Exposure to ultrafine particles from traffic in city streets and the urban atmosphere

Physics Department, University of Manchester Institute of Science and Technology (UMIST), Manchester, UK

Abstract
Mass-based emission controls (such as Euro – I to IV) are successfully reducing emissions of fine particles from road vehicles, but may have actually increased the emission of ultrafine particles and their persistence in the atmosphere just at a time when growing evidence indicates that these ultrafine particles present the greatest threat to health upon inhalation. These particles have so little mass that they barely register in measurements made by the current urban air quality monitoring network, which measures the mass of particulate matter ($\text{PM}_{10}$). Ultrafine particles also have different dynamic and transport properties than larger particles, leading to more variability in concentrations that may rise and fall independently of $\text{PM}_{10}$. Current daily and hourly monitoring of $\text{PM}_{10}$ for the purposes of Air Quality Management fails to represent the wide variation and episodicity in exposure of an urban population to the threat from traffic-sourced ultrafine particles.

In order to quantify and interpret this variability, data from experiments employing sophisticated, high-resolution instrumentation in UK cities will be presented. This data will illustrate the variability on time scales from minutes to hours of ultrafine particle exposure on busy streets, and will indicate how meteorological factors and urban topography determine the exposure to traffic particle emissions in the surrounding urban environment. Such information has important consequences for the assessment of future emission reduction and air quality improvement strategies, especially localised strategies.

Keywords: ultrafine particles, urban background, street canyon
1 Introduction

The regulation of urban air quality with regard to particles is currently achieved using the PM$_{10}$ metric. Automatic monitors have been established in most European cities providing hourly measurements of the mass concentration of particles with an aerodynamic diameter smaller than 10 µm. Emission and dispersion models also deal with particles using the PM$_{10}$ metric, treating it as a single, passive pollutant. However, this measure has many shortcomings because it covers a huge range of substances with different sources, different composition, chemical and physical properties, and different size and shape distributions. Ambient particles of different sizes have a range of dynamical and transport properties and different effects when inhaled. In particular, particles smaller than about 2.5 µm, and especially ultrafine particles (UFP), will penetrate much more deeply into the lung and from there to the circulation, and are increasingly seen as responsible for the principal adverse effects of particles upon health.

In urban air ultrafine particles are most numerous in the tens of nm size range. Because of their minute size, these ultrafine particles contribute very little to particle mass, as measured by PM$_{10}$, but they are far more numerous than larger particles. Being a mass metric, PM$_{10}$ is dominated by the larger particles to which particles advected from the regional background strongly contribute. The result is that the spatial variability of PM$_{10}$ concentrations across an urban area is low and exceedences of the PM$_{10}$ air quality objective are often attributed to increases in the background component, i.e. to processes beyond the city. However, the use of the PM$_{10}$ metric obscures the much stronger local-scale variation in UFP concentrations. Much less data is available on ultrafine particles in urban areas compared to PM$_{10}$, and their concentrations are not currently routinely included in emission or dispersion modelling due to a lack of characterisation. As will be shown, ultrafine particles in urban areas are strongly linked to traffic emission and thus high levels of exposure are determined not by regional background levels but by close proximity to traffic. Short-term exposure, as may be experienced in busy city streets, has been shown to rapidly lead to dramatic physiological effects [1,2,3].

It is against this background that this paper seeks to investigate the nature and variation, spatial and temporal, of ultrafine particle concentrations in the urban street-level atmosphere.

2 Sources and nature of urban ultrafine particles

In most European urban areas, ultrafine particles are emitted principally by traffic. Upon emission, these particles are rapidly mixed and diluted with the ambient air. During daytime periods, coinciding with high traffic flow, one may observe greatly increased concentrations of particles below 300 nm, but especially between 10 nm and about 60 nm.
Measurements of particle composition have been made in a nearby street canyon in Manchester using an Aerosol Mass Spectrometer (AMS, Aerodyne Inc, Billerica, USA, instrument and analysis techniques described by Allan et al., 2003[4]). Particles below 200 nm were mostly associated with locally emitted semi-volatile organic compounds from vehicles [5]. In addition to the direct traffic source, ultrafine particles may form from direct gas to particle conversion.

3 Ultrafine particles in urban centres

Upon emission from vehicle exhausts (or formation in the exhaust plume) ultrafine particles are rapidly diluted and dispersed so that, apart from at locations close to busy roads, a concentration is reached which is fairly even across a city centre, and which diminishes towards the outer edge of the city. Most of the time it is generally assumed that most people are exposed to this ‘urban background’ concentration and, to date, most measurement studies have concentrated on characterising this background level [6,7,8]. A seasonal influence has been noted in several studies, with higher concentrations in winter. This is usually related to the higher rate of particle nucleation from gaseous precursors in vehicle exhaust when temperatures are lower. Weekly variations are regularly observed with higher concentrations on weekdays than at weekends or public holidays, due to the weekly variation in emission, i.e. traffic volume.

It is consistently found that urban background concentrations of ultrafine particles exhibit a diurnal cycle, very similar to the cycle in traffic flow within the city, but moderated by meteorological influences. Daily maxima, which are typically less than two times the daily mean, usually occur during the morning traffic peak. Between the morning and evening traffic peak a daytime concentration exists which is typically 1 to 1.5 times the daily mean.

The Sources And Sinks of Urban Aerosol (SASUA) campaign in Edinburgh in 1999 - 2001 included direct measurements of particle number fluxes made at 70 m above street level in the city centre [9]. It was seen that the morning concentration peak was largely caused by the suppression of vertical turbulent ventilation of the urban canopy layer by the persistence of the stable nocturnal boundary layer. As the morning progressed solar heating and urban heat emission broke down this stable layer, leading to a progressive increase in the vertical flux of particles from the city.

These measurements were conducted in both May and October. In May relatively lower concentrations were measured. Although there was an increase in sensible heat flux above the city, the particle flux was reduced compared to October, indicating a reduced emission. This was presumed to be due to both reduced space heating and an observed reduction in traffic flow. In May, sunrise in Edinburgh is almost four hours earlier than in October, and with stronger morning solar heating we can assume that the nocturnal boundary layer was broken down earlier and faster, leading to a more effective ventilation of morning traffic emissions. This led to a much reduced morning concentration...
peak, as also observed in Leipzig [7] and in Helsinki [10]. Conversely, deeper into winter, and especially at higher latitudes than Edinburgh, we may expect that weak solar heating and late sunrises will prolong the morning concentration peak, leading to increased daily mean concentrations, as has been observed in Helsinki [10].

The UK Automatic Urban Network of air quality monitors was supplemented in 2000 by Condensation Particle Counters (CPCs) at 6 sites, including Manchester Piccadilly. This is classed as an Urban Centre monitor and is located in an open space with a bus station on one side.

Total particle number concentrations ($N$) are available from Manchester Piccadilly in 15-minute averages from 5/4/2000 to 31/8/2000 (although data is missing from the 7th to the 23rd of August). During this period the expected reduction in concentrations on weekends was apparent. The mean of all Tuesday–Friday concentrations was $24\ 500\ \text{cm}^{-3}$. The daily means, however, fell from approximately $30\ 000\ \text{cm}^{-3}$ at the start of April to $20\ 000\ \text{cm}^{-3}$ by the end of July. These daily values mask a considerable variability in the data. Inspection of the time series reveals that numerous spikes, in which concentrations may temporarily increase up to ten-fold, supplement a relatively simple and predictable diurnal cycle.

The spikes were separated from the underlying pattern. Between the hours of 10:00 and 17:00 local time, the mean concentration (of the de-spiked data) was $25\ 300\ \text{cm}^{-3}$ (compared to $32\ 600\ \text{cm}^{-3}$ for the raw data) although this value fell over the period of May to July (Table 1). Therefore, on average, the spikes contributed an extra $7\ 300\ \text{cm}^{-3}$ during this daytime period. The seasonal variation was confirmed by an inverse relationship with temperature (from Manchester Airport). However, a weak, but independent positive relationship was also found with wind speed.

<table>
<thead>
<tr>
<th>month</th>
<th>$N$ (cm$^{-3}$)</th>
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<tbody>
<tr>
<td>April (from 5th)</td>
<td>28 600</td>
</tr>
<tr>
<td>May</td>
<td>29 900</td>
</tr>
<tr>
<td>June</td>
<td>25 000</td>
</tr>
<tr>
<td>July</td>
<td>20 400</td>
</tr>
<tr>
<td>August (only 10 days of data)</td>
<td>20 400</td>
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When the ‘spikes’ were considered separately, a clear dependence on wind direction was seen. One third of all spikes corresponded to southerly winds, i.e. winds blowing directly from the bus station (fig. 1). A further quarter was related to an ENE sector, when the monitor is downwind of the NE side of Piccadilly.
Gardens, which is the next nearest source of traffic (fig. 1). Thus it is suggested that most of the spikes are due to exhaust plumes originating in the local neighbourhood (tens to hundreds of metres from the monitor). The effect of the spikes is illustrated in fig. 2 where the time series of $N$ is shown for one day with northerly winds and one with southerly winds, which is dominated by the influence of the bus station.

The net result of adding the spikes to the estimated background concentrations was a small increase in the daytime mean, but significant increases in the upper percentiles. The spikes were evenly distributed in time during the daytime period. The magnitude of the spikes could not be simply related to any available parameters.

4 Ultrafine particles in street canyons with traffic

The SCAR (Street Canyon Aerosol Research) [11,12] project involved the measurement of ultrafine particle concentrations at 4 m height at the mid-length of a street canyon in Manchester (UK) with busy traffic. Particle measurements were supplemented by measurements of wind and turbulence at several locations within the canyon using ultrasonic anemometers. Measurements were made continuously from Monday to Friday over two weeks (15th - 26th October 2001).

Urban centre background number concentrations $N_{0.1}$ ($4.6 \text{ nm} < D_p < 0.1 \mu\text{m}$) were estimated by assuming a constant dilution ratio between $N_{0.1}$ and NO$_x$, giving values in the range 4 000 – 9 000 cm$^{-3}$.

![Figure 3. Mean number size distributions (SMPS) according to in-canyon flow regime.](image-url)
The mean and median concentrations measured at SCAR were 26 500 cm\(^{-3}\) and 15 600 cm\(^{-3}\), respectively. However, the variation in concentration was much more complex than at Piccadilly. The instrument inlet was a minimum of 5 m from vehicle exhausts and this proximity to traffic meant that traffic plumes dominated the measurements. Thus, wind direction was the dominating factor determining measured concentrations. The anemometric data from SCAR indicated that airflow at street level was mostly channelled along the canyon’s axis. However, in this study the prevailing wind direction in Manchester led to perpendicular flow. 55% of the time this led to a lee vortex such that the instruments were downwind of the road and were largely sampling freshly emitted plumes in an early stage of dilution, leading to greatly increased concentrations. Comparing the number size distributions for these two flow conditions (fig. 3) illustrates that in this cross-canyon flow the higher concentrations were due to increases in ultrafine particles related to the characteristic size range of vehicle emissions.

The frequency distributions of \(N_{0.1}\) concentration were roughly log-normal for each flow regime, although almost an order of magnitude larger in cross-canyon flow (fig. 4). Whereas concentrations within the canyon were typically 4 or more times greater than that in the urban-centre background, during periods of cross-canyon flow mean concentrations increased two and a half times over the mean, leading to values almost 12 times the background. Considering all street canyon data, the upper percentiles were greatly increased compared to the background, with, for instance, the 99\(^{th}\) percentile in the canyon being ten times that in the background (fig. 5).
For the SCAR study, a reference wind speed \( U_{\text{ref}} \) and direction were available from the roof of the UMIST building, 750 m from the SCAR site. It was found that \( N_{0.1} \) was inversely related to \( U_{\text{ref}} \) during parallel flow only. In perpendicular flow there was no clear relationship. \( N_{0.1} \) was inversely related to the wind speed measured at street level, but the effect of the canyon in perpendicular flow was to partially isolate the flow within the canyon from that above it, thus making it impossible to relate dispersion of particles in the canyon to ordinarily available meteorological parameters. Additionally, there was some limited evidence of the action of traffic in increasing turbulent mixing and breaking up any stable vortex flow within the canyon.

## 5 Conclusions

Long-term exposure of citizens to ultrafine particles (UFP) is dominated by a well-mixed urban background concentration. However, this exposure is supplemented by short-term peaks in traffic-influenced locations on the order of 100 m or less from busy roads. Thus a person’s total exposure is strongly influenced by the duration of proximity to traffic in inner-urban neighbourhoods. Whereas PM\(_{10}\) is strongly influenced by regional advection, the regional influence on UFP is exerted by regional variations in the thermal and wind climate affecting the ventilation rate of the urban canopy, and by the relative seasonal variation in thermal and emission diurnal cycles introduced by variation in latitude.

In trafficked street canyons, where source-receptor distances are of the order of metres to tens of metres, the short peaks dominate exposure. Measurements have shown that dispersion in street canyons may be weaker than predicted by models due to flow isolation. Direct exposure to vehicle exhaust plumes at pavement
sites may be typically of short duration but can make a large contribution to an individual’s daily dose. Exposure is especially significant for those spending longer periods in the street, whether for occupational or for leisure (e.g. street cafes) activities. Modelling of PM$_{10}$ currently does not capture the sharp gradients and short-term variability observed in ultrafine particle concentrations. Consequently the modelled effect of a road closure or emission reductions on PM$_{10}$ concentrations within a trafficked street canyon would be minor, whereas the studies discussed indicate that major reductions in exposure to UFP could be achieved.

References


