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Linking Particle Number Concentration (PNC), meteorology and traffic variables in a UK street canyon

Heather D. Price[1][a], Robert Arthur[b], Kelly A. BéruBé[c], Tim P. Jones[a]*

[a]School of Earth and Ocean Sciences, Cardiff University, Park Place, Cardiff, CF10 3YE, UK

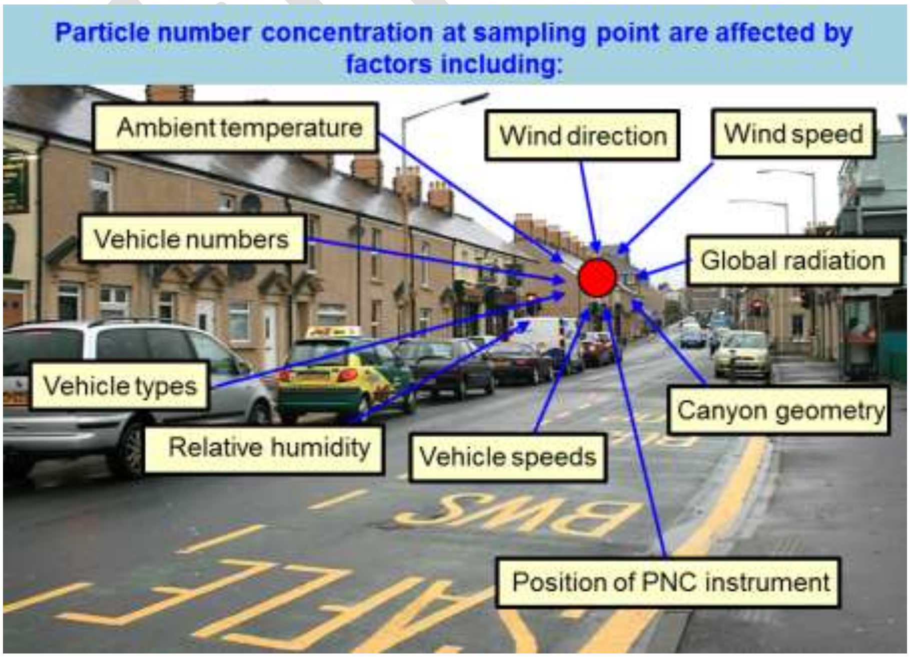
[b]Centre for Health and Environment Research, Department of Primary Care and Public Health, Neuadd Meirionydd, 4th Floor, Heath Park, Cardiff, CF14 4YS, Wales, UK

[c]School of Biosciences, Cardiff University, Museum Avenue, Cardiff, CF10 3US, UK

[1]Present address: Geography and Environment, Shackleton Building 44, University of Southampton, University Road, Southampton, SO17 1BJ

*Corresponding author. Email: h.price@soton.ac.uk, postal address: Geography and Environment, Shackleton Building 44, University of Southampton, University Road, Southampton, SO17 1BJ

Graphical abstract:



Abstract

Ambient particle number concentration (PNC) has been linked with adverse health outcomes such as asthma, reduced lung function and cardiovascular disease. To investigate the relationship between PNC, meteorology and traffic we measured size segregated respirable particles in a busy commuter street in Swansea, UK for ten months using a Dekati Electrical Low Pressure Impactor (ELPI). The ELPI segregates particles into 12 size fractions between 7 nm and 10 µm. The median PNC for the sampling period was 31,545 cm^{-3}. For the ultrafine particles (7 - 93 nm), the highest PNC was found in winter (46,615 cm^{-3}; 15 minute average) and the lowest for that size fraction in summer (29,696 cm^{-3}). For the particles below 93 nm there was a trimodal distribution to weekdays (particularly Monday to Wednesday), with PNC peaks at 09:00, 16:00 and 23:00. Wind direction had a significant influence on PNC and differed between particles in the fine range (below 2.5 µm) and more coarse particles (up to 10 µm). For fine particles, winds parallel to the canyon were associated with higher PNCs which were attributed to the replenishment of traffic particles. For coarse particles, PNCs were higher from winds perpendicular to the canyon and this was linked to source distribution around the sampling site and the recirculation of pollutants within the canyon. During times when vehicle volumes were high and vehicles were exhibiting stop-start behaviour, if this was combined with low wind speeds, ultrafine PNC was highest. This effect was generally observed during the morning rush hour. Current mass-based legislation does not take into account exposure to the number of particles or the change in population exposure diurnally.

Key Words

PNC; ELPI; street canyon; traffic; meteorology

Highlights

- Size-segregated respirable PNCs were monitored for 10 months using an ELPI.
- The sample site was typical of urban centres in the UK.
- Temporal changes in PNCs were found; diurnal, weekday-weekend and seasonal.
- Vehicle volume/speed and meteorology affected UF/fine/coarse particles differently.
- Highest PNC during the morning rush hour (congestion/ low wind speeds).

1 Introduction

Despite decreases in particle emissions over the last fifty years, exposure to airborne particles is currently believed to cause a life length reduction of 6 months averaged across each UK citizen (COMEAP, 2010). Numerous potential health effects have been proposed to result from exposure to particles, including asthma, reduced lung function, and cardiovascular disease (e.g. Link et al., 2013). The investigation into the health effects from exposure to atmospheric particles has traditionally revolved around changes in the mass concentration, in line with legislative standards. Only a very small subset of the studies carried out have investigated the effect of changes in Particle Number Concentrations (PNCs), and results have been mixed. In one bicycle commuting study (Strak et al., 2012), participants were exposed to atmospheric particles for a five hour period. The authors found a consistent relationship between increased PNC and increased acute airway inflammation (measured as fractional exhaled nitric oxide; FE_{NO}). A study across five European cities (Lanki et al., 2006), found a weak association between PNC and acute myocardial infarction. In a multicentre European study (von Klot et al., 2005), cardiac readmissions were associated with same day PNCs (RR 1.026 [95 % confidence interval 1.005 to 1.048] per 10,000 particles cm^{3}). This suggests that even short periods of time spent in locations with high PNCs, e.g. during commuting, could have significant health impacts.

Traffic (through both exhaust and non-exhaust inputs) is a major contributor to PNCs near roads (Padró-Martínez et al., 2012; Quiros et al., 2013). There are complex relationships between vehicle inputs to PNC at the roadsides, and how these concentrations are transformed by factors including the street geometry and the meteorology (Kumar et al., 2008a). While the length of time spent in roadside locations may be relatively low (e.g. during the daily commute), the high number of particles people are exposed to during this time mean that there may still be significant health consequences. In addition, outdoor particles make a contribution to indoor particles, which people are exposed to for longer periods of time. By furthering our understanding of the short term changes in PNCs in different urban areas, we can begin to provide advice and guidance to the public to enable them to make informed choices about their exposure to particles.

There is currently a lack of detailed understanding of the influence of traffic volume and meteorology on particles in the atmosphere (Kumar et al., 2009). Previous studies investigating the response of PNCs to meteorological conditions and traffic variables have often investigated total PNC (Harrison and Jones, 2005), or have been in depth but short term (Pirjola et al., 2012). The aim of this study was to investigate the effect of traffic and meteorology on size-segregated PNC in a street canyon with a high temporal and particle size resolution. Our specific objectives were to collect PNC data in twelve size fractions between 0.007 and 10 μm (respirable particles) over the course of ten months. Meteorological and vehicle data were collected simultaneously, and the variables were investigated statistically. The unique strengths of this campaign were the enhanced particle size resolution and ten month sampling period. This meant that this investigation provided novel and statistically vigorous information on the effects of traffic and meteorology on size-segregated particles in the atmosphere.

2 Experimental

2.1 Sampling location

Sampling took place at Neath Road in Swansea, UK. Neath Road is a main commuting road into and out of Swansea, and traffic was expected to be a significant contributor to local PNCs. Neath Road is a secondary road which runs parallel to the railway line (Figure 1). The site is at the confluence of three valleys and the adjacent hills reduce lateral dissipation and encourage winter inversion layers. In 2001, the area surrounding the site was designated as an Air Quality Management Area (AQMA), deemed unlikely to meet air quality criteria for NO_2 (Wales Air Quality Regulations, 2000). The vehicle composition of the traffic using the street (derived from available data between March and July 2008) was 89.4 % cars, 7.2 % heavy vans, 1.5 % motorbikes, 1.3 % buses and 0.6 % articulated lorries.

Neath Road was NNE/SSW trending with a 30 mph speed limit at the site. The canyon was near-straight with a height/width ratio of between 0.6 and 0.7. The local area consisted of residential housing combined with pockets of commercial activity (Figure 2), and is typical of numerous other housing set ups across the UK. The sampling site was located 20 m from the nearest traffic intervention system; a set of traffic lights which modulated the flow. During peak periods traffic was observed to queue in front of the sampling site.

2.2 Sampling protocol

Sampling took place at the site between January 2008 and October 2008, using an ELPI (Electrical Low Pressure Impactor, Dekati™, Tampere, Finland); an inertial-based cascade impactor. The ELPI consists of three main elements; corona charger, low pressure cascade impactor and multi-channel electrometer, and has been described in detail elsewhere (Keskinen et al., 1992; Price et al., 2010). The sampling head, through which sampled air was drawn, was attached to the wall of a building 5.2 m above the road surface and 0.75 m protruding from the wall on the northern side of the canyon. The head removed particles > 10 μm which were too large to be size-fractionated using the ELPI. The ELPI was positioned inside the building, and the two were connected via 2 m of PTFE tubing (Adtech Polymer Engineering Ltd), i.e. the minimum practicable. No corrections were made for losses in the sampling tubes; particle losses will vary between size fractions, but will be most significant for the ultrafine size range (Kumar et al., 2008b). The ELPI was operated at a flow rate of 30 l/min. Cleaning was carried out on the ELPI regularly and flow rates were checked before and after sampling to ensure that the cut off diameters during sampling were accurate. Aluminium substrates (25 mm; Dekati™, Finland) were used for particle collection. Data in each of the twelve size fractions were collected every 2 s which was then averaged as required (15 minute averages unless otherwise stated).

2.3 Vehicle and meteorological data

Vehicle and meteorological data for the site were provided by the City and County Council of Swansea. Vehicle data included inbound/outbound vehicle counts and speeds to EUR 6 classification, and were measured using a General Packet Radio Service (GPRS) Automatic Traffic Counter (ATC), manufactured by Golden River (Bicester, UK). Temperature and relative humidity were measured using a Hygroclip (Rotronic, Crawley, UK). Rainfall was measured using a tipping bucket approximately 4 km from the particle sampling site. Wind speed and direction were measured by a Wind Sonic instrument (Gill, Lymington, UK) at 5 m and 10 m height above ground level directly above the particle sampling head. At 10 m (i.e. around 2 m above roof height, wind data were considered representative of the synoptic wind conditions, while the 5 m wind data were more reflective of within canyon conditions.

2.4 Statistical methodologies

The particle size fractions are given as the range of particle sizes which collect into that fraction in nm, preceded by the letter N, e.g. $N_{7\text{-}27}$ refers to particles with a lower cut off diameter of 7 nm. The upper limit of the stage, 27 nm, is defined by the lower cut off diameter of the stage above. The cut off diameters are D50 %; aerodynamic particle size with

50 % collection efficiency. All datasets were tested for their conformation to a normal distribution (SPSS, version 16). Spearman's rank correlation coefficient was used for identifying correlation between variables. A value of $p<0.05$ was used to assess significance unless stated otherwise in the text. The Mann-Whitney U test (two variables) or Kruskal-Wallis test (three or more variables) were used for the comparison of the means of datasets. A principal components analysis (PCA) was used to test for relationships between particles and vehicle/meteorological variables and was carried out on all available data across the 10 months of sampling. It offered a means of investigating whether a large group of variables was underlain by a smaller set of variables that could not be directly measured (Belis et al., 2013). The orthogonal transformation method was combined with Varimax rotation, and components with eigenvalues greater than unity were retained.

3 Results and discussion

3.1 Long-term averages

The mean total PNC during the ten month study period (January 2008 – October 2008) was 40,562 cm^{-3}, with a median value of 31,545 cm^{-3} (Table 1). A comparison of total PNC from this study with other UK-based studies shows fairly comparable results (Table 2), though results from Swansea were generally higher than found in other studies. This could be due to the relatively short tailpipe to sampling head distance (due to the sampling head being attached to the wall of a building and a narrow pavement), the wide range of particle sizes sampled in this study compared with the other studies, the different statistical values (e.g. mean, median) quoted in different studies and the averaging periods. In this study 93 % of counted particles were below 100nm, and consequently differences in the lower cut off diameter can significantly affect total PNC. We should also emphasise that relatively little work has been completed to compare instruments manufactured by different companies and utilising different collection and/or counting methodologies (e.g. Asbach et al., 2009). Therefore comparisons between studies which use different instruments must be approached with caution.

There was a clear diurnal pattern to the PNC data for particles below 1 μm during weekdays (Figure 3). Three main peaks identifiable in the ultrafine fraction (N_{7-27}) were at 09:00, 16:00, and 23:00, with the largest during the morning rush hour. Interestingly, this is most clear for Monday to Wednesday. The 09:00 and 16:00 peaks have been identified in previous studies as traffic signals (e.g. Harrison and Jones, 2005; Betha et al., 2013). The exact timing of the peaks vary by location, e.g. a morning peak has been identified between 06:00-07:00 (Mejía et al, 2007), and 08:00-09:00 (Betha et al., 2013), highlighting the importance of variability in traffic, meteorology, and more social factors specific to a geographical area, such as working patterns. The 16:00 peak is another commuting signal, though was generally smaller than the morning peak (discussed in section 3.4). The 23:00 peak may be related to traffic leaving the city following the closing of pubs and restaurants, or may be a domestic heating signal. Some literature currently suggests than number concentration is a more suitable indicator for exposure than mass and should be considered as a supplementary or replacement indicator in air quality legislation. In current legislation particle mass is averaged across 24 hours. This study has shown (Figure 3) that if the same procedure were adopted for PNC, it would fail to capture acute high exposure which is found at certain times of the day, e.g. morning rush hour, and therefore is unlikely to be representative of adverse health effects.

Weekday total PNCs were 23 % higher than weekend total PNCs; a finding which was statistically significant (Mann Whitney U test, $p>0.01$). This replicated similar findings in Brisbane, Australia (Morawska et al., 2002), Rome, Stockholm, Helsinki, Barcelona (Paatero et al., 2005), London (Jones et al., 2008) and Massachusetts, USA (Padró-Martínez et al., 2012). Total PNCs were generally lower during weekends, except for late evenings and early mornings, when leisure traffic was high. A bimodal pattern to PNC was identified on weekend days (Figure 3).

Winter had the highest mean $N_{7\text{-}93}$ at 46,615 cm^{-3} (Table 1), and summer had the lowest mean $N_{7\text{-}93}$ at 29,696 cm^{-3}. The Kruskal-Wallis test determined a statistically significant difference between mean $N_{7\text{-}93}$ in different seasons (H= 62.256 (3 d.f.), $p<0.01$). An increase in PNC during the winter has also been identified in a number of monitoring campaigns (Mejía et al., 2007; Wu et al., 2008; Padró-Martínez et al., 2012). This is due to factors including 1) Atmospheric mixing which encourages pollutant dispersion being generally less effective during the winter months due to lower temperatures (Harrison and Jones, 2005); 2) The lower temperatures during winter leading to greater supersaturation of exhaust gases, encouraging nucleation (Harrison and Jones, 2005); 3) Some natural sources, which may predominantly contribute to the mass measurements, such as sea spraying, have a strong seasonal signal; 4) Social factors will also influence PNCs; summer holidays will reduce commuting traffic and in the winter, colder temperatures will lead to increases in domestic heating (Mejía et al., 2007).

3.2 Principal Components Analysis (PCA)

A PCA was used to investigate the relationships between particle size fractions and vehicle/meteorological data. Data for this part of the study were averaged (15 minutes), based on the availability of the vehicle and meteorological data. Six principal components were identified with eigenvalues >1 (Table 3). Of these components, three explained relationships between particle variables and meteorological/traffic variables (PC1, PC2 and PC5), and three explained the relationships between the meteorological/traffic variables themselves (PC3, PC4, PC6), and are therefore not discussed (as identified in a previous study; Wang et al., 2010). A result of >0.75 in a PC was required by an individual size fraction in order for it to be associated with that PC. Any result > 0.1 for meteorological/traffic variables in a PC was considered of interest due to the large amount of analysed data and the complexities of the environment (including the relationships between different variables) in shaping and transforming PNCs. The 15 minute averaging period may be an important factor in determining the strength of any associations, and it may be that even more highly temporally resolved data could have improved the strength of observed relationships.

The first component (PC1) was associated with N_{7-262}, temperature (-), relative humidity (-), global radiation, wind speed (-), vehicle volume, and vehicle speed (-). These particles are associated both with variables that can lead to increases in PNC (e.g. an increase in vehicle numbers was associated with an increase in PNC), as well as removal processes (e.g. an increase in wind speed was associated with a decrease in PNC). The second component (PC2) was associated with fine particles (predominantly $N_{263-1609}$), wind speed (-), vertical wind (-), wind direction (-). These particles were associated with potential particle removal processes more greatly than the sub-micron particles, likely due to their larger size. PC5 was associated with coarse particles ($N_{1610-10,000}$), rainfall (-), vertical wind, and wind direction. Larger particles were associated with mechanical processes; both wind and rainfall are effective coarse particle removal mechanisms. These associations are discussed further in specific sections below.

3.3 Effect of meteorology

3.3.1 Rainfall

During periods of rainfall (>0.1 mm in 15 minutes; n = 7886), the mean total PNC was 32,000 cm^{-3}, which increased to 42,200 cm^{-3} during non-rainfall periods (<0.1 mm in 15 minutes; n = 479), and the lower mean total PNC during rainfall was found to be statistically significant (Independent t test; t= 7.742, p = 0.05; SPSS version 20). The effect of rainfall on PNC was investigated as a function of particle size (Figure 4a). The largest difference in PNC between rain event days (REDs) with >1 mm rainfall and non-rain event days (NREDs) with no rainfall for the preceding 5 days was found for the $N_{384-1609}$. The lowest PNCs for the different size fractions were found on the day of, and the day following, a rain event (Figure 4b). Generally for all size fractions the PNC increased between two and five days following the rain event, and increased to a maximum over five days after a rain event (five days was the limit of the observed period in this study). These findings replicated those in Figure 4a, with fine particle size fractions the most affected by rainfall. It should be noted that due to low sample sizes and the large number of factors which affect the overall PNC, the spread of data which contributed to the plotted mean in Figure 4b was large. More long term data collection would be required to assimilate enough information to reduce the variance of data. This reduction in PNCs in response to rain suggests a wash-out effect. The rainfall removed particles, reducing the overall PNCs and having the effect of “cleaning” the atmosphere. This effect has previously been identified during a measurement campaign in Göteborg, Sweden (Janhäll et al., 2004), and Raipur city, India (Deshmukh et al., 2013). In these studies, as in the Swansea study, particles >1µm were more significantly affected by rain.

3.3.2 Wind

Results from the PCA indicated that for ultrafine and fine particles, as wind speed increased, PNC decreased. Increased wind speed brings more air parcels to a site which (depending on where those air parcels originate from) will dilute the particles in a street canyon. This effect was not shown statistically (using the PCA) for the coarse size fraction which could be related to the increased scavenging potential of higher winds from road and land surfaces.

Data were grouped into three size fractions representing an ultrafine fraction (93nm and smaller), a fine fraction ($N_{94-2399}$) and a coarse fraction ($N_{2400-10,000}$) to provide more generalised results on the effect of wind on PNC. Mean PNCs were compared from different wind directions at 10m, measured approximately 2m above roof level (Figure 5). Time points with wind speeds below 1 m/s were not included in the analysis (based on previous work, e.g. Cyrys et al., 2008), as at low wind speeds vehicle turbulence becomes more important than ambient wind conditions in modulating PNC. The effect of wind direction upon PNCs was significant and varied by size fraction; for the ultrafine and fine particles the highest number concentrations were from the north and south, near-parallel to the street canyon, while for coarse particles PNCs were highest from the west (perpendicular to the street canyon). The importance of wind direction in modulating PNCs is likely due to 1) the spatial distribution of local sources around the sampling site, and 2) above rooftop wind direction as discussed below.

The highest concentrations of ultrafine and fine particles were observed near-parallel to the street. The main source for these particles in the local area is traffic exhaust, and therefore winds parallel to the street will funnel particles directly towards the sampling equipment. The increase in coarse PNC with winds from the west (i.e. perpendicular to the canyon) may be due to local sources; e.g. “dirtier air” from the west in comparison to cleaner air being brought from the south (from the sea). Another important factor may be the recirculation of the air in street canyons and the positioning of the sampling head on the northern side of the canyon (windward side during westerly winds). Some previous studies have found a build-up of PNC from winds perpendicular to the street (e.g. Dos Santos-Juusela et al., 2013) due to the recirculation of pollutants during these conditions. The height/width ratio of the Neath Road canyon (h/w = 0.6 – 0.7) suggests a wind flow pattern between wake interference flow (h/w = 0.5) and skimming flow (h/w = 1) as described by Vardoulakis et al. (2003). In wake interference flow, winds crossing the canyon perpendicular to the street axis are affected by crossing the rooftops on the windward edge of the canyon and do not have time to recover before reaching the buildings on the leeward edge, while during skimming flow a single vortex is established within the canyon. The simple 2D models are much more complex in the 3D real world situation where, for example, a corkscrew-type wind flow may be set up under flow conditions at an angle that is not quite perpendicular to the canyon and breaks in the buildings can further complicate canyon flow patterns. This suggests that there may be the development of a single vortex (or a recirculation pattern which incorporates this) in the canyon under certain conditions (e.g. perpendicular flow), though during more parallel wind conditions, the in-canyon flow will be even more complicated. In addition, at low wind speeds (e.g. below 1.0 - 1.5 m/s; Cyrys et al., 2008; Kumar et al., 2009), turbulence generated by vehicles within the canyon will be more important than the wind direction in changing the PNC. We measured PNC only on the northern side of the canyon, and therefore we were unable to confirm or deny the presence of a vortex system during sampling, however a comparison of the 5m wind direction and vertical wind speeds (supplementary material; Figure S2) indicated links between easterly winds/ negative vertical wind speeds and westerly winds/ positive vertical wind speeds. This is consistent with the formation of a vortex within the canyon under perpendicular wind flow.

Ultrafine and fine PNCs showed similar concentrations of particles from winds perpendicular to the canyon (i.e. from the east and west). This is in contrast to the coarse particles where a clear difference was noted. Traffic exhaust will be the dominant source of ultrafine/fine particles in a street canyon, while for more coarse particles, though traffic will contribute (e.g. through tyre and brake wear), other sources will also become more important, e.g. mineral dust, biological particles and marine particles. This may explain the differential effect of wind direction on ultrafine/fine and coarse PNC. The low concentration of coarse particles may also be a factor. An additional point of interest is that due to these recirculating winds, PNCs may vary considerably from one side of the street to the other, depending on the wind direction. Therefore when we found very high PNCs, the PNC on the other side of the canyon may have been much lower and when we measured low PNCs, the concentration on the other side of the canyon may have been much higher. The effect of this could be limited by measuring on both sides of the canyon simultaneously in future studies to provide data which is representative of the entire canyon cross section.

3.3.3 Temperature

Temperature varied diurnally, peaking at around midday (see supplementary material; Figure S4a). Results from the PCA suggested that temperature was negatively correlated to PNC for the nanoparticle size range, however this was less clear visually, and was probably a product of the similar diurnal behaviour for the two variables (at least to an extent). Lower temperatures have previously been linked with increases in PNCs (Dos Santos-Juusela et al., 2013). Colder temperatures encourage new particle formation in the atmosphere and decrease the supersaturation ratio which causes an increase in nuclei mode particles (Jamriska et al., 2008). This can occur at the exhaust pipe, where hot exhaust air meets cooler ambient air. Other studies have been unable to identify a clear relationship between the variables (Wu et al., 2008). In this study it is concluded that temperature and PNC were linked (as further exemplified by the seasonal differences observed), but due to the confounding nature of the variables and the complexity of the system the exact influence cannot be quantified.

3.3.4 Relative Humidity (RH)

RH varied diurnally with a minimum at around midday when temperature was highest (see supplementary material; Figure S4b). Peak PNCs coincided with RH lows in this study. It is unclear whether this was causal or an artefact produced from the daily cycling of RH and PNC. It has been suggested that an increase in RH reduces total PNC through condensation (de Hartog et al., 2005). However, RH is likely to have numerous effects on particle processes, both those that increase PNCs, e.g. nucleation, and those that reduce PNCs, e.g. condensation. The relationship between RH and PNC is not straightforward, and not fully quantitatively understood. Some studies have found little or no relationship between PNC and RH (Wu et al., 2008).

3.3.5 Global radiation (GR)

From past work, there is a tentative association between days with peaks in GR and photochemical PNC highs in the UK (Agus et al., 2007), despite lower average global radiation than more southerly parts of Europe. The photochemical peak falls outside of the typical traffic peaks. There have been various methods used to define "photochemical event days" (PEDs) and "photochemical non-event days" in previous work, e.g. Ziemba et al. (2006), PEDs were defined as days with PNC higher than the median PNC averaged for the entire dataset plus twice the standard deviation for two hours or more. In this study, we followed a similar methodology to that set out in Park et al. (2008). We compared PNCs at or around (± 2 hours) midday to the average PNC for that day. If the PNC was twice the 24 hr average PNC or more, the day was termed a PED. The main interest in this study was a comparison between the other measured meteorological and traffic variables on days more likely to be PEDs and those less likely to be PEDs. A low/medium correlation (r_s=0.293; $p<0.01$) was found between N_{7-27} and GR during spring/summer months (whole GR dataset). When considering only the summer months, this increased to 0.388 ($p<0.01$). Traffic variables were similar for both days termed photochemical and non-photochemical days (Table 4). RH was almost 10 % lower on days with photochemical events as determined by our methodology, and the wind speed was higher. GR was significantly higher on days with evidence of a photochemical event. This resulted in a significantly higher total PNC on days with photochemical events. High levels of GR have previously been linked to special nucleation events (Cheung et al., 2012; Wang et al., 2010), and these are poorly understood (Jamriska et al., 2008). This study has shown that a combination of high GR, higher wind speed and lower relative humidity (i.e. "cleaner" atmospheres) were factors which probably increased the likelihood of a photochemical event, though the low numbers of sampled days meeting the criteria means that no definitive conclusions can be made. Given the latitude of the sampling site, the contribution of photochemical event particles to total PNC is likely to be lower in this study than in other parts of the world with higher GR.

3.4 Effect of traffic

Traffic is an extremely important contributor to PNC at urban sites (e.g. Joodatnia et al., 2013; Padró-Martínez et al., 2012; Quiros et al., 2013). On average there were 652 vehicles per hour passing the site, corresponding to a total of 16,000 per day. This vehicle loading is more typical of heavy flow routes in smaller cities rather than main traffic routes in larger cities like London (e.g. Marylebone Road). The vehicle composition during March 2008 – July 2008 (i.e. period of vehicle data availability) was 89.4 % cars, 7.2 % heavy vans, 1.5 % motorbikes, 1.3 % buses and 0.6 % articulated lorries. The vehicular traffic using the street was therefore dominated by light duty vehicles (LDVs). Traffic volumes were 22 % higher on weekdays than weekends (Figure 6a), which was very similar to the 23 % higher weekday PNCs. This was likely a consequence of the working patterns of the majority of the users of the road – car drivers commuting in and out of the city of Swansea.

An increase in total PNC was found with increasing vehicle numbers (Figure 6b). This was reinforced by the PCA, where the vehicle count was positively associated with PNC in PC1 ($N_{7\text{-}262}$; Table 3). Low to medium positive correlation was found between vehicle numbers and ultrafine PNCs ($N_{7\text{-}27}$ $r_s = 0.395$, $N_{28\text{-}55}$ $r_s = 0.324$, and for $N_{56\text{-}93}$ $r_s = 0.353$; $p<0.01$, n=>5700 per analysis, 15 minute data averaging); the majority of PM emitted by the vehicle exhaust is below 100 nm (Hagler et al., 2009). For a site at which the dominant source is traffic, this correlation may be considered low, however some previous studies have been unable to identify a strong relationship between vehicle counts and PNCs (e.g. Kumar et al., 2008a). The products of vehicle exhaust particles, whether formed within the combustion chamber or on leaving the exhaust may be transformed by environmental conditions, and therefore are not solely a result of the traffic count. No clear inbound/outbound effect of traffic was identified, with $r_s = 0.377$ and $r_s = 0.386$ (n= 5701) between total PNCs and inbound/outbound traffic counts, respectively. Here, inbound refers to vehicles heading into the city (i.e. south and on the opposite side of the canyon to the sampling head) and outbound refers to vehicles leaving the city (i.e. north and on the side of the canyon with the sampling head). This suggests effective mixing within the street canyon, a consequence of vehicle-related turbulence and any air recirculation derived from rooftop winds (Wang and Zhang, 2012).

On approximately 63 % of sampled weekdays the afternoon traffic count peak was larger than the morning peak and the $N_{7\text{-}27}$ morning peak was larger than the afternoon PNC peak. This was also observed at other sites (e.g. in Helsinki; Dos Santos-Juusela et al., 2013). This effect is often attributed to improved atmospheric mixing in the afternoon. During the morning PNC peak on days when this was observed, it was generally found that temperatures had only recently begun to rise and relative humidity was beginning to reduce. In addition, wind speeds were just beginning to increase but were still low. These meteorological factors would contribute to stable atmospheric conditions with a lower mixing layer. This discouraged dispersion, thereby contributing to higher PNCs. In contrast, the atmospheric situation in the afternoon was generally different on these days. Wind speeds at the site peaked at around midday, coinciding with the GR and temperature highs, and relative humidity lows. The increase in temperature between morning and afternoon (as was generally observed) promoted air parcel movement, increasing turbulence, and the increased wind speeds encouraged pollutant dispersion.

A rise in vehicle speed was associated with a decrease in total PNCs (Figure 6c). Generally, the PNC output from a vehicle increases with rising vehicle speed (Dijkema et al., 2008). However, slower vehicle speeds, such as during the morning and afternoon rush hours were associated with stop-and-go traffic. These bursts of traffic activity may produce bursts of pollution as the engine works hard to move off from a stationary position (Agus et al., 2007), thereby increasing total emissions (Boogaard et al., 2010). This would be compounded during high volume traffic. The traffic lights located 20m from the site would increase stop-and-go traffic conditions during busy periods, including at the sampling site. Additionally, the efficiency of catalytic converters is temporarily reduced during periods of acceleration (Fruin et al., 2008). Traffic speed is linked to traffic volume; each roadway network will have a capacity. When this is exceeded, e.g. during rush hours, traffic will build up and be forced to queue. As traffic volume increased, traffic speed decreased (R^2=0.86). Vehicle speed and vehicle numbers are highly intercorrelated, and their exact relative contributions are difficult to assess, however the PCA has indicated that the traffic variables were more influential than individual meteorological variables for N_{7-262}.

4. Conclusions

This study has shown that during times when vehicle volumes are high and vehicles are exhibiting stop-start behaviour, if this is combined with low wind speeds, ultrafine PNC will be highest. These conditions were often identified during the morning rush hour. Human exposure to particles was greatest at this time through a combination of highest PNCs and the large group of commuters using the road at these times. Neath Road is a main commuting road into and out of Swansea (16,000 vehicles per day, 90 % cars), and therefore vehicles using the road were a significant contributor to the PNCs that the local commuting population was exposed to.

Improving our knowledge of the complex interactions which occur in urban environments between particles, meteorology and traffic is vital for air quality assessment and city planning. Legislation can be viewed as a 'top-down' approach to air quality management, while a 'bottom-up' approach could include empowering the local population with the knowledge to make their own choices regarding particle exposure. This study has characterised a microenvironment in the UK in terms of PNCs, and is a necessary step towards being able to provide clear advice to the public on how to take control to reduce their exposure.

Acknowledgements

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Supplementary information

A file of supplementary information is provided with this manuscript. In Figure S1, the average wind conditions in the street canyon are shown, in Figure S2 the relationship between vertical wind speed and wind direction in the canyon are exemplified, in Figure S3 the relationship between traffic volume and traffic speed is highlighted and in Figure S4 the relationships between PNC and temperature/RH are explored.

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Figures

Figure 1 Topographical map of the area surrounding the site (site indicated by dot at 25m above sea level). Blue star indicates highest point, 192m above sea level. Arrows indicate valleys which can funnel pollutants to the site. © Crown copyright 2009. An Ordnance Survey/EDINA supplied service.

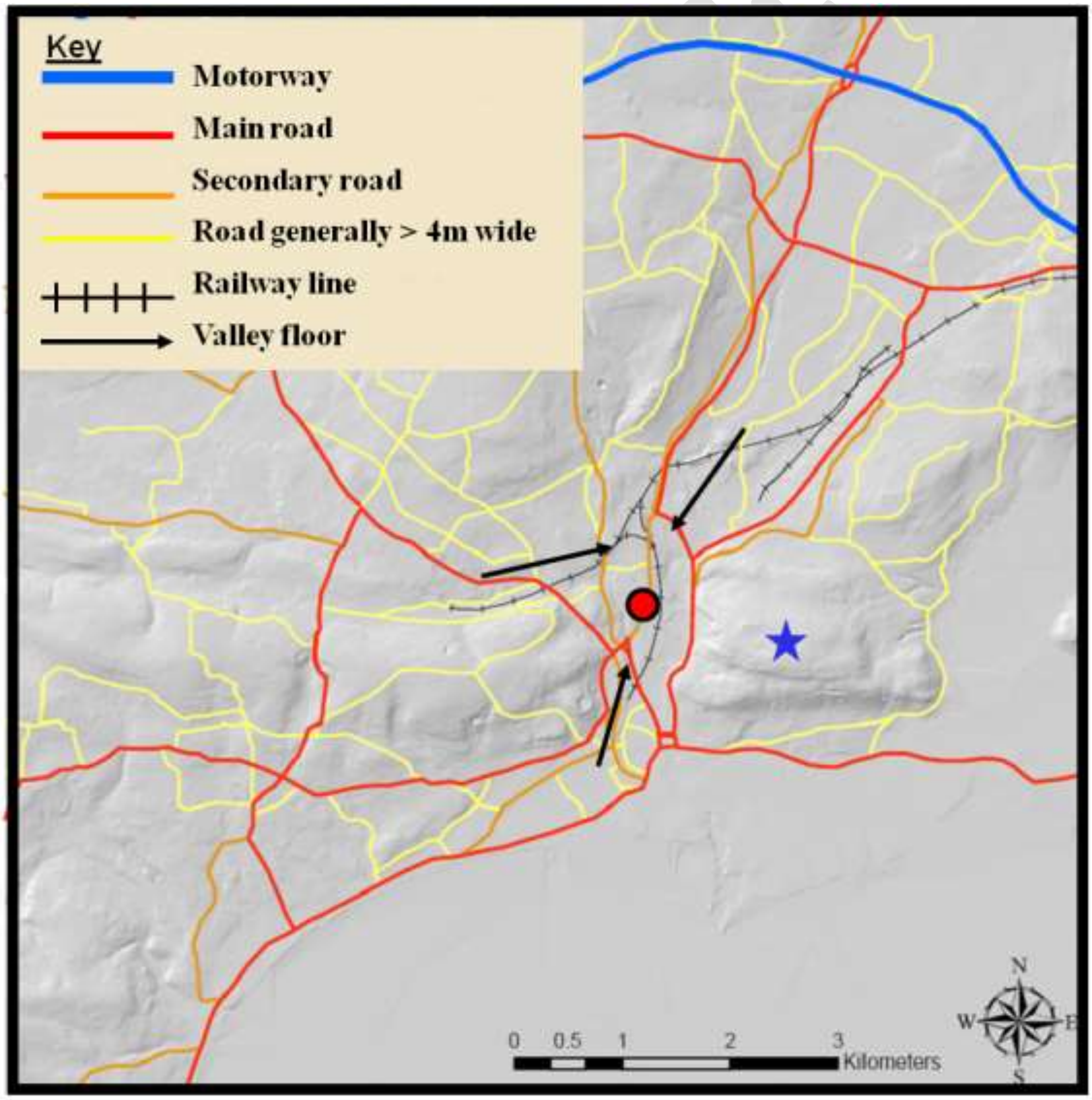


Figure 2 The Neath Road street canyon; a) Photograph looking NNE past the site with sampling head on left hand side (not visible – on photograph edge), b) Local map indicating vehicle flow directions, and c) Schematic showing canyon dimensions (not to scale).

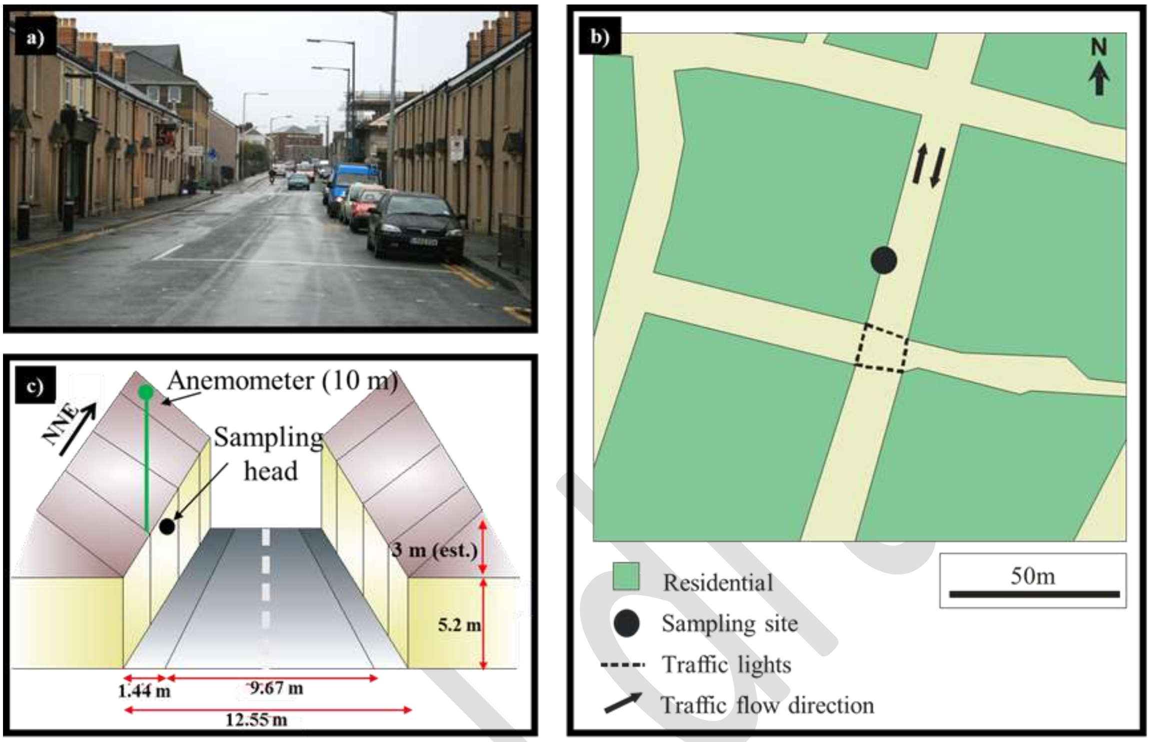


Figure 3 The diurnal pattern of PNCs for each ELPI measured size fraction from Monday-Sunday (24 h mean). The y axis is on a log scale. Calculated from 10 months of semi-continuous particle data (Jan – Oct 2008). Arrows highlight the three peaks in weekday PNC identified in section 3.1.

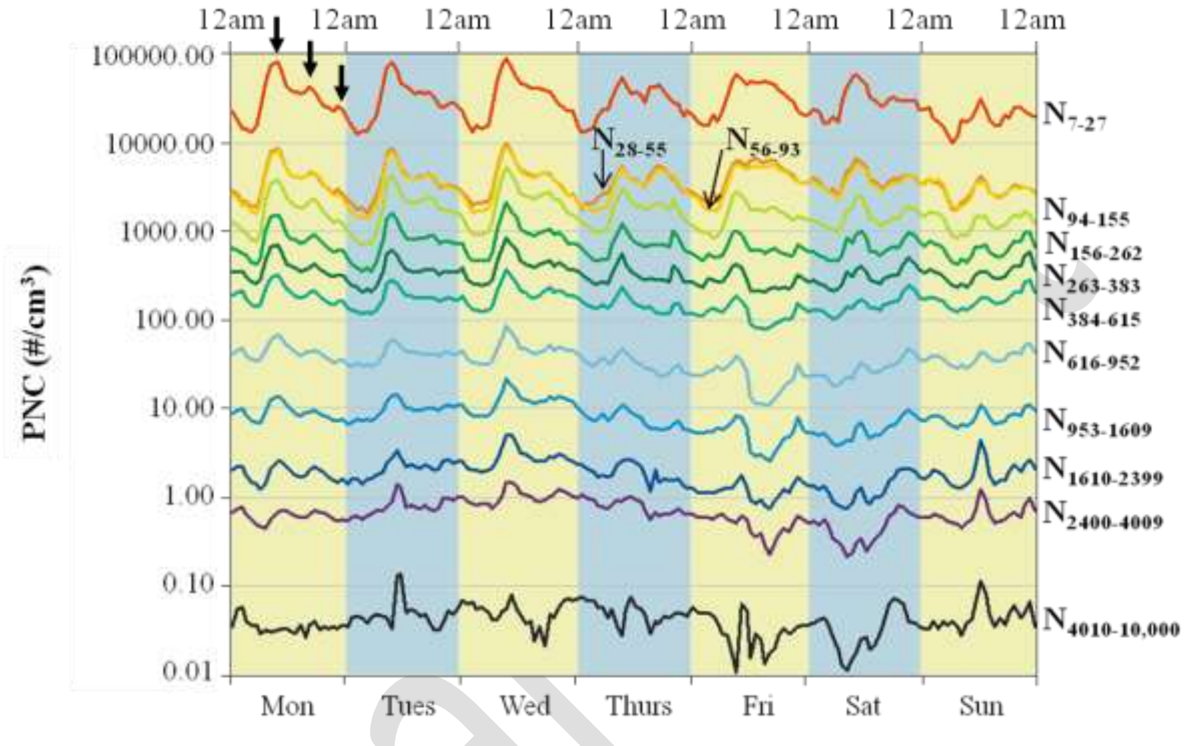


Figure 4 The relationship between PNC and rainfall at the sampling site; a) the change in effect of rainfall by particle size. The graph shows the ratio of rain event days (RED; days >1 mm rainfall)/non-rain event days (NRED; over 5 days without a rain event). Data plotted for the D50 % cut off size for the stage.; b) Comparison of number size distribution of street canyon particles on RED (n= 37) and days following REDs; 1) the day following a RED (R.E.D +1d; n= 9), 2) between 2-5 days following a rain event (R.E.D +2d to 5d; n= 24) and 3) >5 days following a rain event (R.E.D +5d and over; n= 15). Data taken January-June 2008 (based on the availability of rainfall data). Data plotted for the D50 % cut off size for the stage with the two most coarse particle fractions ($N_{2400-4009}$ and $N_{4010-10,000}$) shown in # m^3 (due to low PNCs per cubic centimetre) and all smaller size fractions in # cm^3.

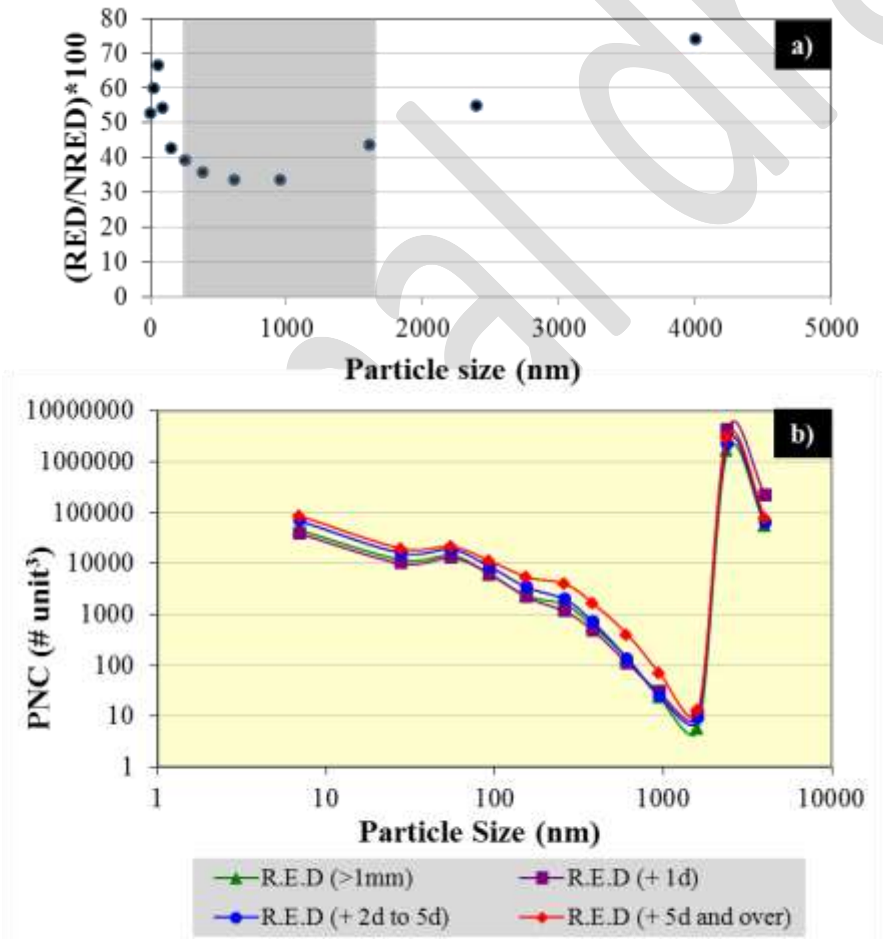


Figure 5 Mean PNC (cm^{-3}) from 10m WDs subdivided by size fraction; a) particles <100nm, b) fine particles (100 nm – 2.5 µm), c) coarse particles (2.5 µm – 10 µm), d) total particles. PNCs during wind speeds below 1 m/s were not included in the analysis. Sampling site plotted as red dot in the centre of each plot, with canyon orientation indicated by red line.

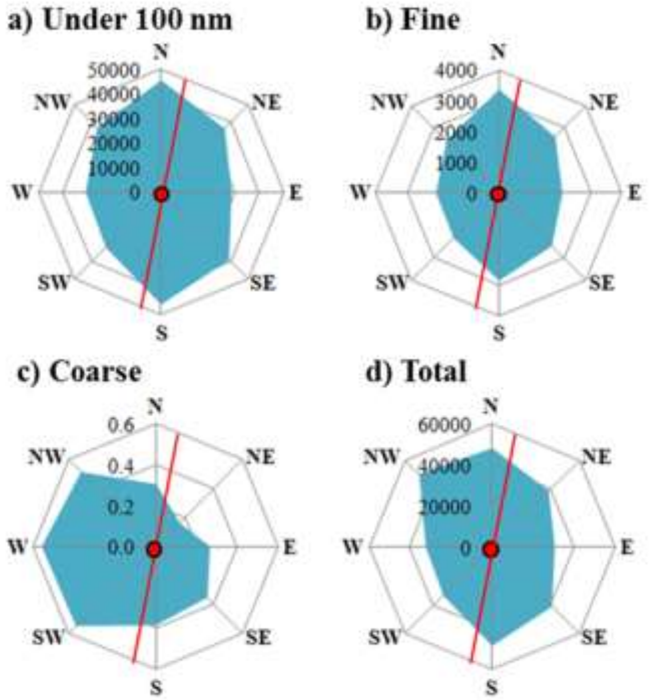
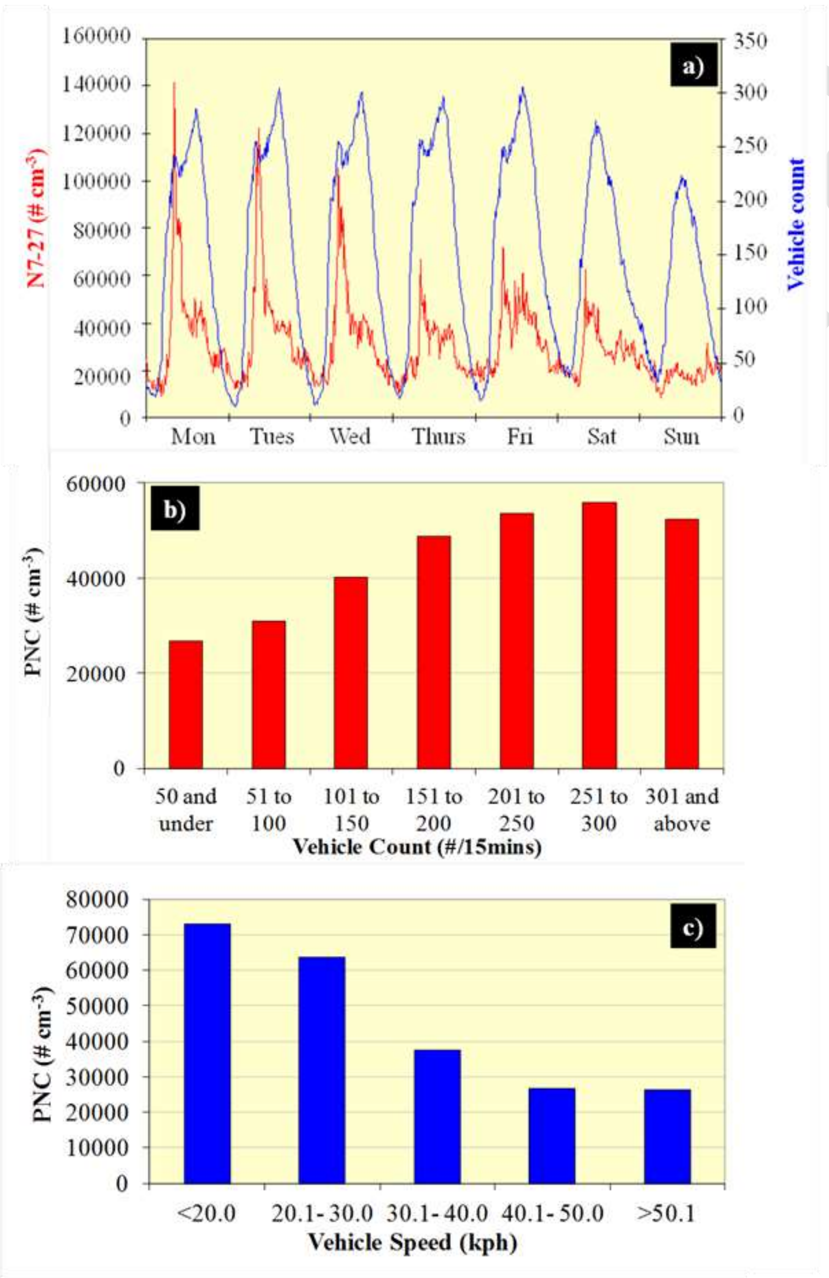


Figure 6 The effect of traffic on PNC; a) Plot comparing N_{7-27} with vehicle number counts over a week. Vehicle counts averaged from 26 weeks of 15 minute data and PNC values from 20 weeks of 15 minute data; spring/summer 2008; b) Change in mean Total PNC with binned 15 minute vehicle counts. Averaged from 7 months of semi-continuous data during 2008 (February-August); c) Averaged Total PNC for five classifications of vehicle speed in km/h; below 20 km/h (n=21), 20.1 to 30.0 km/h (n=2773), 30.1 to 40.0 km/h (n=3329), 40.1 to 50.0 km/h (n=2818), over 50.1 km/h (n=121), spring/summer 2008.



Tables

Table 1 Descriptive statistics of the PNC data at the street canyon. Data averaged from 15 minute PNC data (10,122 data points per size fraction); January 2008-October 2008. Note coarse particle data given as # m^3 due to low numbers.

Size fraction	Total PNC (# cm^{-3})	N_{7-93} (# cm^{-3})	$N_{94-4009}$ (# cm^{-3})	$N_{4010-10,000}$ (# m^3)
Median PNC	31,545	29,025	2520	358,260
Mean PNC	40,562	37,648	2913	608,447
Standard deviation	37,043	34,996	2642	894,784
Min.	724	564	60	13,700
Max.	314,730	305,050	15,770	2,2436,000
Mean Spring PNC		41,212	2696	488,566
Mean Summer PNC		29,696	2287	357,394
Mean Autumn PNC		36,643	3320	757,660
Mean Winter PNC		46,615	4322	1,234,428

Table 2 Comparison of the results from this study with other UK-based studies. The PNC is assumed to be a mean unless stated otherwise.

Reference	**Location**	**Type of site**	**Season**	**Instrument**	**Size fraction (µm)**	**PNC (# cm^{-3})**
This study	Swansea, UK	Roadside	Jan 2008 – Oct 2008	Dekati ELPI	0.007 - 10	31,545 (median)
Reche et al., 2011	Marylebone Road, UK	Roadside	Feb 2009 – Dec 2009	CPC TSI model 3022A	0.007 - 1	22,156
	North Kensington, London, UK	Urban background	Jan 2009 – Dec 2009 (no data for March)	CPC TSI model 3022A	0.007 - 1	12,134
Von Bismarck-Osten et al., 2013	Marylebone Road, UK	Roadside	Jan 2008 – Dec 2010	SMPS TSI 3936 spectrometer (DMA model 3081 with CPC model 3775)	19.2-800	19,620 (median)
	North Kensington, London, UK	Residential	Jan 2008 – Dec 2010	SMPS TSI 3936 spectrometer (DMA model 3081 with CPC model 3775)	19.2-800	5,663 (median)
	Harwell, Oxford, UK	Rural	Jan 2008 – Dec 2010	SMPS TSI 3936 spectrometer (DMA model 3081 with CPC model 3775)	19.2-800	2,857 (median)
Sánchez Jiménez et al., 2012	Hope Street, Glasgow	Roadside	June 2006 – Aug 2006	CPC (TSI Model 3785)	0.005 - 1	23,564 (median)
	St Enoch Square, Glasgow	Urban background	June 2006 – Aug 2006	CPC (TSI Model 3022A)	0.007 - 1	12,851 (median)
	Montrose St, Glasgow	Street canyon (sampling at 12m)	Jun 2006 – Aug 2006	CPC (TSI Model 3785)	0.005 - 1	11,095 (median)
	Marylebone Road, UK	Roadside	June 2006 – Dec 2006 (26 days)	CPC (TSI Model 3022A)	0.007 - 1	109,953 (median)
	North Kensington, London, UK	Urban background	June 2006 – Dec 2006 (26 days)	CPC (TSI Model 3022A)	0.007 - 1	23,407 (median)
Godri et al., 2011	School in North London	Roadside	Mar 6 2008 – Mar 13 2008; Mar 13 2008 – Mar 19 2008	CPC (TSI Model 3022A)	0.007 - 1	13,822; 15,495
	School in East London (a)	Roadside	Nov14 2007 – Nov 21 2007; Nov 21 2007 – Nov 28 2007	CPC (TSI Model 3022A)	0.007 - 1	46,004; 30,549
	School in East London (b)	Roadside	Nov 28 2007 – Dec 5 2007; Dec 5 2007 – Dec 12 2007	CPC (TSI Model 3022A)	0.007 - 1	9,779; 17,859
Lingard et al., 2006	Leeds, UK	Urban roadside (rush hour conditions)	July 2003	TSI Model 3936 L25 SMPS	0.006 - 1	18,000 – 34,000

Table 3 Correlation matrix of a PCA investigating associations between PNC and meteorological and vehicle variables (15 minute average). The shaded components quantify the association between vehicle/meteorological variables and PNC, while the unshaded components quantify the associations between the vehicle/ meteorological variables only.

Variable	**Component**					
	1	**2**	**3**	**4**	**5**	**6**
N_{7-27}	.892		.176		-.126	
N_{28-55}	.934		.134		-.159	
N_{56-93}	.932	.115	.161		-.129	-.102
N_{94-155}	.913	.263	.133			-.101
$N_{156-262}$	.794	.495				
$N_{263-383}$	.526	.770				
$N_{384-615}$	.306	.912		-.154		
$N_{616-952}$		.955		-.117		
$N_{953-1609}$		.894			.271	
$N_{1610-2399}$		.440		.185	.832	
$N_{2400-4009}$	-.143	.189		.191	.911	
$N_{4010-10,000}$	-.124	-.108			.850	
Temperature	-.119		.577	.388		
Relative Humidity	-.156		-.766	.332		
Global Radiation	.154		.826			.107
Rainfall			-.314	.373	-.201	.368
Wind Speed (5m)	-.106		.163	-.110		.917
Wind Speed (10m)	-.100	-.164	.391			.802
Vertical Wind (5m)		-.103	.156	.917	.164	
Wind Direction (5m)			.112	.899	.163	.104
Wind Direction (10m)		-.177	.200	.676	.225	-.328
Vehicle Count	.232		.802	.220		.142
Mean Speed	-.264		-.781	-.231		-.137
% Variance	20.2%	36.7%	51.2%	63.2%	74.3%	82.3%

Table 4 Comparison of the main meteorological and traffic parameters (mean) for the period of June/early July 2008, subdivided by whether evidence of a photochemical event was noted. Data averaged over 24 hour period. For days when photochemical events were noted, n= 7, for days without evidence of photochemical events, n= 19.

	Evidence of a photochemical event	**No evidence of a photochemical event**
Total PNC (cm^{-3})	23547	17846
Rainfall (mm)	0.1	0.1
GR (W/m)	194.3	146.9
Wind Speed (m/s; 10m)	3.2	2.9
WD (degrees; 10m)	209.9	243.8
Temperature (ºC)	14.8	15.4
Relative Humidity (%)	72.6	81.3
Traffic Volume (count/15mins)	165.8	164.6
Mean Vehicle Speed (km/h)	35.0	35.5