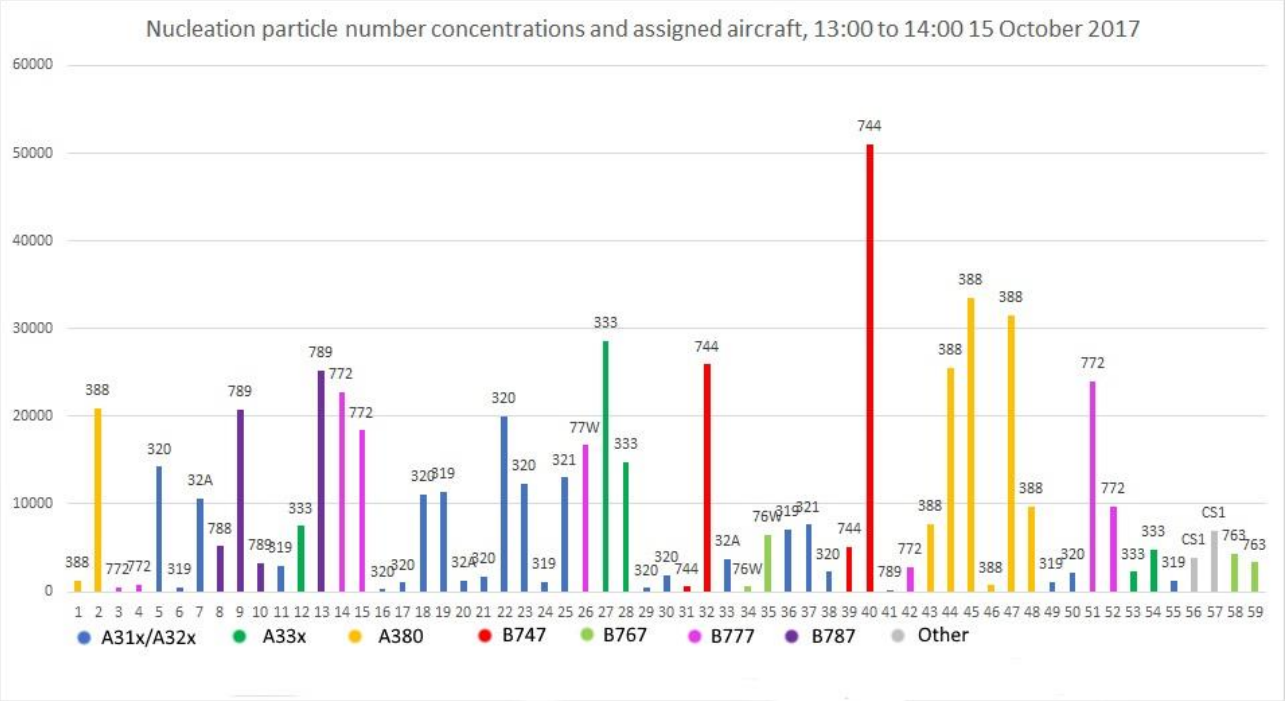

Figure 5 – Nucleation concentrations (#/cm^3) recorded for aircraft departures, 13-14:00 15 October 2017. The aircraft type is labelled at the top of each bar, and colour coded according to the legend.

It is clear from this plot that a significant number of nucleation peaks are very low when compared to other similar aircraft. There are a number of possible reasons for this:

- The departure time of the aircraft is reported at the start of the minute the aircraft "throttles up". It was not possible to determine the exact position of the aircraft on the runway, so assumptions are made about when the emission plume will arrive at the monitoring station.
- High time resolution meteorological data was not available for this survey. All calculations for plume transportation were made using 15 minute averaged wind speed and direction data
- The dataset has not been screened for rainfall. A proportion of plumes will have been negatively impacted during periods of rainfall, but high resolution rainfall data was not available to identify and filter out these periods.

- The 1 minute scan of the SMPS from 6 to 100 nm means that if the exhaust plume arrived at the monitoring station midway through the scan, it is possible that the SMPS would miss the Nucleation particles completely from a departing aircraft.
- It is also possible that, when wind direction was closer to 260 degrees, that the plume from a departing aircraft would be detectable for a longer period, due to the increased distance from the monitoring station, leading to the possibility that the SMPS would record a single aircraft plume over multiple minutes.

Because of the number of mis-assigned plumes, the data were further screened by rejecting identifications when Nucleation particle number concentrations were lower than 4000/cm^3. Using this restriction, 44% of the departures were removed from the analysis. The table below summarises the results from these screened identifications.

Aircraft type	# Aircraft assessed	Nucleation particles, #/cm^3 / RSD, %	BC, µg/m^3	UVPM, µg/m^3	NO, ppb	NO_2, ppb	PM_{10}, µg/m^3	$PM_{2.5}$, µg/m^3
Airbus A31x / A32x series	1188	8060 / 47%	4.18	0.80	42.1	27.9	13.1	8.5
Airbus A33x series	113	11438 / 59%	3.87	0.64	85.0	41.8	12.4	8.0
Airbus A34x series	52	10859 / 60%	4.50	0.51	91.0	31.2	12.4	8.0
Airbus A35x series	21	12266 / 45%	3.34	0.54	99.7	39.9	11.4	7.0
Airbus A380 series	199	13578 / 64%	3.98	0.66	107.9	34.7	13.2	8.3
Boeing 737 series	69	7719 / 46%	3.57	0.73	39.4	24.8	14.3	9.2
Boeing 747 series	202	12734 / 63%	4.12	0.72	95.6	40.0	13.0	8.1
Boeing 757 series	15	7063 / 45%	4.80	0.67	70.9	32.3	13.9	8.9
Boeing 767 series	196	10438 / 57%	4.59	0.79	84.0	38.6	12.7	8.4
Boeing 777 series	477	12422 / 56%	3.56	0.69	112.3	38.3	12.7	8.3
Boeing 787 series	274	12406 / 56%	3.14	0.64	84.1	35.1	12.3	7.9
Others	70	8078 / 47%	3.31	0.72	36.2	26.0	12.9	8.6
All departures	**2876**	**10266**	**3.94**	**0.73**	**71.9**	**33.0**	**12.9**	**8.4**

Table 3 – Summary of average concentrations, separated by aircraft type, screened for Nucleation particle measurements greater than 4000 particles / cm^3. The relative standard deviation (RSD) for nucleation particles is presented to demonstrate the wide variation in the measurements recorded.

The Nucleation particle number data are further assessed in the box and whisker plot below:

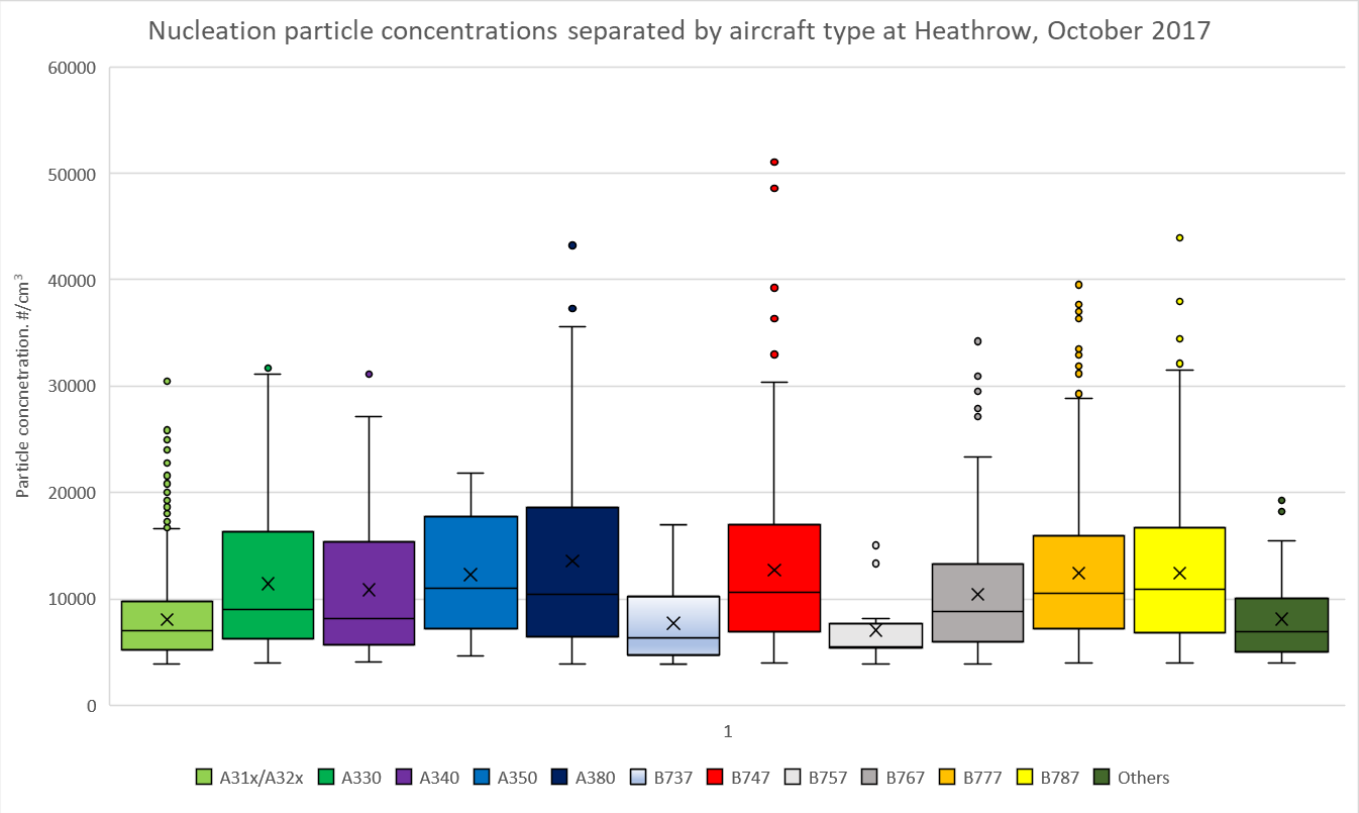

Figure 6 – Box and whisker plot, separating nucleation measurements by individual aircraft type. Average concentrations are represented by a X, median by a line within the box. The box upper and lower limits represent the 25th and 75th percentiles while the whiskers present the 0 and 100% boundaries. Note that the lower whisker ignores data screened by rejecting all results below 4000 particles/cm^3. Outliers are represented by individual dots.

Bearing in mind the varying sample sizes for each aircraft type, the average data in the above table and figure confirms the following:

- Smaller aircraft emit fewer nucleation particles than larger aircraft
- Total NOx concentrations are highest from largest aircraft
- NO_2 concentrations follow a similar pattern to NOx – larger aircraft generally emit higher concentrations than smaller aircraft, though it is likely that the newer fleet of heavy aircraft have lower NO_2 emissions – measured NO_2 from Boeing 747 aircraft is higher than Airbus A380 aircraft, for example. More investigation is required to get a fuller understanding of this observation.

- There is no clear trend in the BC data. Average BC emissions from the Boeing 787 were the lowest recorded for any aircraft type, suggesting that the newer design of this engine may be better for these emissions.
- There is no clear trend in the UVPM data, though average measurements appear to be lower for newer aircraft types – Boeing 787 vs Boeing 767 for example.
- Measurements of PM_{10} and $PM_{2.5}$ appear to be completely independent of aircraft type. Average PM_{10} concentrations recorded during departures of Boeing 747, Airbus A380 and 31x/32x aircraft are essentially identical. This further confirms that background mass concentrations of PM dominate measurements – any additional contribution from aircraft is not likely to be significant.

Further investigation of the nucleation particle count data for each aircraft group was undertaken, using a simple correction to normalise the measurements with respect to wind speed. The data were examined before and after correction, using both the relationship between relative standard deviations within aircraft type and exploring the ratio of average concentrations between aircraft types. Unfortunately, a systematic improvement in the relative standard deviation of each clustered group was not observed, suggesting that the relationship between emissions from aircraft and measured downwind concentrations is more complex than a simple adjustment for one parameter.

5.4.7 Comparison of 2017 particle size distribution with 2016 dataset

A similar study to investigate UFP at the airport was undertaken at Heathrow in the autumn of 2016 by Stacey et al (2020) (21), in direct comparison with other monitoring in the south east of England. The 2016 study configured the TSI SMPS/CPC identically to the comparator monitoring stations, the 2017 study investigated a smaller particle range at a faster time resolution to identify individual aircraft UFP contribution.

There were significant differences identified in the particle distributions and counts between the two datasets, which were a direct consequence of the differences in how the analyser was configured for each campaign. As a result, direct comparison between the 2016 and 2017 data is not possible; this is discussed further in the SI.

5.5 Conclusions

An extensive campaign to monitor UFP at London Heathrow Airport was undertaken in the autumn of 2017. The primary objective was to examine high temporal resolution data to investigate the relationship between individual aircraft and measured concentrations of UFP, PM_{10}, $PM_{2.5}$, NOx and BC.

The SMPS analyser was specifically configured for fast response (1 minute scans) and within a much smaller size range (6-100 nm particles) than in our 2016 campaign. This change in configuration caused a shift in measurements, both in magnitude and peak particle size, meaning that comparison with historic and current UFP data in the UK was impossible.

This study, within 170 metres of a busy runway, shows that nucleation mode particles predominantly originate from the airport, with highest concentrations associated with departing aircraft. This observation is in contrast with some other research, which suggests that UFP concentrations downwind of airports is dominated by aircraft emissions being transported to ground level by wing tip vortices from arriving aircraft.

There is some correlation of nucleation particles with NOx and BC, and these pollutants, together with PM and Aitken particles, also show strong associations with winds from off-airport directions, not associated with nucleation particles. There is some evidence that BC emissions from landing aircraft is higher in UV-active BC, most likely as a result of tyre abrasion upon landing.

Analysis of the 1 minute measurement data with associated aircraft departure information was used to group the data by aircraft type. Larger aircraft departing from the runway recorded higher measurements of nucleation particles and NOx compared to smaller aircraft, but emissions of BC, UVPM and NO_2 appear to be more dependent upon the age of the engine design, rather than the size of the aircraft.

5.6 Data availability

Data supporting this publication are openly available from the UBIRA eData repository at https://doi.org/10.25500/00000535

5.7 Acknowledgements

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CRediT author statement

Brian Stacey: Conceptualisation, methodology, software, validation, formal analysis, investigation, resources, data curation, writing – original draft, writing – review and editing, visualisation, project administration

Roy Harrison: Supervision, writing – review and editing

Francis Pope: Supervision, writing – review and editing

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5.9 Appendix update

A number of amendments have been made to the published paper. These are deemed to be minor in nature and do not require the original papers to be updated. The changes are as follows:

Section	Amendment
5.3.2	Clarified that a PC was used to collect data from both SMPS systems
5.4.1	Clarified visual correlation of datasets is poor, confirmed by calculating r^2
5.4.2	Described the maintenance depot next to the station in further detail
5.4.3	Added observation and description of day vs night Nucleation/Aitken concentrations at LHR2, MRD and N Ken
5.4.5	Added measurement units to the descriptions of Figure 3, 4 and 5
5.4.5	Added detail around BC:UVPM differences on 16 and 20 Oct (also Fig S41)

5.10 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.

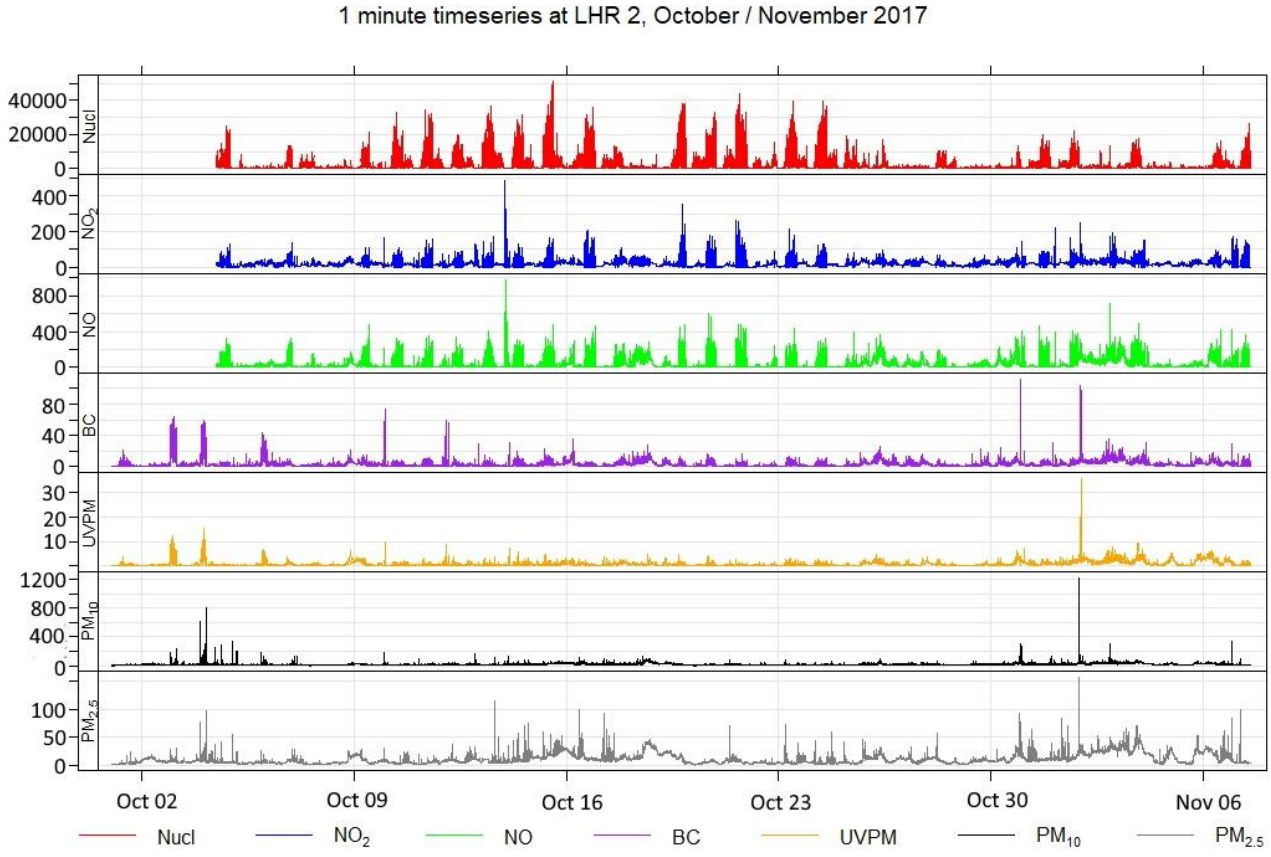

Figure S1 – timeseries plot of 1 minute measurement data at LHR2, 2 October to November 2017. Reported concentrations are ppb for NO and NO_2, µg/m^3 for PM_{10}, $PM_{2.5}$, BC and UVPM, and particles/cm^3 for nucleation particles (labelled Nucl on the plot). The variable nature of the measurements and dependence of concentrations on wind direction is evident in this plot.

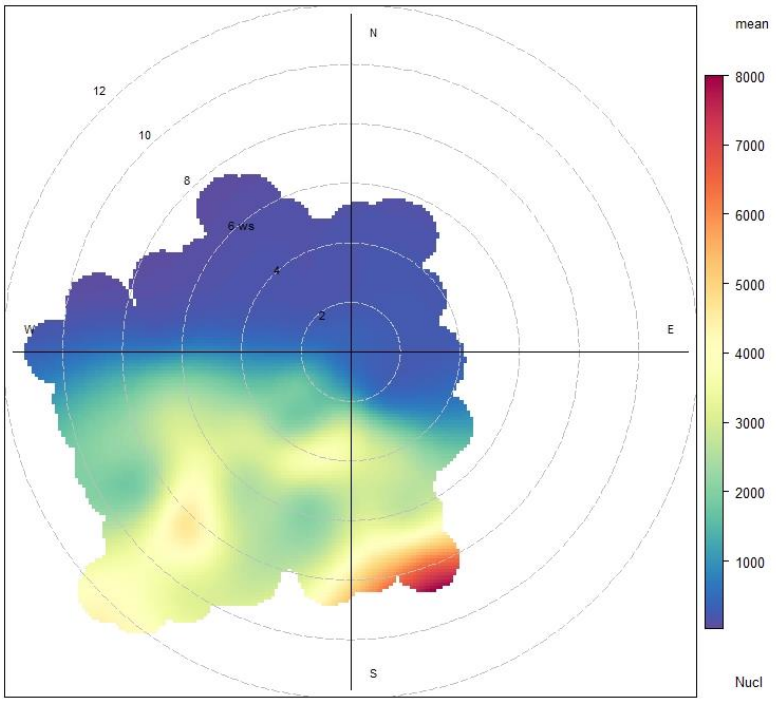

Figure S2 – Polar plot of average nucleation particle concentrations (#/cm^3) at LHR2

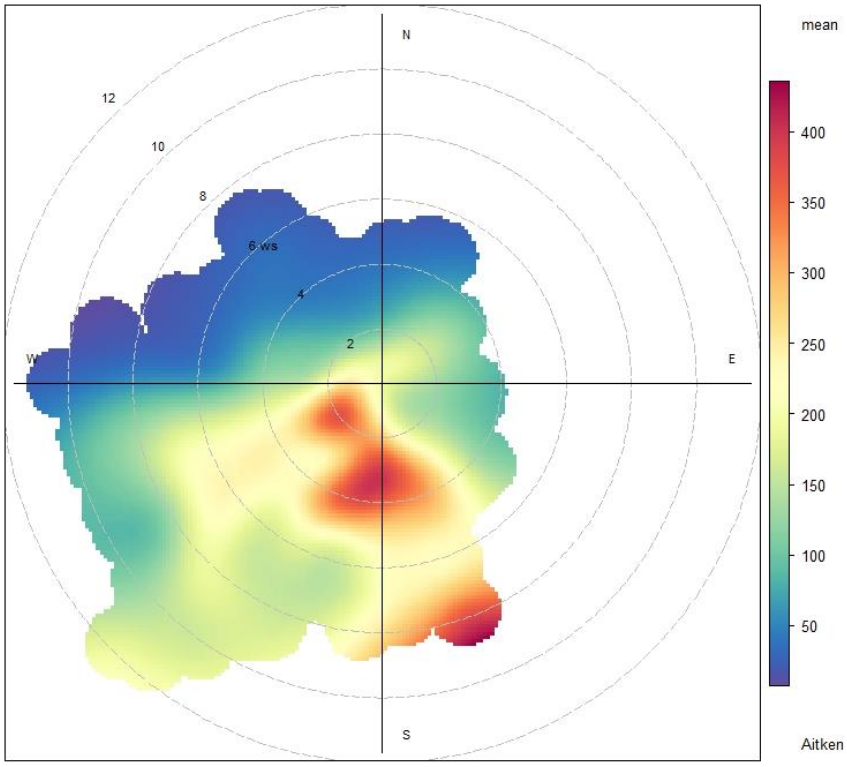

Figure S3 – Polar plot of average Aitken particle concentrations (#/cm^3) at LHR2

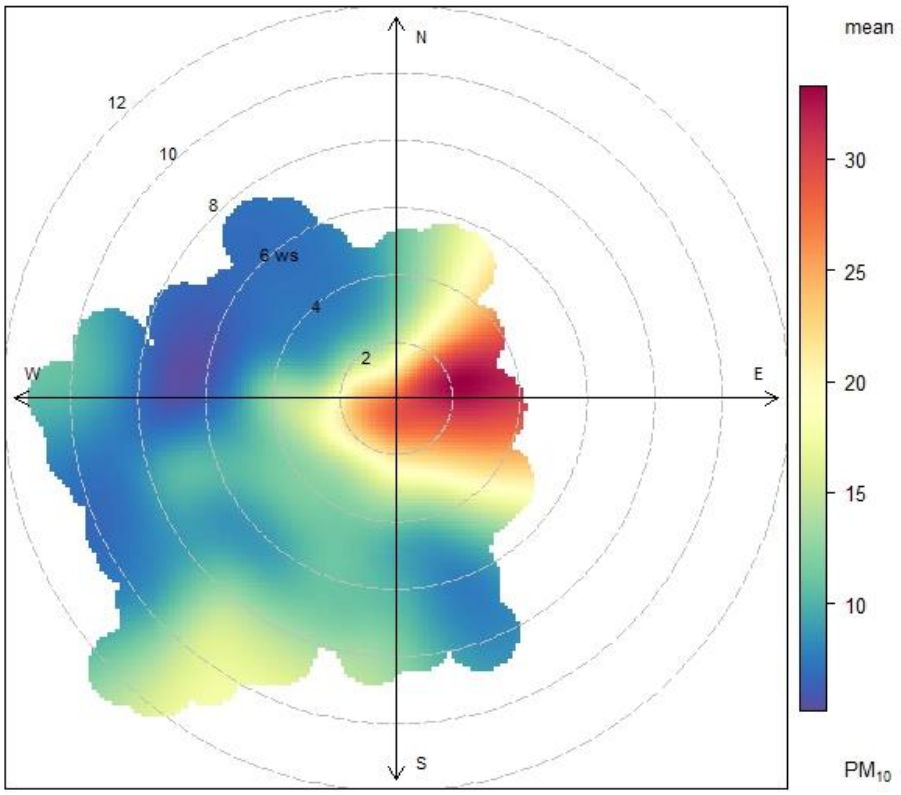

Figure S4 – Polar plot of average PM_{10} concentrations ($\mu g/m^3$) at LHR2

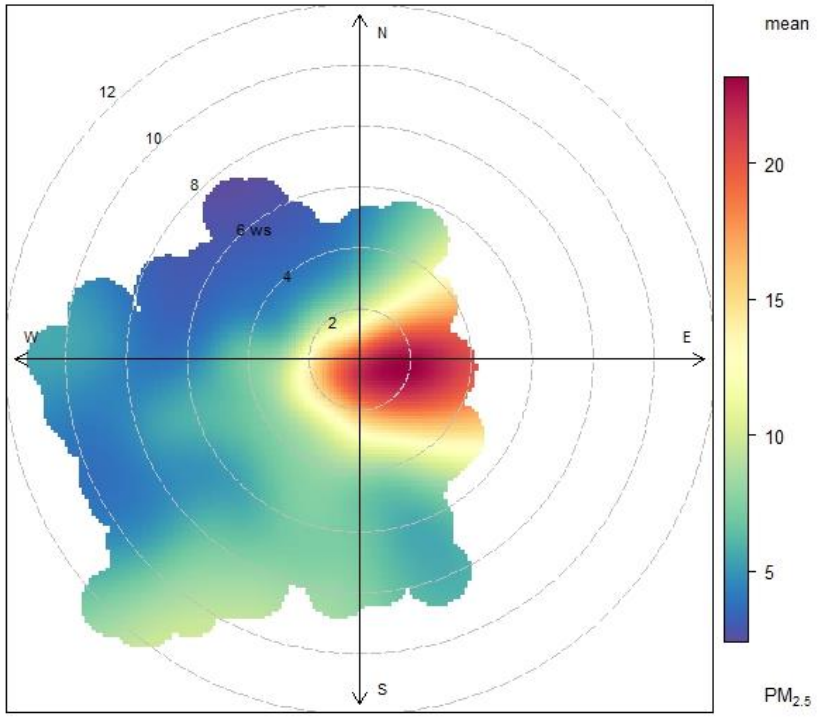

Figure S5 – Polar plot of average $PM_{2.5}$ concentrations ($\mu g/m^3$) at LHR2

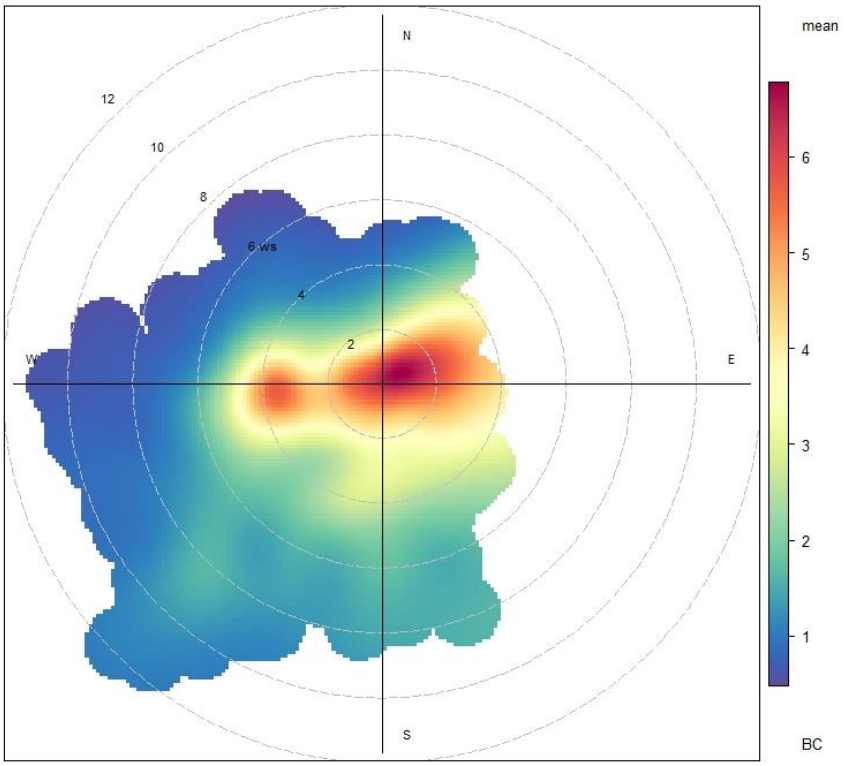

Figure S6 – Polar plot of average BC concentrations (µg/m^3) at LHR2

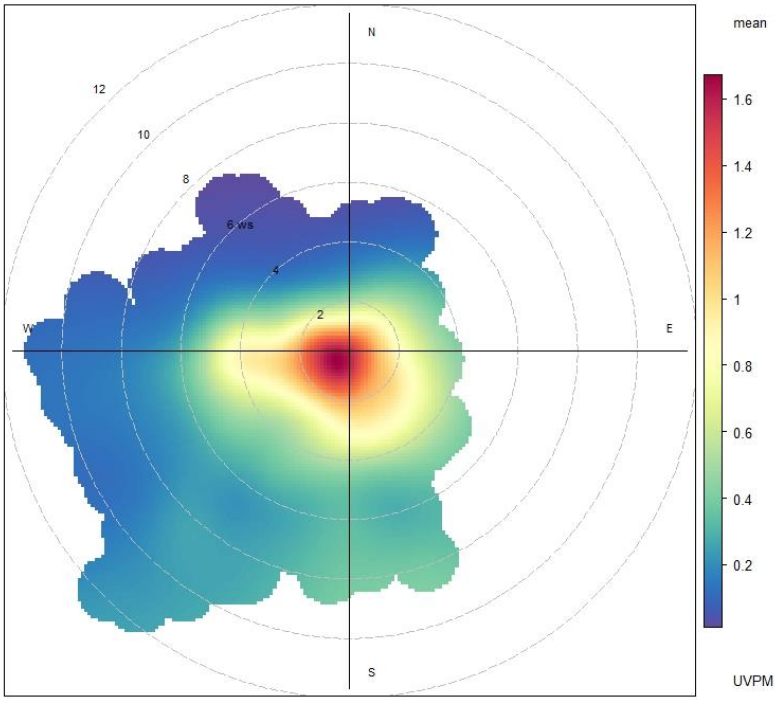

Figure S7 – Polar plot of average UVPM concentrations (µg/m^3) at LHR2

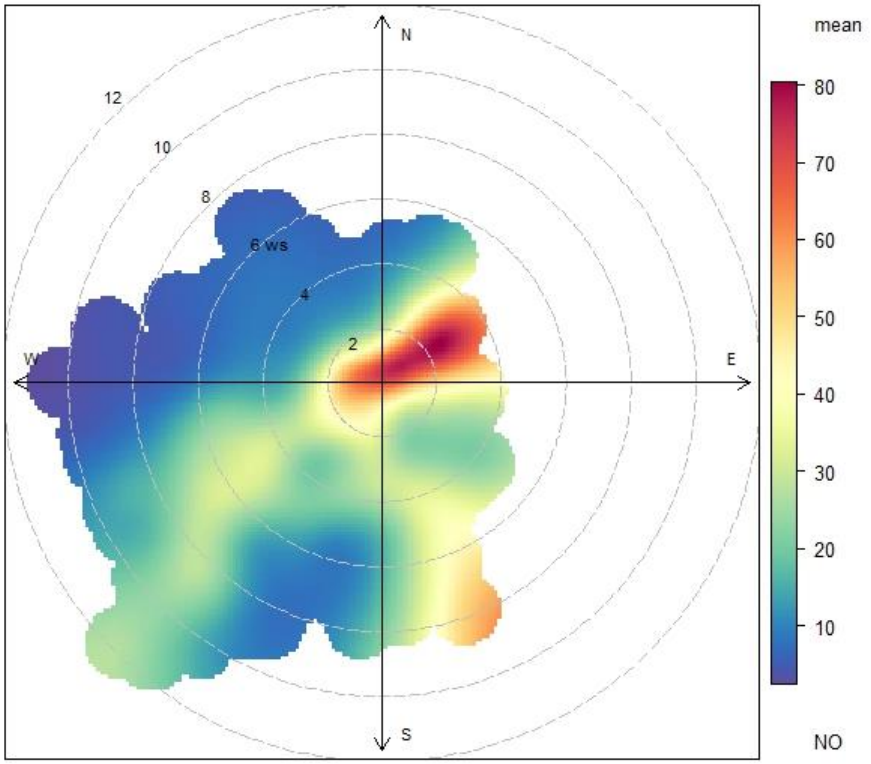

Figure S8 – Polar plot of average NO concentrations (ppb) at LHR2

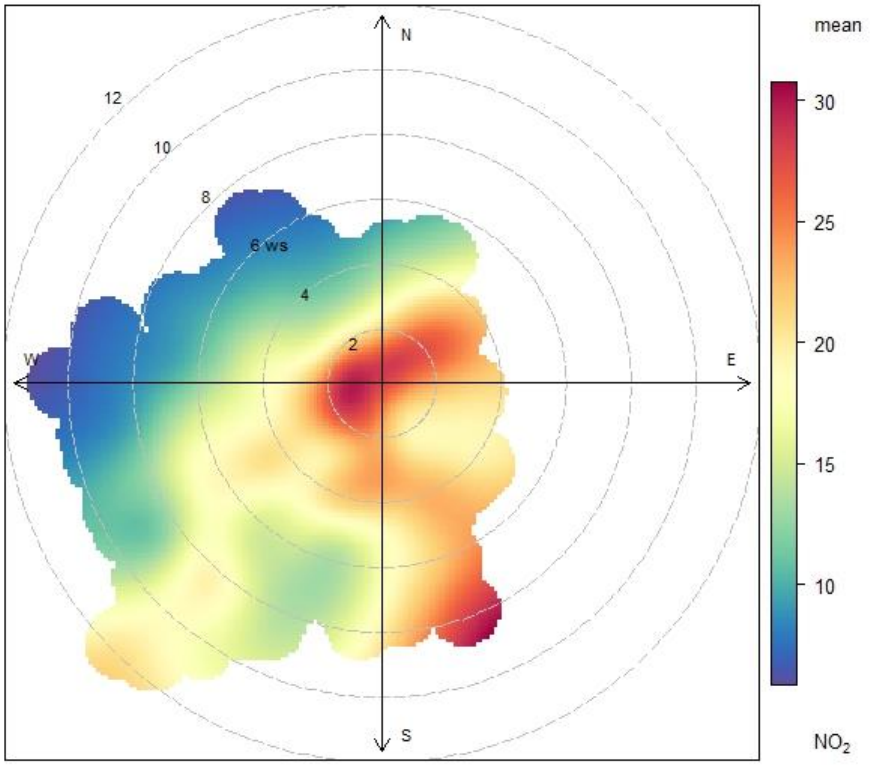

Figure S9 – Polar plot of average NO_2 concentrations (ppb) at LHR2

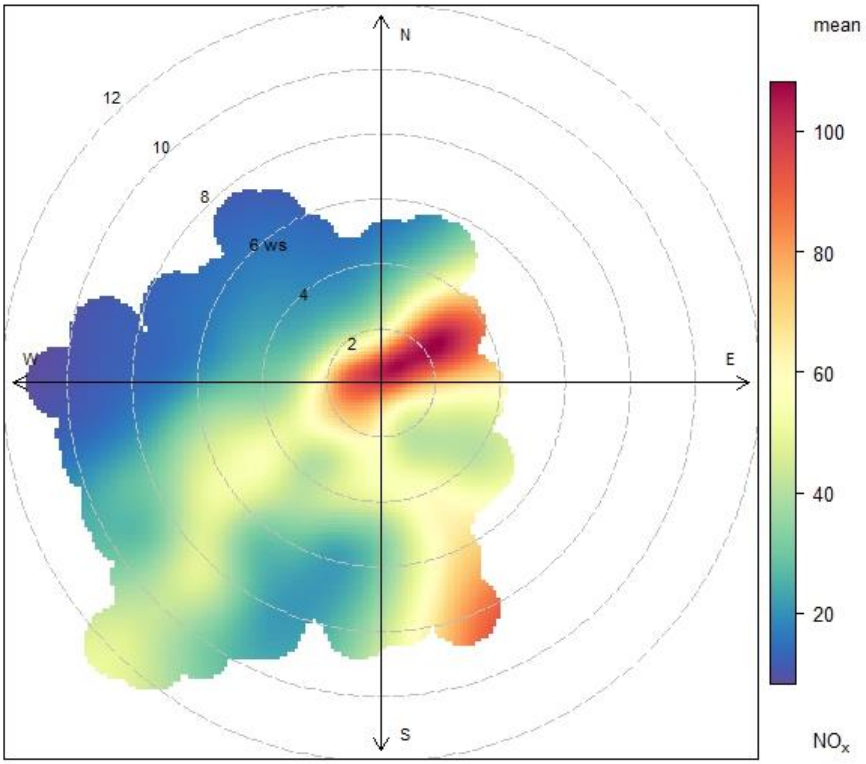

Figure S10 – Polar plot of average NOx concentrations (ppb) at LHR2

Figures S2 to S10 show the dependency of measured concentrations on wind speed and wind direction. Nucleation particles are almost exclusively associated with winds from the airport. Aitken particles follow a similar pattern, but with influences associated with other wind directions. Highest PM_{10} and $PM_{2.5}$ concentrations are not significantly associated with winds from the airport, instead mirroring the pattern seen at many other monitoring stations in the south east of the UK. BC and UVPM show some influence from winds from the airport, but highest concentrations are associated with very light winds. The NO, NO_2 and total NOx polar plots show some airport-related influence, but highest average concentrations are associated with off-airport sources.

Figure S11 shows local construction activity during the measurement campaign. Works were only active from 23:00 to 04:00 and may have influenced these polar plots.

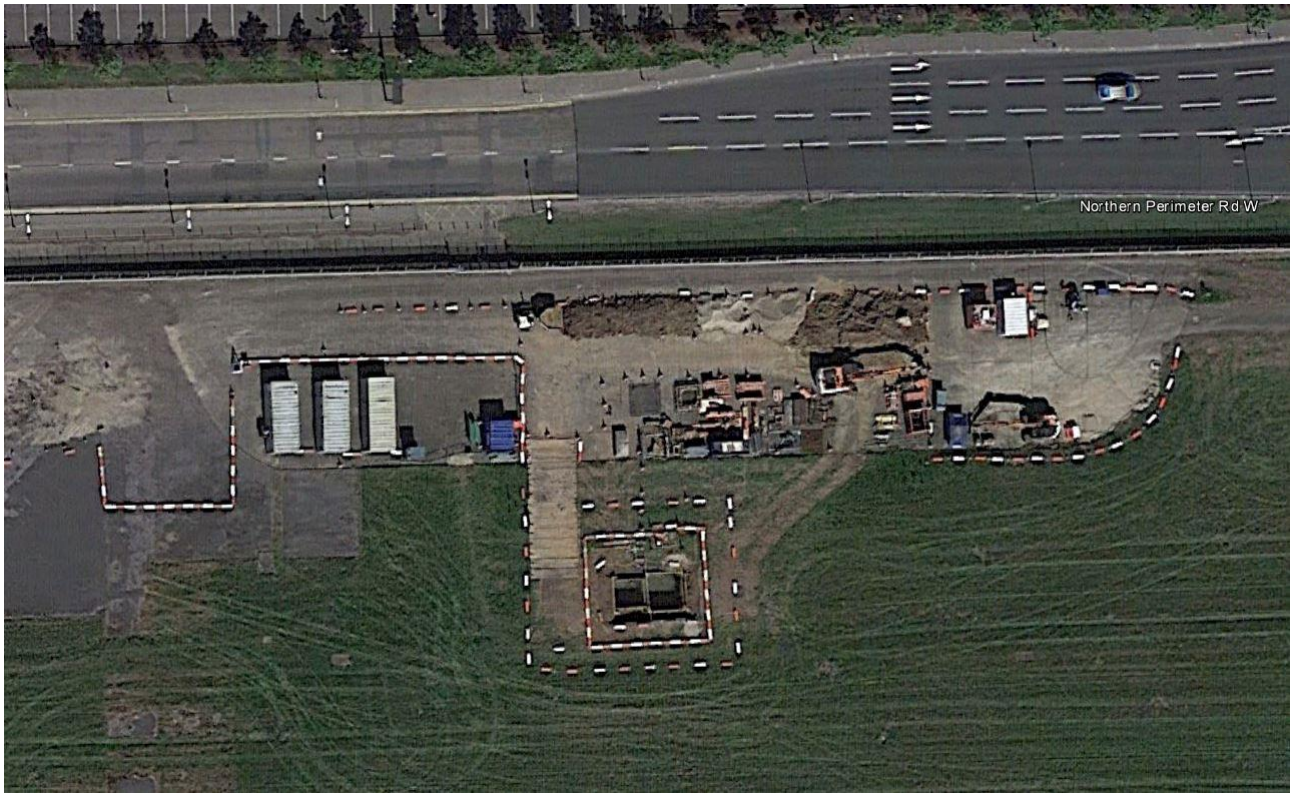

Figure S11 – Construction activity around LHR2 in October 2017. Vehicles were only active between 23:00 and 04:00 each day

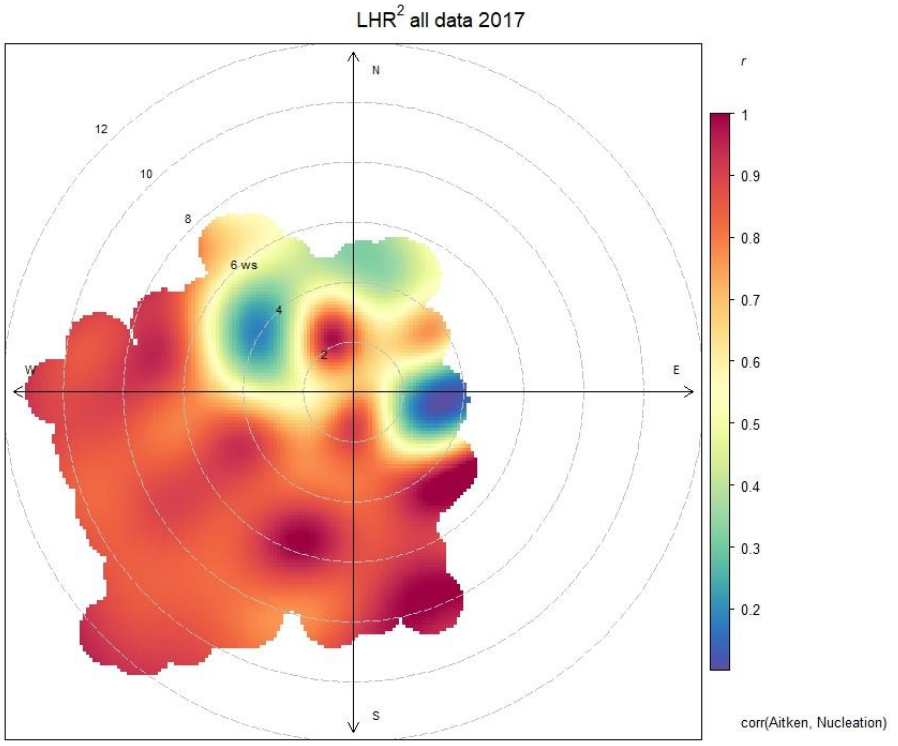

Figure S12 – Bivariate polar plot showing correlation between nucleation and Aitken particles. Highest correlation between pollutants are shown in dark red.

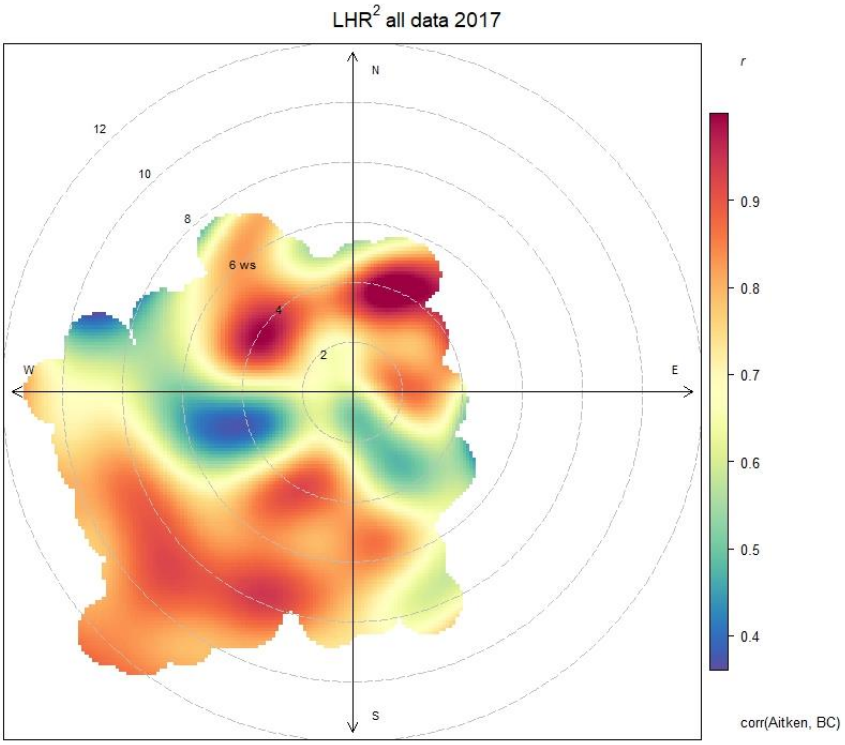

Figure S13 – Bivariate polar plot showing correlation between BC and Aitken particles. Highest correlation between pollutants are shown in dark red.

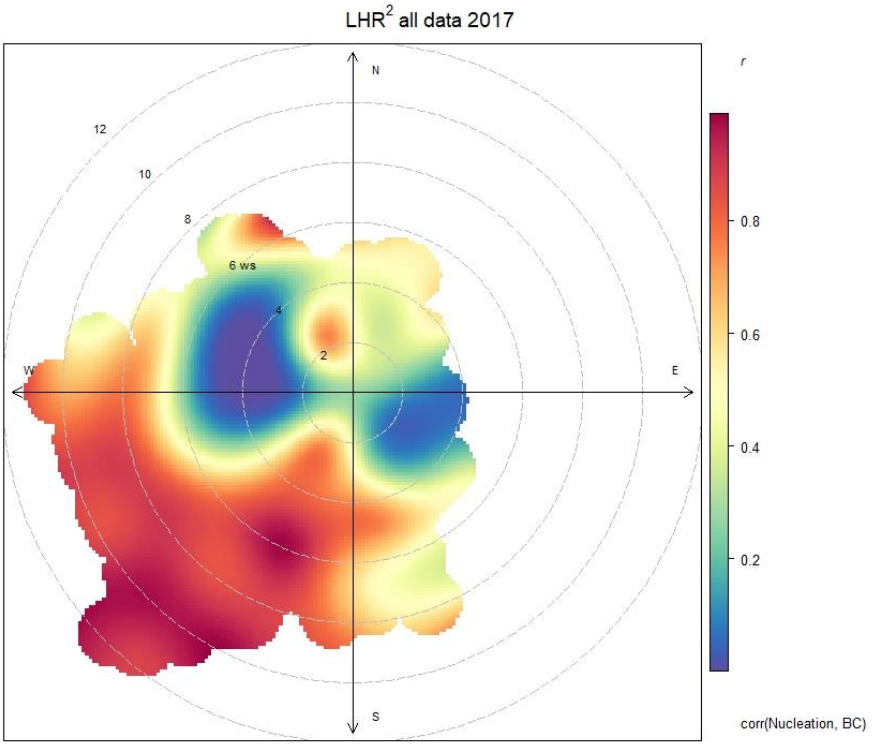

Figure S14 – Bivariate polar plot showing correlation between nucleation and BC particles. Highest correlation between pollutants are shown in dark red.

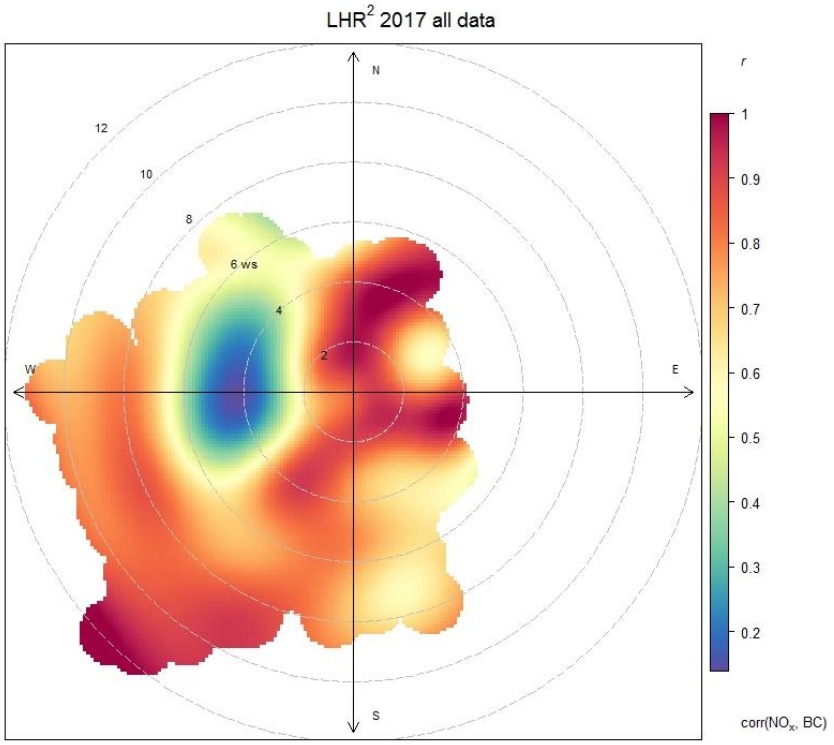

Figure S15 – Bivariate polar plot showing correlation between NOx and BC particles. Highest correlation between pollutants are shown in dark red.

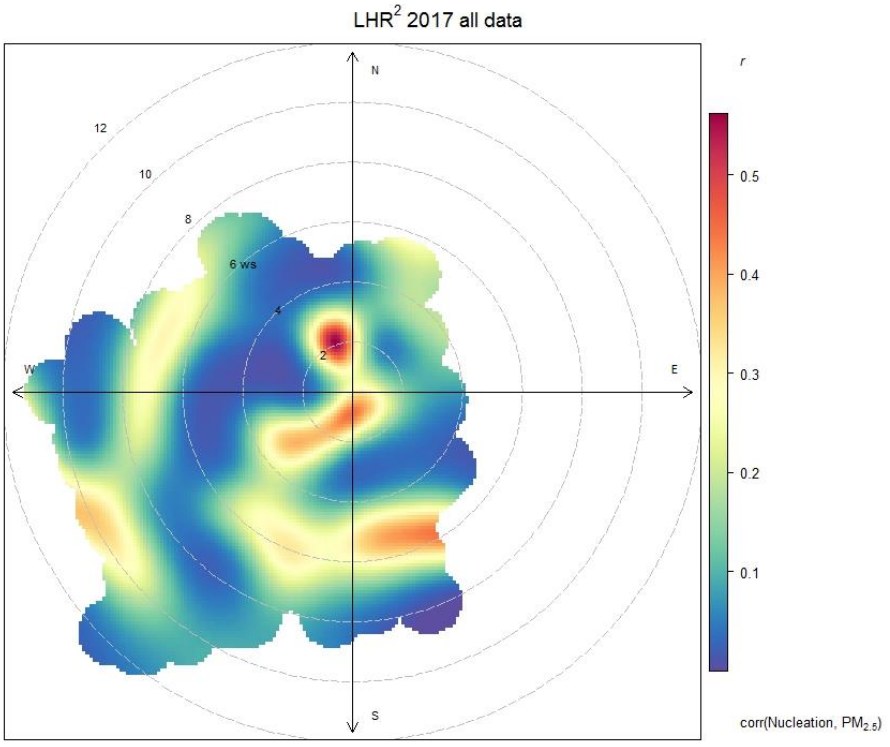

Figure S16 – Bivariate polar plot showing correlation between nucleation and $PM_{2.5}$ particles. Highest correlation between pollutants are shown in dark red.

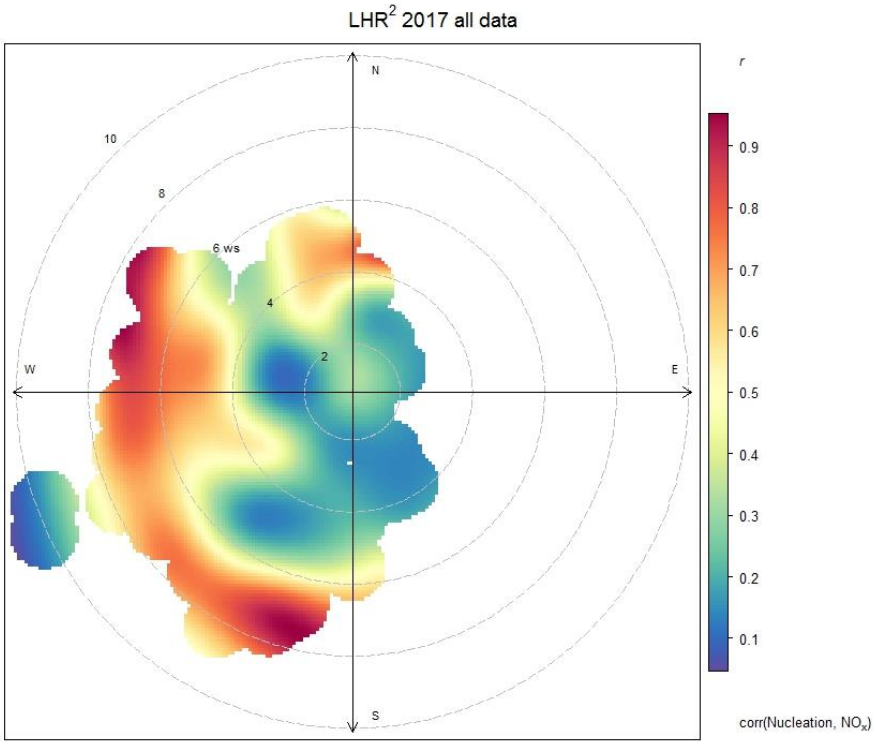

Figure S17 – Bivariate polar plot showing correlation between nucleation particles and NOx. Highest correlation between pollutants are shown in dark red.

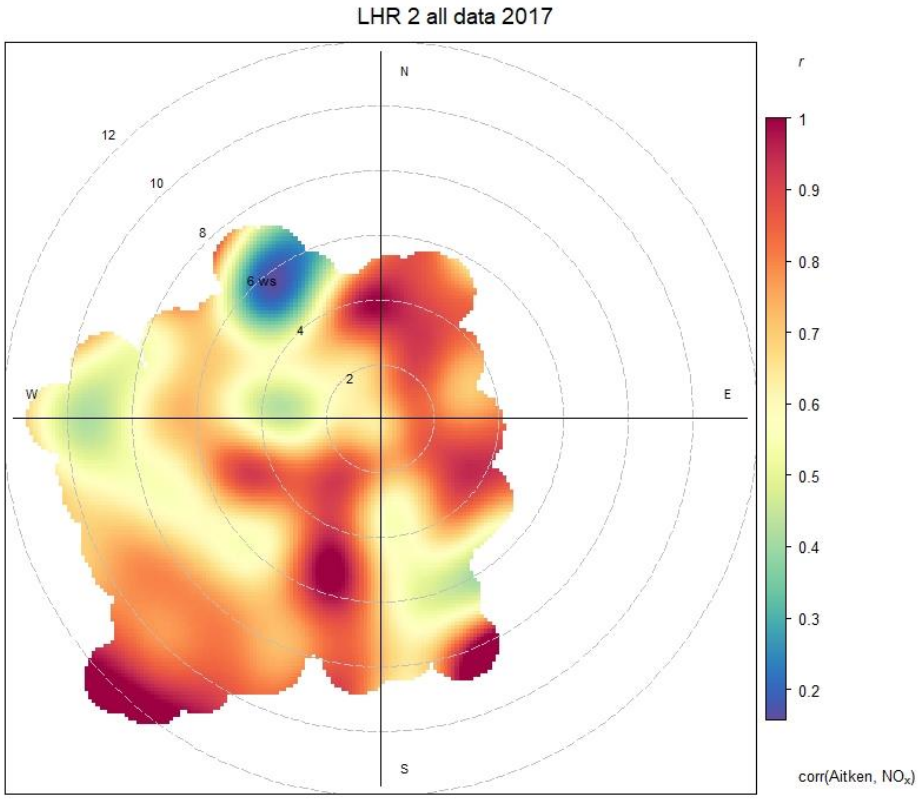

Figure S18 – Bivariate polar plot showing correlation between Aitken particles and NOx. Highest correlation between pollutants are shown in dark red.

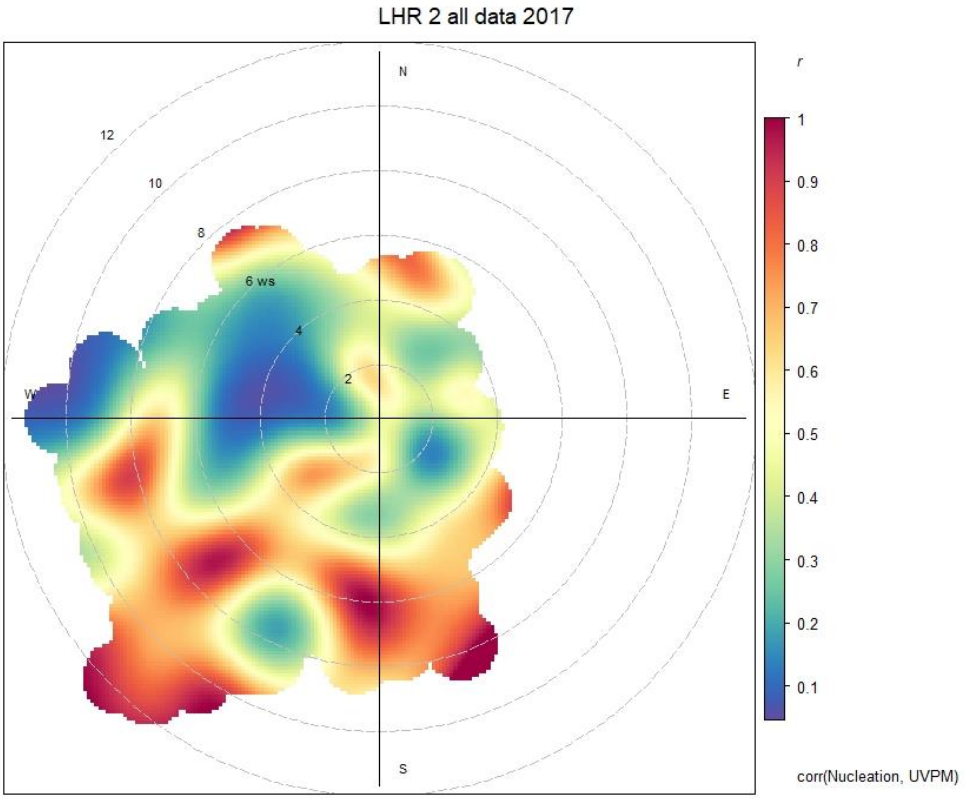

Figure S19 – Bivariate polar plot showing correlation between nucleation and UVPM particles. Highest correlation between pollutants are shown in dark red.

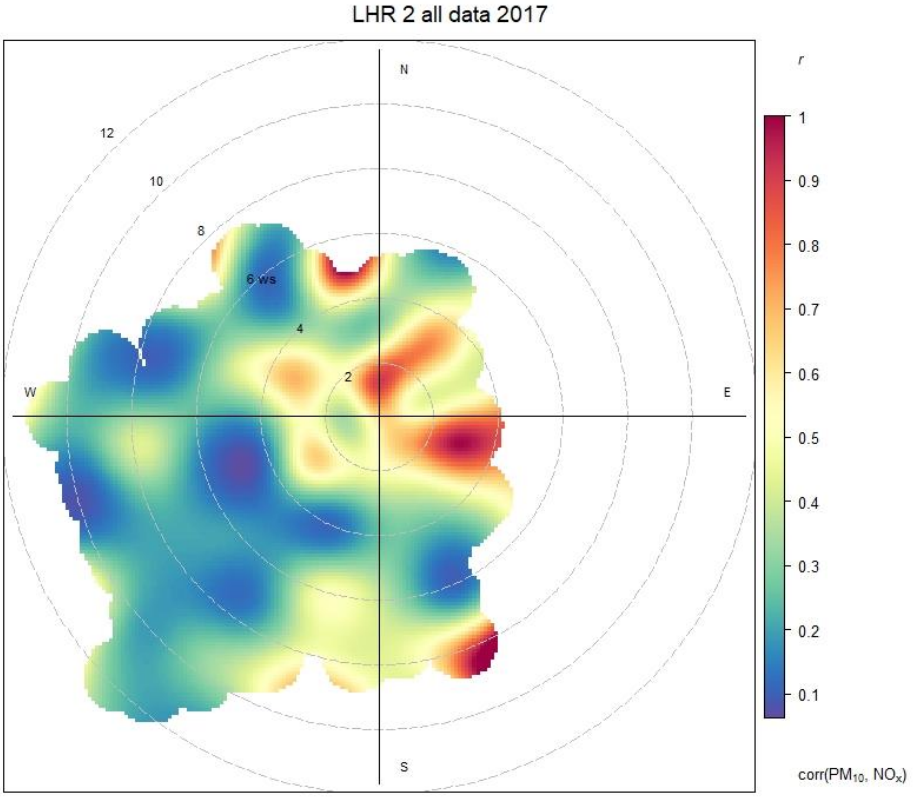

Figure S20 – Bivariate polar plot showing correlation between PM_{10} and NOx. Highest correlation between pollutants are shown in dark red.

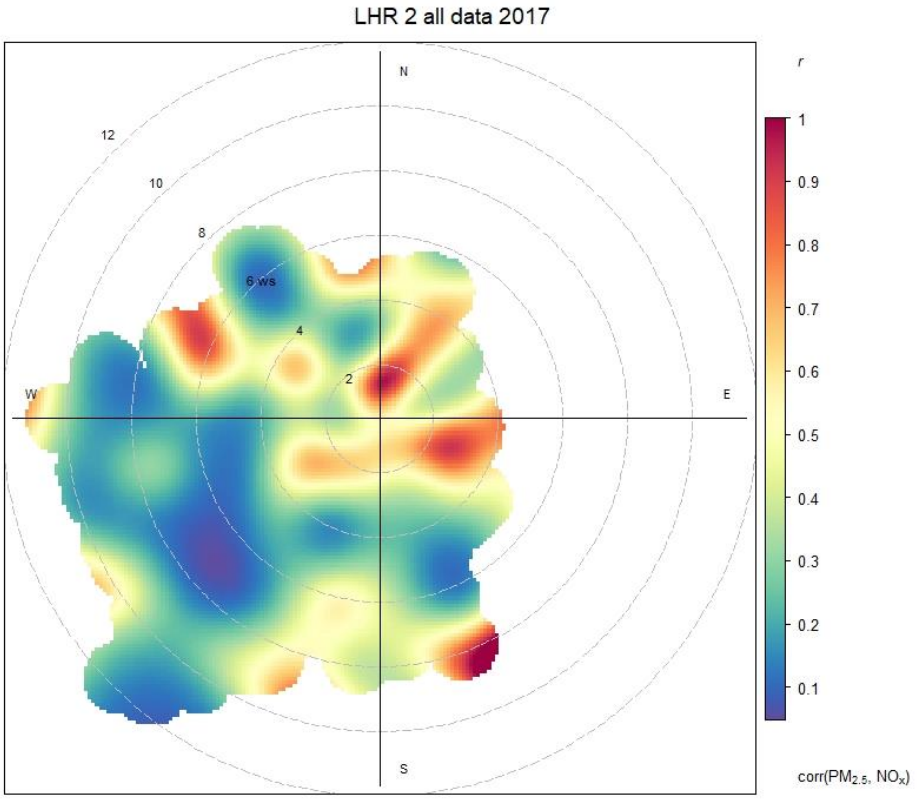

Figure S21 – Bivariate polar plot showing correlation between $PM_{2.5}$ and NOx. Highest correlation between pollutants are shown in dark red.

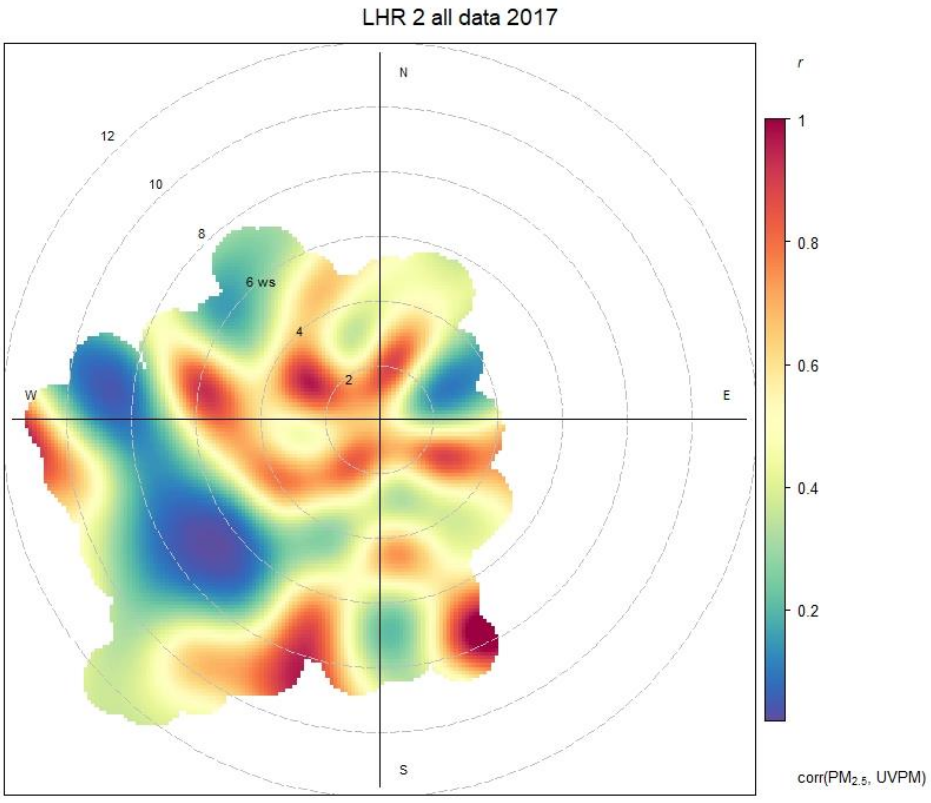

Figure S22 – Bivariate polar plot showing correlation between $PM_{2.5}$ and UVPM. Highest correlation between pollutants are shown in dark red.

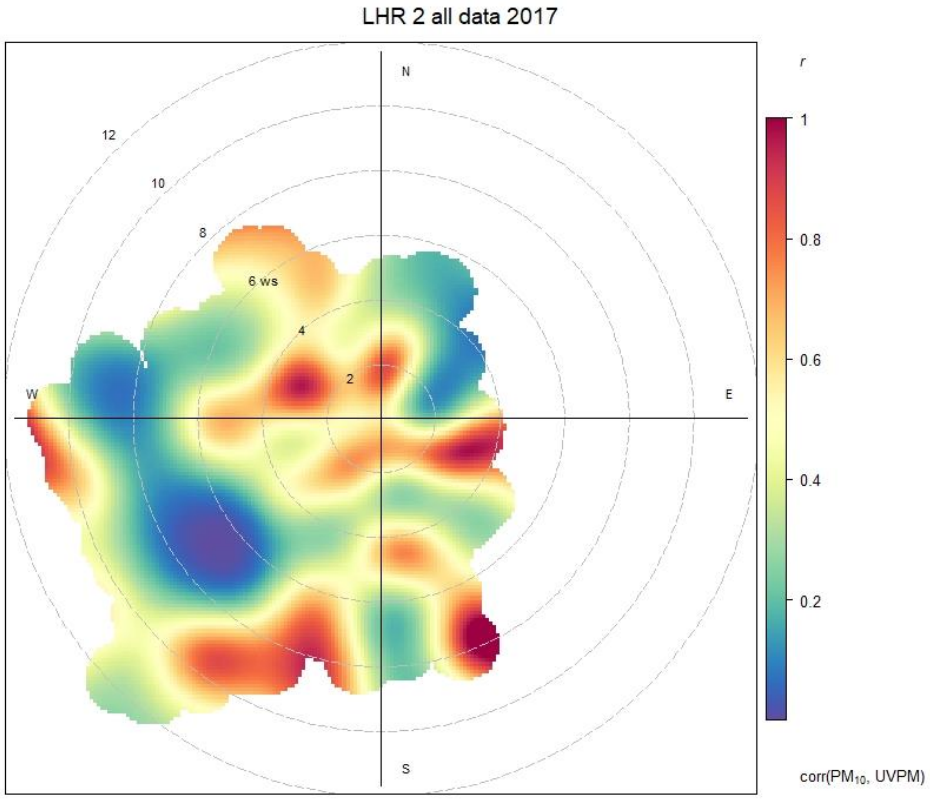

Figure S23 – Bivariate polar plot showing correlation between PM_{10} and UVPM. Highest correlation between pollutants are shown in dark red.

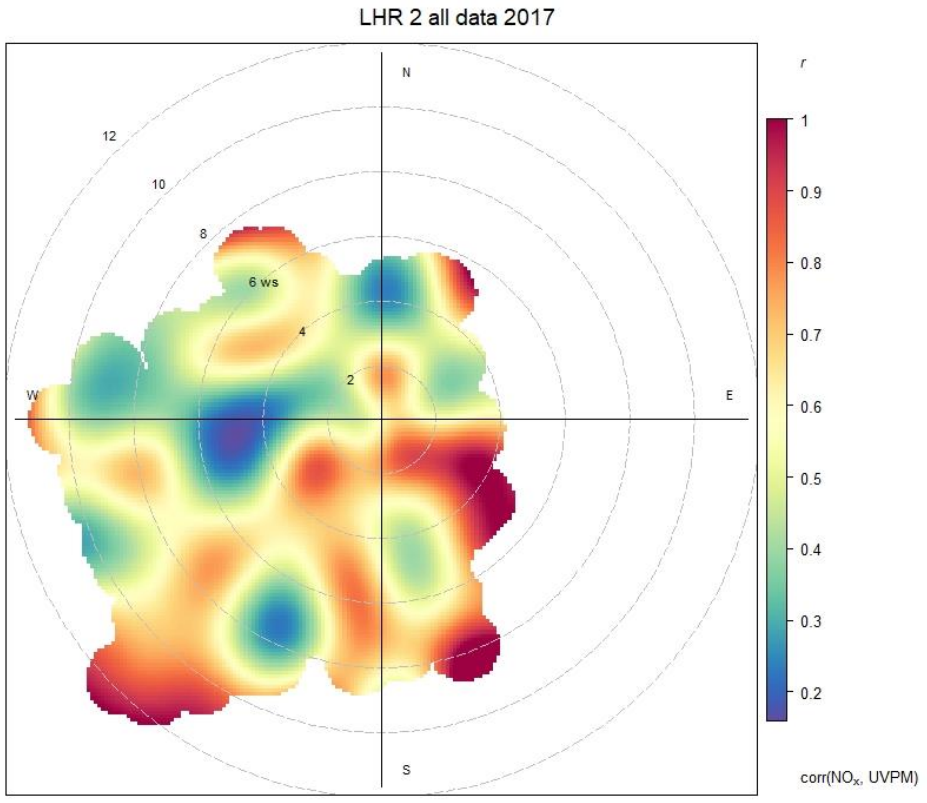

Figure S24 – Bivariate polar plot showing correlation between UVPM and NOx. Highest correlation between pollutants are shown in dark red.

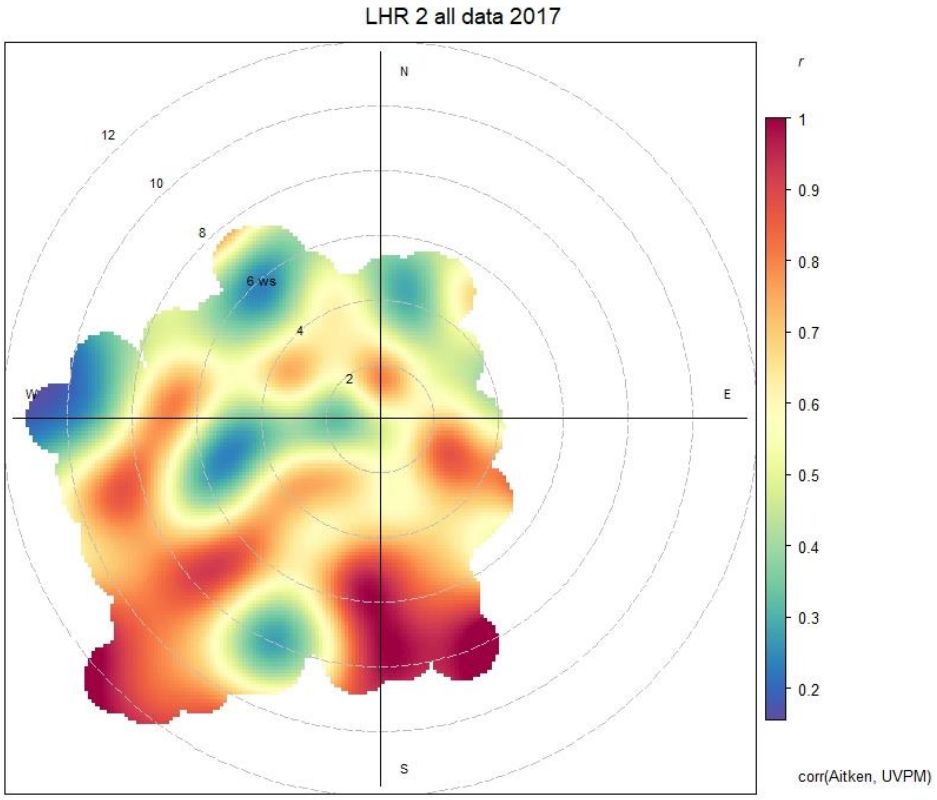

Figure S25 – Bivariate polar plot showing correlation between Aitken and UVPM particles. Highest correlation between pollutants are shown in dark red.

Figures S12 to S25 show the bivariate polar plot correlation for the pollutants measured at LHR2.

For winds associated with the airport, there are clear associations between:

- Nucleation particles with Aitken particles
- Nucleation particles with BC, NOx and UVPM, although there are also weaker associations from other wind directions
- Aitken particles with UVPM, suggestive of a link with tyre wear from landing aircraft, as noted in section 3.5.

There is clear correlation from all wind directions for the following pollutants:

- Aitken particles with BC and NOx
- UVPM with NOx, PM_{10} and $PM_{2.5}$

From wind directions not from the airport, there are clear associations for:

- NOx with PM_{10} and $PM_{2.5}$

There is poor correlation from every wind direction for:

- Nucleation particles with PM_{10} and $PM_{2.5}$

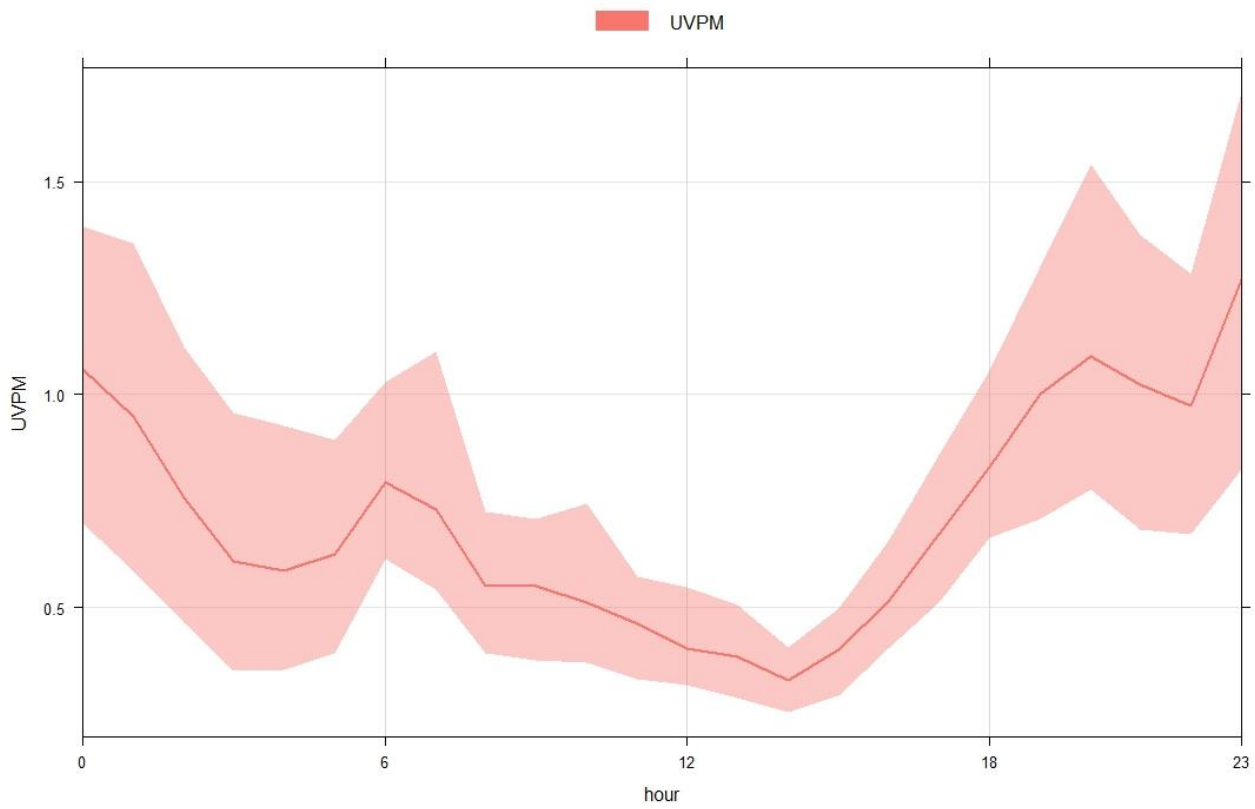

Figure S26 – Diurnal plot for UVPM (µg/m^3) at LHR2, October to November 2017

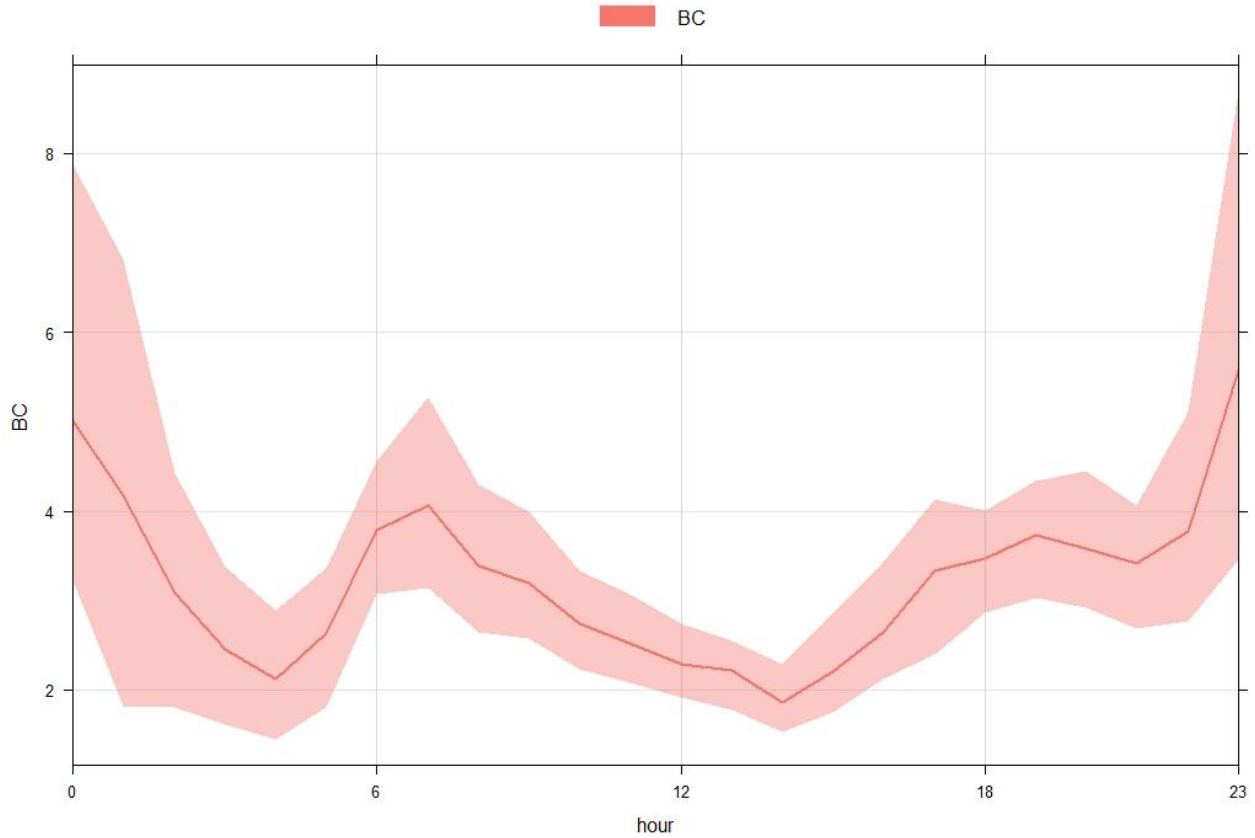

Figure S27 – Diurnal plot for BC (µg/m^3) at LHR2, October to November 2017

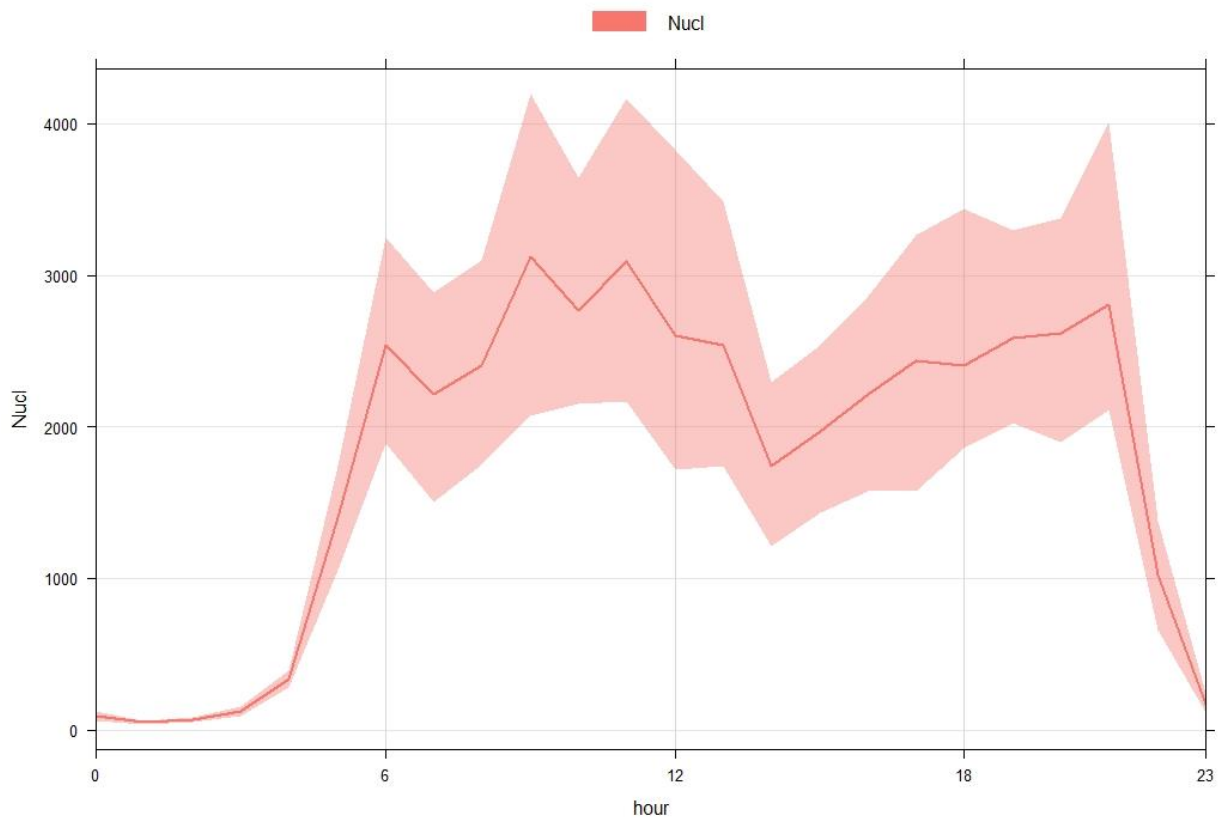

Figure S28 – Diurnal plot for nucleation particles (#/cm^3) at LHR2, October to November 2017

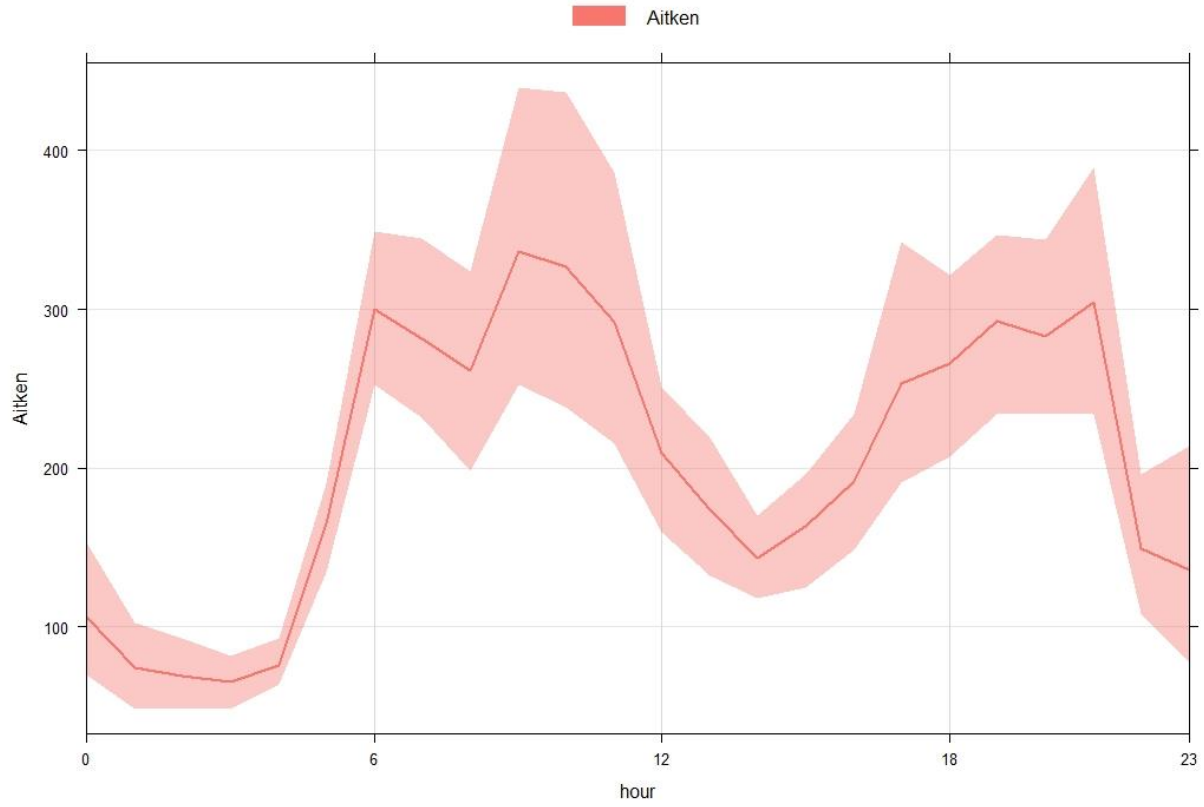

Figure S29 – Diurnal plot for Aitken particles (#/cm^3) at LHR2, October to November 2017

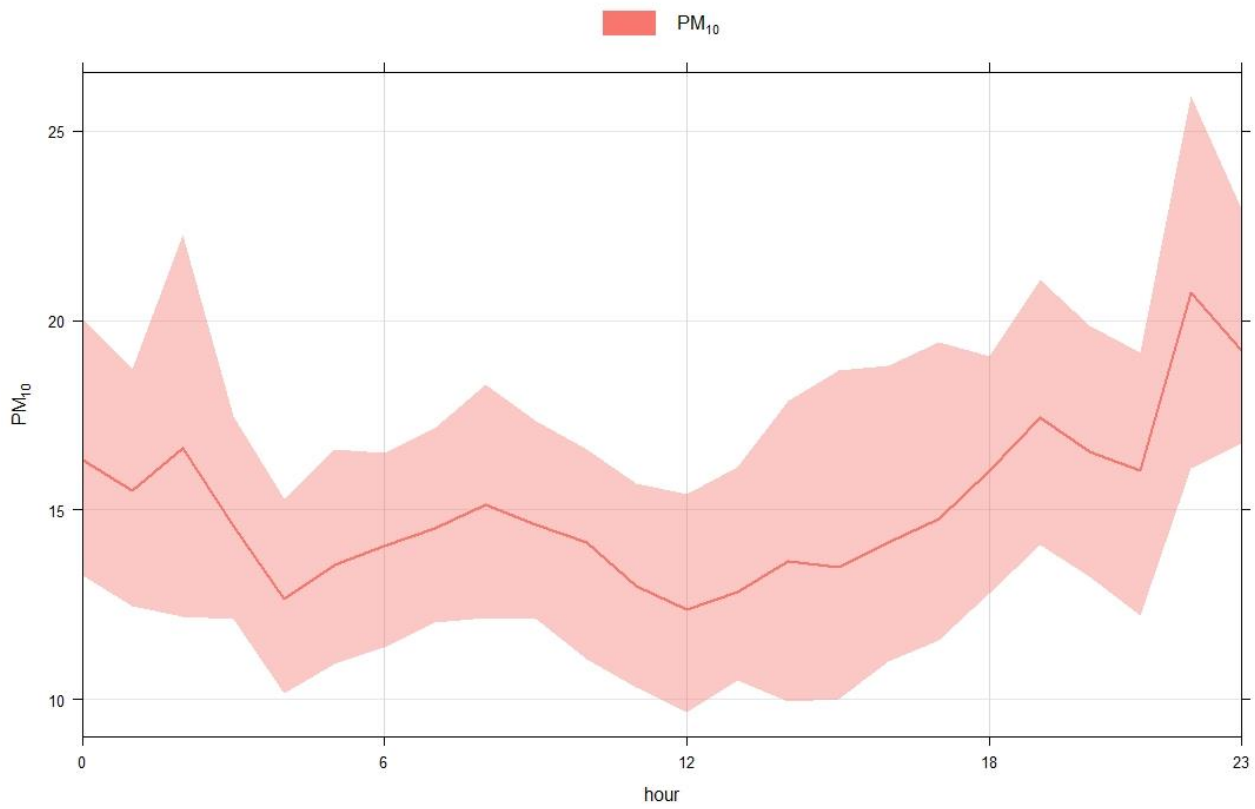

Figure S30 – Diurnal plot for PM_{10} ($\mu g/m^3$) at LHR2, October to November 2017

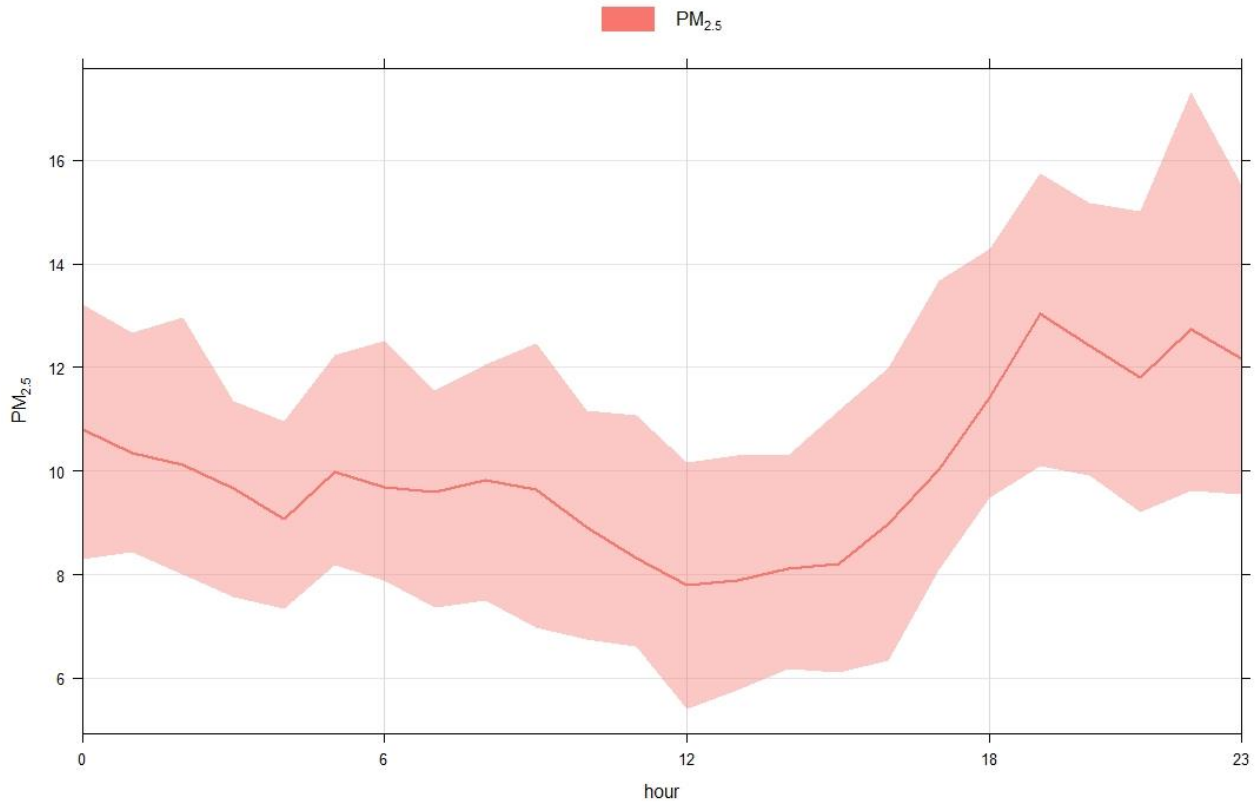

Figure S31 – Diurnal plot for $PM_{2.5}$ ($\mu g/m^3$) at LHR2, October to November 2017

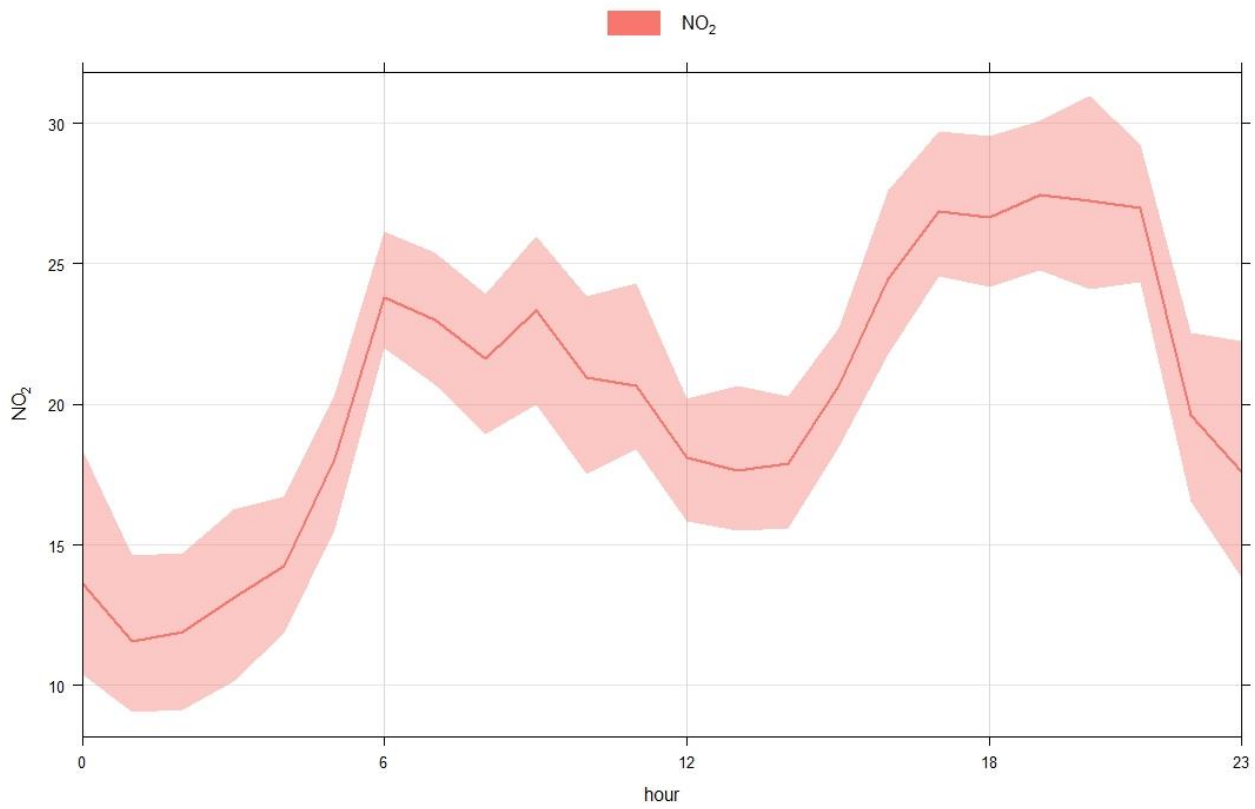

Figure S32 – Diurnal plot for NO_2 (ppb) at LHR2, October to November 2017

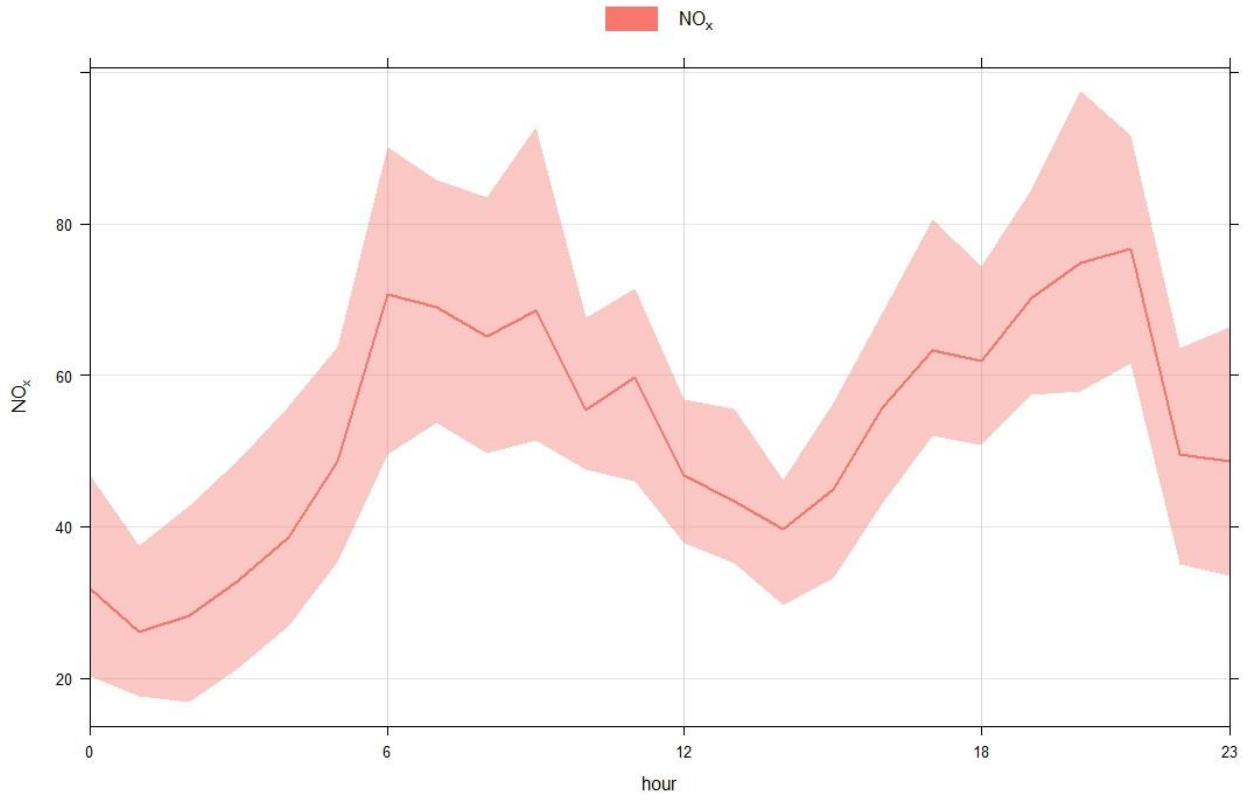

Figure S33 – Diurnal plot for NOx (ppb) at LHR2, October to November 2017

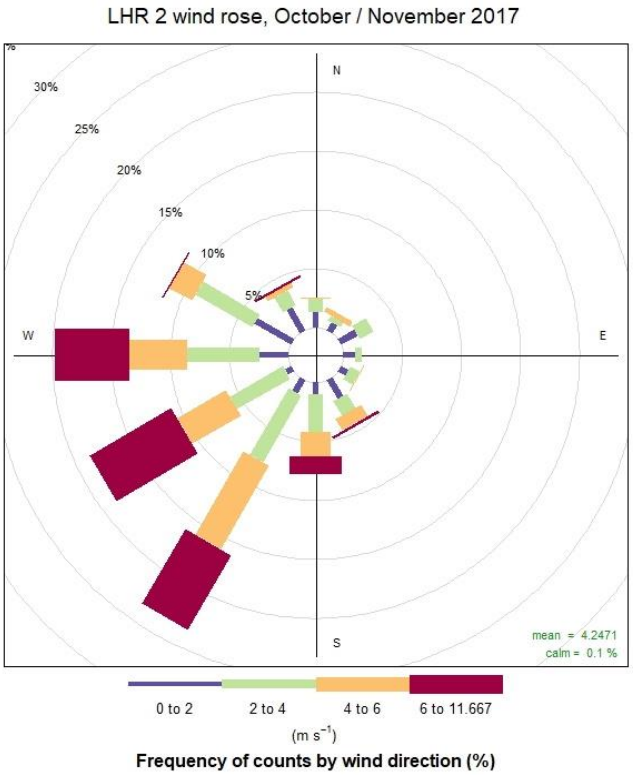

Figure S34 – Wind rose plot for 2017 survey at LHR2

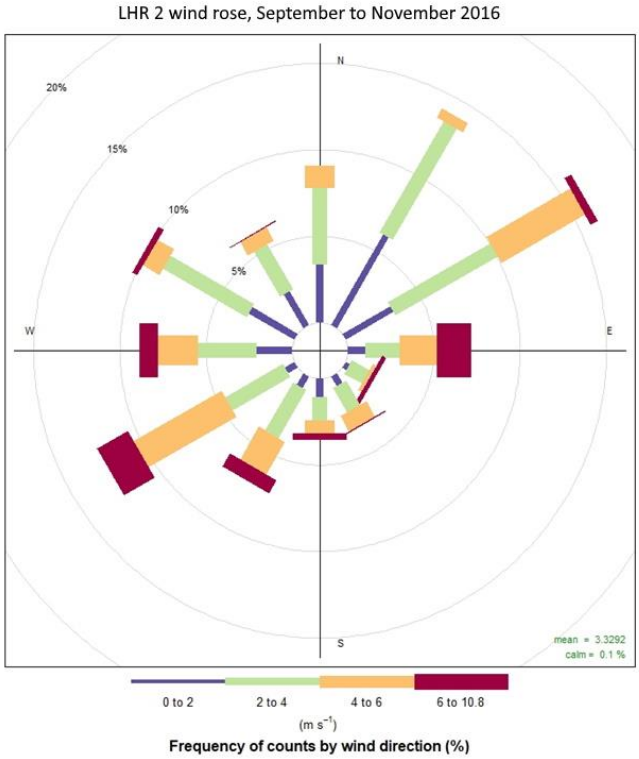

Figure S35 – Wind rose plot for 2016 survey at LHR2

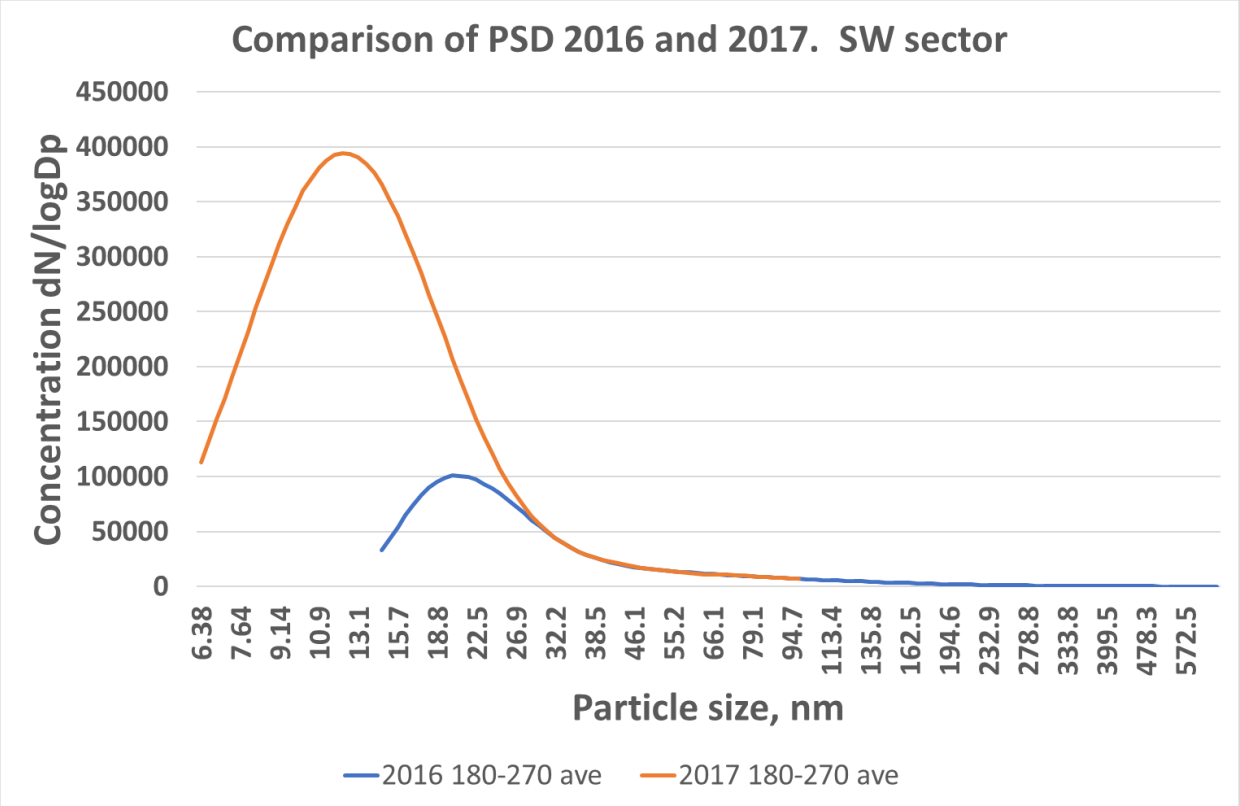

Figure S36 – comparison of 2016 and 2017 average particle size distributions, winds from 180 to 270 degrees

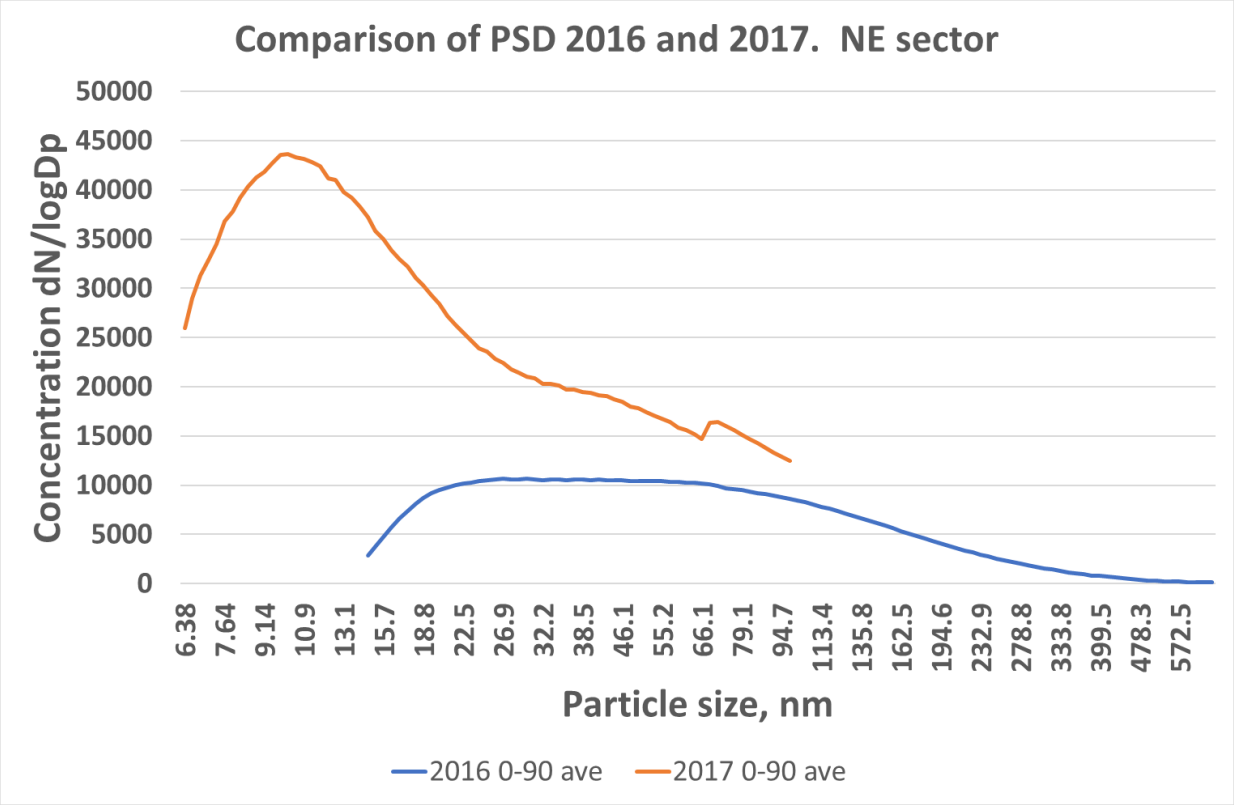

Figure S37 – comparison of 2016 and 2017 average particle size distributions, winds from 0 to 90 degrees

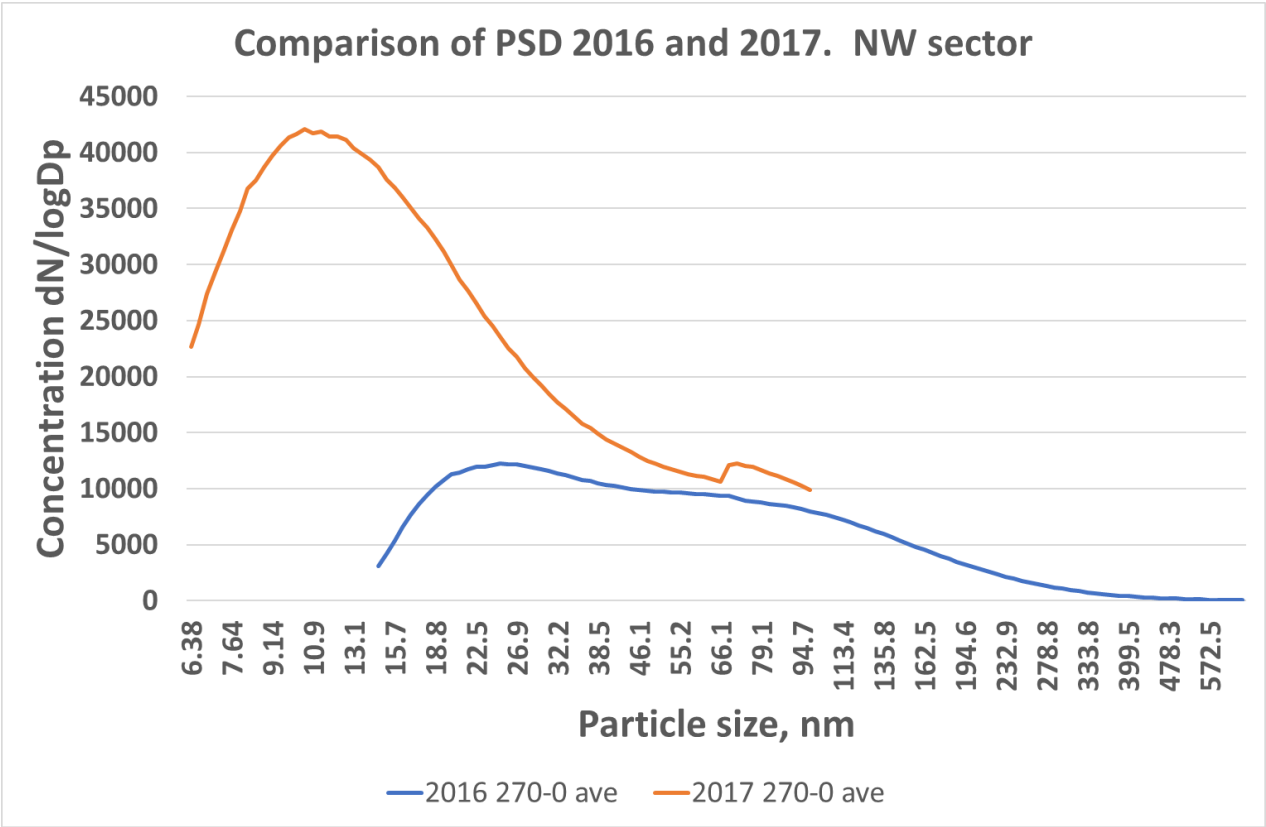

Figure S38 – comparison of 2016 and 2017 average particle size distributions, winds from 270 to 360 degrees

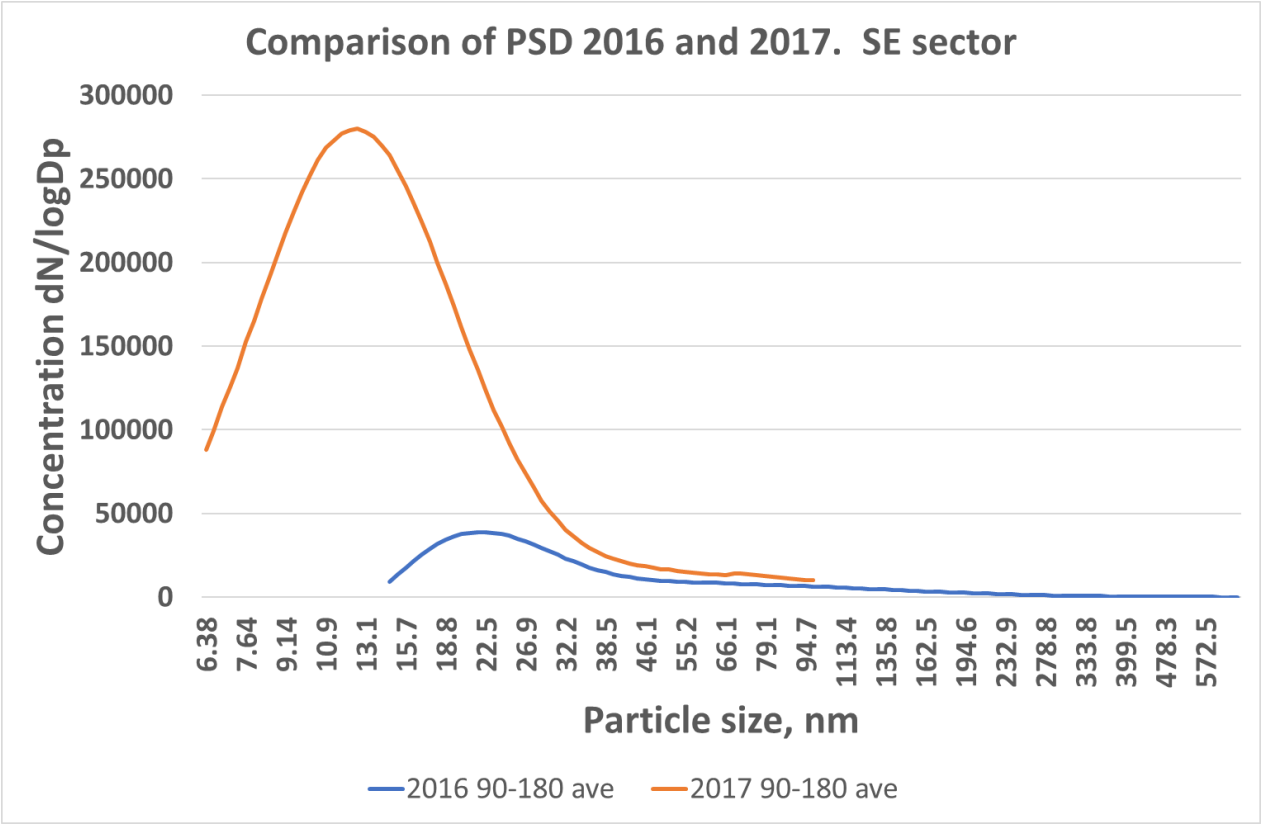

Figure S39 – comparison of 2016 and 2017 average particle size distributions, winds from 90 to 180 degrees

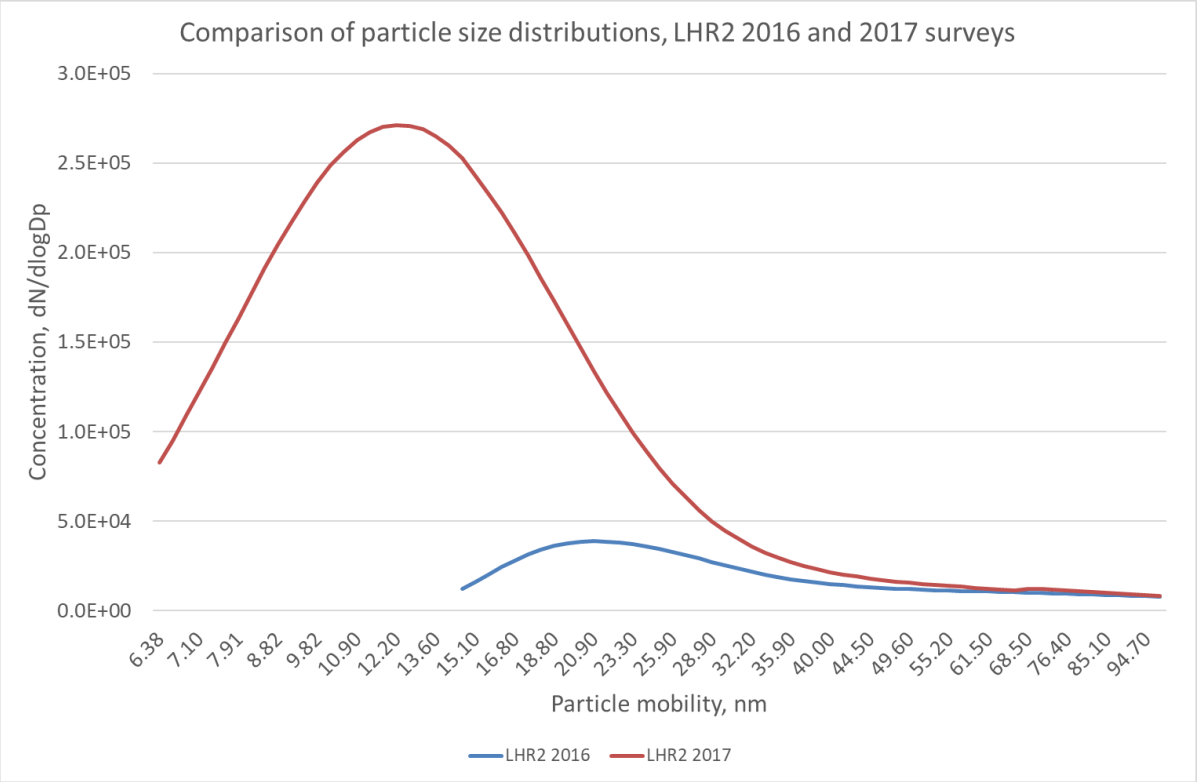

Figure S40 – comparison of 2016 and 2017 average particle size distributions, all data.

A detailed investigation of the relationships between UFP measurements in 2016 and 2017 was undertaken.

Wind direction in 2017 was dominated by stronger SW'ly winds, when compared to the 2016 campaign, shown in plots S34 and S35.

Figures S36 to S40 show the dramatic differences, both in magnitude of concentrations and PSD profile for the 2017 and 2016 datasets. An investigation as to why the datasets are so different was undertaken. The key points are summarised below:

- The physical configuration of the SMPS / CPC was basically the same for both surveys. The inlet was identical, the conductive tubing connecting the inlet, SMPS, CPC and the various auxiliary flows were identical, thus there was no difference in tube lengths between surveys.
- The SMPS and CPC were traceably calibrated for both size and concentration, both before and after each exercise. Each calibration result was within 1 size bin and 1% of the reference standard instrument. The calibrations use monodisperse soot particles and a calibrated CPC.

However, for ambient measurements, the reported PSD from the calibrated SMPS deconvolves the measurements during voltage scanning to reconstruct the polydisperse size distribution, increasing the likelihood of errors in the PSD and size calculations.

- Examination of meteorological data showed winds during the 2017 survey were dominated by south westerly winds, far more than observed in 2016.
- Exploration of the 2016 and 2017 PSD profiles, separated into 4 different wind quadrants, showed that the distribution profile in 2017 does not follow the patterns from 2016. The 2016 PSD associated with northerly winds shows significant measurements of particles from 20-100 nm, clearly different in distribution than the southerly winds. The 2017 data is dominated in all 4 sectors by particles in the 7-30 nm range, with highest concentrations always associated with 7-20 nm particles.
- The average 2017 PSDs show a distinct step change in the data at the 68.5 nm size bin. The step is particularly obvious when nucleation particle numbers are lower; i.e. during periods of northerly winds. This step change is evident in the 1 minute data, the analyser exhibited this behaviour throughout the 2017 survey. In contrast, the same analyser (in a different configuration) did not display this step in the 2016 dataset.

The operational parameters of the SMPS / CPC were markedly different for the 2016 and 2017 surveys. These differences are presented in Table S3:

Parameter	2016 survey	2017 survey
SMPS/CPC/DMA Model	3082/3775/3081	3082/3776/3081
Scan range	14.6 – 668.2 nm	6.38 – 98.2 nm
Time of scan up/down	2:15 / 0:45	0:45 / 0:15
Detector sample flow l/min	0.3	0.05
Detector inlet flow l/min	0.3	1.5
Sheath flow l/min	3	15
Bypass flow l/min	0	0

Table S3 – Comparison of SMPS / CPC configurations in 2016 and 2017 surveys

All reasonable steps to minimise physical and calibration differences between surveys were undertaken. Analysis of the measurement data does not reveal any obvious errors in processing, which leads to a conclusion that SMPS configuration differences are the most likely cause of the discrepancies between the 2016/2017 observations. This, in and of itself, is not a surprise: Andersson (2000) (1), Wiedensohler et al (2012 and 2018) (23, 24) and Takegawa et al (2020) (22) have all demonstrated that even the smallest change in configuration has a dramatic effect on the measurement of the finest particles in a SMPS/CPC setup. Separate personal conversations with Wiedensohler (TROPOS), Tritscher (TSI), Terres (BMW) and Andersson and Marshall (Ricardo) have all confirmed that measurement of particles smaller than 20 nm is significantly influenced by many parameters, not least charge efficiency, diffusive losses, sample tube length and sample flow. A simple calculation of potential diffusive losses at 300cc/min and 1500cc/min flow rates shows two important points:

- A 10nm particle is 35 times more "diffusive" than a 500nm particle in the same air flow
- A particle in a 300cc/min air flow is 5 times more "diffusive" than a particle in a 1500cc/min air flow

These two points combined indicate that diffusive losses in a lower flow rate environment (2016) will be far more significant compared with a higher flow (2017).

Additionally, the reduced operating size range used in 2017 will have an impact on how the analyser processes the results to account for multiple charges. Without particle size data for the larger particles, the calculation algorithm is not able to correctly assign multiple charge correction at the higher end of the restricted range. This is almost certainly the reason for the observed steps in the 2017 data.

All the above evidence means that direct comparison between these two datasets is impossible. In fact, comparison between any two PSD / concentration measurement systems should be viewed with extreme caution unless it is known that the analysers have been configured identically. This is a significant limitation which impacts on all UFP research undertaken to date. Future research should clearly define how instrumentation is configured to improve the value of the data produced.

For the purposes of this paper, the 2017 data is of value for identifying aircraft activities and the emissions from different aircraft types, but because of the configuration differences, detailed comparison with 2016 data or the UFP measurements from UK datasets in 2017 is not possible.

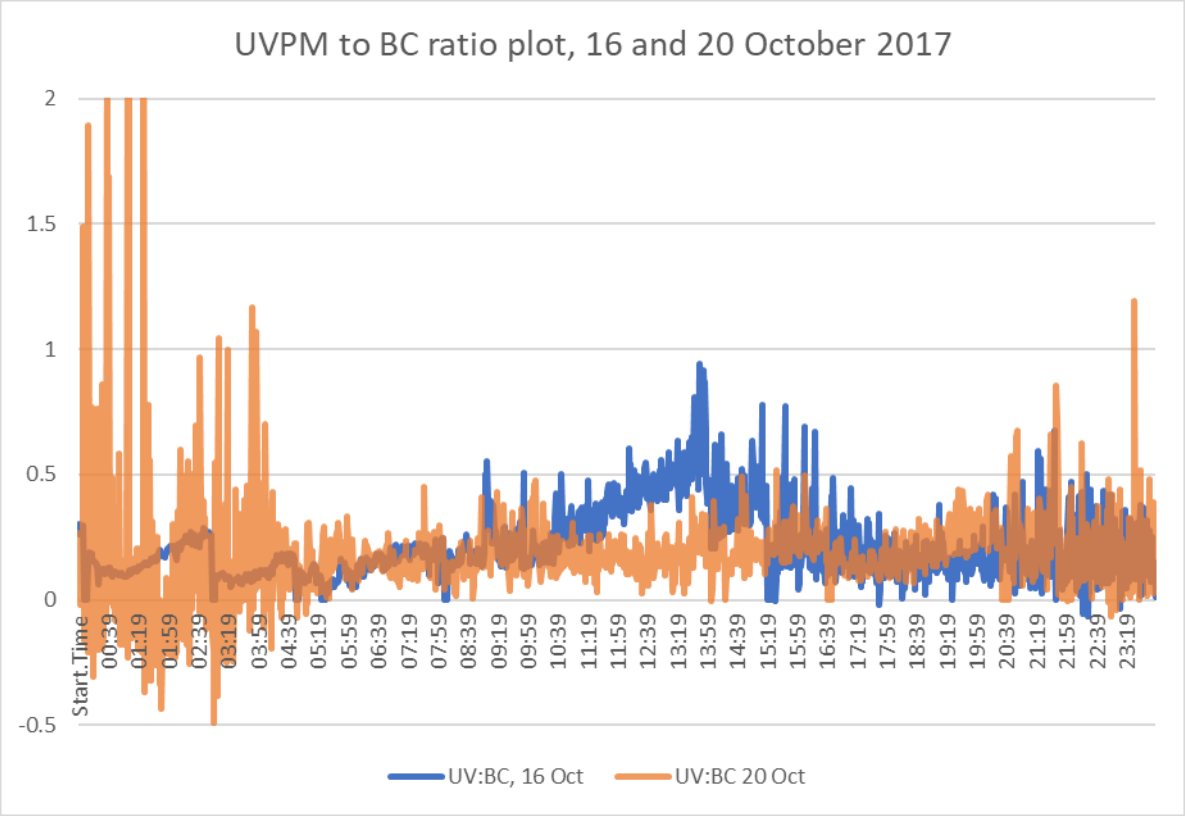

Figure S41 – UVPM ratios, 16 and 20 October 2017

This plot shows how the relationships between BC and UVPM evolve through the day and compare between days. Between 10:00 and 15:00, the ratios are significantly different on 16th and 20th October, suggesting that the measurements are associated with different sources

6 UFP at Heathrow, 2019

This Chapter presents work originally submitted for publication in *Environmental Science and Technology* in April 2022. Following production of this thesis, it will be updated and submitted for publication later in 2022.

6.1 Abstract

Very high concentrations of ultrafine particles were measured at Heathrow Airport London. Exposure to UFP is strongly linked to adverse health effects and guidance for exposure limits has recently been provided by the World Health Organization (WHO). Using 1 second resolution UFP measurements and aircraft GPS data, measurements were assigned to individual aircraft and their operating mode, and this information was used to model UFP emission rates. In all cases, highest emission rates were associated with departing aircraft, with rates for larger aircraft higher than smaller aircraft. Emission rates per passenger is influenced by the number of passengers carried, especially for arriving aircraft. Calculated emission rates are significantly higher than stated literature values, as condensable particles are also included in the measurements. Measured UFP concentrations inside the airport perimeter exceed the WHO guidance, indicating that assessing population exposure around airports will be of increasing importance in future.

6.2 Introduction

There is increasing evidence that exposure to Ultrafine particles (UFP, defined here as particles that are smaller than 100 nm in diameter) is associated with adverse health outcomes (1-10). But there are very few definitive studies and a scarcity of robust measurement data to assess their concentrations and sources. As a result, to date, there are no limit or target values to minimise exposure to UFP. The automotive industry implements assessment of UFP emissions for EURO 6/VI vehicles (11) (maximum permitted particle number concentration per km), but this only relates to particles >23 nm and only for particles which are involatile at 350°C. While the nature of road traffic exhaust emissions as they cool and interact with the ambient environment has been documented, (12, 13) the behaviour of emissions from aircraft is less well characterised or understood.

There is growing interest in measurements of UFP close to airports. London Heathrow Airport is the busiest two runway airport in the world and the largest in the UK. Measurements of NOx and PM have been made at the airport continuously since 1993 and at 5 separate locations currently. Recent studies (14-16, 35, 36) have shown that high numbers of very small particles dominate the environment around airports, and conclusively point to aircraft as the dominant source of these particles. Our previous two studies (14, 15) explored the relationship between UFP at the airport and the three UK monitoring stations, showing that UFP from aircraft are smaller in size than those generated by road traffic, present in higher numbers, and emitted in larger numbers during departures. Stacey et al. (15) provided evidence to suggest that larger aircraft had higher UFP emissions than smaller aircraft. The emissions from Heathrow are readily detectable not only in a local residential area, (17) but also in central London, (18) and this phenomenon is seen in other cities around the world (19).

The research we present here is unique in the sphere of ambient air quality measurements in an airport environment. We are not aware of any other published data that utilises full particle size distribution data sampled at 1 Hz, coupled with exact aircraft type and location data at 1 Hz, along with 1 minute meteorological measurements. Over 10GB of data was collected over a 7 week period in 2019, allowing large clusters of measurements from various wind directions, aircraft types and operating modes to be evaluated. The proximity of the monitoring location to a heavily used runway has historically been very difficult for academic researchers to secure. The airside location used for this study is 170m downwind of the busiest 2 runway airport in the world – for understandable security reasons, airport operators do not usually grant access for monitoring in these locations.

6.3 Materials and Methods

Here we present information about the analysers used, quality control processes, data synchronisation, procedures for data analysis and method used for estimation of UFP emissions. We made use of R, R Studio and the OpenAir suite of tools (20) to analyse the relationships between different aircraft types and operating modes.

Air sampling: The monitoring station used for this study is located in the north eastern corner of Heathrow Airport. The station and measurement data can be viewed online (21). The monitoring

station is 170m from the middle of the northern runway, positioned in the prevailing wind from the runway and main terminal areas. The station has been used to measure NOx and PM continuously since 1993 and for two UFP measurement studies in 2016 (14, 15).

Instrumentation used for the survey: The equipment used at Heathrow Airport for this survey are presented in Table 1 below:

Table 1: List of monitoring apparatus used for the survey at the LHR2 monitoring station. A detailed description of the instruments is provided in the Supplemental Information

Station	Equipment installed
LHR2	Cambustion DMS500 Fast Particulate Spectrometer UFP analyser, configured to sample at 1Hz, 5-1000 nm particle range in 37 different size bins (64 channels per decade), GDI fuel profile algorithm for measurement deconvolution API T200 NO_x analyser, 15 minute average measurements of NO_x, NO and NO_2 FIDAS 200 PM analyser, 1 minute average measurements of PM_{10}, $PM_{2.5}$ and PM_1 Magee AE33-7 Black Carbon analyser, 1 minute average measurements of Black Carbon (BC) and Ultra Violet Particulate Matter (UVPM) Lufft WS-600 weather station (Wind Speed / Wind Direction / Temperature / Pressure / Relative Humidity / Precipitation), 1 minute average measurements Praxis Urban NO_2/NO sensor system, 10 second sampling from Alphasense electrochemical cells

Quality Control Processes: The PM, BC and NOx analysers are all operated according to QA/QC procedures used in the UK national network programmes (QA/QC) (22). All analysers are audited and serviced twice yearly with additional traceable calibration standards. The results from these calibrations, services and audits are all used in the process of data ratification. The procedures used to process the raw data are those used in the UK national monitoring networks, which are fully documented (22).

15 minute data from the PM, BC and NOx analysers are downloaded every hour and stored in the cloud. This provisional data is also published in near real time at https://heathrowairwatch.org.uk

(accessed March 2022). 1 minute data from the PM and BC analysers were stored in the analyser internal storage facilities and downloaded to USB sticks every week.

There is currently no ambient monitoring legislation regarding the performance of UFP analysers. The DMS500 was calibrated for both particle counting and sizing using monodisperse soot particles generated at the ISO17025 accredited calibration facilities at the Ricardo Energy and Environment laboratory. In all cases, the sizing capability of the DMS500 was within 1 size bin of the reference standard and within 2% for particle counting (in the range 5e+03 to 1e+05). There are no recognised test procedures to assess the capability of the DMS500 analyser (or indeed any other analyser with a classifier) to correctly resolve a polydisperse sample, but the philosophies of Wiedensohler et al. (23, 24) for implementing robust QC to UFP measurements were employed for this study.

There are a number of differing philosophies about how to collect samples for UFP measurements. The few ambient monitoring stations in operation across the UK sample at ambient temperature with a nafion dryer to minimise the impact of water on particle size. It should be noted that water is unlikely to have a significant effect on particles smaller than 50 nm diameter (25). The majority of studies of aircraft exhaust emission-related UFP to date have focussed on non-volatile particles, sampling exhaust gases at high temperature to keep volatile / semi-volatile particles in the gas phase. These differences in sampling methodologies, combined with many different techniques and sampling durations used for studies, means that detailed comparisons of number concentrations between different study programmes need to be approached with caution. This was further discussed in the Stacey review of UFP monitoring (16). The DMS500 was configured to undertake regular automatic baseline reprofiling during operation. This operation generally occurred for up to 1 minute at three minute intervals. Data validation of the UFP data involved removal of auto zero data, screening of data where particle number concentrations were below the limit of detection (for the purposes of this study, all measurements were removed when 10nm particles were below 1000 #/cm^3), and where instrument meta data suggested a fault in the measurement process.

The 10 second NO and NO_2 data from the Praxis sensor system required significant processing for baseline drift and temperature sensitivity before any investigation could begin. Once the data had been screened, it was aggregated into 15 minute averages and compared against 15 minute average

data from the reference NOx analyser at the monitoring station. The averaged sensor data showed excellent agreement for both NO and NO_2, providing confidence that the 10 second data could be used to establish relationships between measured NOx from individual aircraft and stated emission rates.

Data synchronisation: 1 second data from the DMS500 was adjusted to agree with the time from the internet (obtained using a mobile phone and verified using https://time.is). 15 minute average measurements from the NOx, PM, BC and Meteorology devices are continuously time synchronised from a similar internet clock source. For processing the 1 minute PM and BC data the analyser internal clocks were checked for accuracy every week against the mobile phone and measurement data time corrected when necessary.

High resolution data for individual aircraft movements and runway operating modes was gratefully received from the Aircraft Operations Team at Heathrow Airport. Every second the position, speed, altitude (for aircraft less than 20 metres above ground level) and identification of every moving aircraft on the airfield is recorded. The accuracy of the time stamp is verified using an internet clock source. For the purposes of this study, only aircraft that were on the northern runway, or on the entrances to the northern runway, were included in the analysis of measurement datasets. It is possible that aircraft taxiing to and from the runway will also contribute to measurements when they are directly upwind of the monitoring station, but the distance and lower thrust settings of these aircraft should minimise this potential impact. During westerly winds, the airport operates the two runways in fixed modes. For half of the day, departures are restricted to one runway and arrivals on the other runway. At approximately 14:00 local time, the operation modes are switched, allowing analysis of AQ measurements during arrivals and departures closest to the monitoring station.

For analysis of arriving and departing aircraft, only data associated with winds in the clockwise sector between 100 and 260 degrees, between the hours of 05:00 and 23:00 BST and only during dry weather were included in the analysis. The position of the aircraft on the runway, together with wind speed and direction was then used to determine the time required for the exhaust plume to arrive at the monitoring station. This delay was then fitted to the measurement data to align the measurements to the aircraft. The aircraft type and position meta data was then added to the measurement database to

allow detailed investigation of measurements. Typically, an aircraft departs or arrives at the airport every 90 seconds.

Data analysis: Data were extensively processed and manipulated using the R and RStudio suite, coupled with OpenAir tools (20) for imaging and data sorting. Data analysis was undertaken using a staged and increasingly detailed approach:

- Clustering the full dataset according to wind directions. Similar analysis of this data has been explored in earlier studies, (14, 15) which identified a clear source of the finest particles from the airport. Undertaking this analysis of the 2019 data and comparing it to previous studies provided reassurance of the validity of the DMS500 data.
- Removing unnecessary data:
    - between the hours of 23:00 and 05:00 (when there is no aircraft activity),
    - during periods of rainfall,
    - when aircraft were departing easterly. Operations when aircraft departed easterly were removed, because aircraft only arrive on the northern runway during easterlies, exit the runway more than 2km from the monitoring station and are therefore assumed to have a minimal contribution to the AQ measurements at the station. Until 2021, aircraft did not normally depart in an easterly direction on the northern runway,
    - data from wind directions clockwise from 260 degrees to 100 degrees. The remaining sector approximates to the zone of Airport contribution to air pollution concentrations.
- Separating the datasets according to runway operating modes, specifically when departing and arriving on the northern runway. Similar analysis of earlier data (14, 15) showed that arriving aircraft emit far fewer particles than departing aircraft. Examination of the 2019 data from the DMS500 confirmed this observation.
- Separating the datasets according to whether an aircraft was on or approaching the northern runway.
- Separating the runway movements into individual aircraft type for both arrivals and departures.

Estimation of UFP emissions from NOx data: For the estimation of UFP emission rates from individual aircraft types, a series of processes were undertaken:

- Verification of the baseline and temperature corrected 10 second NO and NO_2 sensor data, once aggregated to 15 minutes, using the reference NOx analyser.
- Sensor "NOx" data created from the sum of the sensor NO and NO_2 measurements, and verified again using the reference NOx data.
- Separation of the runway only, dry weather NOx data using the procedures described in the data analysis above.
- Subtraction of background NOx concentrations, using the nearby Oaks Road monitoring station. The average 15 minute background NOx concentration was subtracted from every 10 second measurement, assuming that background NOx concentrations would not change significantly during this time.
- 10 second NOx data was incorporated into the 1 second master dataset, again assuming that there was minimal variation in the NOx concentrations during the 10 second period.
- NOx measurements were assigned to individual aircraft and clustered into groups as per the UFP methodology for both departing and arriving aircraft.

Once the NOx measurements / aircraft correlation had been established, the results were compared against the stated NOx emissions rates in the ICAO database (26) (this assumes 85% thrust settings for departing aircraft and 30% thrust settings for arriving aircraft – this decision is discussed later in the text). The average NOx emission rates for each aircraft group were used, rather than further breaking down the results into individual engine types. Where there are significant differences in emission rates, outlier engine types and associated measurements were excluded from the analysis.

For the estimation of emission rates per passenger, we had to decide on a representative value for the number of passengers carried in each aircraft. The manufacturers state a range of seat numbers possible and this will depend on how each company chooses to configure their aircraft. For example, an Airbus A380 can be configured to carry anywhere between 500 and 850 passengers. In practice, the majority of A380s purchased were configured with three classes and approximately 550

passengers, thus we assumed 550 passengers for all A380 aircraft at Heathrow. Similar judgements were made for passenger numbers for the other aircraft types, though the range of minimum and maximum passenger numbers was correspondingly smaller. In order to make the emission rates per passenger comparison, we assumed that the aircraft were operated at these capacities.

Emission rates for Particle Number Concentrations (# / sec) for both departing and arriving aircraft and aircraft type were then calculated using the measured NOx data, measured PNC data and the ICAO stated NOx emissions rates, according to the following equation:

PNC emission rate (#/s) = NOx emission rate (g/s) x PNC (#/m3) / NOx (g/m3) (eq. 2)

6.4 Results and Discussion

For this study, we make use of a UFP analyser (Cambustion DMS500) operating at 1 second resolution at the station closest to the northern runway (LHR2), together with 1 second resolution GPS aircraft movement data, 1 minute resolution meteorology, PM and Black Carbon (BC) data plus 10 second NOx data, to look in detail at the UFP emissions from aircraft, both departing and arriving.

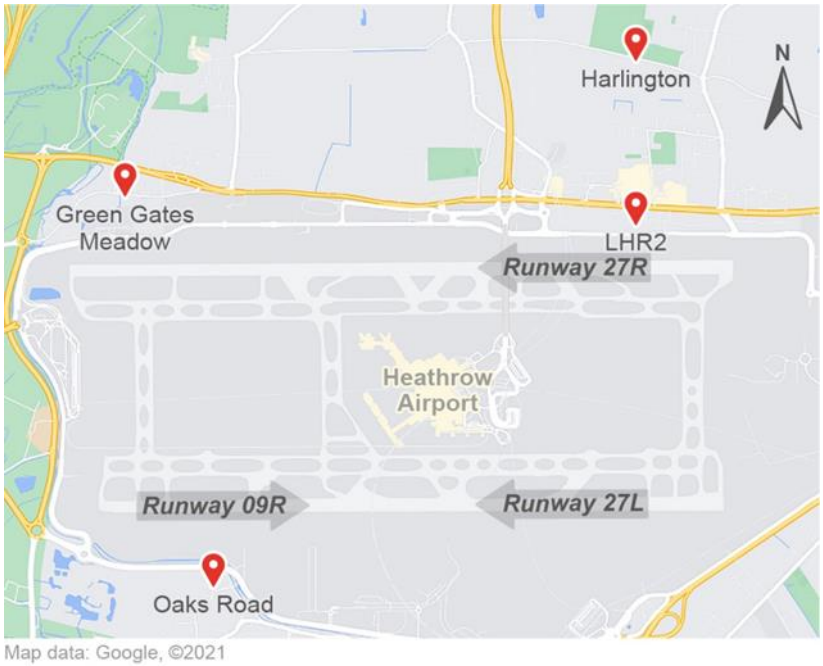

Figure 1: Map of Heathrow Airport with active monitoring station locations.

The map in Figure 1 shows the operating modes of the two runways. Monitoring in 2019 was undertaken at the LHR2 station, 170m from the centre line of the northern runway.

Data capture from the Cambustion analyser was high: of a possible 75,444 minutes of operation, 71,857 (95%) valid measurements were collected. 91.9% of a possible 13,445 operation minutes were collected for arriving aircraft and 95.6% of a possible 13,998 operation minutes were collected for departing aircraft. An additional 94 minutes of measurement data below the limit of detection were rejected during QA/QC.

For analysis of aircraft emission rates, only periods where aircraft were departing or arriving on runway 27R were considered.

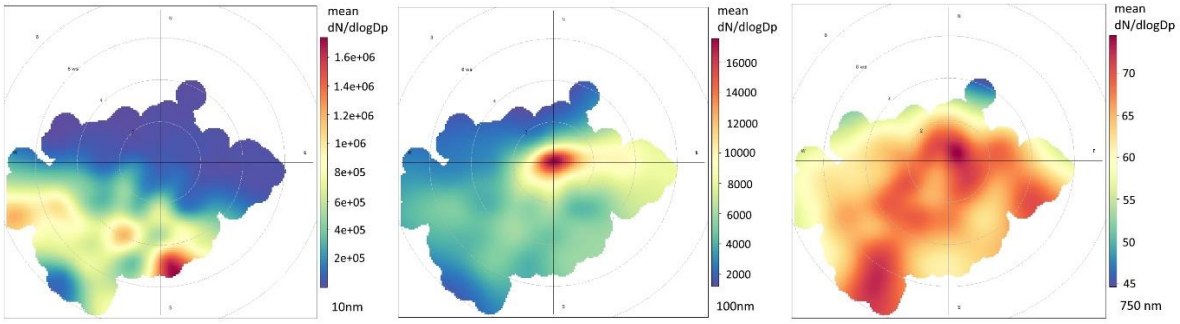

Figure 2: Polar plot of average particle size distribution showing how concentrations are associated with wind direction. Scale is specific for each particle size, ranging from low (blue) to high (red).

The polar plots in Figure 2 provide an indication of the wind directions associated with highest particle concentrations for each size fraction. The plots clearly show how the sources of different sized particles changes. For particles smaller than 75 nm, the airport is the dominant contributing source. Individual details can be picked out from the 10 nm plot – the runway as a line source is plainly visible, as is the large aircraft stand area associated with Terminal 2 directly to the south of the monitoring station. From 75 - ~300 nm, highest concentrations are associated with low winds, mostly to the immediate north of the monitoring station. This suggests the adjacent perimeter road is a significant source of these particle sizes.

At particle sizes from 300 – 1000 nm, the airport contribution increases again, but these larger particles are also associated with other wind directions, suggesting that there are multiple sources of larger particles. This was confirmed in earlier studies (14, 15) that demonstrated larger particles (e.g. $PM_{2.5}$) are not always associated with airport activity.

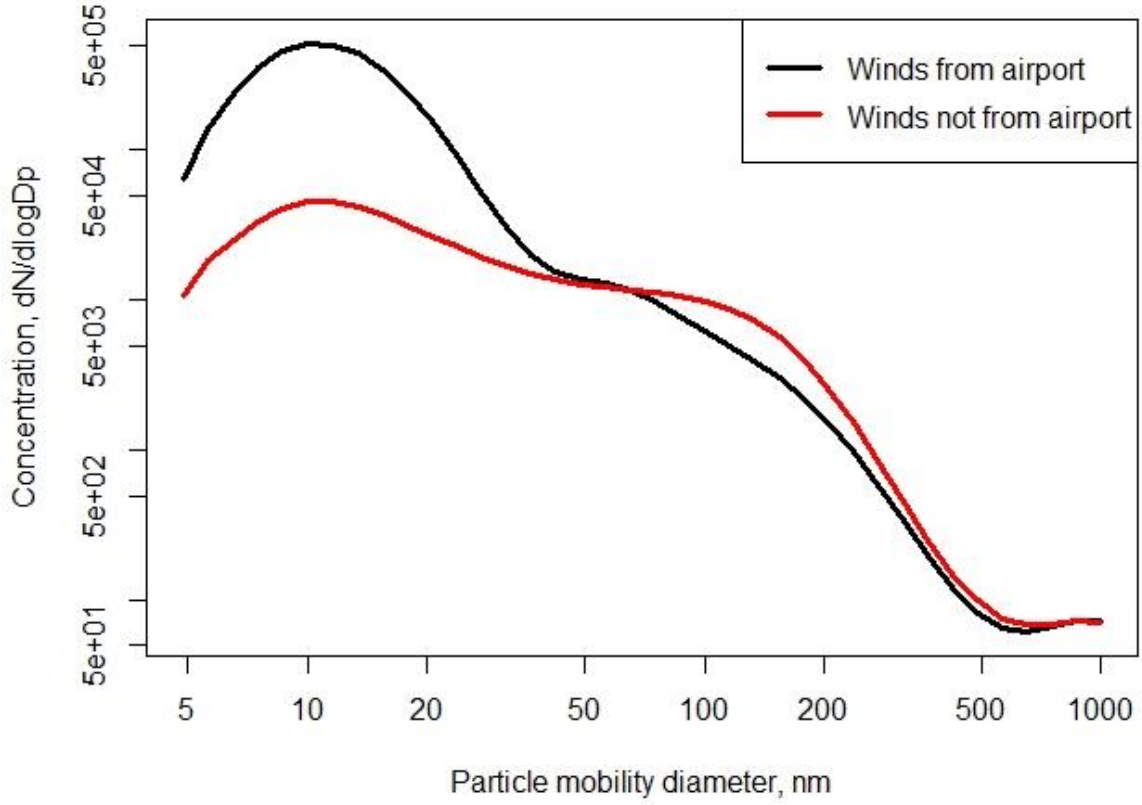

Figure 3: Survey average concentrations for UFP, further split to assess concentrations from on-airport and off-airport wind directions.

The particle size distribution plot in Figure 3 provides additional support for the observations in the polar plots, showing that concentrations for particles larger than ~50 nm are reasonably comparable, regardless of wind direction. For smaller particles, winds from the airport are associated with much higher particle concentrations.

Table 2: Representation of particle size distributions separated by direction. Northerly winds do not originate from the airport, southerly winds are from the airport. UFP concentrations are clearly much higher from winds associated with the airport.

Survey concentrations	PM_{10} µg/m^3	$PM_{2.5}$ µg/m^3	BC µg/m^3	UVPM µg/m^3	Particle Number Count (PNC) #/cm^3
Average	10.4	6.1	1.8	0.4	40517
Northerly winds	13.9	7.9	2.1	0.4	8511
Southerly winds	8.5	5.1	1.6	0.4	58464

Table 2 shows the high UFP concentrations originating from the airport, compared to other particulate species and other directions, reinforcing the question of how this might impact the local communities.

During this survey, BC, PM_{10} and $PM_{2.5}$ concentrations from off-airport (northerly) winds were higher than when winds were from the airport. Examination of PM and BC polar plots from national network monitoring stations in the south east of England are very similar to those seen at Heathrow, suggesting that the airport is not a significant local source of PM or Black Carbon. Total particle number concentrations show the largest wind dependent differences; concentrations are six times higher when the winds are from the airport, compared to when winds are from other directions. This provides clear evidence, repeated at an increasing number of airports, including airports in Zurich, Boston, Los Angeles, Amsterdam and Lisbon, as well as earlier studies at Heathrow, (17, 27-31) that airport activities are a major source of particle number concentrations. However, we have been able to take our research further, to study plumes from individual aircraft.

We examined the 1 second UFP and aircraft movement data to associate UFP measurements with individual aircraft during take off and landing. We found that UFP concentrations from departing aircraft were much higher than arriving aircraft, and that UFP concentrations from larger aircraft departing were higher than smaller aircraft. The particle number count (PNC) differences between arriving aircraft were not as significant, suggesting that the lower thrust settings used for landing have a significantly reduced impact on particle emissions.

For both arriving and departing aircraft, average particle concentrations peaked for particles sized between 10 and 12 nm. For arriving aircraft, particles sized between 100 and 500 nm are measured in larger numbers than seen during departures. This is most likely due to abrasion (tyres, brakes) and resuspension processes during landing. An increase in BC concentrations which attenuate UV light (termed UVPM, and described further in the method section) has also been detected for arriving aircraft, which we attribute to tyre abrasion since rubber smoke has been observed to have a blue-grey tint. Emission factors for aircraft brake and tyre wear are published by the International Civil Aviation Organization (ICAO) (26).

Emission rates of particle numbers from individual aircraft were estimated by associating high resolution NOx measurements with stated ICAO NOx emission rates and then using this association to calculate total particle number emission rates using the PNC measurements. It was not possible to arrange operation of a CO_2 analyser for this study, as these data would have provided a stronger

relationship with the UFP emission rates. The method for evaluating aircraft NOx contribution is explored further in the Supplementary Information. The calculations here assume that NOx and PN measurements are exclusively associated with combustion. Studies by Ungeheuer et al and Fushimi et al (33,34) have identified unburned lubrication oil is present as nucleation and Aitken sized particles in varying proportions in aircraft exhaust, but it is not possible to estimate these contributions from the measurements made at Heathrow.

The table below presents a comparison of stated ICAO particle number emission rates with calculated values from the measurements. The ICAO emission rates are quoted for non-volatile particles. The calculated total particle number emission rates are estimated from the relationship between ICAO NOx emission rates and the NOx and PNC measurements attributed to each aircraft. Measured PNC includes volatile and semi-volatile particles, as well as the non-volatile particles assessed in the ICAO tests. % Standard Error values for the calculated emission rates are provided in brackets, which shows that generally there is strong confidence in the consistency of the reported data.

Table 3: ICAO and calculated total particle emission rates. Airbus A32x refers to all versions of the A320 and A321 series of aircraft. Airbus A318 results are not reported due to the very small sample collected during this part of the campaign. ICAO emissions rate data quoted for engines at 30% thrust (Arrivals) and 85% (Departures), summed for the number of engines on the aircraft.

Aircraft type	Arrivals ICAO non-volatile PN (# / second per aircraft)	Arrivals calculated PNC (# / second per aircraft) (% Std Err)		Departures ICAO non-volatile PN (# / second per aircraft)	Departures calculated PNC (# / second per aircraft) (% Std Err)
Airbus A319	5.13E+14	3.89E+15 (10.3%)		1.41E+15	9.40E+16 (23.4%)
Airbus A32x	1.78E+15	3.29E+15 (2.3%)		3.12E+15	2.98E+16 (4.1%)
Airbus A330	2.00E+15	9.59E+15 (8.4%)		2.32E+15	1.17E+17 (3.8%)
Airbus A340	No data available	8.72E+15 (10.0%)		No data available	1.11E+17 (8.8%)
Airbus A350	3.44E+15	8.60E+15 (4.3%)		2.04E+15	1.82E+17 (10.5%)
Airbus A380	7.36E+15	1.01E+16 (6.4%)		4.84E+15	1.71E+17 (3.3%)
Boeing 737	2.44E+14	4.18E+15 (5.7%)		2.64E+15	2.82E+16 (6.3%)
Boeing 747	No data available	7.28E+15 (3.6%)		No data available	1.71E+17 (7.2%)
Boeing 757	No data available	4.54E+15 (4.4%)		No data available	3.09E+16 (7.5%)
Boeing 767	No data available	9.67E+15 (4.9%)		3.50E+15	1.28E+17 (4.9%)
Boeing 777	No data available	2.03E+16 (3.1%)		No data available	2.63E+17 (12.5%)
Boeing 787	1.49E+15	1.06E+16 (2.4%)		6.84E+14	1.64E+17 (3.2%)

Following informal discussions with airline pilots from Virgin Atlantic and British Airways, it was discovered that operating procedures for take off are often different to the assumptions made when using the ICAO database. These airlines experiment with thrust settings for take off and climb modes to improve fuel economy. The pilots independently stated that they rarely departed at 100% thrust, using reduced settings, resulting in a longer take off, but reduced fuel use. They said that these operating procedures were reviewed and updated monthly. For this reason, emission rates for the ICAO 85% thrust setting has been used in these comparisons.

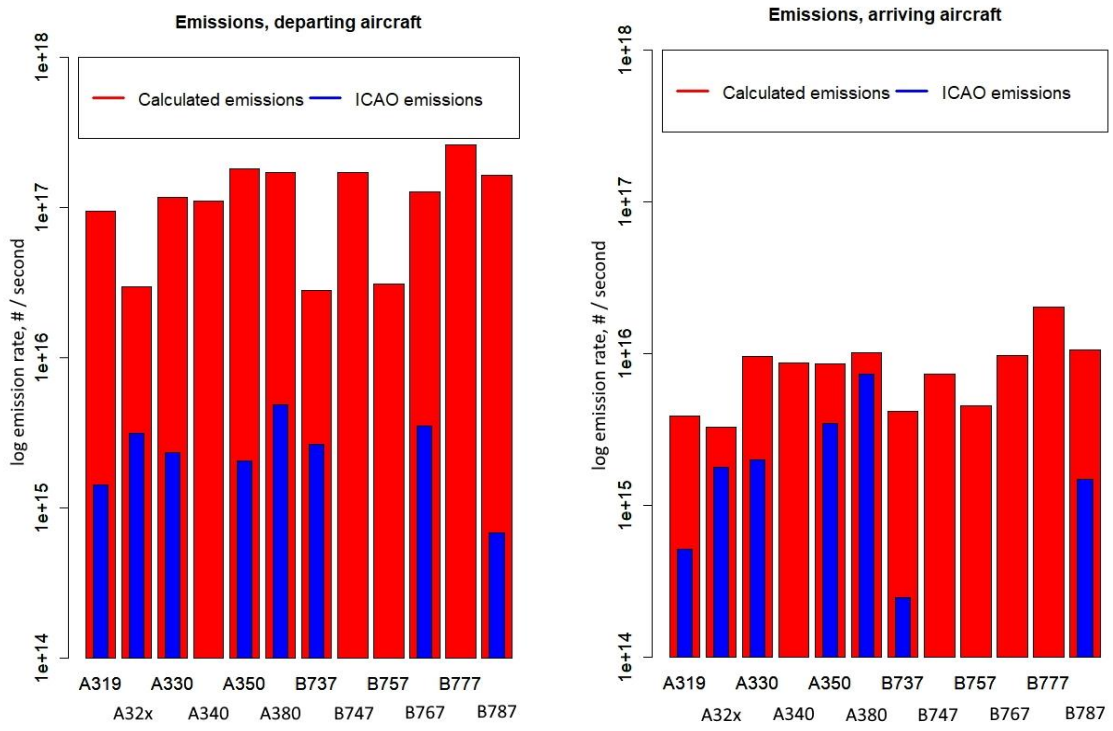

Figure 4: Bar charts of calculated and literature Departure and Arrival particle number emission rates. ICAO emissions data is unavailable for A340, B747, B757 and B777 departures, as well as A340, B747, B757 B767 and B777 arrivals

Figure 4 and Table 3 show a number of different key points. Firstly, and most obviously, the estimated total particle number emission rates are mostly many times higher than the ICAO data for non-volatile particle number emission rates for all aircraft types, both for arrival and departures. This strongly suggests that volatile and semi-volatile particles form the majority of the emissions from aircraft, at least once the exhaust has cooled and mixed in the time taken in transportation of the plume from the runway to the measurement station.

Emission rates from all aircraft are higher for departing aircraft than arriving aircraft. The difference between departing and arriving emission rates is a factor of 2-3 for smaller aircraft and typically 6-8 times higher for the largest aircraft.

The information gathered about UFP for each aircraft type allows us to investigate the data in several different ways. A valuable metric is to look at UFP emission rates per passenger. For this investigation, assumptions are made about the typical seating configurations for each aircraft type, and that aircraft are filled with this allocation of passengers. The assumption that aircraft are always full, together with data screening to remove data where departing and arriving aircraft were not

successfully detected (usually by excluding measurement data below 4000 #/cm^3), will contribute to the uncertainty of the data presented. However, given the size of the measurement dataset, it is likely that the observed differences between aircraft types is genuine.

Table 4: Calculated Total Particle Number emission rates, per passenger for each aircraft type. Airbus A32x refers to all versions of the A320 and A321 series of aircraft.

Aircraft type	Estimated number of passengers	PNC per passenger, Departures, #/sec		PNC per passenger, Arrivals, #/sec
Airbus A319	140	6.7E+14		2.8E+13
Airbus A32x	175	1.7E+14		1.9E+13
Airbus A330	290	4E+14		3.3E+13
Airbus A340	350	3.2E+14		2.5E+13
Airbus A350	330	5.5E+14		2.6E+13
Airbus A380	550	3.1E+14		1.8E+13
Boeing 737	180	1.6E+14		2.3E+13
Boeing 747	420	4.1E+14		1.7E+13
Boeing 757	210	1.5E+14		2.2E+13
Boeing 767	220	5.8E+14		4.4E+13
Boeing 777	330	8E+14		6.1E+13
Boeing 787	275	5.9E+14		3.9E+13

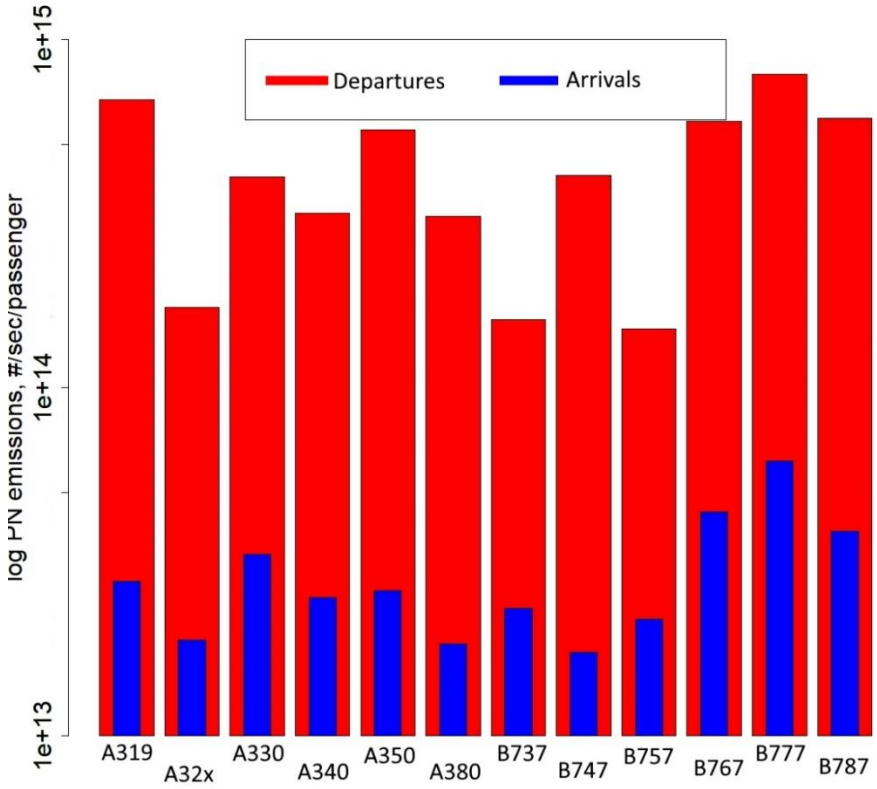

Figure 5: Bar chart plot of emission rates per passenger for arriving and departing aircraft data from Table 3.

Table 4 and the plots in Figure 5 highlight a number of key points. The Boeing 777 aircraft have the highest emission rates per passenger for both arriving and departing aircraft. This is probably due to the age of the fleet in operation at Heathrow. Lowest emission rates from arriving aircraft are seen from the largest aircraft, A380 and 747, a direct result of the large number of passengers carried, though the relative differences between the majority of aircraft types is small (less than a factor of 1.5). Lowest emission rates per passenger from departing aircraft are seen from short haul aircraft, e.g., 737 and A32x. Long haul aircraft have emission rates per passenger at least twice as high as the short haul aircraft. The 787, despite being a modern design, carries relatively fewer passengers and thus a higher PNC emission rate per passenger for both arrival and departure.

This study demonstrates that emissions of UFP from aircraft are a major factor in local air pollution concentrations and emissions near airports (28, 29). Measurements from individual aircraft show that total particle number emission rates per passenger carried are lower for larger aircraft, but calculations show that these aircraft also emit significantly more particles per second. The study also shows that volatile and semi volatile particles, condensing and nucleating as the engine exhausts cool and mix, comprise the majority of the total particle numbers that the general public will be exposed to downwind of the airport.

In September 2021, the World Health Organization (WHO) published updated guidelines for global air quality, (32) which includes a "good practice" statement about UFP exposure levels. "High" levels of UFP constitute concentrations above 10,000 #/cm^3 for a 24 hour average, or above 20,000 #/cm^3 for an hourly mean. The average results for this survey in 2019 indicate that, while the monitoring location is not representative of population exposure, UFP concentrations downwind of the airport are likely to challenge this guidance, especially if they are adopted into target or limit values in future legislation.

6.5 Data Availability

Data supporting this publication are openly available from the UBIRA eData repository at https://doi.org/10.25500/edata.bham.00000749

6.6 Supporting Information

This includes further details of the sampling site and instruments, and some original data, as collected and before processing.

6.7 Conflict of Interest

The authors declare no conflict of interests: The employers of Brian Stacey (Ricardo) are contracted by Heathrow Airport Limited (HAL) to provide air quality monitoring and advice services. HAL also paid the tuition fees of Brian Stacey as a part-time Ph.D. student at the University of Birmingham. HAL did not fund the work reported in this paper, but allowed access to the monitoring sites used to house specialist instruments.

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6.8.1 Author Contributions

BS designed and implemented the measurement campaign, collected and processed the data, undertook the analysis and interpretation of the data and wrote the paper. RMH and FDP provided direction for the analysis and interpretation, reviewed and edited the manuscript.

6.9 Appendix update

A number of amendments have been made to the paper originally submitted for publication. These changes, together with feedback received from reviewers, will be incorporated into the resubmitted paper. The changes are as follows:

Section	Amendment
Throughout	Clarification that emission rates were calculated
6.2	Clarification of ICAO and thermal denuder operating temperatures
6.4	Added information about data capture and minor text clarifications
6.4	Added text to recognise that not all PN may be associated with NOx
6.4	Added some thoughts about how the assumptions about no of passengers and potential for missed aircraft, etc, might impact on confidence in analyses
6.4	Added text to justify choice of 85% thrust setting used for comparison

6.10 Supplemental information

6.10.1 Background information

The monitoring station used for this study is located in the north eastern corner of Heathrow Airport. The station and measurement data can be viewed online (1). The monitoring station is 170m from the middle of the northern runway, positioned in the prevailing wind from the runway and main terminal areas. The station has been used to measure NOx and PM continuously since 1993 and for two UFP measurement studies in 2016 (2) and 2017 (3). Measurement of UFP was undertaken at the Heathrow Airport LHR2 monitoring station between 7th September and 29th October 2019. A Cambustion DMS500 was installed at the station, which also measures NOx, PM_{10}, $PM_{2.5}$, Black Carbon and meteorological measurements. Further details of the analysers and data collected are presented below.

Measurement Techniques

1 minute averaged PM_{10} and $PM_{2.5}$ are measured using a Palas Fidas 200.

1 minute averaged meteorological conditions are measured using a Lufft WS600. Measurements of wind speed/direction, temperature, humidity, atmospheric pressure and rainfall were transmitted to and recorded by the Fidas 200 datalogger.

1 minute averaged Black Carbon (BC) data is measured using a Magee Instruments AE33-7 aethalometer. The analyser collects particles on a glass fibre filter tape while continuously measuring transmission of light through the tape at 7 different light wavelengths (from 370nm to 950nm). Particles are deposited on the filter and analysed continually by the analyser using a dual flow, twin spot approach, that moves the tape before it becomes excessively loaded, eliminating the need for post collection data correction for optical non-linearity. BC measurements are reported 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 – 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 (4):

UVPM = ConcATT370 – ConcATT880 (eq. 1)

Our study at Heathrow in 2017 (3) suggested that tyre smoke from landing aircraft could be detected in the UVPM fraction under certain conditions.

15 minute averaged NOx, NO and NO_2 data are measured using a Teledyne API T200. The analyser uses ozone chemiluminescence to measure NO and a thermal converter to measure NOx, thereby allowing measurement of NO_2 by difference. This is the standard method for measurement of NO_2, described in EN14211 (5). These pollutants are measured at the airport as part of a long term monitoring programme that has been in place since 1992.

1 second UFP data was collected using a Cambustion DMS500 analyser. The analyser uses a corona charger to charge particles before they enter the classifier column, which consists of a central high voltage electrode and a series of 22 grounded electrometers, positioned sequentially in a detector tube. The calculation algorithm chosen to deconvolve the signals and calculate particle number concentrations was the profile for exhaust from Gasoline Direct Injection vehicles, which was expected to be the closest match for the airport environment. It assumes particles are spherical, with two distinct size modes, one for particles larger than 35 nm and one for particles smaller than 35 nm. The analyser was set up to sample undried, undiluted air at 8 L/min and sampled at ambient temperature. The sample will therefore be a broad mixture of volatile, semi-volatile and non-volatile particles, providing a reasonable representation of human exposure to UFP. UFP data was collected over 7 weeks in September and October 2019.

10 second NO and NO_2 data was collected from a South Coast Science Praxis Urban low-cost sensor system. The system uses electrochemical cells from Alphasense, the data were used to produce high resolution NOx measurements to allow the estimation of PNC emissions from aircraft to be undertaken. Measurements were collected for a 2 week period in October 2019.

Analysers used for the survey were operated and calibrated using documented procedures and/ or current accepted best practices. Measurement accuracies and detection limits for each pollutant are presented in Table S1 below:

Pollutant	Accuracy	Limit of detection
NO (API)	±14.0%	±2ppb
NO_2 (API)	±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%	50 particles /cm^3
NO (Praxis)	30%	±5ppb
NO_2 (Praxis)	30%	±5ppb

Table S1 – Accuracy and detection limits for instruments used for the survey.

Fig S1 ¦ Location of Heathrow LHR2 monitoring station in relation to the Northern runway

Fig S2¦ Instrumentation at LHR2. Clockwise from centre left: Magee AE33-7 Aethalometer, Nano-MOUDI cascade impactor sampler, Teledyne API T200 NOx analyser, Fidas 200 PM analyser, Cambustion DMS500 UFP analyser.

6.10.2 Measurement data

Hourly averaged data for the survey is presented in the timeseries below:

Fig S3 ¦ Hourly timeseries data for LHR2, Sep-Oct 2019. PM_{10}, $PM_{2.5}$ Black Carbon and UV Particulate Matter (UVPM) are all in units of µg/m^3, Particle Number Concentrations in #/cm^3

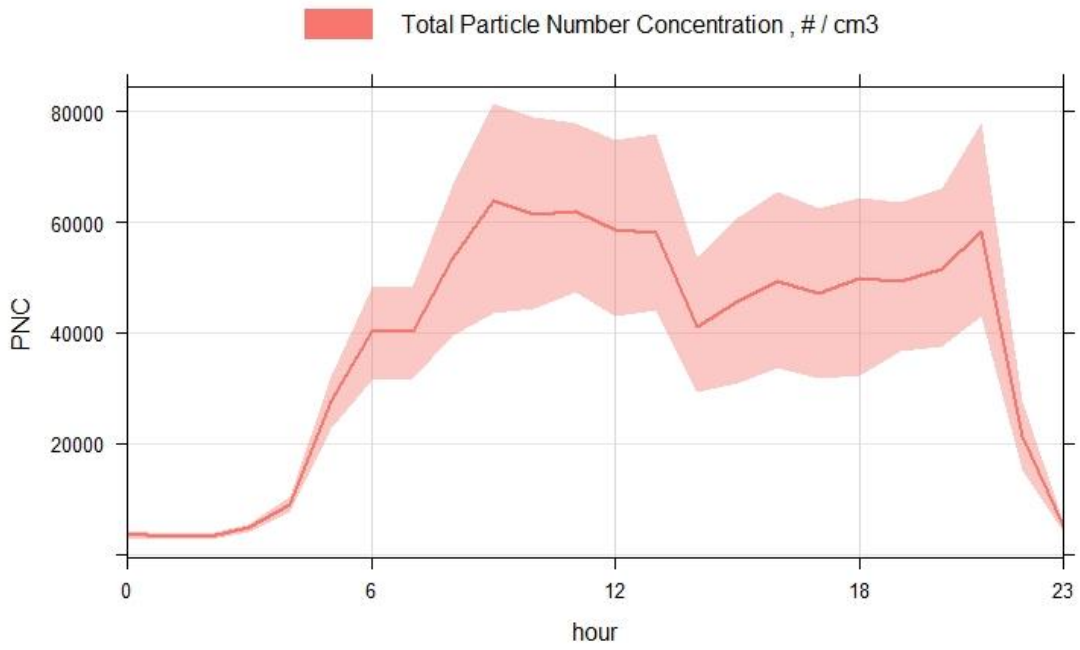

Fig S4¦ Diurnal plot of Particle Number Concentrations at Heathrow, September and October 2019

Figure S3 shows that PNC concentrations are elevated when winds are from the direction of the airport, when aircraft are on the northern runway and highest when aircraft are departing. The periods where aircraft are operating closest to the monitoring station can clearly be seen in the PNC plots, along with key features such as overnight (when there is no aircraft activity, shown more clearly in Figure S4) and when aircraft departed during easterly winds (e.g. 15 to 22 Sep). The PNC concentrations show no significant relationship to the PM measurements recorded at LHR2, demonstrating that PM concentrations at Heathrow are not typically driven by aircraft emissions.

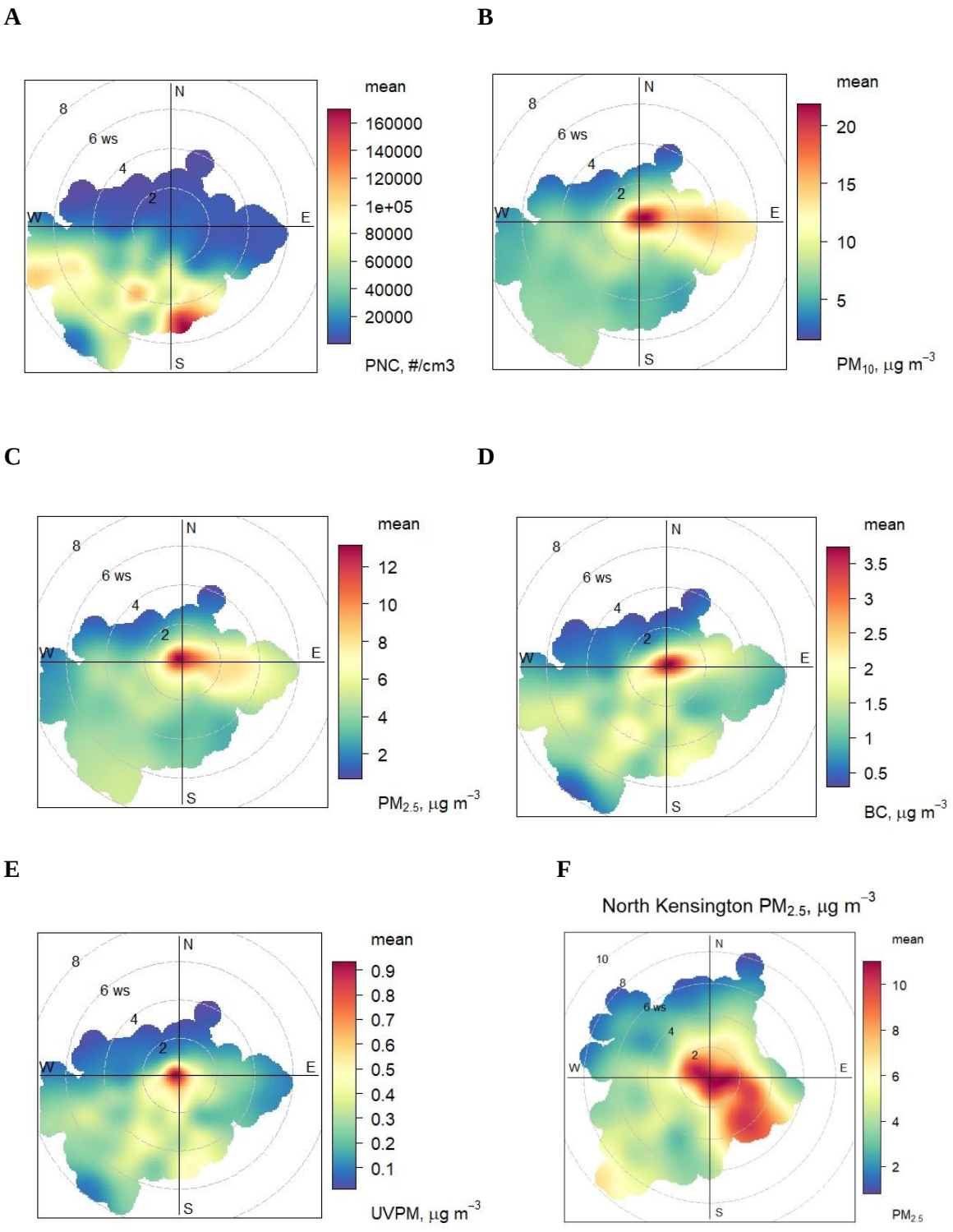

Fig S5 ¦ Polar plots of pollutants at LHR2 **A** Particle Number Concentration, **B** PM_{10}, **C** $PM_{2.5}$, **D** Black Carbon, **E** UV Particulate Matter, **F** PM2.5 at the London North Kensington monitoring station

The above polar plots show the wind speeds and directions associated with the highest pollutant concentrations. PNC is clearly dominated by the contribution from the airport, while PM_{10} and $PM_{2.5}$ have largest influences from the east of the airport and when windspeeds are very low. Plot F shows

the $PM_{2.5}$ polar plot at North Kensington, which shows highest concentrations are also largely associated with easterly winds. BC and UVPM both have clear influence from the airport, but concentrations are highest when wind speeds are very low.

10 second measurements of NO and NO_2 from a South Coast Science Praxis Urban sensor system were used to correlate NOx and PNC measurements with individual aircraft. Data from the sensors was collected between 13 and 26 October 2019. The 10 second measurements from the NO and NO_2 sensors were first processed and scaled, aggregated to 15 minute averages, then compared against the ratified reference NOx data from the monitoring station. The timeseries plot below shows the correlation between the two datasets.

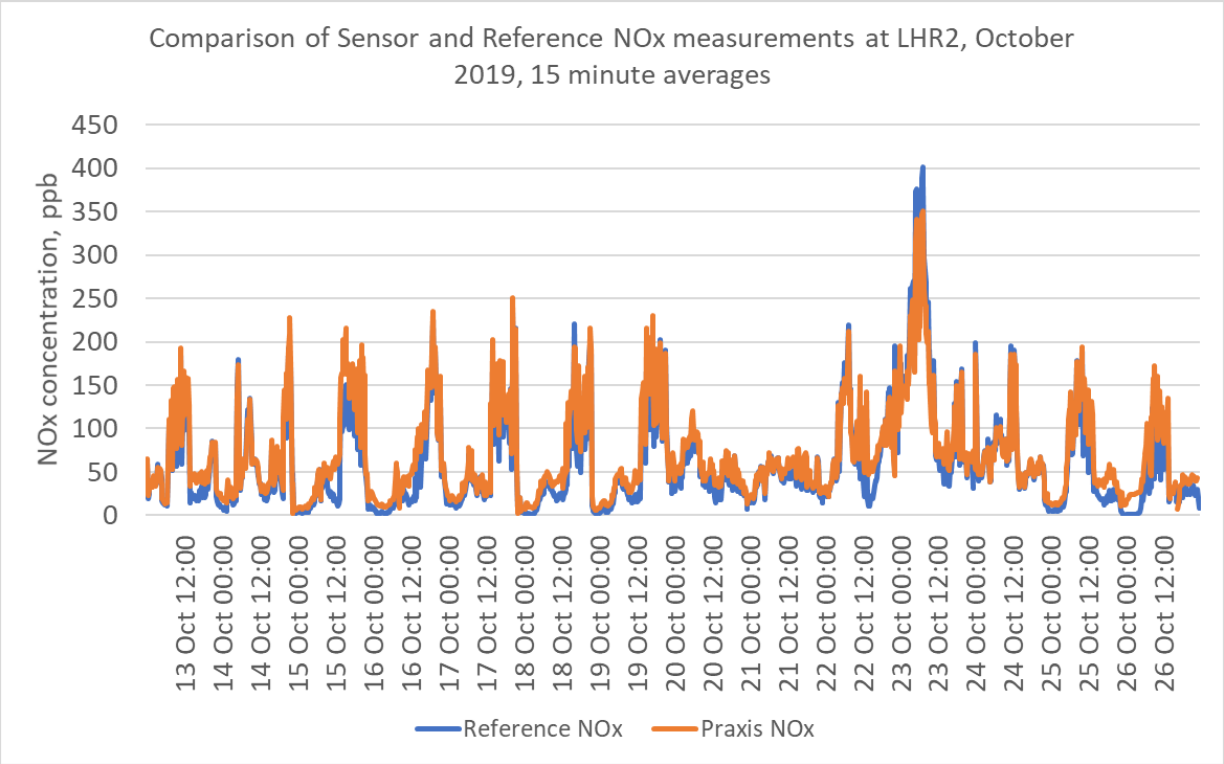

Fig S6 ¦ Timeseries plot comparing NOx measurements from Teledyne API T200 and South Coast Science Praxis Urban analysers.

The agreement between these two different measurement techniques is sufficiently strong (r^2 = 0.93, slope = 0.93) to allow the 10 second NOx data from the sensors to be used to link measurements to emission rates from individual aircraft, this point is discussed fully in the main text.

6.10.3 Analysis of measurement data

The UFP, meteorology and aircraft movement data were investigated to assess differences between aircraft types and arrival / departure modes. As noted in the main text, only arrival and departure movements on the northern runway, 27R, were used for the analysis, during dry weather and when winds were directly from the runway to the monitoring station. Measurements were uniquely assigned to individual aircraft and then clustered by operating mode and aircraft type.

Aircraft type	Average PNC Arrivals, #/cm^3 (Std E)	Average PNC Departures, #/cm^3 (Std E)
Airbus A32x	54100 (255)	113739 (639)
Airbus A330	61405 (1118)	167513 (3700)
Airbus A340	54324 (4722)	161878 (5411)
Airbus A350	53751 (1234)	194349 (6832)
Airbus A380	60843 (1519)	221497 (5214)
Boeing 737	67454 (1548)	126117 (3669)
Boeing 747	58328 (1147)	220923 (3523)
Boeing 757	68367 (3066)	116812 (6773)
Boeing 767	57875 (1086)	144115 (5483)
Boeing 777	56164 (666)	183505 (1819)
Boeing 787	56511 (715)	184916 (2601)

Table S2¦ Table of measured UFP concentrations, separated into arrivals and departures. Airbus A32x refers to all versions of the A318, A319, A320 and A321 series of aircraft. Total particle number concentrations reported are calculated from the sum of all UFP channel measurements uniquely assigned to a specific aircraft type active on the northern runway, once all potential sources of interferences are removed. Standard Error associated with the measurements is reported in brackets. The low standard errors, as a proportion of the total concentrations, provides confidence in the quality of the measurements.

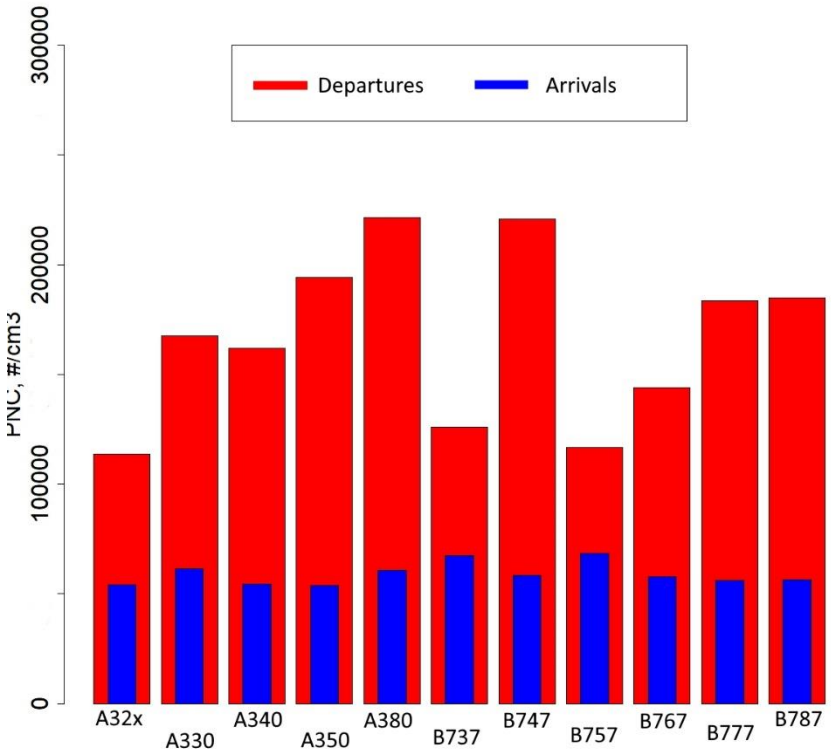

Fig S7¦ Bar chart plot of measured UFP concentrations from Table S2., sorted by largest averaged measurements on departure.

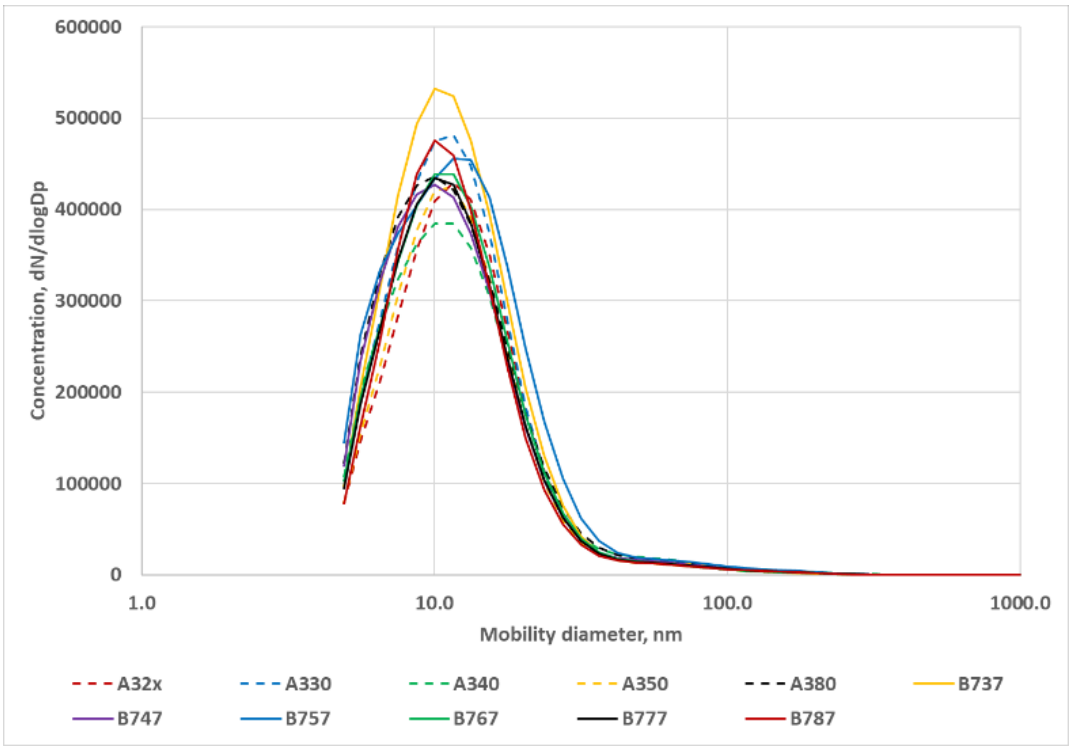

Fig S8¦ Particle Size Distribution (PSD) plots for arriving aircraft, separated by aircraft type.

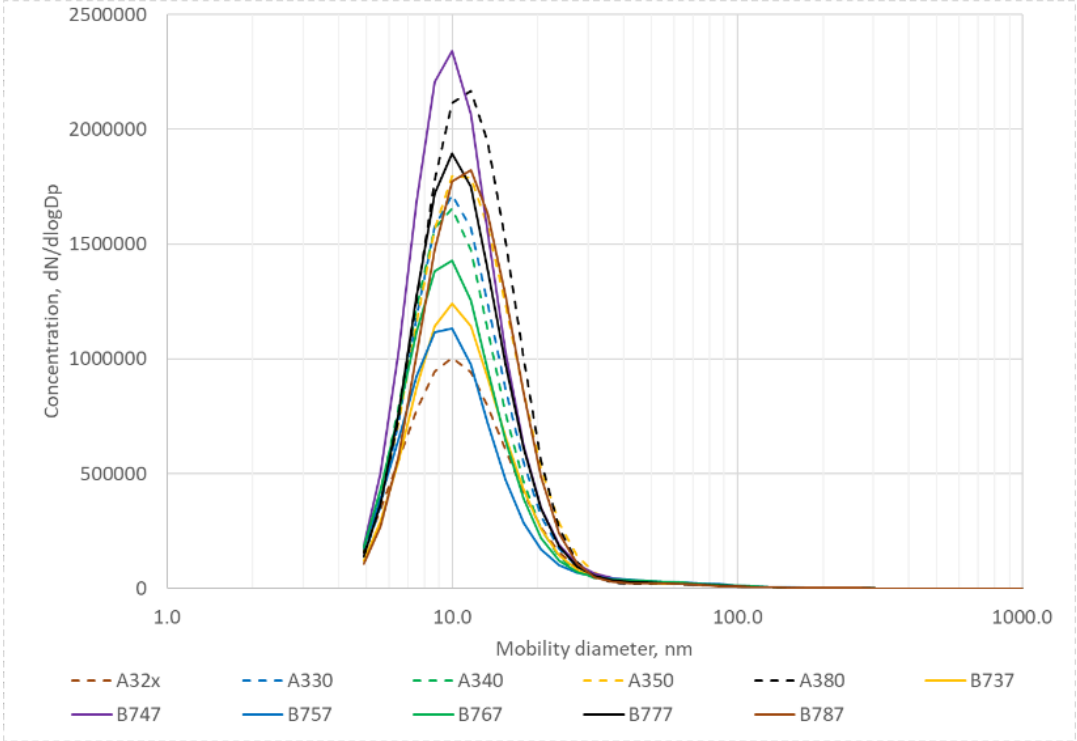

Fig S9¦ PSD plots for departing aircraft, separated by aircraft type.

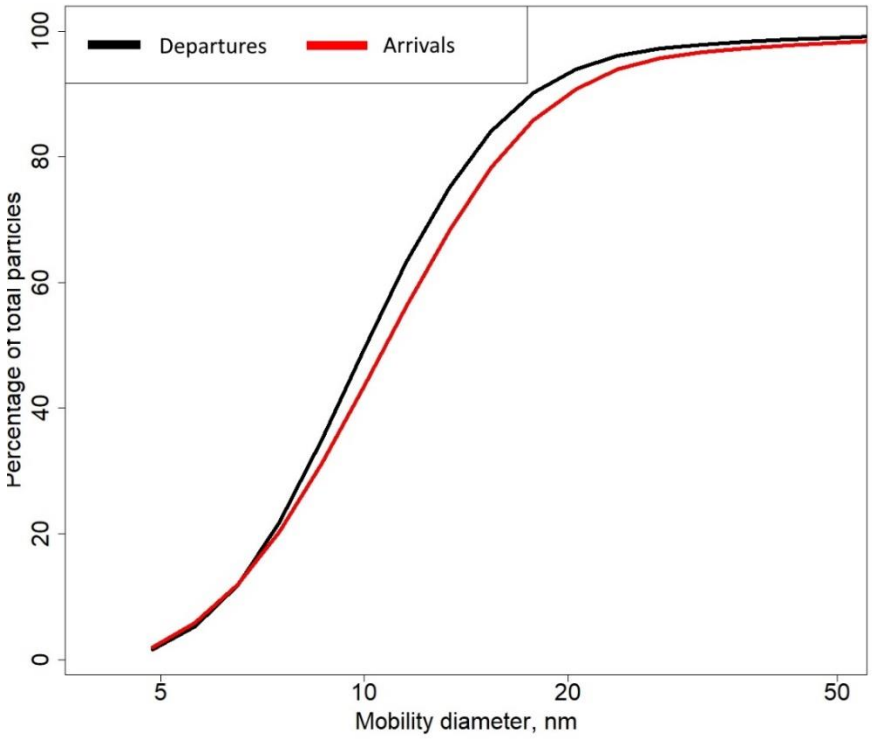

Fig S10¦ Cumulative frequency plot of UFP from departing and arriving aircraft. The plot shows that departing aircraft are not only associated with higher concentrations, but the particles are smaller than those measured from arriving aircraft

Table S2 shows how total particle number concentrations differ between arriving and departing aircraft. The data confirm earlier observations by this research team at Heathrow that departing aircraft are associated with higher UFP concentrations than arriving aircraft. For short haul aircraft, measured concentrations from departing aircraft are approximately twice as high as arriving, whereas for long haul aircraft, the difference between arrival and departure is 3-4 times. When viewing all aircraft on arrival, the spread of PN concentrations between aircraft types is reasonably small (average ~59000 #/cm^3, +/- 9000). For departing aircraft, the spread is much greater (~163000 #/cm^3, +/- 60000). This is certainly an indication of the increased energy required for departure of a larger aircraft, and the observation is a likely artifact of the proximity of the monitoring station to the emission sources. It is likely that dispersion and dilution would remove the ability to resolve the measurement differences between aircraft type and operating modes. Figure S7 presents the results in Table S2, sorted in order of largest measurements for departing aircraft, where the higher measurements from larger aircraft can clearly be seen.

Figure S8 shows the particle size distribution for arriving aircraft. There are clear similarities between aircraft types, both in the particle size distributions, reaching maximum concentrations at a particle size between 10 to 12 nm and in the magnitude of peak concentrations, all essentially reaching maximum concentrations between 40 - 50,000 #/cm^3.

Figure S9 shows the particle size distribution for departing aircraft. There is again clear similarity in the particle size distributions, reaching maximum concentrations at a particle size between 10 to 12 nm. The dependency of concentration with departing aircraft size is also clear here, Boeing 747 and Airbus A380 measurements are associated with the highest particle number concentrations. Both the 747 and A380 are wide-bodied (two aisle) aircraft with four wing mounted engines.

Figure S10 provides summary analysis of the particle size distribution profiles. It shows that in all cases, 90% of the total particles of departing aircraft are smaller than 17nm, whereas for arriving aircraft, particles are measurably larger: 90% of the particles are smaller than 21nm. This is further evidence that engine thrust settings have a marked effect on the nature of emitted particles.

6.10.4 References

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7. Conclusions

7.1 UFP near airports is higher than typical urban areas

My work in 2016 demonstrated that UFP at airports is comparable to measurements at the busiest kerbside location in Central London. But this only tells half the story – Airport UFP is dominated by the particles in the range 15-30 nm, whereas the kerbside site particles are larger. The particle size distribution profiles for kerbside, urban and rural backgrounds all follow a very similar profile to each other (differentiated by concentration), but the airport environment profile follows a completely different profile for particles smaller than 50 nm. Because the instrumentation and QA/QC for all five monitoring stations was effectively the same, these concurrent measurements identify the airport environment as significantly different for UFP.

7.2 Aircraft are the source of high UFP

The UFP and meteorological data collected from my work in 2016 and 2017 shows that aircraft are the dominant source of UFP close to the airport. Polar plot analyses showed that the airport was the dominant source of <50 nm particles. This was seen at both locations in 2016 – when winds were south westerly, UFP concentrations at LHR2 were high. When winds were northerly, UFP concentrations at the Oaks Road station were high. This anti-correlation between stations is a further demonstration of the main source of UFP. The airport contribution was confirmed further in 2019. In addition to the main runway contribution, a further source was identified to the south of the monitoring station. This was found to be due to the opening of a large aircraft stand in 2018, close to the Terminal 2 building. This highlights the ever changing nature of operations at a major airport, and a need to maintain knowledge of these activities.

7.3 UFP from departing aircraft is higher than arriving aircraft

The LHR2 monitoring station permitted further analysis of the 2016 and 2017 data polar plots, accounting for time of day and aircraft operating modes, showed that UFP measurements from

departing aircraft is higher than the measurements for arriving aircraft. When aircraft were not operating overnight, UFP measurements returned to "typical" background concentrations. This observation provides clear evidence that aircraft are the source of the elevated UFP measurements.

7.4 Larger, older aircraft are (usually) associated with high UFP measurements

Investigation of the measurement data with aircraft movements on the runway showed that highest UFP concentrations are typically associated with aircraft with 4 engines, larger aircraft and older aircraft fleet. This is most obvious for measurements associated with departing aircraft. The trend for arriving aircraft is not as clear, it is likely that smaller aircraft need to operate at a higher engine thrust to improve maneuverability on arrival, compared to larger aircraft. Even when the number of passengers is considered, UFP measurements per passenger still follows a similar profile, with the exception of the Airbus A380, which has a very large passenger capacity compared to all other commercial airliners.

7.5 Aircraft UFP emissions are higher than literature values

High resolution NOx measurements were used to correlate to NOx emission rates from individual aircraft on the runway. This correlation was used to calculate UFP emission rates with measurements, which showed that, generally, measured UFP from aircraft is approximately an order of magnitude higher than the quoted values in the CAEP engine emission database. This is explained in the different measurement strategies – The latest CAEP standard (CAEP/10) requires the sample temperature to be maintained at 165 °C and close to the engine exhaust. Volatile removal systems for analytical equipment tend to follow the definition that non-volatile particles remain solid at 350 °C. In contrast to this non-volatile metric, The studies in this thesis were undertaken at ambient temperature and report both volatile and non-volatile particle numbers. The condensable aerosols were seen to form the vast majority of particles measured at the airport perimeter and are not currently part of any assessment of aircraft engine emissions.

7.6 UFP measurements near airports exceed WHO guidance

WHO "best practice" suggests that particle number concentrations (> 10 nm) are considered to be high for daily averages higher than 10,000 #/cm^3 and high for hourly averages higher than 20,000 #/cm^3. Measurements recorded at Heathrow at all three campaigns have exceeded these recommendations, suggesting that UFP near airports is going to become a key focus for air quality in the near future.

7.7 Lack of comparability between measurements hampers research

The biggest challenge associated with measuring UFP is the choice of instrument to make the recordings. There are a wide range of techniques available to "measure" UFP, a number of different ways to report the data, a multitude of possibilities for conditioning (or not conditioning) the sample before measurement, a range of methodologies and materials to "calibrate" the instruments and a range of methodologies to deconvolve the measurement signals to report particle size distribution. All of these permutations, together with the absence of documented quality control in many published papers to date, means that it is virtually impossible to confidently compare measurements from different researchers, as reported in my review paper of 2019. If data comparisons are difficult, this has bigger implications for health impact assessments.

These difficulties are obvious within the context of the three studies presented in my thesis. The 2016 and 2017 studies used notionally the same instrumentation, but changes to the scanned particle size range, scan times and flow rates had such a marked effect on particle behaviour, that the 2 datasets could not be directly compared, either for size distribution or particle numbers. Similarly, the 1Hz measurements from the Cambustion in 2019 are of limited value in quantitative comparison, because the sample conditioning and detection principles are so different and very little work has been undertaken to harmonise these measurements in a real world environment. Despite these shortcomings, research studies – this thesis included – show qualitative similarity in particle size distribution and particle number and continue to demonstrate that UFP emissions and measurements associated with the aviation industry are significant. Further quantitative and harmonisation work is

required to better understand the airport environment. These are important observations that should be considered in detail for future measurement campaigns if context is a major driver in reporting. Currently, the availability of reference-quality PNC and PSD data is very low. In the UK, for example, at the start of 2022 there were three national network monitoring stations measuring PNC and PSD, with a fourth measuring PNC only. There are expansion plans for data from three research stations to be incorporated into the national network in 2022, which will employ similar QA/QC procedures. However, none of these seven sites target measurements near airports, and only one measures close to roadside. The situation is similar across Europe and is not helped by the significant investment required to buy and operate this equipment. Until UFP measurements are available at appropriate data quality and in sufficient density, the ability to conduct high quality health impact research is going to be limited.

8. Future Research

It is clear is that UFP near airports is an ongoing topic of considerable interest for the aviation industry and health impact assessment.

More work and research is required in the pursuit of a definitive understanding of the nature of UFP near airports. This work could include:

- High time resolution CO_2 data combined with UFP and aircraft movements to verify the UFP emissions calculations derived from NOx data.
- Measurement of Total PNC, PNC at 350 °C and thereby measurement of condensable PNC. High concentrations of condensable particles are expected from the 2019 data and emissions calculations, but this has not yet been confirmed with a measurement campaign. The nature of these particles will be of enormous interest for assessing health impact.
- Particle speciation. As with PM_{10} and $PM_{2.5}$, the size-specific composition of UFP is poorly understood, especially close to airports and more research is warranted.
- Harmonisation of data from different instruments and measurement techniques. Unlike calibration of more conventional instrumentation, where standard gas mixtures can be used to reliably scale measurements, UFP analyser data presents some unique challenges. Even before the robust application of Quality Control, different analysers – and even different configurations of notionally identical analysers – will have a radical effect on reported results. Research and publications into these differences is going to be essential if the data are to be meaningfully applied to health assessment studies.
- Impact of alternative fuels on UFP emissions. It is clear that sulphur plays a significant role in the formation of UFP, demonstrated in roadside measurements made during the introduction of Ultra Low Sulphur petrol and diesel in 2007 in the UK and in aircraft engine trials using alternative fuels and blends. Removal of sulphur from aviation fuel is not straightforward: it is an expensive process and the sulphur currently plays a key role in lubrication of seals in the fuel lines. Research to investigate the impact of new fuel blends

and reduced sulphur formulations, along with impacts to the safe storage and supply of fuel to the engines will be of significant value to the aviation industry.

- Behaviour of condensable particles in the atmosphere. Formation and dispersion mechanisms for these particles in the uncontrolled conditions of the ambient environment are not well understood at this time, either chemically or physically. Research into these particles, from aviation and road transport, will play a significant role in driving forward understanding and building robust exposure policies.
- Better understanding of the links between aircraft location, engine thrust settings, engine types and exhaust emissions is needed. At present, information is typically limited to rigid test environments, which may not be an accurate reflection of real world aircraft usage, which is likely to be driven more by economics than any emission reduction strategies.