A MULTI-SITE STUDY OF PROCESSES INFLUENCING PARTICLE NUMBER CONCENTRATIONS IN URBAN AIR

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ABSTRACT

Particle number concentration data are reported from a total of eight urban site locations in the United Kingdom. Of these, six are central urban background sites, whilst one is an urban street canyon (Marylebone Road) and another is influenced by both a motorway and a steelworks (Port Talbot). The concentrations are generally of a similar order to those reported in the literature, although higher than in some of the other studies. Highest concentrations are at the Marylebone Road site with lowest at the Port Talbot site, with the central urban background locations lying somewhere between with concentrations typically around 20,000 cm^{-3}, with a seasonal pattern affecting all sites with highest concentrations in the winter months and lowest in the summer. Data from all sites show a diurnal variation with a morning rush hour peak typical of an anthropogenic pollutant. When the dilution effects of windspeed are accounted for, the data show little directionality at the central urban background sites indicating the influence of sources from all directions as might be expected if the major source were road traffic. At the London Marylebone Road site there is high directionality driven by the air circulation in the street canyon, and at the Port Talbot site different diurnal patterns are seen for particle number count and PM_{10} influenced by emissions from road traffic (particle number count) and the steelworks (PM_{10}) and local meteorological factors. Hourly particle number concentrations are generally only weakly correlated to NO_x and PM_{10}, the former showing a slightly closer relationship. Correlations between daily average particle number count and PM_{10} were also weak, which suggests that adverse health effects which are related to PM_{10} concentration may not be well predicted by particle number. Episodes of high PM_{10} concentration in summer typically show low particle number concentrations consistent with transport of accumulation mode secondary aerosol, whilst winter episodes are frequently associated with high PM_{10} and particle number count arising from poor dispersion of local primary emissions.

Key Words: Particulate matter, particle number, PM_{10}, urban air quality

INTRODUCTION

During the early 1990's, PM_{10} (the mass concentration of particles with aerodynamic diameters less than 10 μm) was selected as the preferred metric for the measurement of airborne particulate matter in United Kingdom air quality monitoring systems. Material within the PM_{10} size fraction contains a wide range of different chemical and biological species from numerous different sources. The sources may however be separated into two major groups. The first comprises high temperature sources which typically result in nucleation particles with sub-micron diameters formed either from the condensation of combustion products in the vicinity of the combustion process, or later as the result of chemical reactions involving gaseous combustion products. The second includes mainly fugitive material sources where predominantly larger material is raised from surfaces either by activity or by turbulent airflow across the surface. In addition, the PM_{10} size fraction contains material in the 'accumulation mode' (0.1 - 1μm diameter) where the combined effects of size selective deposition processes – Brownian motion, impaction and sedimentation – are at a minimum [1]. Relatively small numbers of larger particles may have a large influence on PM_{10} mass relative to far greater numbers of much smaller particles.

The diversity of PM_{10} with respect both to sources and chemical composition has led to efforts to define the chemical or physical characteristics of PM_{10} that may determine its effect on health. Despite difficulties due to the concentrations of the different components of PM_{10} tending to be modulated in similar ways by meteorological factors, various studies [2, 3] have suggested that the smaller material in the PM_{10} size range may have a greater significance for health. In 1997 US EPA introduced a requirement to monitor $PM_{2.5}$ [4]. It has also been proposed that ultrafine particles may play a special role in inducing adverse effects [5], and if this were demonstrated to be of importance, a better measure of particulate matter would be the particle number concentration, as PM_{10} mass is dominated by the larger particles.

To date, many of the published studies of particle number concentrations have reported measurements carried out over limited periods of time at a single location. In the work reported here, measurements were carried out over periods of up to four years, at eight sites across the United Kingdom, allowing seasonal effects and the consequences of the local distributions of sources to be evaluated.

EXPERIMENTAL

Particle Number Concentration

Condensation Particle Counters (TSI Model 3022A) were co-located at existing air monitoring stations from December 1999 onwards. The locations were chosen to provide a range of different sites within London, and a wide geographical range of sites across the remainder of the United Kingdom. The sites chosen, the type of site, the date of commencement of operation of the particle counters at each site and percentage of data captured, are presented in Table 1. After 15 months operation the instrument in use at the Marylebone Road site was transferred to the London Bloomsbury site.

The Model 3022A Condensation Particle Counter detects all particles with diameters greater than 15 nm, with a minimum detectable particle diameter of 3 nm. Fifty percent of 7 nm particles are detected. The instrument can record particle concentrations of up to 10^7 cm^{-3} with an accuracy of 20%. Instruments were operated to provide 15-minute mean values of particle number concentration. The instruments were serviced and re-calibrated by TSI (UK) at the recommended intervals.

Other Data

Hourly values of PM_{10} and NO_x were obtained from the co-located instruments at each of the sites to allow comparison of particle number concentration to be compared with these two pollutants. PM_{10} is the most widely used metric for airborne particulate matter in the United Kingdom, while

NO_x is a major component of high temperature emissions and its presence indicates the presence of local combustion sources. Hourly values of $PM_{2.5}$ were also obtained from the Marylebone Road and Bloomsbury sites where this metric is also measured. All data used had been ratified by the networks standard procedures except for the earlier $PM_{2.5}$ data for which there was no ratification procedure prior to January 2003.

The measurements of PM_{10} and $PM_{2.5}$ are made with Rupprecht and Patashnick Tapered Element Oscillating Microbalance (TEOM) instruments. In these instruments the inlet airflow and the filter on which the sample is collected are heated to 50^0 C in order to prevent changes in relative humidity having an effect on the mass of particulate matter measured. This heating of the sample results in the loss of semi-volatile material from the sample including ammonium nitrate [6]. In the United Kingdom, TEOM samplers of both size fractions are operated with an in-built data correction algorithm:

$$\text{Reported concentration } (\mu g\ m^{-3}) = 1.03 \times \text{Measured concentration} + 3\ \mu g\ m^{-3}$$

Representative wind directional and speed data, for a nominal height of 10 metres, was obtained for each location from a nearby meteorological station. The location of each meteorological station and its distance from the air quality measurement location are given in Table 1. The meteorological stations were chosen for their proximity to the air quality measurement location, the completeness of the meteorological data, and their positioning outside the urban area so that wind flow is not likely to be influenced by the presence of large buildings, and the data are likely to provide a reasonable estimation of wind direction above the pollutant sampling sites. Manning et al. [7] found broad agreement between the wind direction measured at 'roof top' level beside an urban street canyon and an airfield meteorological station some 40 km away, although wind speeds were substantially modified probably due to differences in surface roughness.

RESULTS AND DISCUSSION

Seasonal Particle Count Concentrations

The mean monthly particle number concentration for each site is shown in Figure 1. Due to the shorter sampling periods and instrumentation problems no data were available for the month of March at London Bloomsbury, or for the month of December at Marylebone Road. All available data obtained at each site from the date of the installation of the instrument to the end of December 2003 was used in the calculation of these means. Plots of the frequency distributions of number concentration were approximately log-normal and therefore geometric standard deviations have been calculated. For particle number these ranged between 1.96 at Glasgow and 2.85 at Manchester. Other than for the Manchester site, the highest geometric standard deviation was 2.17 at Belfast indicating that generally the spread of concentrations is broadly similar with a geometric standard deviation of around 2.0. This was compared with the geometric standard deviations for PM_{10} and NO_x. In the case of PM_{10}, these ranged from 1.68 at Bloomsbury to 2.11 at Port Talbot and for NO_x from 1.97 at Bloomsbury to 2.58 at Belfast. These values are indicative of the spread of data at individual sites being somewhat larger for NO_x than for particle number and larger for particle number than for PM_{10} mass. Overall, however, the variables show a broadly similar geometric standard deviation indicating that the spread of the frequency distributions are not greatly different.

Mean values of particle number concentration by other authors are listed in Table 1. These studies have shown higher concentrations to occur in urban areas and close to highways [8,9] with rapid decreases in concentration as the distance from the highway increases [10, 11], particularly in smaller size ranges [8, 9, 12, 13]. Strong correlations have been observed between particle number concentration and NO_x or CO [13, 14]. These gases are typically seen as indicators of traffic pollution. The weekday diurnal profiles of particle number concentrations with peaks in the morning around 08:00 hours, and in the late afternoon during winter months, at the times of

maximum traffic flow are also typical of traffic pollutants [8, 9, 12, 15, 16, 17, 18, 19]. Wind direction can have a significant effect on particle number concentrations [16, 17].

In addition to the association between particle number concentration with the location and periods of maximum traffic a number of studies have also identified the growth in numbers of small particles around midday. Nilsson et al. [20] and Ketzel et al. [8] observed the formation of particles in rural areas where the concentrations of pre-existing particles was low. Nilsson et al. [20] suggest that the presence of turbulence mixing the air was more important than the midday increase in radiation in causing particle formation. In studies in urban areas Wehner and Wiedensohler [13] associated the post noon particle formation with solar radiation, while an association with upwind sources of SO_2 was noted by Jeong et al. [19]. In a review of various studies on the formation of particles Kulmala et al. [21] observed that particle formation rates are higher in urban areas. However, new particle formation from homogeneous nucleation is relatively infrequent in Birmingham, UK [22] and is thought unlikely to have affected our data appreciably.

Particle number concentrations were generally similar between all the sites (Figure 1) with the exception of Marylebone Road where the site is on the kerbside of a road with traffic flows in excess of 80,000 vehicles per day within a street canyon. At all sites the monthly particle number concentrations are at a minimum during the summer months and highest during the winter. The values of mean monthly particle number shown in Figure 1 for the background sites (10,000 cm^{-3} to 20,000 cm^{-3} in the summer months, and up to 42,000 cm^{-3} in the winter months) are similar to those found in under similar conditions in the studies listed in Table 2, with the exception of those measured by Ketzel et al. [8] and Molnar et al. [16]. At the site within the street canyon at Marylebone Road, mean monthly particle number concentrations of up to 117,000 cm^{-3}, are of a similar magnitude to those measured near heavily trafficked roads in other studies presented in Table 2 [9, 10, 11, 12, 23]. This study, and those listed in Table 2, were carried out over different ranges of particle size. While differences in the maximum size of particle measured may not have a

significant effect on the particle number concentration, this may not be the case where there are differences in the minimum size measured.

It was observed in December 2003 that the pedestrianised area around the Glasgow air quality monitoring site was being used for the operation of fairground equipment. The substantially higher concentrations of particle number observed in December at Glasgow compared to the other months of the year may be the result of the use of mobile electricity generation equipment associated with the fairground activities during the Christmas period.

Diurnal Particle Count Concentrations

The mean diurnal profiles of particle number concentration measured between January 2000 and December 2003 at Belfast (the site with the greatest amount of data) on weekdays (Monday to Friday), and on Sundays, are shown in Figures 2(a) and 2(b) respectively. In each case the data is split into four groups: the winter period (December to February); the approximately month long periods from the beginning of March to the introduction of British Summer Time (BST) on the last weekend in March and from the end of BST on the last weekend in October to the end of November; the periods from the start of BST to the end of April and from the start of October to the end of BST; and the summer period (May to September). The error bars shown in the graphs are the standard error in the mean of each 15-minute mean value of particle number concentration.

The weekday diurnal profiles of particle number concentration (Figure 2a) are typical of the diurnal profiles which have been described for anthropogenic urban pollutants, such as black smoke ascribed to domestic combustion [24] or PM_{10} due to traffic [25], with a large morning peak and some evidence of a secondary late afternoon peak. Similar weekday diurnal profiles of particle number concentration, with a major morning peak and in some instances a late afternoon peak, have been identified by others (e.g. [14, 16, 18, 26]). In this study the 15-minute time base of the particle number concentration measurements allows a one hour difference in the time of occurrence of the

morning peak, before and after the change from Greenwich Mean Time (GMT) to British Summer Time (BST), and vice versa, to be identified, confirming that the particle number metric is driven by anthropogenic sources. Seasonal plots of weekday diurnal particle number concentrations presented by Gomiscek et al. [26] for winter and summer show the summer peak concentrations to occur one to two hours earlier than during the winter.

The weekday diurnal profiles of particle number concentration (Figure 2a) confirm the seasonal effects seen previously (Figure 1). After sunrise, solar heating of the ground surface will heat the adjacent atmosphere and progressively erode any nocturnal inversion, and over time completely destroy the inversion [27]. During summer months the earlier dawn will result in a greater period of time for this process to take place before the major anthropogenic sources become active. The lower particle number concentrations in the morning peak during April and October and during the summer months is probably due to the greater development of the boundary layer before anthropogenic material is released into it.

The diurnal particle number concentration profiles obtained on Sundays (Figure 2b) show a much reduced morning increase in concentration indicating that the bulk of the particulate matter measured on weekdays comes from anthropogenic sources that are dominant on weekdays – probably traffic as opposed to domestic heating. This weaker diurnal profile of particle number concentration at weekends has been observed by other authors (e.g. [14, 18]).

Relationships Between Particle Count and PM_{10} and NO_x Concentrations

Hourly mean values of particle number concentration were calculated from the 15-minute values and the paired values of particle number, PM_{10} and NO_x were compared at each site. Values of the square of the correlation coefficient, gradient and intercept for the regressions of particle number against PM_{10}, and particle count against NO_x, are presented in Table 3 for each site. The standard errors in the values of gradient and intercept are given in parentheses. At the two sites, London

Bloomsbury and Marylebone Road, where $PM_{2.5}$ data were available, the square of correlation coefficient, gradient and intercept for the regressions are given in Table 4. The reduced major axis regression was used rather than a standard linear regression due to the variability of both sets of values in the regression [28].

The regressions of particle number against PM_{10} and NO_x at each site (Table 3) show a substantially better correlation of particle number with NO_x than with PM_{10}, with the exception of the Manchester data. At the Port Talbot and North Kensington sites correlations are particularly poor between particle number and either PM_{10} or NO_x. Only in the case of the kerbside site at Marylebone Road is there good correlation between particle number and NO_x. This better correlation of particle number concentration with NO_x than with PM_{10} was also found by Cyrys et al. [14], Ketzel et al. [8] and Noble et al. [17] which they take to indicate that particle number concentration is dominated by material from traffic sources. Values of r^2 for regressions of 24-hour data for particle number count and NO_x, also shown in Table 3, are also weak. This suggests that health outcomes which correlate with PM_{10} concentration may be far less well predicted by particle number count, although this needs to be investigated directly.

The sites can be separated by the gradient of the regression of particle number concentration against PM_{10} to identify the importance of traffic related sources. The greatest gradient is found at Marylebone Road - within a street canyon containing a heavily trafficked road; whilst the smallest gradient is at Port Talbot – a site close to heavy industry with fugitive dust sources. At the two sites where $PM_{2.5}$ data were available the correlations between particle number concentration and $PM_{2.5}$ are similar to those between particle number concentration and PM_{10}.

It may be seen from Table 3 that for the majority of the urban background locations, the relationship between particle number count and PM_{10} concentration was highly scattered. It was attempted to clarify this situation by binning the data for PM_{10} and calculating a mean particle number

concentration for each integer value of PM_{10}. The typical pattern observed was of an approximately linear increase in particle number concentration with PM_{10} up to about 50 µg m^{-3}. Above this concentration the data bifurcated with one path showing a continuing steady increase of particle number concentration as PM_{10} increased whilst in the other group of data the particle number concentration decreased as PM_{10} increased to values of in excess of 200 µg m^{-3} (hourly mean). When the data were disaggregated by season as appears in Figure 3 for the London Bloomsbury site, it is clear that the seasons show significantly different behaviour. In the June to August data, there is relatively little increase in particle number count as the PM_{10} concentration increases. The opposite extreme of behaviour appears in the December to February data where the particle number count to PM_{10} concentration was consistently higher than in the June to August data with a tendency for the particle number count to increase quite sharply at PM_{10} concentrations above about 50 µg m^{-3}. The March to May data lay just above the June to August data, whilst the September to November data generally lay just below the December to February data. This is interpreted as reflecting different ratios of primary traffic emissions (high number concentration to PM_{10} ratio) and accumulation mode secondary particles (low particle number to PM_{10} concentration ratio). Thus, in the summer months the influence of secondary accumulation mode aerosol is much increased relative to the primary traffic emissions which tend to have their greatest influence in the winter months when the formation conditions for secondary particles are less conducive and there is a tendency towards more stable atmospheres with limited dispersion of primary ground-level emissions. The divergence is greatest at high PM_{10} concentrations where summer episodes tend to be the result of long-range transport of accumulation mode secondary aerosol whilst winter episodes are more often the result of local trapping of primary emissions as a result of poor dispersion conditions.

The one exception to this pattern was Port Talbot where, after an initial increase in particle number count to about 20,000 cm^{-3} with PM_{10} to about 50 µg m^{-3}, the average particle number count remains at about 20,000 cm^{-3} despite PM_{10} increasing to 290 µg m^{-3}. This probably reflects the

influence of coarse dusts from the steelworks causing an increase in PM_{10} but having little effect on particle number.

A more detailed analysis of the data for Belfast revealed that high ratios of particle numbers/PM_{10} at $PM_{10} > 65$ µg m^{-3} occurred during the morning rush hour (local primary emissions) or were associated with lower relative humidity, lower temperatures and lower wind speeds, conditions likely to be associated with poor dispersion of local emissions. An examination of airmass back trajectories for hours with high PM_{10} concentrations showed a tendency for the highest ratios of particle number to PM_{10} in slow-moving airmasses with a large clean sector component, whilst those trajectories with faster moving air from the European mainland generally showed much lower ratios.

Effect of Wind Direction

At each site, the hourly paired values of particle count, PM_{10} and NO_x were grouped by each 10^0 segment of wind direction, and mean concentrations were calculated for each wind direction. The normalised mean concentration of each pollutant is plotted against wind direction and examples appear in Figures 4(a) to (c). In each of these graphs the inner circle represents a normalised concentration of unity, and the outer circle represents a normalised concentration of two times unity. The Marylebone Road sampling site is within a street canyon containing a heavily trafficked road with nearby traffic lights. The approximate alignment of the road (and canyon) is shown in Figure 4(c).

The regressions of particle number against PM_{10} and NO_x take no account of the non-isotropic distribution of sources around the air quality monitoring sites. Plotting the mean normalised concentration of particle number, PM_{10}, and NO_x (and when available $PM_{2.5}$) against wind direction, allows comparisons to be made between the pollutants, and interpreted in relation to the known geography of each site. Differences in concentration in different wind directions may be a

result of the positions of local sources, the long distance transport of material, or differences in the mean wind speed in winds from differing directions affecting the dispersion of material. Most of the plots showed little directionality in particle number concentration, PM_{10} or NO_x. Some showed a small bias towards higher concentrations arriving from the easterly sector. At Belfast and Manchester this easterly bias was less pronounced for particle number concentration than for PM_{10} or NO_x. Other specific examples were:

Birmingham Centre

The plots of particle number, PM_{10} and NO_x (Figure 4(a)) all show higher concentrations in easterly winds and lower concentrations in westerly and south-westerly winds. When the mean particle number concentration is plotted against the mean wind speed for each wind direction the data generally lie on a line giving a close fit to an equation of the form;

$$\text{Particle number concentration} = \text{constant} (- \ln (U/2) + 3)$$

where U is the wind speed in knots. This form of equation is typical of the concentration predicted by the ADMS dispersion model for sites immediately downwind of a large area source in neutral atmospheric conditions. Higher concentrations of particle number than predicted by the above equation are found at wind directions of 80^0 to 150^0, as is actually observed in Figure 4(a).

The main commercial area of the city is to the east of the monitoring site, with a major highway running in tunnel past the site with portals to the north east and south east of the site. The major station in the city lies approximately 700 metres to from the monitoring site in a direction of 110 degrees, the direction of maximum NO_x concentrations, and higher than average concentrations of particle number. At directions other than between 80^0 and 150^0, the primary effect on directionality of mean concentrations appears to be the dilution by the wind.

Port Talbot

The particle number concentration shows little variation with wind direction (Figure 4(b)), while the PM_{10} plot shows the existence of a strong source of PM_{10} to the south west, and the NO_x plot shows a similarly strong source of NO_x to the north east. The monitoring site is approximately 700 metres away from a large steelworks to the south west, a likely source of fugitive dust. Such dust will have a relatively large diameter and hence dominate PM_{10} measurements, while having a negligible effect on the values of particle number measured. To the north east of the monitoring site an inter-urban motorway carrying in excess of 50,000 vehicles on a typical weekday lies at a distance of approximately 75 metres. The presence of this motorway is apparent in the measurement of NO_x but not in the measurement of particle number. Zhu et al. [11] observed a rapid reduction in the concentrations of particles within 150 metres of a highway carrying over 12,000 vehicles an hour, with concentrations of particles in the size range most influenced by the highway being indistinguishable at 300 metres downwind of the highway from that measured upwind of the highway. At a distance of 75 metres from the less busy highway at Port Talbot it may be that the concentration of particle number of material from vehicles on the motorway has already fallen to a level that is low compared to the background particle number concentrations although the NO_x concentrations may still be significant compared to the background levels of NO_x.

The weekday normalised diurnal concentration profiles of particle count, PM_{10} and NO_x are shown in Figure 5. The profiles of particle number and NO_x show the morning peak typical of traffic-related urban pollutants indicating that both particle number and NO_x are influenced by this type of source. The diurnal profile of PM_{10} has an afternoon peak indicating a strong influence of a source with a different directionality and/or diurnal pattern of emissions. Fugitive dust emissions are likely to be greatest around mid-afternoon when surface temperatures are at a maximum, surface moisture content is at a minimum, and mean wind speeds are highest due to thermally induced air movement. Kuhlbusch et al. [29] observed maximum concentrations of coarse particulate matter ($PM_{2.5-10}$) during the afternoon which they associated with higher mean wind speeds. The issue of location is

important as the sampling site is inland of the steelworks and any local land-sea breeze circulation is likely to favour steelworks emissions reaching the site during the afternoon sea breeze.

London, North Kensington

There is a strong correlation between the directional plots of particle number and PM_{10} with a squared correlation coefficient of 0.86 compared to a value of 0.75 when particle number is regressed against NO_x. Lower concentrations of all pollutants are seen in south-westerly winds which tend to be stronger leading to better dispersion. The monitoring site is in a residential area to the west of central London and is passed by a major radial route into the city centre some 500 metres to the south and south east. The concentrations of particle number, PM_{10} and NO_x are all greater when the wind is from the east, the direction of central London and the European mainland.

Marylebone Road

This site is within a street canyon on the southern kerb of a major radial route into central London with traffic flows in excess of 80,000 vehicles per day. The plots of normalised concentrations against wind direction (Figure 4(c)) show higher concentrations of all pollutants when the wind is from southerly directions. This is consistent with the cross street component of the wind being reversed at street level in the street canyon so that a southerly wind produces a flow from the north at street level carrying material from the passing traffic onto the monitoring site, as described by Nakamura and Oke [30] and observed in PM_{10} data by Harrison et al. [31]. Wehner et al. [12] found similar high concentrations of particle number concentrations in rotating flow in a street canyon.

It is also apparent in Figure 4(c) that there are two different patterns of pollutant concentration with wind direction. The distributions of the normalised concentrations of PM_{10} and $PM_{2.5}$ are symmetrical about a line normal to the axis of the road (and street canyon). In contrast, the distributions of the normalised concentrations of particle number and NO_x are asymmetrical, with higher concentrations occurring between wind directions of 150^0 and 260^0. Regressing the mean

particle number concentration against mean NO_x, PM_{10} and $PM_{2.5}$ concentrations for each wind direction gives squared correlation coefficients of 0.96, 0.84 and 0.88 respectively, confirming this closer relationship between particle number and NO_x, than between particle number and PM_{10} or $PM_{2.5}$.

While the eastbound traffic flow on Marylebone Road is free flowing throughout the day, the west-bound flow is congested with reduced traffic speeds during the working day, particularly during the afternoon. This is a result of a light-controlled pedestrian crossing 50 metres to the west of the monitoring site, which leads to stationary traffic to the west of the monitoring position, which when traffic numbers are high, results in traffic queuing past the monitoring position. A light-controlled junction 125 metres to the west of the pedestrian crossing causes further congestion at times of peak traffic numbers. The higher concentrations of particle number and NO_x when the wind direction has a westerly component may be due to material from the exhausts of stationary vehicles waiting at these traffic lights or from the exhaust of vehicles accelerating from stationary. The corresponding lack of an asymmetric distribution of PM_{10} and $PM_{2.5}$ implies that both these pollutants are associated with moving traffic, and that material directly from the exhausts of stationary vehicles does not contribute significantly to either of these measures. While $PM_{2.5}$ is generally taken as a measure of smaller particulate matter, when there are large concentrations of resuspended coarse particulate matter, the coarse particulate matter has been found to dominate the $PM_{2.5}$ [32].

OVERVIEW OF DATA

Particle number concentrations were similar at all urban sites, except at a site located on the kerbside of a heavily trafficked road in a street canyon. There is a seasonal trend in particle number concentrations with the highest values occurring during the winter months. The weekday diurnal trend of the mean particle number concentration in an urban area is typical of that for other anthropogenic pollutants with a morning peak.

At urban background monitoring sites there is only a weak correlation between particle number and either PM_{10} or NO_x concentrations. There is a better correlation between particle number and NO_x concentrations, than between particle number and PM_{10}, or $PM_{2.5}$, concentrations. A clear correlation can be found between particle number and NO_x concentrations at the kerbside monitoring location.

Comparison of the particle number and NO_x concentrations measured at different wind directions can enable the identification of some sources in the vicinity of the monitoring position, although particle number concentration tends to be less dependent on wind direction than either PM_{10} or NO_x. In many central urban locations concentrations of particle number show little directionality which cannot be explained by a simple windspeed dilution effect. At a distance of as little as 75 metres from a highway carrying 50,000 vehicles per day the presence of the highway does not result in any apparent increase in particle number concentrations. At the kerbside of a highway carrying 80,000 vehicles per day in a street canyon, particle number concentration is closely related to NO_x concentration and the presence of stationary traffic makes a contribution to both pollutants but not to PM_{10} or $PM_{2.5}$.

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Table 1: Sampling sites and meteorological stations

Sampling site	Type of site	Start of particle count sampling	Data capture [%]	Sampling site location	Meteorological station	Meteorological station location	Distance between sampling site and meteorological station [km]
Belfast Centre	Urban centre	Jan 2000	78.5	339744	Aldergrove	147798	19.9
Glasgow Centre	Urban centre	Feb 2001	48.8	NS 589650	Bishopton	NS 418711	18.2
Manchester Piccadilly	Urban centre	Jun 2000	53.6	SJ 843983	Ringway	SJ 814844	14.2
Birmingham Centre	Urban centre	Feb 2000	61.0	SP 064868	Elmdon	SP167841	10.6
Port Talbot	Urban background	Jan 2000	68.1	SS 780882	Mumbles Head	SS 627870	15.3
North Kensington	Urban background	Apr 2000	70.0	TQ 240817	Heathrow	TQ 077767	17.0
Marylebone Road	Kerbside	Jan 2001	69.3	TQ 281820	Heathrow	TQ 077767	21.1
London Bloomsbury	Urban centre	Apr 2002	56.9	TQ 302820	Heathrow	TQ 077767	23.1

Table 2: Particle number concentrations reported in other studies

Authors	**Location**	**Description of site**	**Minimum size [nm]**	**Maximum size [nm]**	**Mean concentration [cm^{-3}]**	
Cyrys et al. [14]	Erfurt	urban	10	2500	18000	
Gomiscek et al. [26]	Vienna, Linz, Graz	urban	7		29300-31100 16200-20600	winter mean summer mean
	near Vienna	rural	7		10200 10500	winter mean summer mean
Harrison et al. [9]	Birmingham	background	7		28600, 36600	at 2 sites
	Birmingham	2m from kerb of highway	7		96000	
Hussein et al. [18]	Helsinki	urban	8	400	10500, 14500	at 2 sites
Jeong et al. [19]	Rochester, NY	urban	10	470	8160	
Ketzel et al. [8]	S Sweden	rural	3	900	2500	
	Copenhagen	near city	3	900	4500	
	Copenhagen	urban	3	900	7700	
Longley et al. [23]	Manchester	street canyon	4.6	100	27000	
Molnar et al 2002	Gothenburg	roadside	10	368	2000	
Monkkonen et al. [34]	New Dehli	urban background	10		61000	weekdays
Nilsson et al. [20]	Finland	over forest	3	500	8000	mid afternoon
Noble et al. [17]	El Paso	urban	20	20000	15200, 16700	at 2 sites
Ruellan and Cachier [23]	Paris	5m from major road	7	3000	220000	
Ruuskanen et al. [35]	Alkmaar	urban background	7		26000	weekdays
	Erfurt	urban background	7		27900	weekdays
	Helsinki	urban background	7		21700	weekdays
Shi et al. [10]	Birmingham	roadside	9.6	352	160000, 190000	on 2 days
	Birmingham	background	9.6	352	21000, 24000	on 2 days

Vakeva et al. [15]	Lahti, Finland	Urban street canyon	6	300	39000	
	Lahti, Finland	Rooftop	10		10800	
Wehner et al. [12]	Leipzig	Street canyon	3	800	32000 110000	opposite rotating flows in canyon
Wehner and Wiedensohler [13]	Leipzig	urban	3	800	21377 14278	Winter weekday Summer weekday
Zhu et al. [11]	Los Angeles	17m downwind from centre of highway	6	220 for SMPS	200000	
	Los Angeles	200m upwind of highway	6		48000	

Table 3: Reduced Major Axis regressions of particle number against PM_{10} and NOx for all paired data

Sampling site	Number of hourly samples	Regression of particle number against PM_{10} (hourly data)			Number vs PM_{10} – daily data r^2	Regression of particle number against NO_x (hourly data)		
		r^2	gradient (std err) [cm^{-3} / µg m^{-3}]	intercept (std err) (hourly data) [cm^{-3}]		r^2	gradient {std err) [cm^{-3} / ppb]	intercept (std err) [cm^{-3}]
Belfast	24067	0.14	1518 (15)	-2022 (361)	0.17	0.37	661 (4)	7986 (162)
Glasgow	9816	0.31	1303 (12)	-1173 (270)	0.39	0.39	225 (2)	11530 (178)
Manchester	14473	0.26	1012 (8)	-2810 (282)	0.18	0.18	475 (4)	1393 (278)
Birmingham	17725	0.20	1035 (8)	-1823 (172)	0.46	0.46	390 (2)	4602 (100)
Port Talbot	21084	0.02	602 (5)	1887 (164)	0.09	0.09	575 (5)	5126 (129)
North Kensington	20032	0.05	1167 (10)	-3759 (235)	0.16	0.16	334 (3)	7326 (138)
Marylebone Road	6419	0.29	2546 (31)	-4813 (1232)	0.63	0.63	450 (4)	2213 (776)
Bloomsbury	6360	0.11	964 (14)	-4746 (425)	0.43	0.43	321 (3)	4414 (225)

Table 4: Reduced Major Axis regressions of particle number against $PM_{2.5}$ for all paired data when $PM_{2.5}$ data available

Sampling site	Number of hourly samples	Regression of particle number against $PM_{2.5}$		
		r^2	gradient [cm^{-3} / µg m^{-3}]	intercept [cm^{-3}]
Marylebone Road	6312	0.24	3446 (44)	-2405 (1289)
Bloomsbury	6297	0.12	1654 (24)	-1287 (379)

FIGURE LEGENDS

Figure 1: Monthly mean particle number concentrations at the eight sampling sites

Figure 2 Seasonally disaggregated averaged diurnal profiles of particle number concentration at the Belfast site, for (a) weekdays, (b) weekends

Figure 3 Mean hourly particle count for each integer value of PM_{10} as a function of PM_{10} concentration and season for the London Bloomsbury site.

Figure 4 Normalised directional average of particle number count (CPC), PM_{10}, NO_x and $PM_{2.5}$ (Marylebone Road only) for (a) Birmingham, (b) Port Talbot and (c) London Marylebone Road.

Figure 5 Average weekday diurnal profiles for particle number, PM_{10} and NOx concentrations at Port Talbot

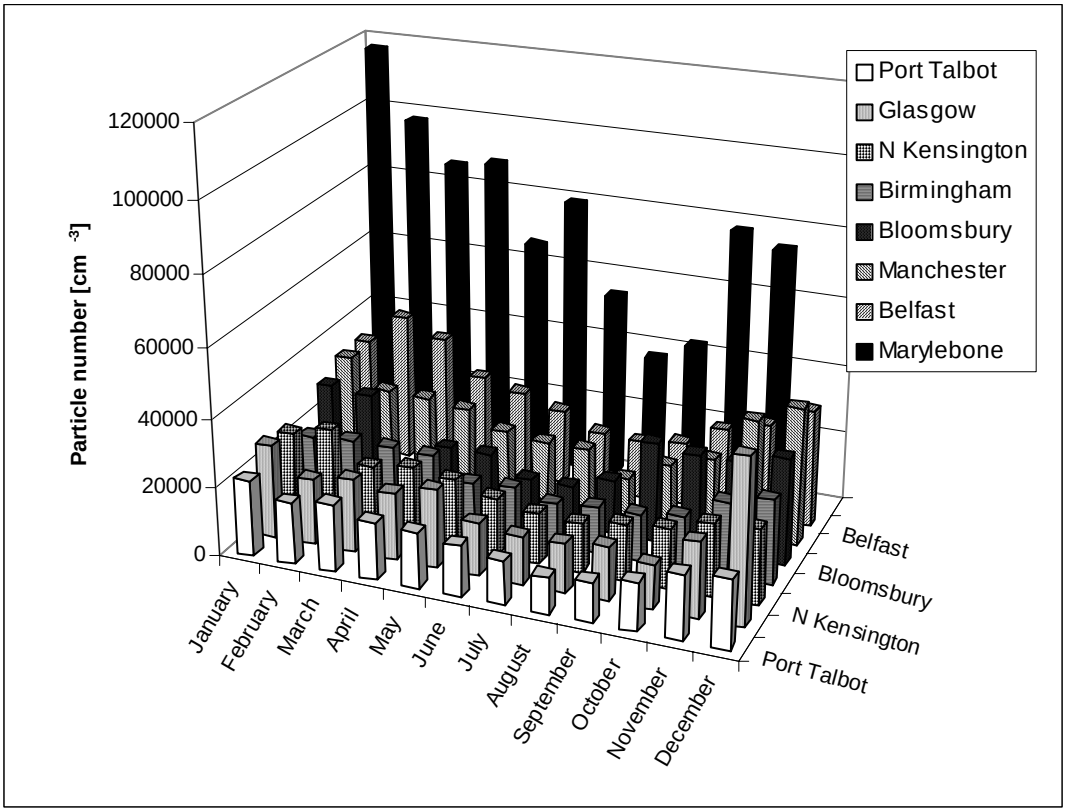
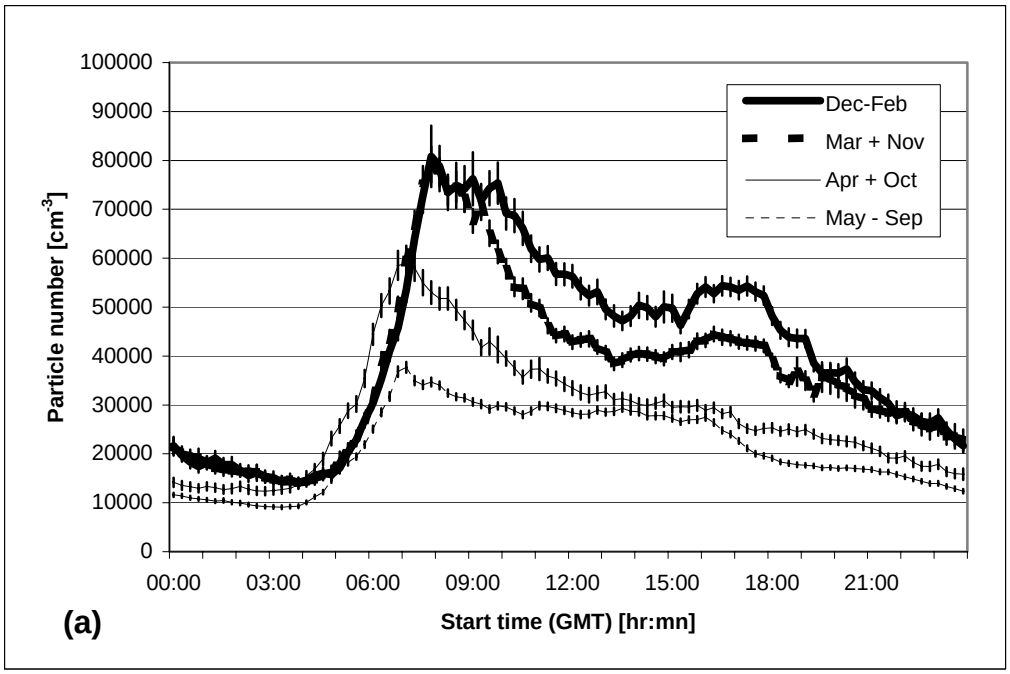

Particle number [cm -3]
120000
100000
80000
60000
40000
20000
0
Port Talbot
Glasgow
N Kensington
Birmingham
Bloomsbury
Manchester
Belfast
Marylebone
January
February
March
April
May
June
July
August
September
October
November
December
Belfast
Bloomsbury
N Kensington
Port Talbot

Figure 1


100000
90000
80000
70000
60000
50000
40000
30000
20000
10000
0
Particle number [cm-3]
Dec-Feb
Mar + Nov
Apr + Oct
May - Sep
00:00
03:00
06:00
09:00
12:00
15:00
18:00
21:00
Start time (GMT) [hr:mn]
(a)

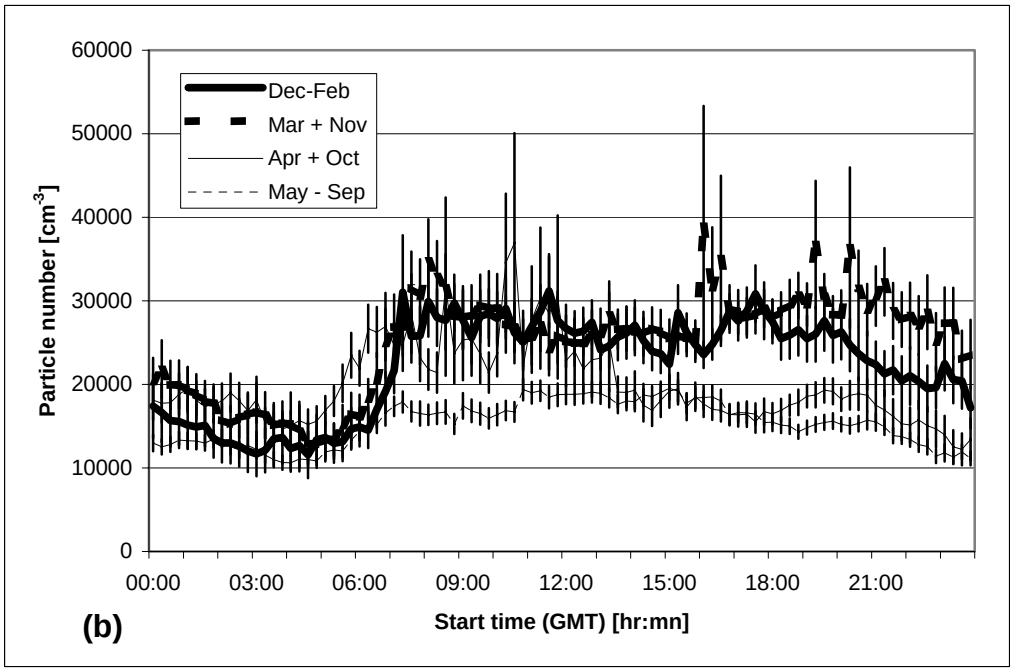

60000
50000
40000
30000
20000
10000
0
Particle number [cm-3]
Dec-Feb
Mar + Nov
Apr + Oct
May - Sep
00:00
03:00
06:00
09:00
12:00
15:00
18:00
21:00
Start time (GMT) [hr:mn]
(b)

Figure 2

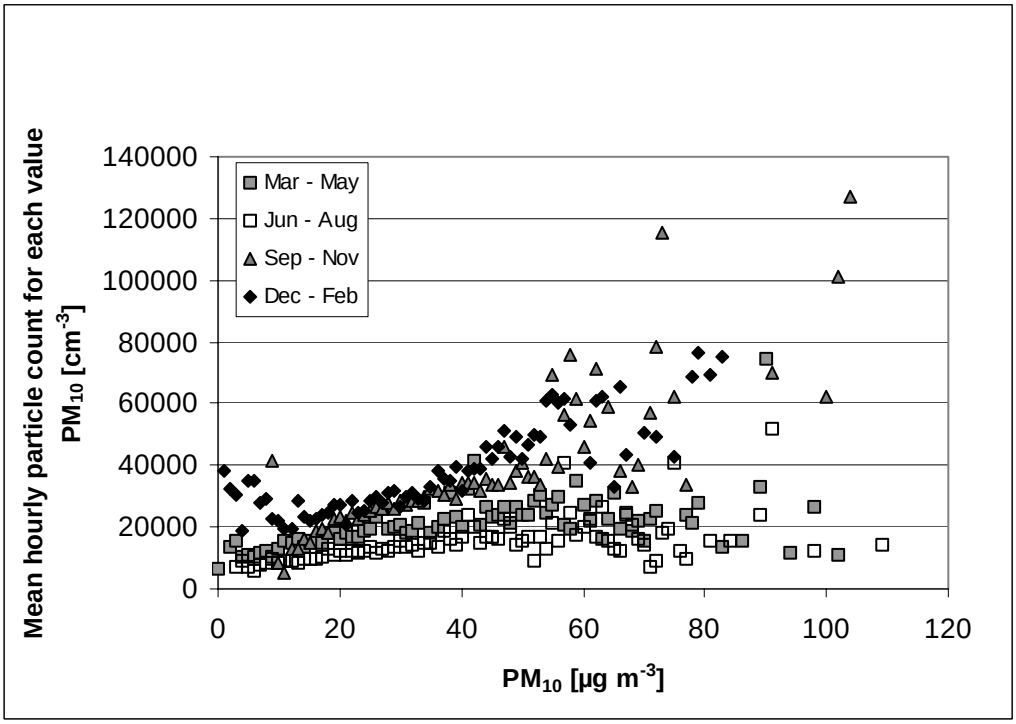

Mean hourly particle count for each value
PM10 [cm-3]
140000
120000
100000
80000
60000
40000
20000
0
Mar - May
Jun - Aug
Sep - Nov
Dec - Feb
0
20
40
60
80
100
120
PM10 [µg m-3]

Figure 3

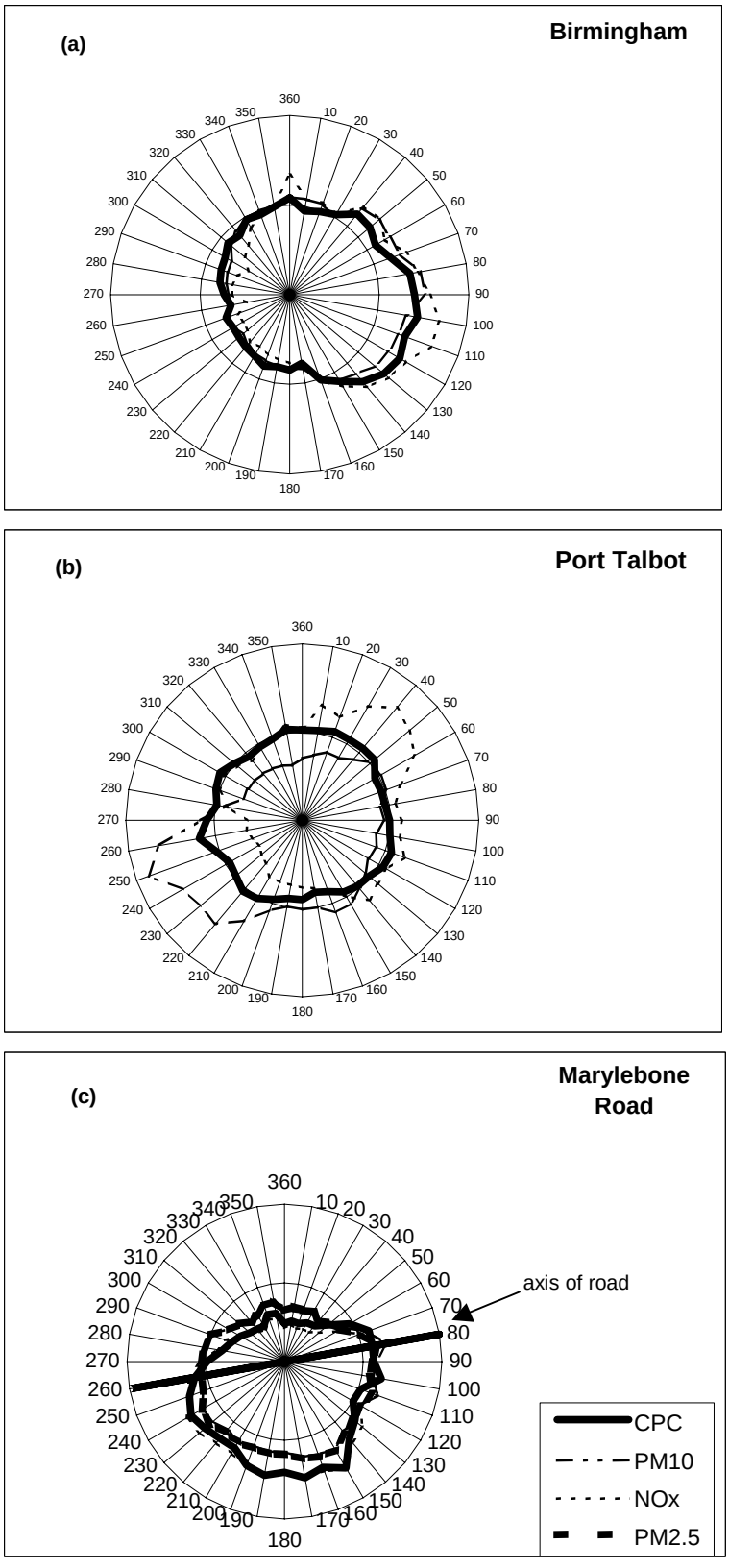

(a)
Birmingham
(b)
Port Talbot
(c)
Marylebone Road
axis of road
CPC
PM10
NOx
PM2.5

Figure 4

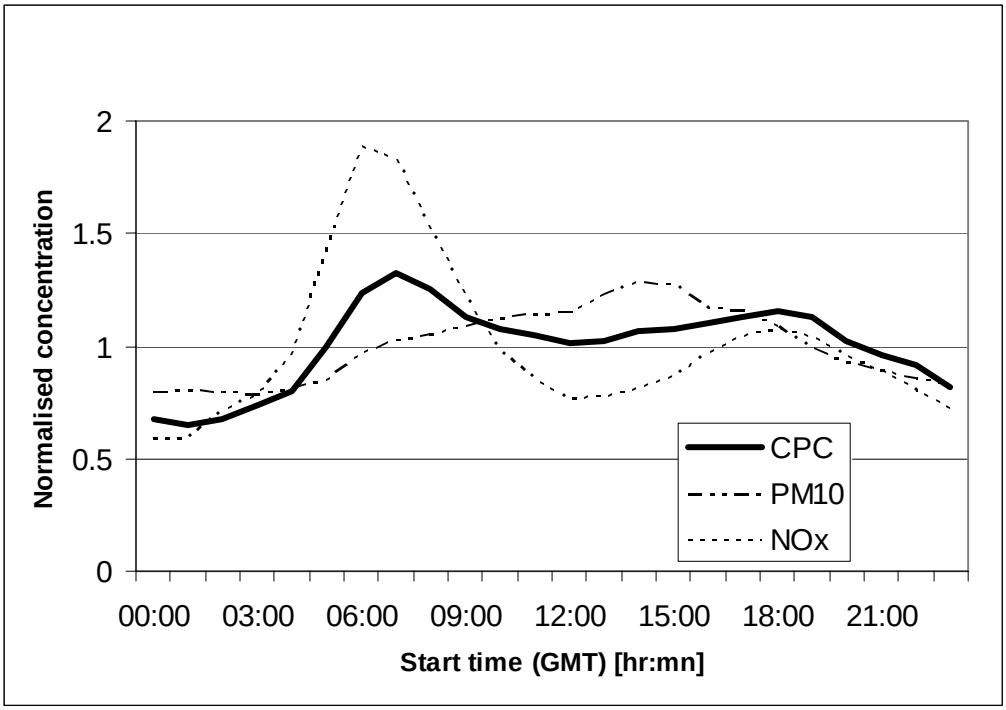

Normalised concentration
2
1.5
1
0.5
0
00:00
03:00
06:00
09:00
12:00
15:00
18:00
21:00
Start time (GMT) [hr:mn]
CPC
PM10
NOx

Figure 5