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Atmospheric Chemistry and Physics Discussions

Interpretation of Particle Number Size Distributions Measured across an Urban Area during the FASTER Campaign

Roy M. Harrison[1*†], David C.S. Beddows[1]
Mohammed S. Alam[1], Ajit Singh[1], James Brean[1],
Ruixin Xu[1], Simone Kotthaus[2] and Sue Grimmond[2]

[1]Division of Environmental Health and Risk Management,
School of Geography, Earth and Environmental Sciences
University of Birmingham
Edgbaston, Birmingham B15 2TT
United Kingdom

[2]Department of Meteorology
University of Reading, Reading RG6 6BB
United Kingdom

[*] To whom correspondence should be addressed.
Tele: +44 121 414 3494; Fax: +44 121 414 3709; Email: r.m.harrison@bham.ac.uk

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

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ABSTRACT

Particle number size distributions have been measured simultaneously by Scanning Mobility Particle Sizers (SMPS) at five sites in Central London for a one month campaign in January – February 2017. These measurements were accompanied by condensation particle counters (CPC) to measure total particle number count at four of the sites and aethalometers measuring Black Carbon (BC) at five sites. The spatial distribution and inter-relationships of the particle size distribution and SMPS total number counts with CPC total number counts and Black Carbon measurements have been analysed in detail as well as variations in the size distributions. One site (Marylebone Road) was in a heavily-trafficked street canyon, one site (Westminster University) was on a rooftop adjacent to the Marylebone Road sampler, a further sampler was located at Regent's University within a major park to the north of Marylebone Road. A fourth sampler was located nearby at 160 m above ground level on the BT tower and a fifth sampler was located 4 km to the west of the main sampling region at North Kensington. Consistent with earlier studies it was found that the mode in the size distribution had shifted to smaller sizes at the Regent's University (park) site, the mean particle shrinkage rate being 0.04 nm s^{-1} with slightly lower values at low wind speeds and some larger values at higher wind speeds. There was evidence of complete evaporation of the semi-volatile nucleation mode under certain conditions at the elevated BT Tower site. Whereas SMPS total count and Black Carbon showed typical traffic-dominated diurnal profiles, the CPC count data typically peaked during nighttime as did CPC/SMPS and CPC/BC ratios. This is thought to be due to the presence of high concentrations of small particles (2.5 – 15 nm diameter) probably arising from condensational growth from traffic emissions during the cooler nighttime conditions. Such behaviour was most marked at the Regent's University and Westminster University sites and less so at Marylebone Road, while at the elevated BT Tower site the ratio of particle number (CPC) to Black Carbon peaked during the morning rush hour and not at nighttime, unlike the other sites. An elevation in nucleation mode particles associated with winds from the

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West and WSW sector was concluded to result from emissions from London Heathrow Airport, despite a distance of 22 km from the Central London sites.

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1. INTRODUCTION

The adverse health consequences of air polluted by particulate matter are now well recognised (WHO, 2006). While the main focus has been on the public health impact of exposure to fine particulate matter measured by mass ($PM_{2.5}$), there has also been concern over the possible contribution of ultrafine particles of less than 100 nm diameter to adverse health outcomes. While such particles contribute little to the total mass of particles in the atmosphere, they dominate particle number (Harrison et al., 2000) and authoritative reviews have concluded that although evidence is currently highly incomplete, they may contribute to the toxic hazard associated with ambient particulate matter (HEI, 2013; WHO, 2013). There have also been suggestions that particle surface area plays a major role in health impacts and this resides largely in the accumulation mode which is typically centred around 100-200 nm diameter (Harrison et al., 2000). Consequently, there is a strong interest from a health perspective in sub-micrometre particles and there are many reports of their concentrations and size distributions within the atmosphere.

In addition to concerns over human health, there are other reasons for the study of the size distribution of airborne particles. Not only does this strongly influence their location and efficiency of deposition in the human lung, the particle size distribution can also be a strong indicator of particle source, with there being some clear differences between the modal diameter of particles arising from different sources (Vu et al., 2015a). The clearest distinction is between particles arising from combustion and other high temperature sources, which tend to be predominantly very small, and particles generated by attrition processes which are typically far more coarse. However, even within the particles generated from combustion and other high temperature sources, there may well be different modal diameters associated with different sources or even multiple modes associated with an individual source (Vu et al., 2015a). For example, exhaust emissions from diesel engines typically comprise both a nucleation mode and an overlapping Aitken mode, reflecting in the former case particles comprised mainly of condensed lubricating oil formed after the

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combustion process, and in the latter case, solid carbonaceous particles formed within the combustion process (Shi and Harrison, 1999; Alam et al., 2016).

After their emission, particle size distributions are also liable to change through dynamic processes. These include evaporation which causes particles to shrink without changing the overall number, condensation which causes particles to grow without a change in total number, coagulation which also causes growth but reduces the total particle number, and deposition which causes a reduction in number and is a strong function of the particle size.

Within this study, particle number size distributions were measured simultaneously by electrical mobility spectrometers at five separate sites across London and the size distributions are compared with a view to gaining a better understanding of the sources and processes affecting particles in the urban atmosphere.

2. EXPERIMENTAL

Data were collected from 27 January 2017 to 16 February 2017 as part of the second campaign of the FASTER project. Data recovery was high at all sites except Westminster University, where good SMPS data were collected on only three days, January 30 and 31 and February 1, 2017.

2.1 Sampling Sites

Data were collected at five sampling sites in total, three of which were established specifically for the FASTER campaign, Westminster University, Regent's University and BT Tower. The other two sites (London Marylebone Road and London North Kensington) collect data as part of the national Automatic Urban and Rural Network. The site locations (seen in Figure 1) and characteristics are as follows:

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- *Marylebone Road*. Air sampling equipment is housed in a large kerbside cabin on the sidewalk of a busy central London street canyon with an inlet approximately 2.5 m above ground-level (agl). The adjacent six-lane highway carries around 80,000 vehicles per day. The highway is relatively straight and runs almost due east-west (angle 80º from north). The buildings on either side of the highway are around six storeys in height giving a street canyon aspect ratio of approximately 1:1.
- *Westminster University*. Air sampling instruments were located on the roof of the Westminster University building, almost directly above the Marylebone Road air sampling site on the southern side of the street. The instruments were housed in a temporary enclosure located approximately 26 m above street level and 4.5 m from the front edge of the roof where it overlooks the road, and with an inlet 1.5 above the roof.
- *Regent's University*. A temporary enclosure for the instruments was located on the roof of Regent's University which is an isolated building within Regent's Park due north (i.e. 360°) of the Marylebone Road and Westminster University sites. The only highway lying between Marylebone Road and the Regent's College site is a lightly trafficked road within Regent's Park. The distance between the Westminster University and Regent's University sites is estimated at 380 m. The instruments were located 16 m agl and 1 m from the edge of the roof.
- *London North Kensington*. Instruments were sited in a permanent cabin located within the grounds of a high school in a lightly trafficked suburban area of central London, with an inlet approximately 2.5 m agl. The air pollution climate at this site, often taken as representative of the background air quality within central London, has been characterised in detail by Bigi and Harrison (2010).
- *BT Tower*. Instruments were sited on level T35 at approximately 160 m agl on a narrow tower which rises well above the surrounding buildings on a quietly trafficked street approximately 380 m to the south of Marylebone Road. The site was used extensively in the REPARTEE experiment (Harrison et al., 2012a).

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2.2 Sampling Instruments

The instruments (Table 1) were operated according to Wiedensohler et al. (2012) guidelines and calibrated and intercompared both before and after the sampling campaign. Small correction factors (< 5%) were applied to CPC (condensation particle counter) data as a result of the intercomparison. SMPS (scanning mobility particle sizers) data were analysed using the AIM9 and AIM10 software provided by TSI as appropriate to the instrument. The national network sites (Marylebone Road and North Kensington) are fitted with diffusion dryers according to EUSAAR/ACTRIS protocols (Wiedensohler et al., 2012), but the other sites were not. The particle size ranges measured were 14.9-615.3 nm at Westminster University, Regent's University and BT Tower, 16.55-604.3 nm at Marylebone Road and North Kensington, and a further system with a short DMA (differential mobility analyses) gave 4.96-145.9 nm at Regent's University.

It was not possible to use identical SMPS systems at each site. The variants used are shown in Table 1. We expect little difference between the long column classifiers (TSI 3081) used at all sites but with different platforms (TSI 3080 and TSI 3082) and CPCs (TSI 3775 and 3776). Differences are expected to be minimal as platform-specific software was used to invert the data and both the CPC are butanol-based, with only slightly different lower cut-points which were well outside of the range of measured particles. At the Regent's University site, both a long DMA (3081) and short column DMA (3085) were utilised and the data were merged to give a single continuous size distribution from 6 nm to 650 nm. A possible cause of divergence is the fact that two of the sites (Marylebone Road and North Kensington) used diffusion dryers according to the EUSAAR/ ACTRIS Protocol. The dryers were tested when installed and showed very low particle losses (less than 5%) and no significant change to particle size distributions (NPL, 2010). The dryer may, however, affect the particle size distribution due to the hygroscopicity of certain kinds of particles. Vu et al. (2015b) reviewed hygroscopic growth factors for submicron aerosols from different sources. Their data are difficult to extrapolate to this study as measurements of hygroscopic growth

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are typically made at very high relative humidities, normally around 90%. Even at 99.5% relative humidity, the growth of particles of less than 100 nm sampled from the atmosphere is relatively low (Vu et al., 2015b). Consequently, a reduction in humidity from 88% typical of the campaign to the values of 30-40% achieved in the dryer would be expected to have only a small effect on particle sizes especially as fresh traffic-generated particles which comprise a large proportion of the sub-micrometre particulate matter in the urban atmosphere are hydrophobic and therefore undergo zero or very limited growth in humid atmospheres.

2.3 Weather Conditions During the Campaign

Wind speed and direction data were taken from Heathrow Airport to the west of London to reflect the synoptic flow minimally affected by local building effects. At the start of the campaign (27 January 2017) the wind direction was easterly and moved to southerly by January 29th, briefly passing through northerly before returning to a southerly circulation between January 31 and February 3rd. During this time, wind speeds were typically around 4 m s^{-1} and temperatures mild for the time of the year (mostly 6-10 ºC). From February 4th to 8th there was a period of lower wind speeds (1-4 m s^{-1}) with variable wind directions and low nocturnal minima temperatures (down to 1ºC). From Feburary 8 – 12th, a period of northerly winds (speeds of 3-5 m s^{-1}) and lower temperatures (1-3ºC) without appreciable diurnal variation occurred. After February 12th, the winds came from the east moving to south-westerly by February 17th, with wind speeds variable (between 0 and 6 m s^{-1}) and temperatures steadily rising to daily maxima of 12ºC.

The mixed layer heights (MLH) were determined from Vaisala CL31 ceilometer data collected at the Marylebone Road site (Figure 1, Table 1). The observed 15 s (10 m gates) aerosol attenuated backscatter profiles were pre-processed (Kotthaus et al., 2016) prior to using the CABAM algorithm (Kotthaus and Grimmond, 2018) to determine 15 min intervals MLH. The multiple aerosol layers (e.g. nocturnal residual layers) in the atmosphere are detected (Kotthaus and

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Grimmond, 2018; Kotthaus et al., 2018). Here the lowest detected layer is analysed. At times the MLH cannot be detected (e.g. during rain or very weak gradients in attenuated backscatter), but a residual layer might still be indicated. The ceilometer detects periods of precipitation, including events that may not be recorded by ground-based stations (e.g. insufficient to trigger a tipping bucket rain-gauge).

During the campaign the observed MLH varied from a daily minimum of 45 m agl to a daily maximum of 1312 m agl with an overall 15 min average (median) of 421 (382) m agl. The daily average (median) maximum MLH was 777 (695) and minimum was 194 (197) m agl. The daily range and the amount of data available per day are shown in Figure S10.

2.4 Modal Analysis of Size Distributions

Modes were fitted to the 15 min data obtained at Marylebone Road, Regent's and Westminster Universities using curve fitting and data analysis software "Fityk (version 1.3.1)" developed by Wojdyr (2010). In the present analysis, a standard peak function (equation 1) was used to disaggregate the size distributions into lognormal modes:

$$P_i = A_i \cdot exp\left[-\left(\frac{\ln(D/c_i)}{W_i}\right)^2\right] \quad (1)$$

By fitting linear a combination of n peaks ($P_1 + P_2 + \ldots + P\text{i} + \ldots + P\text{n}$) to the number size distributions, the following information was calculated: 1) amplitude A_i and location of dN/dlogD at the mode of the distribution c_i, 2) area under the curve (nm cm^{-3}), and 3) width of the lognormal curve W_i.

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3. RESULTS AND DISCUSSION

3.1 Particle Size Distributions

A time series of total particle number concentrations from the SMPS instruments appears in Figure 2. A strong diurnal variation is seen at all sites and is exemplified by the average daily variation shown in Figure 3.

The data stratified by LHR wind direction (Figure 4) were used to perform the modal analysis. The log normal modes fit to the size distribution were used to provide insights into the separate modes contributing to a measured size distribution. Although most measurements could be fit with three separate modes some distributions were best fit with only two modes. An example of a three mode fit of a size distribution from North Kensington appears in the data for the 270º wind sector at this site (Figure 5). It may be seen that using three modes gives a very good overall fit to the data. The details of the modes fitted and their relative magnitude and breadth appear in Table S1.

The Marylebone Road sampling site is located in a heavily trafficked (approx. 80,000 vehicles per day) street canyon. The canyon is aligned almost east-west and the sampling site is at kerbside on the southern side of the street. The canyon has a height to width ratio of ~1 consequently we expect skimming flow when flow is perpendicular, with one or more vortices established in the canyon (Oke et al. 2017). When there is one vortex, the sampler is exposed to freshly emitted traffic contaminants when the wind above the canyon is from the south (Figure 6). Particle number concentration on Marylebone Road is highest for the 225º and 270º wind sectors (Figure 4a) when traffic-generated pollutants are carried efficiently to the sampler. When winds have a northerly component such as those for 0º and 45º in Figure 4a, the air reaching the sampler is typical of background air from north London and peak concentrations fall by a substantial margin. The particle size data from Marylebone Road (Table S1) show no strong effect of wind direction on the modal diameter for the first fitted mode in the distribution. The average diameter for the 180 and

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225º wind sectors are 21.4 nm while for the 0 and 45º sectors they are 22.9 nm. The second and third mode in the distribution are far more sensitive to wind direction, with the southerly traffic-dominated wind directions showing modes at around 32 and 76 nm as opposed to 56 nm and 263 nm for the northerly mode data. The former values compare well with modes in the number distribution of around 20 nm and 50 nm previously attributed to the nucleation mode and Aitken mode particles respectively from engine exhaust when sampled at Marylebone Road, with data analysed by Positive Matrix Factorization (Harrison et al., 2011).

The Westminster University sampling site is 26 m higher and slightly displaced (~8 m) horizontally from the Marylebone Road air sampling station. The observations at roof level are influenced by the flow separation over the roof, if the air is entering or exiting the canyon, and the background concentrations. The particle size data (Table S1) indicate a nucleation mode very similar in size to that observed within the street canyon at the Marylebone Road site. Concentrations are elevated for the 135 and 180º wind bearings suggesting that enhanced concentrations occurring within the canyon on southerly winds are also elevated at the Westminster University sampler but the dataset is very small and hence not included in Figure 4. The second mode appears to be broadly similar in size to that at Marylebone Road and falls within the range of modal diameters measured at Marylebone Road. Similarly, the third mode falls within the rather variable range also seen at Marylebone Road.

The North Kensington site is widely taken as representative of the background air pollution climate in central London (Bigi and Harrison, 2010; Bohnenstengel et al., 2015). At this site, the size of the first mode in the size distributions is remarkably constant at 22-26 nm which is slightly larger than that observed at Marylebone Road. The second mode is also less variable than at most other sites and broadly within the range of the second mode sizes at Marylebone Road (see Table S1). The third mode is highly variable in size with wind direction but again broadly comparable to the data

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from Marylebone Road. The Beddows et al. (2015) Positive Matrix Factorization of particle number size distributions data from this site identified four factors contributing to the particle number size distributions: a secondary component accounting for 4.4% of particle number with a mode at around 250 nm, an urban background factor (43% of particle number) peaking at around 50 nm, a traffic component (44.8% of particle number) peaking at around 30 nm and a regional nucleation component (7.8% of particle number) peaking at 20 nm. The regional nucleation component showed a strong seasonality with greatest prevalence in the summer months and is thought unlikely to have contributed significantly during the period of this campaign. Consequently, the first mode observed in our current study is very comparable to the traffic mode observed by Beddows et al. (2015), and the second mode corresponds strongly to the urban background factor identified by Beddows et al. (2015) who associated this factor with aged traffic emissions and wood smoke, the latter of which is unlikely to have influenced the size distribution at Marylebone Road significantly.

3.2 Particle Shrinkage

Previous London work has shown the tendency of nucleation mode traffic-generated particles sampled within Regent's Park to have shrunk by evaporation at rates of on average 0.13 nm s^{-1} (Harrison et al., 2016) while particles in the regional atmosphere typically undergo condensational growth at a rate of about 0.6-0.9 nm h^{-1} (Beddows et al., 2014). This reflects an initial local rapid loss of more volatile hydrocarbons, followed by a subsequent slower condensation of low volatility species formed by atmospheric oxidation in the regional atmosphere.

Under southerly flows the Regent's University site is downwind of Marylebone Road (Fig. 1). The modal diameters measured at Regent's University in the nucleation mode (Table S1) are clearly indicative of a shrinkage of particle diameter for the wind sectors 180º, 225º and 270º, corresponding to air having passed over Marylebone Road. These data show that the nucleation

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284 mode is shrinking from a diameter in the range of 21-24 nm at Marylebone Road, and 22-24 nm at
285 Westminster University to a diameter of 14, 9 or 12 nm at the Regent's University site. In this case,
286 particle shrinkage seems to be limited to those three wind sectors, with possibly some shrinkage in
287 the 45º wind sector, but particles in other wind sectors retain broadly similar diameters to those
288 measured at Marylebone Road and Westminster University. The second particle mode and third
289 particle mode (where identifiable) at Regent's University are broadly similar and considerably
290 larger than those measured at Marylebone Road or in the limited dataset at Westminster University.
291
292 In our earlier studies of the evolution of particle sizes between Marylebone Road and Regent's Park
293 (Harrison et al., 2016), the nucleation mode in the Marylebone Road size distributions lay between
294 20-24 nm (i.e. very similar to this study). In Regent's Park this had reduced to within the range of
295 6-11 nm with the largest sizes measured in the 0º wind sector and the smallest in the 180º wind
296 sector. The current data show a similar general pattern, although the extent of size reduction is
297 smaller. The travel distance to the Regent's University site is shorter, hence accounting in part for
298 less shrinkage, but the overall shrinkage rate in the current study (0.04 nm s^{-1}) was smaller than
299 previously (0.13 nm s^{-1}) (Harrison et al, 2016). This is probably explained by two factors. Firstly,
300 with warmer mean air temperatures (12-18ºC) evaporation would be enhanced, and secondly, as the
301 site used for collection of the data described in the Harrison et al. (2016) study was in the centre of
302 the park and further from any major highways than the Regent's University site, it may have
303 experienced lower vapour concentrations. Consequently, the two datasets appear highly consistent
304 with one another.
305
306 Previous BT Tower site observations have reported loss of < 20 nm particles (Dall'Osto et al.
307 2011). This loss was greatest when atmospheric turbulence levels were lowest and hence the time
308 for ground to sampling height (160 m) transport greatest. That analysis is not repeated in this study.
309 However, the nucleation mode size (Table S1) has grown slightly from the sizes measured at

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Marylebone Road for the nucleation mode. It is notable that unlike the earlier results, the amplitude of this mode at the BT Tower was substantial and slightly larger than that observed at the ground-level background North Kensington site suggesting that there was generally good coupling between ground-level and the Tower site. It is notable that the first mode diameter with greatest amplitude was for the 270° sector (Figure 4d); this is discussed later. The particle size distribution associated with the 225° wind sector had only one mode at 40 nm suggestive of the second solid particle mode with complete evaporation of the semi-volatile nucleation mode.

Earlier studies have shown that particle number concentrations (< 100 nm) in a street canyon (Olivares et al., 2007) and urban air (Hussein et al., 2006) increase with reducing temperature. This is consistent with the semi-volatility of nucleation mode particles from road traffic (Harrison et al., 2016), and consequently it would be expected that the particle size distribution as well as the number concentration would be affected by ambient temperature. To investigate this, the size distributions collected in the lowest quartile of air temperatures (1.1 to 3.8ºC) were compared with those in the highest quartile of temperature (9.1 to 11.8ºC). This showed generally higher concentrations associated with the higher temperatures, and a clearer nucleation mode at higher temperatures, at all sites, and most notably at Marylebone Road. Such behaviour is contrary to expectations, as greater evaporative losses would be expected at higher temperatures, reducing the magnitude of the plot, or shifting the mode to smaller sizes. To understand this effect more clearly, wind directions with the coldest and hottest quartiles of temperature are analysed. The coldest periods all occurred during northerly flows (270 to 90º) and >85% of highest quartile of temperatures occur during southerlies (90 to 270º). The behaviour, especially at Marylebone Road and Regent's University therefore appears to be determined predominantly by synoptic wind conditions. For Marylebone Road, the street canyon flow (Figure 6) is the dominant influence and at Regent's University the traffic sources are most proximate with southerly flows.

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3.3 Particle Number Concentration (CPC) Data

Average diurnal variations of total particle number count derived from the Condensation Particle Counters produced using the Openair Software Package (Carslaw and Ropkins, 2012) appear in Figure S1. Perhaps surprisingly, at both Marylebone Road and Westminster University, these show a peak occurs between midnight and 6 am before reducing and then rising to a second peak in the afternoon. CPC concentrations at these sites far exceed those at Regent's University and the BT Tower, whereas integrated counts from the SMPS instruments were considerably smaller and showed a diurnal variation broadly similar to that expected for road traffic emissions (Figure 3). While it is quite normal for the CPC to give a higher count than the SMPS since it measures over a wider size range and may have lower internal losses (although the SMPS data analysis software corrects for internal losses), the ratio of CPC to SMPS is typically around two, but this value was significantly exceeded episodically, especially at Westminster University (Figure S2). The overall pattern of CPC to SMPS ratios (Figure 7) shows that some of the highest ratios were at Regent's University with two individual occasions exceeding 13. Some high peak values were observed at Westminster University during the short SMPS time series. Wood burning is recognised as an influential source of particles in London (Harrison et al, 2012b; Crilley et al., 2015), and has a diurnal profile with higher concentrations typically at night. During the ClearfLo winter campaign the BT Tower was influenced substantially by wood smoke irrespective of boundary layer depth (Crilley et al, 2015). Since the BT Tower site was predominantly within the mixed layer during the 2017 campaign (Figure S10) and the CPC/SMPS average ratios at the Tower show little nocturnal elevation, we consider it unlikely that wood smoke explains our observations. Furthermore, particle size distributions associated with biomass burning are typically larger than those from road traffic, and outside of the sub-15 nm size range (Vu et al., 2015a).

To evaluate this phenomenon more closely, the Black Carbon data were examined. These are typically taken as a good tracer of diesel exhaust which is expected to be the main source of the

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362 particle number count. The diurnal variation in Black Carbon (Figure S3) conformed reasonably
363 well to that expected for a traffic-generated pollutant with Marylebone Road concentrations far
364 exceeding those at the other sites and showing a typical traffic-associated pattern. Particle number
365 (derived from the CPC) to Black Carbon ratio (Figure S4) shows huge diurnal variability similar to
366 that seen in the ratio of particle number count from the CPC to that derived from the SMPS. We
367 infer from this behaviour that a large number of particles smaller than the lower limit of the SMPS
368 and above the lower limit of the CPC (i.e. 2.5-14.9 nm for the 3776 instrument at Westminster
369 University and Regent's University; 4-14.9 nm for 3775 instrument at BT Tower; and 3-16.55 nm
370 for 3025 instrument at Marylebone Road) were present in the atmosphere. Both the mean ratio of
371 CPC to SMPS (Figure S2) and CPC to Black Carbon (Figure S5) have ratios that are greatest in the
372 early morning (midnight to 6 am). This is unexpected for the CPC/SMPS ratio, as the contribution
373 of traffic relative to regional aerosol is expected to be least and the coarser regional aerosol contains
374 few particles in the size range below the lower limit of the SMPS instrument. Similarly, for the
375 Black Carbon data, one would expect that if traffic is the main source of particles measured by the
376 CPC, the latter would show a diurnal fluctuation like that of Black Carbon, which in London arises
377 mostly from traffic emissions. Consequently, it seems likely that nucleation processes favoured by
378 the cooler temperatures and lower condensation sink in the early hours of the morning are creating
379 large numbers of particles in the range of 2.5-15 nm mobility diameter. These are forming as air
380 moves away from the traffic source and hence are greatest at the rooftop Westminster University
381 site and have diminished to some extent by coagulation or re-evaporation by the time they reach the
382 Regent's University site which still shows a marked elevation in particle number to Black
383 Carbon ratio in the earlier hours of the morning compared to the Marylebone Road site.
384
385 Such behaviour is somewhat unexpected and a review of papers in which vertical gradients in
386 particle number count have been measured above roadside sites showed no earlier evidence of such
387 behaviour (Lingard et al., 2006; Agus et al., 2007; Nikolova et al., 2011; Ketzel et al., 2003;

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388 Longley et al., 2003; Kumar et al., 2008a,b; Kumar et al., 2009; Li et al., 2007; Vakeva et al.,
389 1999; Zhu et al., 2002; Wehner et al., 2002). However, evidence is seen in some of Villa et al.'s
390 (2017) observations, particle number count increased with height up to around 10 m above a multi-
391 lane highway. The authors reported this unexpected pattern for some ascents/descents and
392 attributed it to exhaust tubes of heavy duty trucks tending to project vertically upwards and to be
393 located at a height of several metres above ground. They suggest this is not the case in urban
394 canyons.
395
396 Another possibility arises from the report of Rönkkö et al. (2017) that large numbers of sub-4 nm
397 particles are observed in the exhaust of some diesel engines and the observation by Nosko et al.
398 (2017) of substantial numbers of similarly sized particles amongst emissions from brake wear.
399 Kontkanan et al. (2017) reported observations of sub-3 nm particles from many sites, the highest
400 concentrations being in urban locations. The diurnal and regional variations did not relate clearly to
401 photochemistry and it was concluded that sub-3 nm particle concentrations are affected by
402 anthropogenic sources of precursor vapours. The correlation of sub-3 nm particle concentrations in
403 Helsinki with nitrogen oxides suggested a link with traffic emissions. Shi et al. (2001) measured
404 particles of >9.5 nm by SMPS, >7 nm by CPC and >3 nm by ultrafine CPC, finding large numbers
405 of particles in urban air in the ranges 3-7 nm and 3-9.5 nm by differences of counts. Ratios of CPC
406 (>3 nm):SMPS (>9.5 nm) were highly variable, but typically around 4. Clear links to road traffic
407 were seen, with drive-by experiments showing large numbers of particles in the 3-7 nm range in the
408 exhausts of both diesel and gasoline vehicles (Shi et al., 2001). Nanoparticles were also produced
409 in the plume downwind of a stationary combustion source (Shi et al., 2001). Herner et al. (2011)
410 measured the size distribution of particles emitted from vehicles equipped with diesel particle
411 filters, and with diesel filters and selective catalytic reduction. The dominant mode in the size
412 distribution was at 10 nm diameter and comprised particles with a high fraction of sulphate. In
413 highway and roadside measurements in Helsinki, Enroth et al. (2016) measured particle size

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distributions with a dominant mode at 10 nm diameter. Such particles would be largely below the lower threshold for counting by the SMPS but not the CPC. It is plausible that during the cooler hours of the night a tail of <2.5 nm particles might be subject to condensational growth if the co-emitted vapour were to be supersaturated in the atmosphere within the street canyon. The dominance of a 10 nm mode in the size distribution would appear to be the most plausible explanation for the high number concentration of particles observed at the Westminster University rooftop location and the apparent transport of a substantial proportion of such particles to the Regent's University measurement site. While this can explain the typically high CPC/SMPS ratios observed, it does not explain their diurnal variation. This appears to require growth of sub-2.5 nm particles into the range measured by CPC in the cooler, more humid nocturnal conditions. Rönkkö et al. (2006) and Schneider et al. (2005) studied the formation of mechanisms and composition of diesel exhaust nucleation particles in the laboratory and during car chasing. They conclude that formation of nucleation mode particles depends upon formation of sulphate nuclei upon which hydrocarbons condense, consistent with earlier studies of Shi and Harrison (1999) and Shi et al. (2000) conducted in our laboratory. Factors favouring nucleation mode particle formation were found to be low temperature and high humidities, consistent with field measurements made on Marylebone Road (Charron and Harrison, 2003). Both factors prevail at nighttime, probably contributing to the relative increase in 2.5–15 nm diameter particles seen most notably between midnight and 6am (Figure S2). Salimi et al. (2017) reported nocturnal new particle formation events in Brisbane, Australia, finding that air masses associated with nocturnal events were typically transported over the ocean before reaching their sampling site, but the relevance to our study is unclear, although the maritime air might sometimes be expected to show lower temperature and higher humidity than that from the land.

Support for our observations also comes from the very detailed measurement and modelling study of Choi and Paulson (2016). Measuring particle number size distribution downwind of a major

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highway, they found a positive anomaly in particle number within the first 60 m of the plume peak, as the peak for the small particles appeared further downwind than the peak in accumulation mode particles. They attributed this to growth of unmeasured sub-5.6 nm particles into the smallest measurable size range and suggested condensational growth or self-coagulation as the mechanism (Choi and Paulson, 2016). Kerminen et al. (2007) measuring near a major road in Helsinki reported particle growth by condensation to be a dominant process during the road-to-ambient evolution stage at nighttime in winter. They inferred that under such conditions (low wind speeds with a temperature inversion), traffic-generated particle numbers were enhanced and could affect submicron particle number concentrations over large areas around major roads. The distance scales for such processes in both studies (Choi and Paulson, 2016; Kerminen et al., 2007) were within 100 m of source under the conditions of measurement but might conceivably extend over greater distance scales. Similar processes of particle evolution within an aircraft exhaust plume have been reported by Timko et al. (2013).

Pushpawela et al. (2018) report a phenomenon of hygroscopic particle growth at nighttime, which can potentially be mistaken for new particle formation. This phenomenon was observed between 0.5-5.0 hours after sunset, peaking at 3.5 hours (Pushpawela et al., 2018). This would not appear to explain our observations, where the peak in N/SMPS and N/BC plots (Figures S2 and S5) is greatest at 3-4 am local time, which in London in winter is some 10-11 hours after sunset. Additionally, such a phenomenon would be expected to be unrelated to local traffic emissions, and hence more uniform across the various sites.

3.5 Spatial Distribution of Particles – Horizontal and Vertical

Figure 2 shows the time series of particle concentrations from the SMPS instruments throughout the campaign. Clearly, as expected, the Marylebone Road site shows the highest concentrations through the campaign period due to its proximity to the road traffic source. The other sites tend to track one

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466 another quite closely with no consistent ranking of concentrations. There are periods such as
467 February 1st to 3rd when Regent's University well exceeds North Kensington, but at other times,
468 they are very similar (e.g. 10 – 12 February), or periods when North Kensington exceeds Regent's
469 University (e.g. 7 February) but these are few. In the former period (1 – 3 February), winds were
470 southerly and concentrations at Regent's University would be enhanced by passage of air across
471 Central London, including Marylebone Road. In the situation where concentrations were similar (10
472 – 12 February), winds were in the northerly sector, giving relatively low concentrations at all sites,
473 and rather little spatial variation. The temporal pattern at all sites showed substantial similarity
474 overall (Figure 2), including diurnal patterns (Figure 3), although the magnitude of concentrations
475 varied.
476
477 A time series of CPC particle number concentrations (Figure 8) showed that under most conditions ,
478 the number count was lowest at the BT Tower site, and that the number count at Westminster
479 University frequently exceeded that at Marylebone Road, with Regent's University lower, but
480 above the concentration at the BT Tower (Figure 8). During the period of northerly winds (8 – 12
481 February), all sites showed low concentrations with Regent's University and BT Tower similar for
482 much of the time, as for the SMPS data (Figure 2). The highest CPC count concentrations during
483 the latter were measured at Westminster University (Figure 9) which was downwind of Marylebone
484 Road at those times. The similarity seen between Westminster University and Marylebone Road
485 for much of the campaign, with concentrations far in excess of those at BT Tower is strongly
486 suggestive of continuing particle growth into the size range 2.5–14.9 nm at Westminster University
487 with re-evaporation occurring before reaching the elevated BT Tower site, as previously observed
488 by Dall'Osto et al. (2011). Elevations in N/BC data were seen at the BT Tower site (Figure S4 and
489 S5) but these occurred mainly during the morning rush hour period, presumably due to fresh traffic
490 emissions, rather than overnight as at the other sites (Figure S5).
491

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492 Figure 2 suggests that vertical gradients between the proximate Regent's University and BT Tower
493 sites were small in SMPS count (Figure 2), but at certain times were substantial in the CPC count
494 (Figure 9). The particle size distributions measured at the BT Tower (Figure 4d) differ from
495 Marylebone Road and North Kensington (Figure 4a and b) in having no obvious mode in the
496 nucleation size range at 20 – 30 nm, a feature shared with Regent's University (Figure 4c). Only
497 during westerly winds (270°) does the BT Tower show such a mode (Figure 4d), while at Regent's
498 University (Figure 5) the 270° wind direction also shows differences from the others with a mode at
499 below 20 nm. Anomalous behaviour in this wind sector is also observed at North Kensington
500 (Figure 4b), and at Marylebone Road. The most pronounced nucleation mode peak is associated
501 with the 270° and 225° wind directions. In the Marylebone Road case, these wind directions are
502 almost parallel to the highway, which might explain the high concentrations and pronounced
503 nucleation mode, but this explanation does not work for the other sites. A more likely explanation is
504 that all sites are affected by emissions from Heathrow Airport which is to the west of London and
505 has been recognised as a major source of nucleation mode particles associated with aircraft and road
506 traffic emissions (Masiol et al., 2017). At a site 1 km from the northern boundary of Heathrow
507 Airport, PMF factors attributed to aircraft (mode at <20 nm) and fresh road traffic emissions (mode
508 at 18–35 nm) accounted respectively for 31.6% and 27.9% of particle number count in the warm
509 season and 33.1% and 35.2% in the cold season (December 2014 – January 2015) data (Masiol et
510 al., 2017). Heathrow Airport is located approximately 22 km from our Central London sites on a
511 bearing of 255°. Keuken et al. (2015) measured a large elevation in concentrations of particles of
512 10–20 nm diameter attributed to aircraft emissions (emission studies are reviewed by Masiol and
513 Harrison, 2014) at a site 7 km east of Schiphol Airport (Netherlands) and have shown by modelling
514 and measurement that concentrations are elevated to considerably greater downwind distances.
515 Similarly, Hudda et al. (2014) reported PNC to have increased 4 to 5 fold at 8 – 10 km downwind
516 of Los Angeles International Airport (USA).
517

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518 The size distributions have also been analysed according with mixed layer height (MLH),
519 determined by ceilometer (Kotthaus and Grimmond, 2018). Both Marylebone Road (Figure S6) and
520 Regent's University (Figure S7) have the highest concentrations associated with the deepest MLH
521 class (>1000 m). This seems likely to be due to an association with southerly winds and the street
522 canyon circulation. Whereas, North Kensington (Figure S8) has the highest concentrations during
523 shallow MLH (< 100 m and 100 – 200 m) when dispersion is limited for the low altitude emissions.
524 The most interesting behaviour is seen at the elevated (160 m) BT Tower site, which is consistent
525 with Harrison et al. (2012a) and Dall'Osto et al. (2011). During the shallowest MLH (< 100 m) the
526 measurement site is above the inversion and the size distribution lacks an obvious nucleation mode
527 (Figure S9). As the MLH deepens, a nucleation mode appears which dominates the size
528 distribution for the deepest MLH categories (900 – 1000 m and >1000 m) with a mode at 20 – 30
529 nm, similar to that seen at Marylebone Road for the same MLH depths (Figure S6). The gradual
530 transitioning of size distribution as the MLH deepens is consistent with the surface source (mainly
531 road traffic) of nucleation mode particles, and their evaporative loss which increases with the
532 timescale of vertical mixing to the height of the sampler, as reported by Dall'Osto et al. (2011), and
533 the ultimate isolation of the sampler from ground-level emissions at the shallowest boundary layer
534 heights, as observed by Harrison et al. (2012a).
535
536 **3.6 Detailed Comparison of Marylebone Road, Westminster University and Regent's**
537 **University**
538 Unfortunately, a full dataset for the Westminster University site was only collected over the period
539 January 30th to February 1st due to a late set-up of the instrument and a malfunction after February
540 1st. This period however merits closer examination as it is the only period where SMPS data were
541 available for all three sites. For much of the time the SMPS data for the Westminster University
542 site looks surprisingly similar to that of the Marylebone Road site despite the former being on the
543 rooftop and the latter being within the street canyon. A detailed analysis hour by hour showed that

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544 out of 51 hourly observations, in 23 the amplitude of the mode (dN/dlogD) at Westminster
545 University was within ± 20% of that at Marylebone Road while in 25 cases the amplitude was
546 greater at Westminster University than at Marylebone Road, and in just two cases the amplitude
547 was smaller at Westminster University. In an attempt to explain this observation, the
548 meteorological data for the periods of similar magnitude and of different magnitudes were
549 compared but no systematic difference was seen in wind direction, air temperature or relative
550 humidity between any of the periods. Wind directions were generally in a south-easterly to easterly
551 sector, mean temperatures around 8ºC and relative humidity high (85 and 99%). The maximum
552 MLH were low and there was a lot of rain (Figure S10).
553
554 In order to gain further insight, the time series of observations were plotted for this period and
555 appear in Figure 9. The SMPS integrated number counts shown in Figure 9(a) show a remarkable
556 similarity between Marylebone Road, Westminster University and Regent's University. For the
557 first two days, Regent's University concentrations are lower than those from the other two sites,
558 although on the third day they are very similar to those at Westminster University. On the first and
559 last days, the peak concentrations at Marylebone Road exceed those at Westminster University but
560 on the middle day (January 31st) the differences between these two sites are very small. The CPC
561 particle number counts shown in Figure 9(b) are very similar to those at Marylebone Road on the
562 first and last day but exceed those at Marylebone Road on January 31st. Concentrations at Regent's
563 University are typically only around half or less of those measured at Westminster University. The
564 magnitude of the CPC concentrations peaking at over 40,000 cm^{-3} is close to double the integrated
565 SMPS counts which peak at a little over 20,000 cm^{-3} indicating a large number of particles in the
566 size range below 14.9 nm.
567
568 However, the Black Carbon data (Figure 9c) have daytime concentrations at Marylebone Road that
569 far exceed those at Westminster University and Regent's University, the latter sites tracking each

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other and having very similar concentrations. Since Black Carbon can be viewed as a conserved tracer of vehicle emissions over these small time and distance scales, the inference is that particle production must be continuing as the vehicle exhaust mixes upwards from the street canyon Marylebone Road site to the Westminster University rooftop site. The southerly wind directions likely associated with upward flow on the Westminster University canyon wall (Fig. 6) would carry vehicle exhaust past the Marylebone Road measurement station (south side of the road).

Air leaving the canyon and being entrained by the complex building roof flows could expose the Westminster University sampler to air exiting the street canyon and to the general flow towards Regent's University site (Fig. 6 and 1). Such behaviour is consistent with the observations of particle growth in the sub-SMPS size ranges reported in the previous section extending into the SMPS size range. This is similar to behaviour observed by Kerminen et al. (2007) in Helsinki who observed not only possibly evaporation of some particles in the 7–30 nm range, but also on apparent growth of nucleation mode particles into the 30–63 nm size range between sampling points at 9 m and 65 m downwind of a highway. The results in Figure 9 are suggestive of a substantial growth of nuclei into the range of the CPC at Westminster University.

4. CONCLUSIONS

The measurement of particle number size distributions in the atmosphere is resource intensive and there have been rather few studies in which more than two samplers have been operated within a city. Typically if there are two sites, one is a traffic-influenced site and the other urban background. In this study, data have been collected at a total of five sites, although unfortunately the dataset from the Westminster University site is limited to only a few days. Nonetheless, the dataset allows some deep insights into the spatial distribution of particle sizes and number counts not only horizontally but in the vertical dimension. Not unexpectedly, concentrations of particles at the street canyon Marylebone Road site considerably exceed concentrations at other sites, but there are

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nonetheless considerable similarities in diurnal profiles and the magnitude of concentrations at the other, background sites.

One of the main motivating factors for this study was to confirm earlier observations of shrinkage of the nucleation mode particles between traffic emissions on Marylebone Road and the downwind site at Regent's University within Regent's Park. Particle shrinkage was observed within the current study although at a slower mean rate (0.04nm s^{-1}) than in the earlier study (Harrison et al., 2016) in which the mean shrinkage rate was 0.13nm s^{-1}. However, temperatures in the current study all fell below those in the earlier work of Harrison et al. (2016). Other factors may also have been influential. There have been marked changes in the road vehicle fleet in London between the two measurement campaigns. The earlier dataset as reported by Dall'Osto et al. (2011) and Harrison et al. (2016) was collected in 2006 at which time the sulphur content of diesel fuel was regulated at below 50 ppm. Between the two campaigns, the sulphur content of both gasoline and diesel motor fuels was reduced to below 10 ppm sulphur in order to facilitate the introduction of diesel particle filters from 2011 onwards. The incorporation of a diesel particle filter on EURO 5 and EURO 6 vehicles leads to a substantial overall reduction in particulate matter emissions but also a change in the hydrocarbon content of the particles. Secondly, the Regent's Park sampling site used for the 2006 measurements was at about double the distance from Marylebone Road compared to the Regent's University used in the latest study. This would allow for greater dilution of the traffic plume from Marylebone Road and other adjacent highways, leading to a greater reduction in vapour phase hydrocarbons at the more distant site causing an accelerated evaporation process. The reduction in fuel sulphur content in 2007 was accompanied by a marked change in the size distribution of particles emitted from road traffic, including a reduction in the nucleation mode particles (Jones et al., 2012). The work of Dall'Osto et al. (2011) also analysed data from the BT Tower, showing increasing evaporative loss of nucleation mode particles as the travel time from ground level to the sampling site on the Tower became longer with reduced atmospheric turbulence

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levels. Although that phenomenon has not been studied in detail in the latest dataset, the results are clearly consistent with such a process, and with an apparent total loss of the nucleation mode in particles associated with regional pollution sampled when the boundary layer top was below the sampling height on the tower.

Although the phenomenon of particle shrinkage had been seen in earlier work, there were two further major observations made in the current study which were not anticipated. The first, was the clear influence of a major source to the west of London, almost certainly Heathrow Airport, upon concentrations of nucleation mode particles. The association of an enhanced nucleation mode in the 270º or 225º sector is indicative of a major source of very fine particles, and the work of Masiol et al. (2017) at a sampling site close to Heathrow Airport provides strong evidence for major emissions both from aircraft engines and the large volumes of road traffic attracted by the airport. Earlier research by Keuken et al. (2015) and Hudda et al. (2014) gives a clear precedent for measurement of strongly elevated concentrations of very fine particles several kilometres downwind of a major airport, but to our knowledge this is the first observations of concentrations above urban background at a distance of 22 km from the centre of the airport.

The other observation which was wholly unexpected was of the very poor relationship between total particle numbers measured by the Scanning Mobility Particle Sizers and the total particle numbers measured by co-located condensation particle counters. While both the SMPS counts and co-located Black Carbon measurements show a typical road traffic diurnal profile, the CPC data show a quite different diurnal profile peaking at night. This is most evident in the ratios of CPC/SMPS and CPC/BC seen at all sampling sites, with the exception of CPC/BC at the elevated BT Tower site which does not show a nocturnal maximum, but peaks during the morning rush hour period. Earlier studies such as that of Choi and Paulson (2016) and Kerminen et al. (2007) have reported data consistent with such a phenomenon, but with very modest elevations in particle count

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compared to those in the current data. The implication is of the presence of large numbers of particles within the range of 2.5 – 15nm and hence observable with the CPC but below the lower cut of the SMPS. It seems likely that such particles grow at night from vary small nuclei and it seems possible that the exceptional magnitude of this process within London results from the high density of diesel traffic leading to substantial nocturnal concentrations of condensable vapours close to the traffic source. A common feature to such observations appears to be its association with still conditions on winter nights which lead to poor dispersion of vehicle emissions and a pool of vapour co-emitted with traffic particles which becomes supersaturated as it cools in the ambient atmosphere, leading to condensation on small nuclei when the general particle concentrations and hence the condensation sink are relatively low in magnitude.

These very abundant particles within the 2.5 – 15 nm range are likely to prove ephemeral as they would be expected to re-evaporate as the air mass dilutes away from source. However, the health effects of exposure to particles within this range are poorly known and no recommendation can be given as to whether health-related studies would be best to measure the particle size range covered by the SMPS as is most typically performed at present, or whether CPC data going down to smaller particles sizes would be more appropriate.

There are some additional general conclusions from the work. Firstly the results demonstrate the dynamic behaviour of traffic-generated (and other) particles within the urban atmosphere. Our earlier paper (Dall'Osto et al., 2011) referred to "remarkable dynamics", and further remarkable dynamic processes have been observed in the current study. Secondly, as this work has revealed sources and processes that were not originally anticipated, although with the benefit of hindsight it might have been possible to predict them, there is clearly a need for further detailed observational studies of the behaviour of sub-100 nm particles within the urban atmosphere.

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939 **TABLE LEGENDS**
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Site Name	**Marylebone Road**	**Westminster University**	**Regent's University**	**BT Tower**	**North Kensington**
Lat (° N), Long (°W)	51.522530, 0.154611	51.522322.0.15515	51.525542, 0154570	51.521426, 0.138924	51.521082, 0.213403
Height of ground msl (m)	26	26	30	25	23
Height of inlets agl (m)	4	26	17	160	3
Instruments installed	Long_DMA_SMPS/ CPC Vaisala CL31	Long_DMA_SMPS/CPC/ (Micro)Aethalometer/Anemometer	Long_DMA_SMPS/ Short_DMA_SMPS/ CPC/Aethalometer/Anemometer	Long_DMA_SMPS/CPC/ (Micro) Aethalometer/Anemometer	Long_DMA_SMPS Vaisala CL31
Particle spectrometer type	3080+3081+3775	3080+3081+3776	(3082+3081+3775)/(3082+3085+3776)	(3080+3081+3775)	(3080+3081+3775)
Aerosol dryer	Yes	No	No	No	Yes
CPC type	TSI 3025	TSI 3776	TSI 3776	TSI 3775	None

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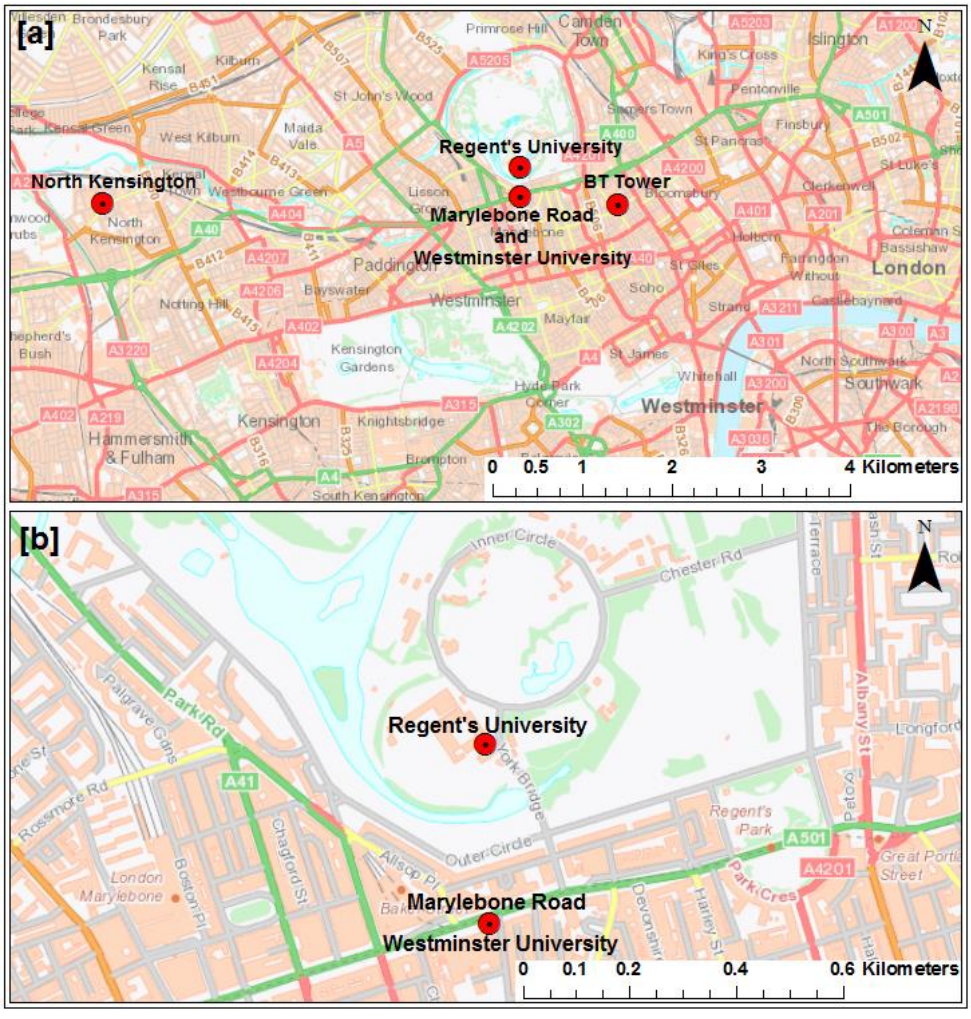

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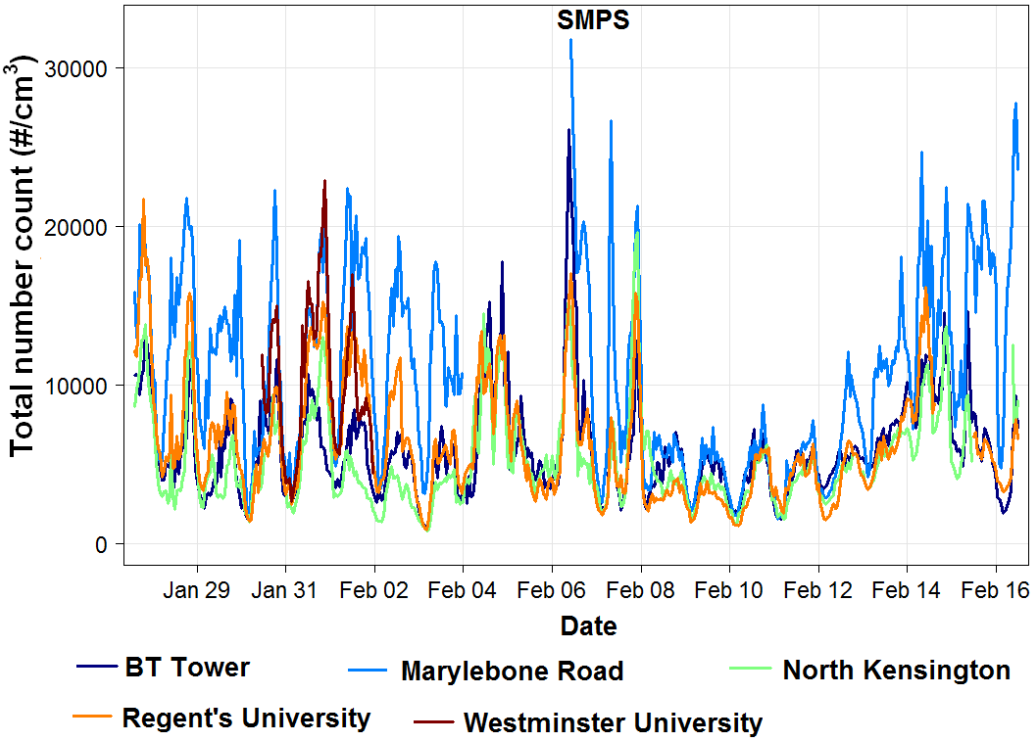

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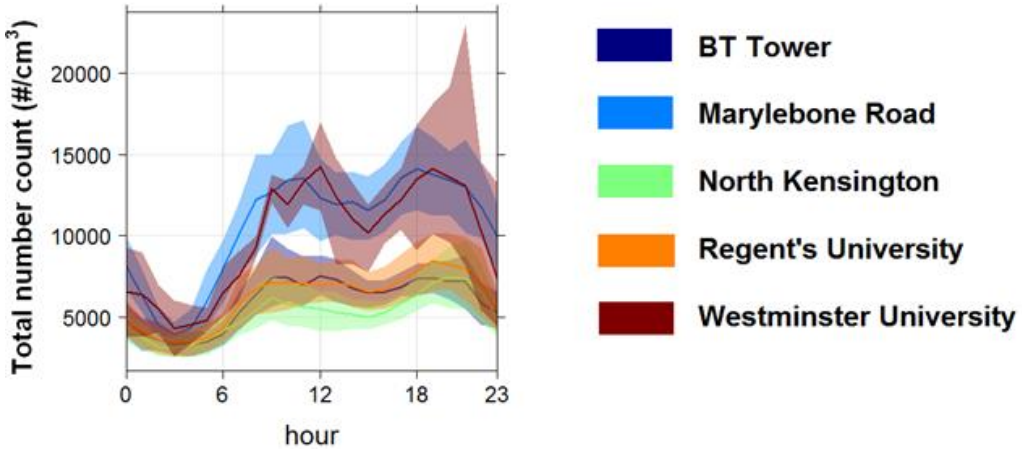

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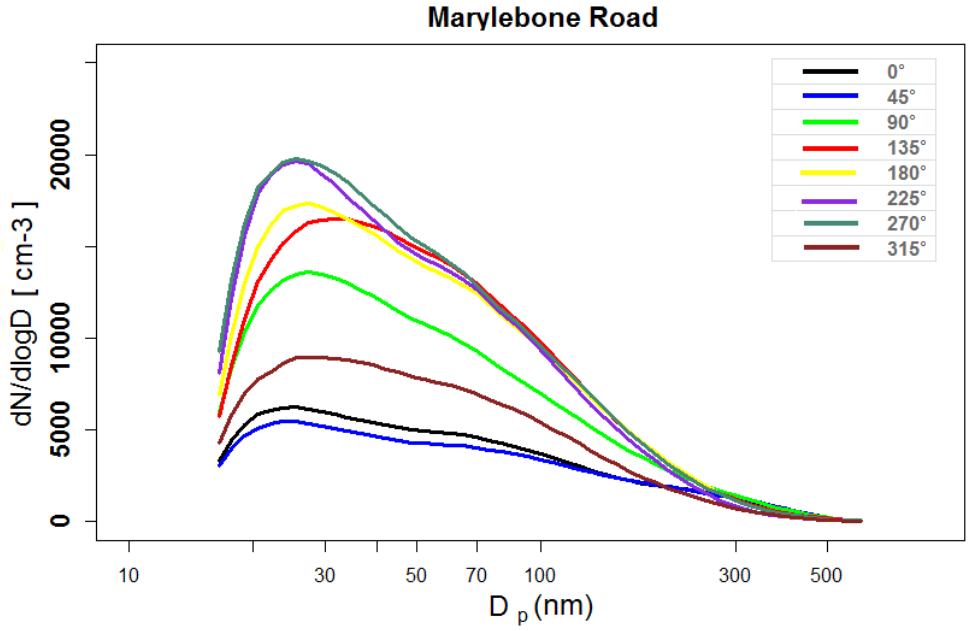

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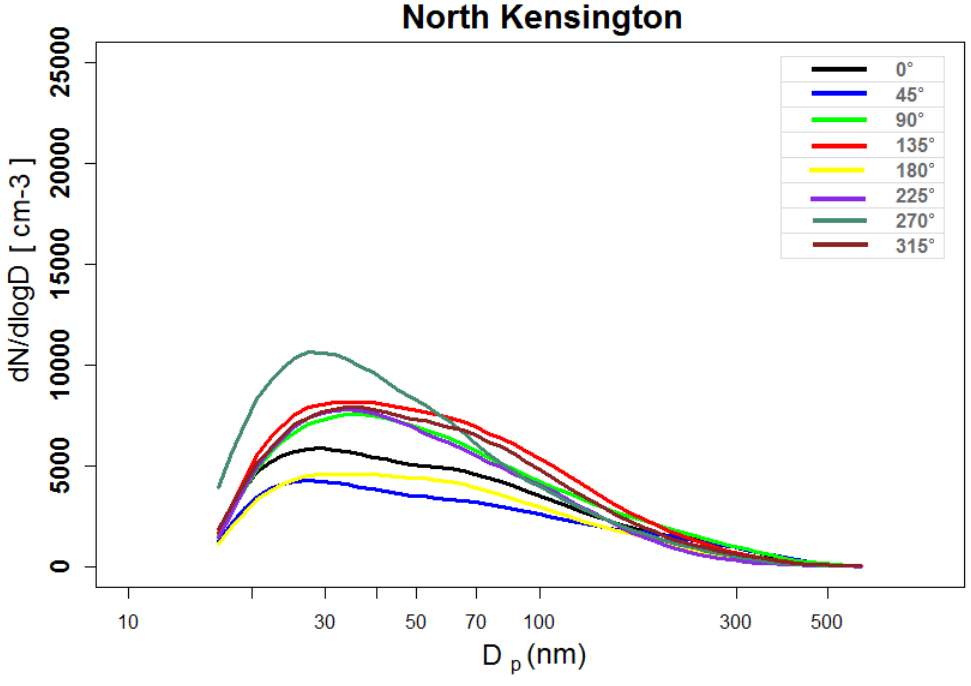

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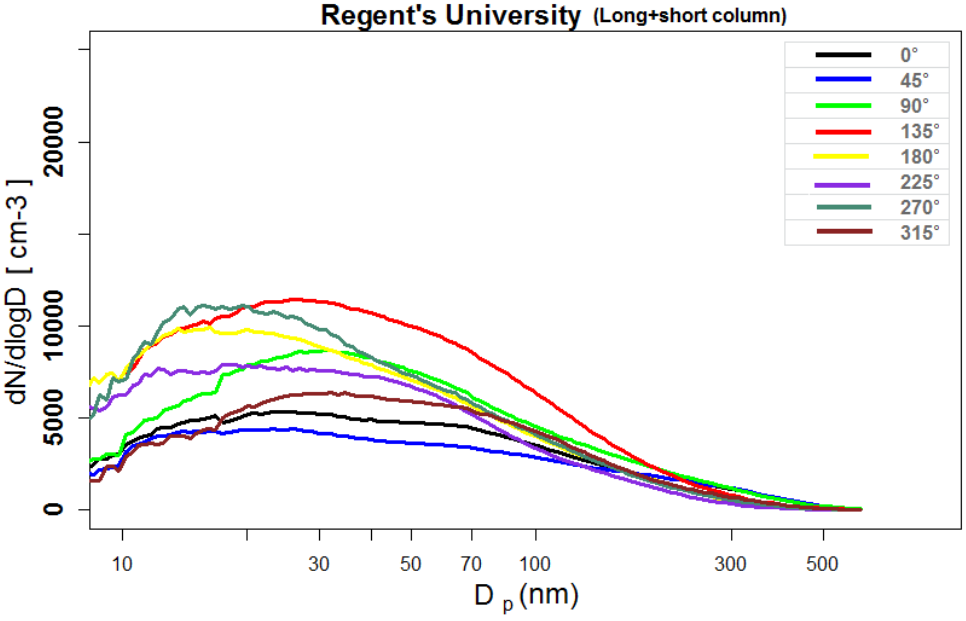

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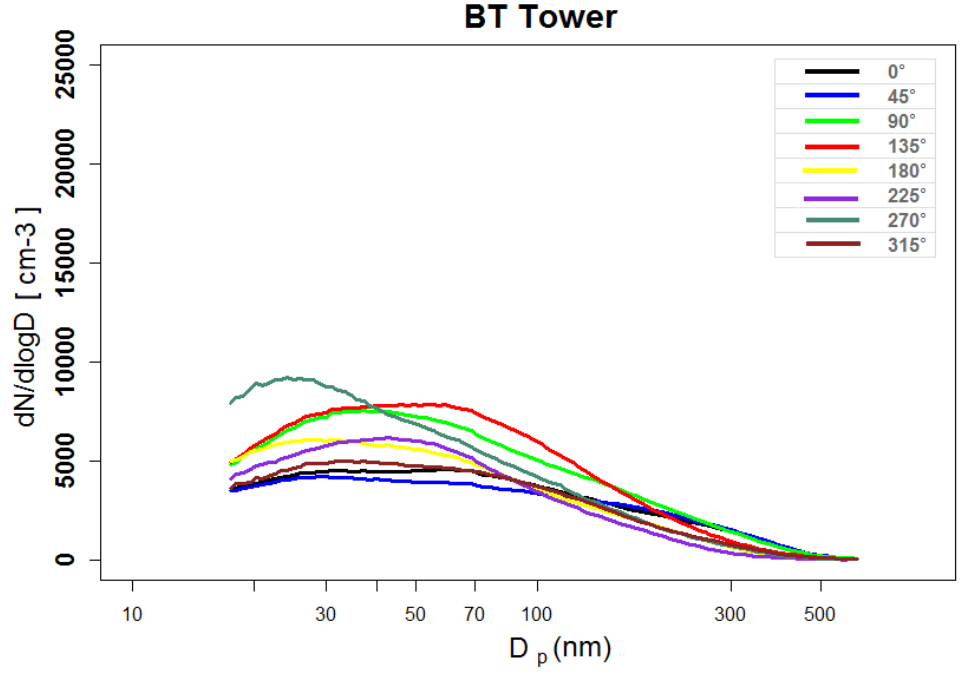

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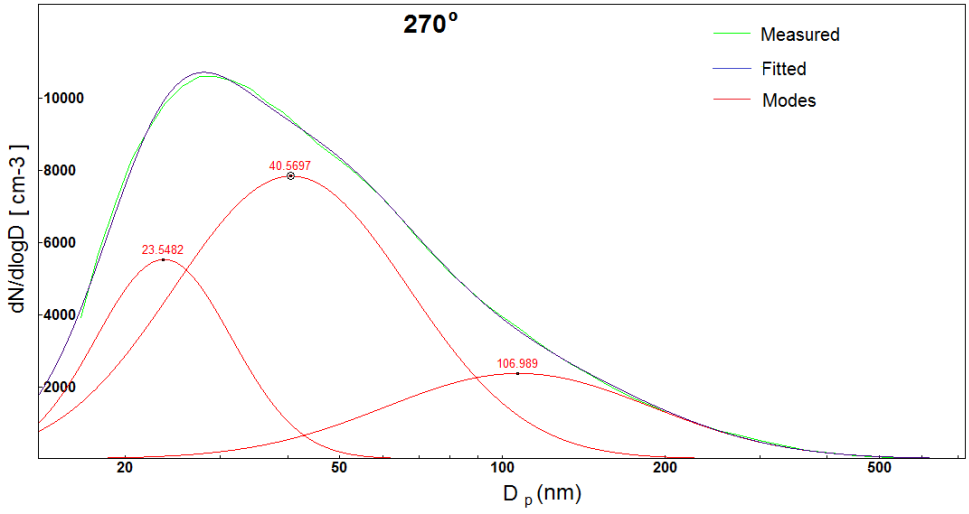

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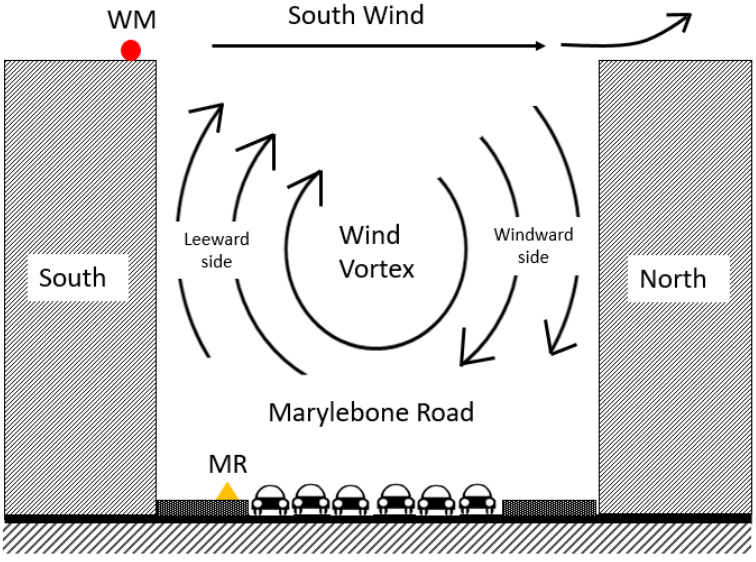

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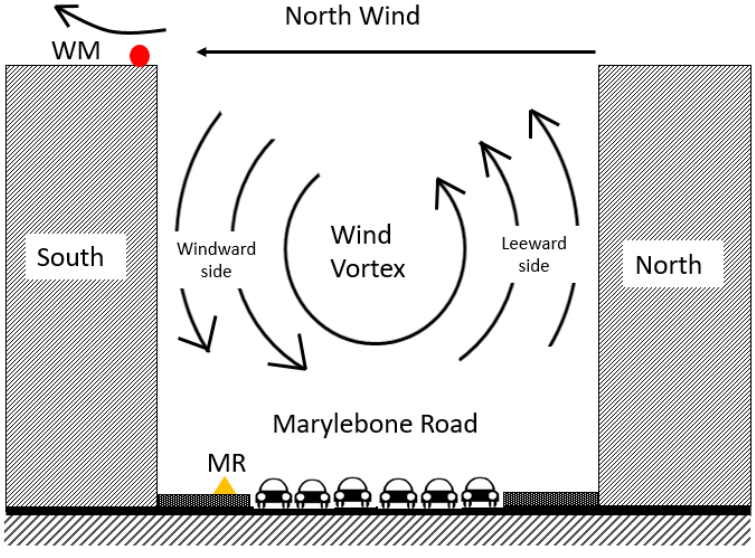

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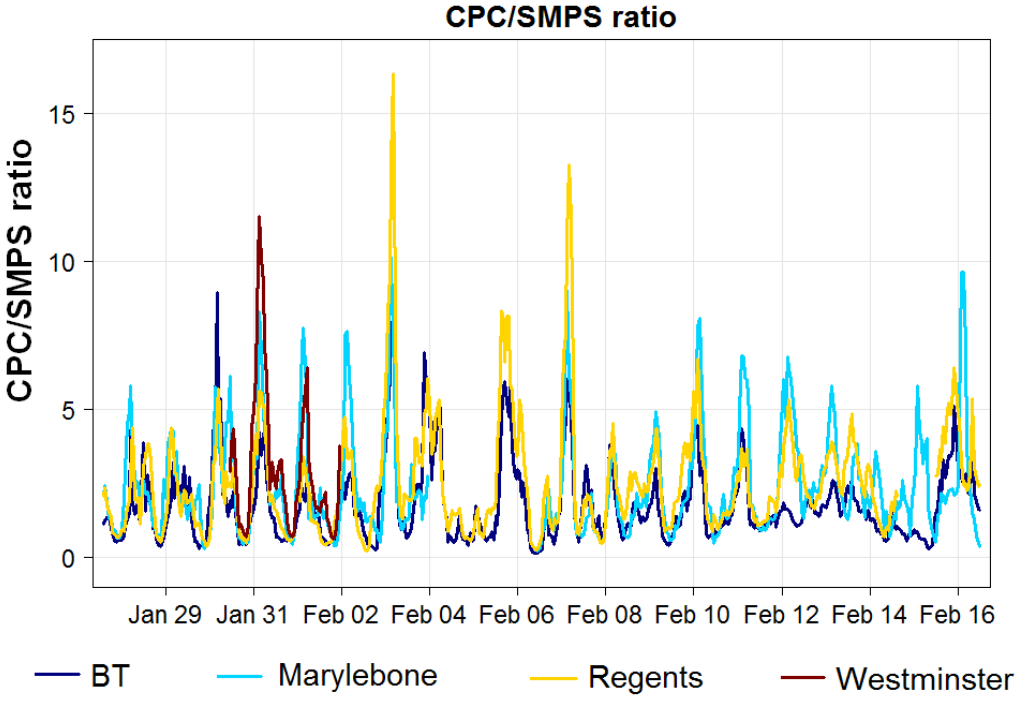

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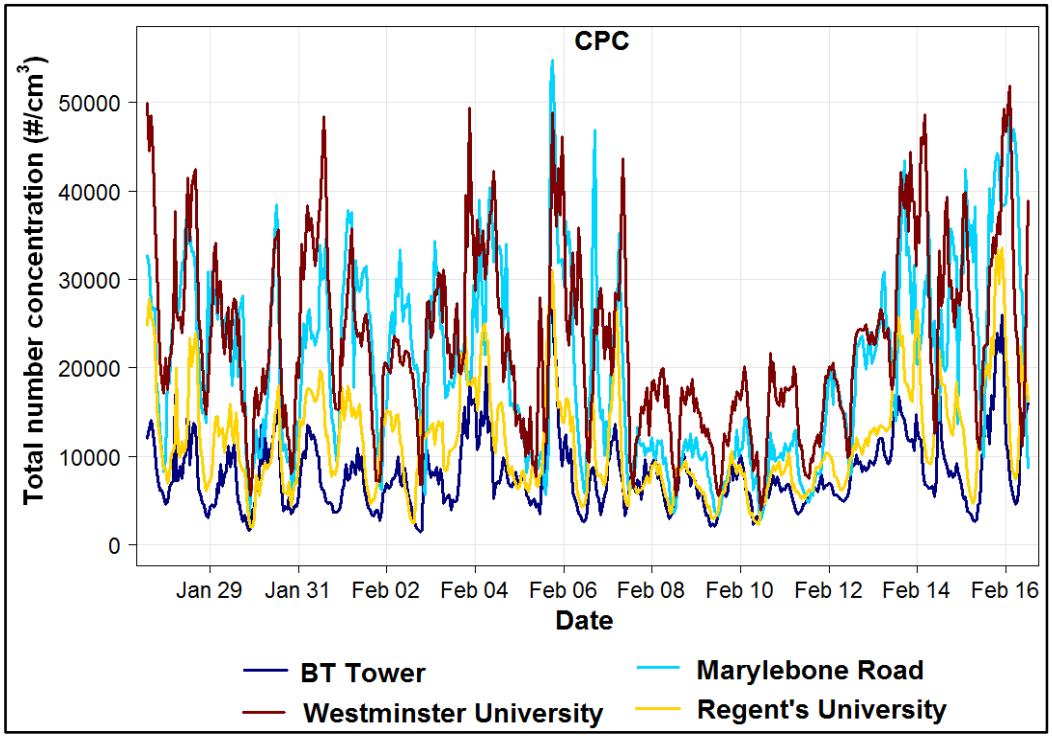

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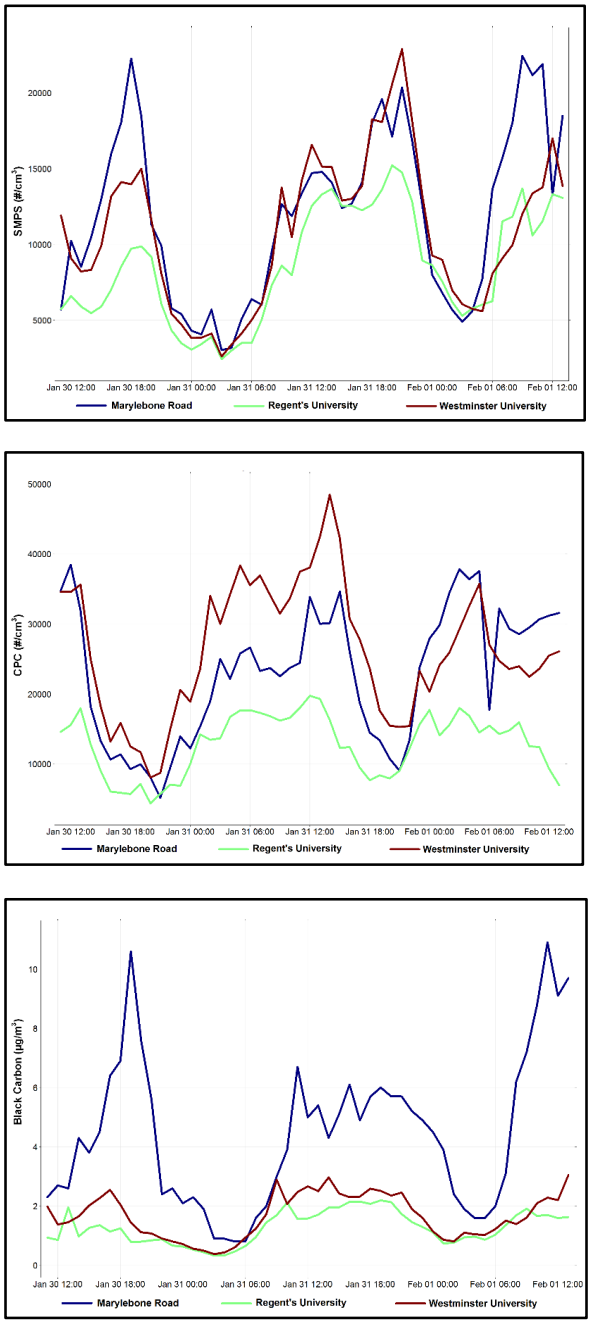

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